Collective Light-matter Interactions via Emergent Order in Cold Atoms

by

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Dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Department of Physics in the Graduate School of Duke University

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ABSTRACT

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Collective behavior in many-body systems, where the dynamics of an individual element depend on the state of the entire ensemble, plays an important role in both basic science research and applied technologies. Over the last twenty years, studies of such effects in cold atomic vapors have lead to breakthroughs in areas such as quantum information science and atomic and condensed matter physics. Nevertheless, in order to generate photon-mediated atom-atom coupling strengths that are large enough to produce collective behavior, these studies employ techniques that intrinsically limit their applicability. In this thesis, I describe a novel nonlinear optical process that enables me to overcome these limitations and realize a new regime of collective light-matter interaction.

My experiment involves an anisotropic cloud of cold rubidium atoms illuminated by a pair of counterpropagating optical (pump) fields propagating at an angle to the trap's long axis. When the pump beam intensities exceed a threshold value, a collective instability occurs in which new beams of light are generated spontaneously and counterpropagate along the trap's long axis. In order to understand the physical mechanism responsible for this behavior, I study first the system's nonlinear optical response when driven below the instability threshold. I find that the incident optical fields produce an optical lattice that causes the atoms to become spatially organized on the sub-wavelength length scale. This organization corresponds to the formation of an atomic density grating, which effectively couples the involved fields to one another and enables the transfer of energy between them. The loading of atoms into this grating is enhanced by my choice of field polarizations, which simultaneously results in cooling of the atoms from $T \sim 30 \, \mu K$ to $T \sim 3 \, \mu K$ via the Sisyphus effect. As a result, I observe a fifth-order nonlinear susceptibility $\chi^{(5)} = 1.9 \times 10^{-12} \, (m/V)^4$. 
that is 7 orders of magnitude larger than previously observed. In addition, because of the unique scaling of the resulting nonlinear response with material parameters, the magnitude of the nonlinearity can be large for small pump intensities (i.e., below the resonant electronic saturation intensity 1.6 mW/cm$^2$) while simultaneously suffering little linear absorption. I confirm my interpretation of the nonlinearity by developing a theoretical model that agrees quantitatively with my experimental observations with no free parameters.

The collective instability therefore corresponds to the situation where the cold vapor transitions spontaneously from a spatially-homogeneous state to an ordered one. This emergent organization leads to the simultaneous emission of new optical fields in a process that one can interpret either in terms of mirrorless parametric self-oscillation or superradiance. By mapping out the phase diagram for this transition, I find that the instability can occur for pump intensities as low as 1 mW/cm$^2$, which is approximately 50 times smaller than previous observations of similar phenomena. The intensity of the emitted light can be up to 20% of the pump beam intensity and depends superlinearly on the number of atoms, which is a clear signature of collective behavior. In addition, the generated light demonstrates temporal correlations between the counterpropagating modes of up to 0.987 and is nearly coherent over several hundred $\mu$s. The most significant attributes of the light, though, are that it consists of multiple transverse spatial modes and persists in steady-state. This result represents the first observation of such dynamics, which have been shown theoretically to lead to a rich array of new phenomena and possible applications.
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Chapter 1

Introduction

Collective behavior, where the dynamics of an individual element depend on the state of an entire multi-element system, is ubiquitous and can lead to new and often unexpected phenomena. Examples arise in almost every discipline of science, from the study of neural nets and mob mentality to ferromagnetism, lasing, and the stock market. The basic requirement for the emergence of collective behavior is a mechanism by which the constituent elements interact. While there is always some degree of interaction between elements in any physical system, the interaction must be sufficiently strong to give rise to a qualitative change in the system’s behavior.

One prominent example of collective phenomena is Dicke superradiance (or coherent spontaneous emission), where couplings between emitters modifies the spontaneous emission rate of the ensemble. Dicke superradiance involves a collection of $N$ identical atoms located within a volume of less than an optical wavelength, where some of the atoms are initially in their excited state but no macroscopic dipole moment exists. As the atoms decay randomly via spontaneous emission, the radiated electromagnetic fields create a quantum interference between neighboring atoms’ probability amplitudes for the emission of a photon. This modifies the spontaneous emission characteristics of the sample and causes the ensemble to decay more rapidly than the spontaneous decay time for a lone atom and leads to the emission of an intense burst of directed, coherent radiation. Thus, the collective inter-atomic interactions cause the ensemble to synchronize and behave like a single ‘superatom’ with properties quite different from its constituents. Dicke superradiance has been studied extensively both
experimentally and theoretically due to its conceptual simplicity and potential applications, such as enabling trace detection of impurities in the atmosphere [2], providing a new source of coherent radiation for optical metrology [3], and aiding in quantum information processing [4-6].

More recently, advances in atomic cooling and trapping have enabled the discovery of a new type of superradiance, referred to as superradiant Rayleigh scattering (SRyS) [7], which has lead to a resurgence of interest in the field. In contrast to Dicke superradiance, which is based on synchronization of the internal atomic states, SRyS is based on the collective center-of-mass motion (i.e., external state dynamics) of an atomic ensemble. In the simplest conception of SRyS, one considers an ensemble of initially-stationary atoms illuminated by an optical field. As the atoms scatter photons, their momenta change. In an ultracold atomic vapor, matter-wave interference between the moving and stationary atoms produces an atomic density grating that modifies how the vapor scatters light. As in the case of Dicke superradiance, one observes the emission of an intense pulse of light.

Superradiant Rayleigh scattering belongs to a broader class of collective phenomena in which an atomic system undergoes a spontaneous transition from a spatially uniformly-distributed state to an ordered one when it is illuminated by an optical field. This emergent, dynamical organization is distinct from previous works involving static, externally-imposed atomic order [8-10], and can lead to reduced optical instability thresholds [11] and new phenomena [12-15]. In these processes, thermal atomic motion washes out the density grating and causes decoherence. Thus, previous observations of such phenomena in free space required ultracold temperatures ($T < 3 \mu K$) and employed optical fields detuned far from atomic resonance in order to avoid recoil-induced heating [7, 16]. Collective scattering in these systems is inherently transient, though, because the recoil associated with repeated scattering events eventually
causes a Doppler shift such that the atoms no longer interact with the incident light. When one places the atoms in an optical cavity with a single transverse mode, the cavity enhances the duration of the light-matter interaction, which effectively increases the coherence time and enables the self-organized state to occur at temperatures of up to several hundred µK \[^{17}\]. Because the atoms in this case have a spread of momenta, the issue related to the Doppler shift mentioned is largely avoided. By including an auxiliary cooling mechanism, recent studies have demonstrated steady-state behavior \[^{18, 19}\]. Nevertheless, single-mode cavities are more technically challenging to work with than free-space systems, impose additional constraints on the allowed optical frequencies, and are incompatible with multi-mode fields, which are necessary for realizing recently-proposed spin glass systems \[^{15, 20, 21}\] and multi-mode quantum information processing schemes \[^{22–24}\].

My contribution to the field of collective light-matter interactions involves the experimental realization of a novel approach to this problem that allows me to circumvent the aforementioned issues and study the superradiant transition using cold, thermal atoms \((T \sim 30 \, \mu K)\) in the absence of an optical cavity. I observe steady-state superradiance in which new, multimode optical fields arise via the emergence of global spatial atomic organization in the form of an atomic density grating. The process shows a distinct threshold, below which the vapor remains unorganized and I observe no superradiant light. To confirm the role of atomic organization, I study the system below the superradiant transition and, in doing so, discover a new nonlinear optical (NLO) process that leads to large light-matter interaction strengths and demonstrates a unique dependence on material parameters.

In this first chapter, I discuss in further detail the advantages and disadvantages associated with using cold atoms as a platform for studying collective behavior. In addition, I review previous studies on the topic of collective light-matter interactions.
in cold atom systems in order to provide additional background and motivation for my work. Finally, I briefly present the main results presented in this thesis.

1.1 Why cold atoms?

Atomic vapors represent an almost ideal system for studying quantitatively collective effects. One can easily experimentally realize ensembles with a large and controllable number of identical atoms that can be well-isolated from external environmental perturbations. Also, the dynamics of individual atoms and their interactions with one another are generally well-understood, which enables precise control at the quantum level and allows one to test quantitatively theoretical predictions made via first-principles. In addition, one can exploit a variety of tunable atom-atom interaction mechanisms, such as atomic collisions and radiation-based forces. While control of the atomic collision cross section has played an important role in atomic physics, the interactions are inherently short-ranged. Instead, I will focus on photon-mediated atom-atom interactions, which enable atomic couplings over arbitrarily large spatial extents.

In order for the atoms to interact strongly with one another via the exchange of photons, they must first couple strongly to the optical field. One approach for realizing large coupling strengths, known as resonant enhancement, involves tuning the frequency of the incident optical fields close to an atomic resonance \[25\]. One very simple experimental method for employing resonant enhancement involves shining near-resonant laser beams on a warm atomic vapor cell. This approach has enabled the observation of collective instabilities based on NLO processes in which the atomic ensemble spontaneously synchronizes and scatters light coherently \[26, 27\]. Unfortunately, resonant enhancement is often accompanied by large linear absorption, which
corresponds to spurious scattering that negatively affects the intended NLO process and reduces the desired atom-atom interaction strength. Doppler broadening due to atomic motion in a warm vapor exacerbates this problem by increasing the spectral region over which appreciable absorption occurs from several MHz (i.e., the natural linewidth $\Gamma$) to several hundred MHz. One must therefore typically choose moderate detunings from resonance to balance the deleterious effects of absorption with the enhancement of the NLO coupling strength.

One straightforward technique for mitigating the problem of absorption involves using cold atoms, where one can easily achieve typical atomic temperatures $T \lesssim 1 \text{ mK}$. In this case, Doppler broadening is negligible and one can work close to resonance (i.e., within several natural linewidths) without facing strong absorption. Cold vapors also typically have vastly reduced collisional rates compared to warm vapors, which increase the available interaction and coherence times. Although these advantages suggest that cold atomic systems should vastly outperform warm vapor cells when one studies identical processes, this is generally not found to be true in experiments. Instead, experiments show only minor differences between warm and cold vapors to date. There are two main reasons for this comparable performance between the two systems.

First, scientists have developed a number of techniques for circumventing the problematic aspects of warm vapors. For example, several methods exist for avoiding unwanted absorption. Placing atoms in an optical cavity modifies the atoms’ absorption and emission characteristics and can effectively reduce the negative effects of absorption [28], although this comes with its own technical challenges and limitations. Instead, one can choose a Doppler-free beam geometry such that the relative Doppler shifts of the photons involved in the NLO process cancel out. A third, and by far the most widely-used, technique involves the coherent preparation of the atomic ensem-
ble, such as in electromagnetically induced transparency (EIT), which make use of quantum interference effects to considerably reduce absorption while simultaneously enhancing the NLO coupling strength \[29\]. Methods also exist for avoiding the problems of short coherence times due to collisions in a warm vapor, such as simply using a buffer gas and using an anti-relaxation coating \[30\] (such as parraffin) in the vapor cell.

While clever techniques have improved the competitiveness of warm atomic systems, cold atomic vapors present their own challenges. Because radiation pressure due to incident laser beams can destroy the cold atomic sample by pushing atoms out of the interaction volume, one must either choose a beam geometry that balances radiation pressure forces or accept operation limited to the transient regime. Also, it is typically difficult to create a cold atomic sample with the same number of total atoms available for interaction with the optical fields as in a vapor cell. This is partially due to the fact that one can easily make vapor cells tens of centimeters long while most cold atomic samples are between a few hundred microns and a few centimeters in length. In addition, the maximum density in a cold sample is typically several orders of magnitude smaller than in a warm vapor. To make matters worse, even if one produces large atomic densities, it can be self-defeating since the reabsorption of spontaneously-emitted photons inside the cold cloud (known as rescattering) can either overwhelm the particular interaction one wants to study or destroy the sample. While one can reduce the negative impact of these effects by using dark state techniques \[31\], transient compression \[32, 33\], or spatially-deformed cold atom geometries \[34–36\] (such as the anistropic, pencil-shaped sample that I use in this thesis), the final results for equivalent experiments in warm and cold atoms are not dramatically different from one another.

It is natural, then, to wonder why anyone would choose to work with cold atoms
in the first place. The answer is simple: one can manipulate the center-of-mass motion of cold atoms, which is impossible in warm atomic systems. Thus, rather than trying to simply recreate experiments in a cold atom system that were performed originally with warm atoms, one should exploit the inherent, unique characteristics associated with cold atoms. As I discuss in the next section, this tactic results in a wealth of novel phenomena with a range of potential applications.

### 1.2 Collective light-matter interactions in cold atoms via atomic recoil

Although it may sound tautological, one of the most significant attributes of cold atoms is the fact that they move slowly (i.e., have small momenta). In many cases, the photonic recoil associated with optical scattering is comparable to the average atomic momentum in a cold vapor. Therefore, light-matter interactions in cold atomic systems necessarily play an important role in the dynamics of both the internal and external atomic states and lead to a rich array of collective phenomena. Here I give a brief review of the relevant literature in this area.

The study of recoil-based collective effects in cold atoms began in the early 1990's, very soon after the experimental realization of efficient cooling and trapping techniques. In an effort to achieve large, dense traps (as discussed in Sec. 1.1), Walker et al. [160] observed that the spatial distribution of cold atoms in their magneto-optical trap (MOT; see App. A) changed dramatically beyond a critical atom number. For example, their spherical MOT transformed into a stable ring of atoms surrounding a central atomic core. They attributed this effect to a long-range, repulsive radiation pressure force caused by photon rescattering between trapped atoms. Additional re-
lated effects, such as vortical radiation pressure and radial oscillations of the MOT, have also been observed in dense MOTs. Multiple-scattering can also lead to modifications to the fundamental radiation pressure force, the localization of light, and random lasing, depending on the degree of disorder in the atomic ensemble.

Beyond these phenomena based on multiple scattering, new NLO processes arise due to collective atomic motion. One example of such an effect is collective atomic recoil lasing (CARL), which was proposed by Bonifacio et al. in 1994. In CARL, one considers a coherent optical field, which we refer to as a pump field, that is detuned far from resonance and incident on a collection of spatially uniformly-distributed two-level atoms in free space. A counterpropagating probe field arises spontaneously from fluctuations, which can take the form of either spontaneously-emitted light or atomic density variations that backscatter the pump field. The weak probe field interferes with the pump light and creates a periodic dipole potential that modifies the atomic center-of-mass motion and results in atomic bunching (i.e., the formation of an atomic density grating) with a spatial period on the order of the optical wavelength. The pump light then backscatters off of this density grating into the probe mode, resulting in a positive feedback loop that amplifies both the intensity of the probe beam and the grating contrast. The evolution of the system from a disorganized to self-organized state corresponds to a synchronization transition analogous to the Kuramoto model, and is reminiscent of both inversionless lasers and the free electron laser (FEL). The process is inherently transient, though, as the intensity imbalance of the pump and probe beams results in a net acceleration of the atoms that eventually shifts the atoms out of resonance with the optical fields due to the Doppler shift.

While studies in warm vapors purported to have observed CARL, Brown et al. showed that these results could be interpreted without invoking atomic recoil.
Kruse et al. performed the first unambiguous demonstration of CARL in 2003 using a cloud of cold atoms \((T = 100’s \text{ of } \mu \text{K})\) placed within a high-Q optical ring cavity (finesse \(F \sim 100,000\)). They pump the atoms along the cavity axis using an intense optical field far-detuned from the atomic resonance and measure the generated, counterpropagating field emitted from the cavity (see Fig. 1.1a). In order to distinguish this result from the original, free-space formulation of CARL, I refer to this approach as cavity CARL (CCARL). Using cold atoms greatly increases the fraction of atoms that bunch in the self-generated optical potential compared to the case of warm atoms. In addition, the presence of the cavity increases the effective coherence time of the system because the photons make many round-trips between the cavity mirrors before ceasing to interact with the atoms. This enhances the back-action of the cavity field on the atoms and enables the density grating to form before thermal atomic motion washes it out. In agreement with predictions, the light is emitted transiently and displays a time-dependent frequency due to the atomic acceleration. By including an optical molasses (see App. A.2) to provide a damping force, the system reaches a steady-state in which it continuously emits light. The inclusion of a cooling mechanism \((i.e., \text{dissipation channel})\) to realize steady-state operation represents an important modification of the original CARL concept and is crucial to the results that I present in this thesis.

In a later experiment conducted by the same group, Slama et al. [43] study the case in which the finesse of the optical cavity is low \((i.e., \text{the ‘bad cavity’ regime with } F \sim 5000)\). This situation more closely resembles the original spirit of CARL, where cavity feedback is explicitly not included. Due to the reduced lifetime of the photons in the cavity, the coherence time of the system decreases, thermal diffusion overwhelms the buildup of the density grating, and CCARL does not occur for atomic temperatures of a few hundred \(\mu\text{K}\). Alternatively, one can understand the reduced cavity finesse in terms of an increase in the amount of loss of the system. Thus, because one requires that the
Figure 1.1: Previous studies of collective interactions. a) Schematic for CCARL \[17\], where cold atoms in a traveling-wave cavity driven by a pump field become bunched and scatter light into the cavity mode. b) SRyS \[7\] geometry in which an ultracold vapor in free space illuminated by single pump field simultaneously gives rise to recoiling atoms and new beams of light. c1) The experimental scheme and c2) possible resulting atomic organization structures for the configuration used by Baumann et al. \[14\]. d1) The experimental scheme and c2) possible resulting atomic organization structures for the multimode configuration proposed by Gopalakrishnan et al. \[42\]. All figures come from the respective cited papers.
gain exceed the loss in order for the initial fluctuations that seed CARL to experience a net amplification, one must correspondingly increase the single-pass gain. By using much colder atoms (e.g. \( T < 40 \mu K \)), thermal atomic motion is greatly suppressed and one obtains longer atomic coherence times and larger gain. The gain becomes so large, in fact, that the system enters the superradiant regime and emits intense bursts of coherent light. I refer to this process as cavity-enhanced SRyS (CSRyS). While CCARL and CSRyS rely on the same gain mechanism, the physical distinction between them stems from whether the cavity optical field or the atomic sample stores the coherence. Alternatively, one can discuss the difference between these two processes in terms of whether the single-pass gain is small (CCARL, where one requires many passes) or large (CSRyS, where only a few passes are sufficient).

For sufficiently cold temperatures \( (T < 3 \mu K) \), superradiance occurs in the absence of a cavity. Inouye et al. [7] first demonstrated SRyS by shining a single, far-detuned laser beam on an anisotropic, pencil-shaped Bose-Einstein condensate (BEC, see Fig. 1.1b). Later, Yoshikawa et al. demonstrated similar results using an ultracold, thermal vapor [16]. In these experiments, a fraction of the atoms undergo Rayleigh scattering, which causes them to recoil. The interference of the stationary and recoiling atoms gives rise to a spatial modulation of the atomic density distribution. This density grating leads to stimulated Rayleigh scattering that occurs preferentially along the atomic sample's long axis (i.e., the direction of largest gain).

In 2002, Domokos et al. [13] introduced a twist to CCARL and CSRyS that lead to the prediction of new phenomena. Rather than using a single laser to pump the atoms along the axis of a ring cavity, Domokos considered the case where a pair of counterpropagating laser beams illuminate atoms along the direction perpendicular to the axis of a standing-wave cavity with a single transverse mode (see Figs. 1.1c1 and c2). Light scattered from the atoms into the cavity mode creates a standing wave field
that combines with the orthogonal pump fields to create a total optical field that varies spatially in two dimensions. This spatially-varying field is commonly referred to as an optical lattice, and exerts a force (known as the dipole force) on the atoms that affects their spatial distribution.

As the cavity field grows, the atoms transition from an initially spatially homogeneous state to a self-organized one. Depending on the phase of the cavity field, the atoms can assume one of two possible checkerboard patterns that maximize scattering into the cavity. The system’s selection of which pattern to occupy corresponds to the breaking of discrete translational symmetry and can be directly experimentally measured by recording the phase of the light emitted from the cavity \[19\]. In addition, this technique gives rise to a new mechanism for cooling atoms due to the dissipation channel introduced by irreversible photon loss from the cavity. In contrast to standard cooling techniques based on spontaneous emission and depend on a particular atomic level structure, this method works on a broad range of atomic species and avoids the problem of rescattering that can limit the cooling efficiency \[44\].

The original experimental verifications of the mechanism proposed by Domokos et al. \[13\] were carried out with a thermal atomic cloud \[19, 44\]. These studies relied on inferring details of the atomic behavior from the light exiting the cavity because the contrast of the generated density grating in the vapor was too low to image directly. A recent study by Baumann et al. \[14\] circumvented this problem by using a BEC, which allowed them to simultaneously monitor the evolution of the atomic and optical fields. Indeed, they found that emission of superradiant light coincided with the emergence of atomic organization. In addition, for the case of a BEC, one can show that the Hamiltonian describing this system maps onto the Dicke Hamiltonian \[14, 45, 46\], which allows one to experimentally investigate the role of discrete symmetry breaking in a superradiant quantum phase transition as well as study the boundary between the
unorganized and organized (superradiant) states.

Despite the success of this approach, Gopalakrishnan et al. [47] pointed out that the observable phenomena in cavities containing only a single transverse mode are inherently limited because the cavity geometry predefines the structure of the generated lattice. In contrast, multimode cavities allow the collective light-matter interaction to determine the shape and location of the resulting spatial organization through the spontaneous breaking of continuous translational symmetry (see Figs. 1.1d1 and d2). This opens the door to a variety of new phenomena, such as the emergence of dislocations, frustration, supersolidity, and quantum spin glass phases [15, 20, 47]. This cross-over between ultracold atomic physics and condensed matter physics represents a very promising research direction; nevertheless, these collective scattering phenomena are also intimately connected to well-known effects in the field of nonlinear optics that play an important role in photonic technologies.

While most of the previously-mentioned studies focus on the atomic dynamics rather than the emitted light, Inouye et al. sought to highlight the equivalent role of the optical and matter fields in SRyS [7]. In particular, they discuss the gain mechanism underlying SRyS in terms of a four-wave mixing process in which the standing matter wave (i.e. density grating) and involved optical fields are mutually amplified. Similarly, the Dicke-like Hamiltonians used by Baumann et al. [14] and Nagy et al. [21] explicitly include four-wave mixing terms in their description of superradiance. In this context, one can view SRyS as a wave mixing instability, where gain from a NLO process combines with a feedback mechanism to generate new, macroscopic optical fields from initial fluctuations. Such processes have been studied in the NLO community for over forty years [48–50]. In the parlance of the nonlinear optics community, such an instability is known as parametric self-oscillation (PSO). Parametric self-oscillation is very similar to oscillation in a standard laser except that stimulated emission gain in
a laser is replaced by a parametric NLO wave mixing process in PSO. Also, as in the case of a laser, PSO requires a feedback mechanism, which can be provided either externally (e.g., via a cavity) or internally (e.g., via distributed feedback). When no cavity is present, one typically refers to the process as mirrorless PSO (MPSO).

In a 2008 paper, Saffman et al. studied the situation where counterpropagating, near-resonant beams are incident on a cold atomic vapor. For sufficiently strong pump intensities and atomic densities, it is well-known that this configuration can give rise to MPSO in warm atoms \[26,27,49\] through the scattering of the pump beams off spontaneously-created internal state gratings (e.g., population, polarization, or spin gratings). In addition to creating internal state gratings, the involved optical fields also give rise to mechanical forces that can produce density gratings in cold atoms. For proper choice of the pump beam parameters, Saffman et al. predicted theoretically that the emergent density gratings lower the MPSO instability threshold compared to the warm atom case. This result is important because it implies the possibility of more efficient devices based on MPSO for use in all-optical networks \[27\]. In addition, MPSO via four-wave mixing has been shown to generate quantum states of light \[51\], which are important for quantum information processing applications \[52\].

In general, this connection between superradiance and MPSO is extremely important and has been typically overlooked by the atomic and optical physics communities. I attempt to rectify this by considering the collective scattering mechanism reported in this thesis both in terms of superradiance and a wave mixing instability, and compare my results to those of SRyS and MPSO when appropriate. To this end, I typically use the words 'superradiance', 'wave mixing instability,' and 'MPSO' in this thesis.
1.3 Overview of thesis

This thesis describes my investigation of collective light-matter interactions in a cloud of cold atoms. In it, I study experimentally and theoretically a new nonlinear optical wave mixing process in which atomic bunching and cooling act in concert to produce an extremely large, higher-order nonlinear optical response at low-light-levels. The nonlinearity can become so strong that it coherently amplifies noise in the system (i.e., density or electromagnetic fluctuations) into a macroscopic signal. This signal can act back on the system and leads to the generation of global spatial atomic order as well as the generation of new optical fields. This process enables me to demonstrate for the first time steady-state, multi-mode superradiance and study the system as it transitions from the single-atom to the collective regime.

In Chapter 2, I introduce the fundamental mechanisms through which light and matter interact, where I assume that the atoms do not interact with one another. In particular, I show how one can use light to control both the internal and center-of-mass states of an atom. By using spatially-varying optical fields, one can create an optical lattice that gives rise to atomic cooling (known as the Sisyphus effect) and spatial localization. I describe in detail a method for calculating the resulting atomic phase space distribution when a gas of atoms moves in a one-dimensional lattice with a spatially-varying polarization. One finds that the gas transforms into a non-thermal system consisting of two distinct components: a cold component composed of atoms localized within the optical lattice, and a hot component consisting of nearly free atoms (see Fig. 1.2). As the depth of the optical lattice $U_0$ increases, population is transferred from the hot to the cold component, resulting in atomic cooling. This result plays an important role in the development of a theoretical model for my system.

I extend this discussion in Ch. 3 to include the possibility for photon-mediated
Figure 1.2: Predicted scaling of hot and cold components. a) Temperatures of the hot ($T_h$) and cold ($T_c$) components in terms of the recoil temperature $T_r = 362 \, \text{nK}$ as a function of the lattice well depth $U_0$. b) Fraction of the atoms in the hot ($f_h$) and cold ($f_c$) components, where the solid line corresponds to a phenomenological fit.

atom-atom interactions. In this case, it is possible for an entire ensemble to synchronize and scatter light collectively when the interaction strengths are sufficiently large. I focus on two, closely-related collective phenomena: MPSO and superradiance. I present first a theoretical description of MPSO in terms of nonlinear wave mixing and show that, beyond a critical NLO coupling strength, the system becomes unstable to perturbations such that vacuum fluctuations are sufficient to produce new, macroscopic optical fields. I then introduce superradiance and discuss its realization both in terms of the synchronization of an ensemble's internal states (i.e., collective spontaneous emission) and center-of-mass states (i.e., collective emission via atomic recoil). After reviewing a semi-classical model of SRyS, I point out the correspondence between MPSO and superradiance and emphasize the advantages of cold over warm vapors.

Having completed my review of the existing literature, I present in Ch. 4 experimental results for my system below the superradiant transition (i.e., in the limit of small gain). Starting with a highly-anisotropic, pencil-shaped cloud of cold atoms, I shine a pair of counterpropagating pump beams and a weak signal beam on the vapor and study its NLO response (see Fig. 1.3). The optical lattice formed by the interfer-
Figure 1.3: Beam geometry for collective wave mixing. I apply a pair of weak, counterpropagating pump beams to a pencil-shaped cloud of cold atoms at an angle $\theta$ relative to the $\hat{z}$ axis. To study the nonlinearity below the instability threshold, I also include an incident signal beam. Through NLO wave mixing, a reflected idler field is produced. Above the instability threshold, the signal and idler fields are generated spontaneously and propagate along the trap's long axis. I detect the temporal and spatial profiles of the emitted beams using fast detectors and a camera, respectively.

The presence of these fields causes the atoms to spatially organize such that a density grating forms. The resulting atomic density grating efficiently couples the signal and pump fields and results in the amplification of the signal field and the production of a counterpropagating idler field. For a proper choice of the optical field polarizations, this process is enhanced by Sisyphus cooling and leads to a large NLO response at low-light-levels with high transparency (i.e., small linear absorption). The resulting nonlinearity displays a unique dependence on the optical fields and gives direct evidence for the existence of a hot and cold component of the atomic momentum distribution.

In Ch. 5 I develop a theoretical model of the NLO interaction studied in Ch. 4. My approach combines established techniques for describing the propagation of optical fields in a nonlinear medium and atomic motion in a dissipative optical lattice. By introducing a phenomenological description of the dependence of the fraction of atoms in the hot and cold components (see Fig. 1.2), I obtain an analytic expression for the intensities of the resulting signal and idler fields. The predictions of the model show quantitative agreement with the experimental results with no free parameters, and therefore reinforce my interpretation of the involved physical mechanism.
For sufficiently large gain, the vapor spontaneously crystallizes and emits new, intense optical fields via collective scattering when illuminated by only the pump beams (see Fig. 1.3). I investigate this superradiant transition experimentally in Ch. 6 and map out the superradiant phase diagram as a function of various system parameters. I also show that the generated light displays strong temporal correlations between the counterpropagating fields and contains multiple transverse modes (see Fig. 1.4a and b, respectively). In addition, I demonstrate that the system can operate continuously under certain circumstances, which represents the first realization of steady-state superradiance (see Fig. 1.5). Finally, I compare my results with theoretical predictions for MPSO, SRyS, and steady-state superradiance. General agreement between the observed and expected properties of the superradiant light confirm my understanding of the collective instability.

In Ch. 7 I review the main results of the thesis and discuss future research di-
Figure 1.5: Steady-state superradiance. Typical time series showing the intensities of the superradiant a) signal and b) idler modes when I turn the pump beams on at $t = 0$ and include a weak confining potential in the $\hat{x}$-$\hat{y}$ plane.

In order to tell a clear story in the body of the thesis that is focused on the observation of new physics, I defer a complete description of my experimental apparatus to the appendices. Appendix A describes the physical mechanisms responsible for my anisotropic MOT, including an effect that plays an important role in the operation of MOTs with large optical depths, which I refer to as absorption-induced trapping. Finally, App. B describes the technical details associated with realizing and characterizing my MOT, as well as its relevant properties.
Chapter 2

Introduction to the mechanical effects of light on atoms

In this chapter I discuss the basic physics underlying the interaction between light and atoms. I first derive the optical Bloch equations for a stationary two-level atom, which describe the evolution of the atom’s internal atomic states. I then consider the mechanical effects of light by calculating the force experienced by an atom in an optical field. The situation in which multiple optical fields interfere to create a spatially-varying force is known as an optical lattice and can lead to atomic cooling and spatial localization. I describe a specific cooling mechanism that occurs in an optical lattice known as Sisyphus cooling, which plays an important role in the central results that I present in this thesis (see Chs. 4-6). After qualitatively discussing this effect, I present my calculations of the atomic phase space distribution that this process produces, which aid me in developing a theoretical model for my system (see Ch. 5).

2.1 Stationary two-level atom in an optical field

I begin by considering the interaction of an optical field and a stationary two-level atom. While one can find the material presented in this section in standard textbooks (e.g., Refs. [25] and [53]), I summarize the results here in order to define my notation and develop a self-contained theory. As shown in Fig. 2.1, I look specifically at the case of a monochromatic optical field (as produced by a laser in the ideal case) with frequency $\omega$ interacting with an atom with a ground (excited) state $|g\rangle$ ($|e\rangle$) and
energy \( E_g = \hbar \omega_g \) (\( E_e = \hbar \omega_e \)). The Hamiltonian for this system is

\[
\hat{H} = \hat{H}_0 + \hat{V}(t),
\]

(2.1)

where \( \hat{H}_{0,nm} = \langle n|\hat{H}_0|m \rangle = E_n \delta_{nm} \) is the atomic Hamiltonian in the absence of an optical field and \( \hat{V}(t) \) corresponds to the interaction between the atom and optical field. In the electric dipole approximation,

\[
\hat{V}(t) = -\hat{\mu} \cdot \tilde{E}(t),
\]

(2.2)

where \( \hat{\mu} \) is the dipole operator,

\[
\tilde{E}(t) = E e^{-i\omega t} + c.c.
\]

(2.3)

is the electric field, \( E = E \hat{e} \) is the component of the electric field that varies slowly in time (compared to \( 2\pi/\omega \)), \( \hat{e} \) is the electric field polarization, and \( c.c. \) denotes the complex conjugate. Assuming that the atomic states have a definite parity implies that \( V_{gg} = V_{ee} = 0 \) and

\[
V_{eg} = V_{ge}^* = -\mu_{eg} \cdot \tilde{E}(t).
\]

(2.4)

To describe the temporal evolution of the atomic states in the presence of the optical field, I use a density matrix representation given by

\[
\dot{\rho}_{nm} = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}]_{nm},
\]

(2.5)

where \( \dot{A} \equiv \partial A/\partial t \) and \( \rho_{nm} = \rho_{nm}^* \) because \( \hat{\rho} \) is Hermitian. Substituting Eq. (2.1) into
Eq. (2.5) one finds that the equations of motion for the density matrix are given as

\[ \dot{\rho}_{eg} = -i \omega_{eg} \rho_{eg} + \frac{i}{\hbar} V_{eg}(\rho_{ee} - \rho_{gg}) \]
\[ \dot{\rho}_{ee} = -\frac{i}{\hbar} (V_{eg} \rho_{ge} - \rho_{eg} V_{ge}) \]
\[ \dot{\rho}_{gg} = -\frac{i}{\hbar} (V_{ge} \rho_{eg} - \rho_{eg} V_{ge}) \]

where \( \omega_{eg} = \omega_e - \omega_g \) is the atomic transition frequency.

Equation (2.6) describes the conservative evolution of the system. To take into account the effects of dissipation, one can include phenomenologically additional relaxation terms in Eq. (2.6). The modified equations of motion are given by

\[ \dot{\rho}_{eg} = - \left( i \omega_{eg} + \gamma \right) \rho_{eg} + \frac{i}{\hbar} V_{eg}(\rho_{ee} - \rho_{gg}) \]
\[ \dot{\rho}_{ee} = -\Gamma \rho_{ee} - \frac{i}{\hbar} (V_{eg} \rho_{ge} - \rho_{eg} V_{ge}) \]
\[ \dot{\rho}_{gg} = \Gamma \rho_{ee} + \frac{i}{\hbar} (V_{ge} \rho_{eg} - \rho_{eg} V_{ge}) \]

where the excited state decays at a rate \( \Gamma = 1/T_1 \) and the dipole moment dephases at
a rate $\gamma = 1/T_2$. These rates are related to one another such that $\gamma = \Gamma/2 + \gamma'$, where $\gamma'$ corresponds to damping caused by additional effects (e.g., elastic collisions between atoms). For simplicity, I take $\gamma' = 0$ in the following.

Since the total population is conserved in this closed system (i.e., $\rho_{gg} + \rho_{ee} = 1$), one can rewrite Eq. 2.7 in terms of the population difference $w \equiv \rho_{ee} - \rho_{gg}$ as

$$
\dot{\rho}_{eg} = -\left(i\omega_{eg} + \gamma\right)\rho_{eg} - \frac{i}{\hbar}\mu_{eg} \cdot E e^{-i\omega t} w
$$

$$
\dot{w} = -\Gamma(w + 1) + \frac{2i}{\hbar}(\mu_{eg} \cdot E e^{-i\omega t} \rho_{ge} - \mu_{ge} \cdot E^* e^{i\omega t} \rho_{eg}),
$$

where I have included the matrix elements of the interaction Hamiltonian and made the rotating-wave approximation (i.e., I take $V_{eg} \approx -\mu_{eg} E \exp[-i\omega t]$). The steady-state solutions to Eq. 2.8 are given by

$$
\begin{align*}
W &= -\frac{\left[1 + (2\Delta/\Gamma)^2\right]}{1 + (2\Delta/\Gamma)^2 + s_0} \\
\rho_{eg} &= \frac{\mu_{eg} \cdot E e^{-i\omega t} w}{\hbar(\Delta + i\Gamma/2)},
\end{align*}
$$

where $\Delta \equiv \omega - \omega_{eg}$ is the detuning of the optical field from the atomic resonance and $s_\Delta = 1/I_{sat}(\Delta)$ is the detuning-dependent saturation parameter. Here, $I = 2\epsilon_0 c|E|^2$ is the optical intensity, $I_{sat}(\Delta) = I_{sat}[1 + (2\Delta/\Gamma)^2]$ is the detuning-dependent saturation intensity, $I_{sat} \equiv \epsilon_0 c(\hbar \Gamma/2\mu_{ba})^2$ is the on-resonance saturation intensity (i.e., the intensity at which $w$ drops to one-half of its on-resonance value), $c$ is the speed of light in vacuum and $\epsilon_0$ is the permittivity of free space.

The single-atom polarization $\tilde{p}$ is related to the off-diagonal density matrix elements according to

$$
\tilde{p}(t) = \langle \mu \rangle = \text{Tr}(\rho \hat{\mu}) = (\mu_{gs} \rho_{eg} + \mu_{eg} \rho_{ge}).
$$

23
To isolate the rapidly-varying temporal components, one can introduce a complex, time independent polarization amplitude $p$ such that

$$\tilde{p}(t) = p e^{-i\omega t} + c.c. \quad (2.11)$$

This enables one to define the atomic polarizability $\varphi$ according to

$$p = \varepsilon_0 \varphi E. \quad (2.12)$$

From Eqs. 2.9 and 2.10, $\varphi$ is given as

$$\varphi = -\frac{\hbar \Gamma}{2 I_{sat}} \left( \frac{2\Delta/\Gamma - i}{1 + (2\Delta/\Gamma)^2 + s_0} \right). \quad (2.13)$$

Extending this single-atom polarizability to a macroscopic polarization (i.e., dipole moment per unit volume) for a sample of non-interacting atoms yields

$$P = \eta p, \quad (2.14)$$

where $\eta$ is the atomic density. This enables one to define the susceptibility $\chi$ according to

$$P = \varepsilon_0 \chi E. \quad (2.15)$$

From Eqs. 2.9 and 2.14, the susceptibility is given as

$$\chi = \eta \varphi = -\frac{\alpha_0}{\omega_{ba}/c} \frac{2\Delta/\Gamma - i}{1 + (2\Delta/\Gamma)^2 + 1/I_{sat}}. \quad (2.16)$$

Here $\alpha_{\Delta} \equiv \alpha_0/[1 + (2\Delta/\Gamma)^2]$ is the absorption coefficient experienced by a weak opt-
cal field and
\[ \alpha_0 \equiv \frac{\omega_{ba}}{c} \left[ \frac{2\eta|\mu_{ba}|^2}{\epsilon_0 \hbar \Gamma} \right] = \frac{\omega_{ba} \Gamma \hbar}{2 I_{sat}} \] (2.17)
is the on-resonance absorption coefficient.

### 2.2 Radiation pressure and the optical dipole force

While massless, photons carry a definite momentum and can, therefore, exert a force on matter. I take a simple, semiclassical approach to calculate the force imposed by an optical field on a stationary atom. While this may seem artificial given that a localized atom can never be stationary and a net force will instantly cause the atom to begin moving, it is a useful exercise that approximates reality in many situations. In later sections, I treat the problem of a moving atom in various optical field configurations using a more complete model.

The force experienced by an atom in an optical field is [53]

\[ F = \frac{d}{dt} \langle \hat{p} \rangle = \frac{i}{\hbar} \langle [\hat{H}, \hat{p}] \rangle, \] (2.18)

where \( \hat{p} \) is the momentum operator and \( \langle A \rangle = \text{Tr}(\hat{\rho}A) \) represents the expectation value of \( A \). By evaluating the commutator between \( \hat{H} \) and \( \hat{p} \) using the relationship \( \hat{p} = -i\hbar \nabla \), one finds that the force is simply the gradient of the optical potential

\[ F = -\nabla \langle \hat{H} \rangle. \] (2.19)

In the dipole approximation, the relevant portion of the Hamiltonian is given by Eq. [2.2] where we now consider the electric field to have a spatial dependence. Evaluating
the expectation value in Eq. 2.19 gives

\[ F = \nabla \text{Tr}(\rho \hat{\mu} \cdot \hat{E}) \]

\[ = \nabla [\hat{E} \cdot \text{Tr}(\rho \hat{\mu})] \]  \hspace{1cm} (2.20)

In the limit of low saturation (i.e., when \( s_0 \ll 1 \), one can ignore the position dependence of the atom-field coupling and Eq. 2.20 simplifies to

\[ F = (\hat{p} \cdot \nabla)\hat{E} \]

\[ = (2 \text{Re}[\epsilon_0 \hat{E} e^{-i\omega t}] \cdot \nabla)2 \text{Re}[\hat{E} e^{-i\omega t}]. \]  \hspace{1cm} (2.21)

Assuming that the electric field has the general form

\[ E(r) = E_0(r) e^{i\phi(r)} \]  \hspace{1cm} (2.22)

and averaging the force over a time large compared to \( 2\pi/\omega \), one finds that the total force can be written as \( F = F_{sc} + F_d \) \[55\]. The first term, known as the scattering or radiation pressure force, is

\[ F_{sc} = 2\epsilon_0 \phi'' E_0^2(r) \nabla \phi(r), \]  \hspace{1cm} (2.23)

where \( \phi = \phi' + i\phi'' \). The second term, typically called the dipole or gradient force, is

\[ F_d = 2\epsilon_0 \phi' E_0(r) \nabla E_0(r). \]  \hspace{1cm} (2.24)

While it is possible to tailor the relative amplitudes of these two forces by controlling the optical field, the ratio \( F_d/F_{sc} \) is on the order of \( \phi'/\phi'' \sim |\Delta|/\Gamma \) for the case where
$E_0$ and $\phi$ vary over comparable spatial scales. Thus, one can control the relative
importance of these two forces by choosing the detuning from resonance. To gain
insight into the physical mechanisms underlying these forces, it is instructive to look
at simple, limiting cases for each force.

First, I consider the scattering force. For a single, traveling wave with wave vector
$k = k \hat{z}$, one can write the electric field as

$$E(z) = E_0 e^{ikz}. \quad (2.25)$$

The electric field therefore has a constant amplitude and spatially-varying phase, and
gives rise to a force

$$F_{sc}(z) = \hbar k \frac{s_0}{2} \frac{1}{1 + (2\Delta/\Gamma)^2 + s_0} \hat{z}. \quad (2.26)$$

One can understand this force by considering the recoil imparted to an atom via an
absorption and spontaneous emission cycle. Because absorption occurs along $\hat{k}$ and
spontaneous emission is isotropic (and therefore transfers no net momentum), the
atom gets an average momentum kick of $\hbar k$ per scattering event. The rate at which
these scattering events occurs is $\gamma_{sc}$, which one can calculate using Eq. $2.9$ and conser-
vation of population as

$$\gamma_{sc} = \frac{\Gamma \rho_{ee}}{2} \frac{s_0}{1 + (2\Delta/\Gamma)^2 + s_0}. \quad (2.27)$$

Thus, in agreement with one’s intuition, the scattering force given in Eq. $2.26$ simply
corresponds to $F_{sc} = \hbar k \gamma_{sc}$. The role of spontaneous emission in this process is critical.
It limits the maximum amplitude of the force to $\hbar k \Gamma/2$ for intense beams, because the
largest possible scattering rate is $\gamma_{sc} = \Gamma/2$ when the excited state occupancy reaches
its maximum value of $\rho_{bb} = 1/2$. In addition, spontaneous emission provides an irreversible loss mechanism that can lead to atomic cooling, although the minimum achievable temperature is limited to the Doppler temperature $T_D = \hbar \Gamma / 2k_B$ due to the inherent randomness of spontaneous emission (see App. A.2). Typical values for $T_D$ are on the order of 100 $\mu$K.

In contrast, the dipole force is conservative and cannot directly lead to cooling (although, as I discuss later in this chapter, cooling schemes involving the dipole force do exist). The physical mechanism underlying the dipole force is the AC Stark effect in a position-dependent optical field, which produces a spatially-varying modulation of the ground state energy (or light shift). One simple example in which such a light shift arises is a standing wave composed of two copolarized, counterpropagating traveling waves along $\pm k \hat{z}$. One can write the electric field as

$$E(z) = A(e^{-ikz} + e^{ikz}) = 2A\cos(kz), \quad (2.28)$$

where the amplitude varies spatially but the phase is constant. From Eq. 2.24, this produces the force

$$F_d = \frac{2\hbar k \Delta \sin(2kz)I_A/I_{sat}}{1 + (2\Delta/\Gamma)^2 + s_0(z)} \hat{z}, \quad (2.29)$$

where $I_A = 2\varepsilon_0 c |A|^2$ is the single-beam intensity. Unlike the scattering force, which saturates due its dependence on spontaneous emission, the dipole force does not saturate at large intensities.

In addition, because the dipole force is conservative, one can write $F_d = -\nabla U_d$, where $U_d$ is the effective dipole potential. For the case of a standing wave, the potential
becomes

\[ U_d = \frac{\hbar \Delta}{2} \log \left( 1 + s_\Delta \right). \]  

(2.30)

The potential minima correspond to the locations of lowest atomic energy; therefore, the atoms are attracted to the potential minima. For red (blue) detuned optical fields \([i.e., \Delta < 0 (\Delta > 0)]\), these minima occur at the locations of maximum (minimum) optical intensity. As I discuss in Sec. 2.3, the ability to spatially arrange atoms via the dipole force has many applications.

One can generalize the results of Eqs. 2.26 and 2.29 for the case where the total electric field arises from the superposition of multiple plane waves as

\[ F_{sc} = 2\varepsilon_0 \mu'' \left[ \sum_l E_l^2 k_l + \frac{1}{2} \sum_{l \neq j} (k_l + k_j) E_l E_j (\mathbf{e}_l \cdot \mathbf{e}_j^*) e^{i((k_l - k_j) \cdot r + (\phi_l - \phi_j))} \right], \]

(2.31)

\[ F_d = \varepsilon_0 \mu' \sum_{l \neq j} i(k_l - k_j) E_l E_j (\mathbf{e}_l \cdot \mathbf{e}_j^*) e^{i((k_l - k_j) \cdot r + (\phi_l - \phi_j))}, \]

(2.32)

where \(E_j(r), \mathbf{e}_j, k_j, \) and \(\phi_j\) are the amplitude, polarization, wave vector, and phase of field \(j\).

2.3 Optical lattices and sub-Doppler cooling

The idea of using light to spatially confine atoms in sub-wavelength regions of space, called microtraps, was suggested as early as 1968 [54]. An array of optical microtraps is known as an optical lattice. The standing wave described in the previous section corresponds to a one-dimensional (1D) lattice, where the spatially-varying light shift gives rise to a potential that modifies the atomic center-of-mass motion. One can create more complicated, multi-dimensional lattices by interfering multiple optical fields. By varying the optical fields’ relative intensities, wave vectors, and phases, one can con-
trol the structure and depth of the lattice. Consequently, one can use optical lattices to
effectively build materials from the ground up and manipulate them at the quantum
level. Thus, optical lattices have become an integral part of a variety of areas, including atomic and molecular physics [55], condensed matter physics [56], and quantum
information science [57].

2.3.1 Two-level atom in an optical lattice

Before tackling more complex situations, I focus first on the 1D optical lattice discussed
above, which is formed by co-polarized, counterpropagating optical lattice fields with balanced intensities. In this case, the radiation pressure force vanishes and one obtains
the dipole potential given in Eq. 2.30. If one works in the limit of low saturation such
that \( s_\Delta \ll 1 \), the dipole potential reduces to the simple form

\[
U_d \sim U_0 \cos^2(kz)
\]

where

\[
U_0 \sim \frac{2 \hbar \Delta I_A}{I_{\text{sat}}(\Delta)}
\]

is the depth of the potential wells. As shown in Fig. 2.2, the resulting potential is
sinusoidal, has a spatial extent of less than an optical wavelength, and is periodic with
period \( \lambda/2 \), where \( \lambda \) is the wavelength of the light.

When the depth of the potential wells exceeds the average atomic energy, the atoms
become spatially trapped wells. In many cases, typical atomic momenta are on the
order of the recoil momentum \( (p_r = \hbar k) \), as discussed below), which implies that their
deBroglie wavelengths are on the order of an optical wavelength. Thus, one must
describe the atomic motion quantum mechanically. For atoms at the tops of the wells,
tunneling from one well to another can occur. This gives rise to a band structure
of allowed and forbidden energy levels and allows one to observe phenomena well-
known in solid-state physics, such as Bloch oscillations [58]. The observation of such
Figure 2.2: Vibrational transitions in a dipole potential. Resulting dipole potential wells due to a standing optical wave of depth $U_0$. The energy levels deep in the well are spaced by $\hbar \omega_{\text{vib}}$ whereas, near the top of the well, energy bands develop. Transitions between different vibrational levels give rise to resonances at integer multiples of $\omega_{\text{vib}}$.

effects illustrates that, even for unconfined atomic states in such an optical lattice, one must consider the quantized nature of the center-of-mass motion.

For colder atoms near the bottom of the wells, the energy bands become very narrow. By expanding $U_d$ in the vicinity of the potential minima, one recovers a purely quadratic potential of the form $U_d \sim U_0(1 - k^2z^2)$. The atoms therefore exhibit oscillatory motion with frequency

$$\omega_{\text{vib}} = \sqrt{\frac{2U_0k^2}{m}} = \frac{2}{\hbar} \sqrt{U_0E_r}, \quad (2.34)$$

where $E_r = (\hbar k)^2/(2m)$ is the recoil energy associated with a photon. The atomic wavefunctions correspond to harmonic oscillator states, where the state $|n\rangle$ has an energy $E_n = \hbar \omega_{\text{vib}}(n + 1/2)$. The quantized nature of the bound states in an optical lattice was observed directly via two complimentary methods. In the first technique, one
mixes the fluorescent light emitted by the trapped atoms with a weak optical field and records the beatnote \[59\]. Fourier transforming the beatnote yields the power spectrum of the emitted light, which displays a very narrow peak at \(\omega_{\text{beat}} = 0\) due to elastic scattering (i.e., transitions involving \(\Delta n = 0\)) and sidebands at \(\omega_{\text{beat}} = \pm \omega_{\text{vib}}\) due to transitions in which atoms gained or lost a single vibrational quanta (i.e, \(\Delta n = \pm 1\), see Fig. 2.2). The amplitudes of the sidebands correspond to the relative populations of the vibrational levels, and therefore give direct information about the equilibrium atomic temperature. An alternative technique involves shining a pump and probe beam on the atoms. By scanning the frequency of the probe beam, one can drive two-photon transitions between the vibrational levels, which give rise to resonances in the transmitted probe spectrum at multiples of \(\omega_{\text{vib}}\).

One might expect that the spontaneous scattering of photons from atoms in the optical lattice, which represents the most rapid decay mechanism, would limit the narrowest observable spectra features to \(\gamma_{\text{sc}}\) (i.e., the optical scattering rate, given by Eq. 2.27). While this holds for free atoms, a mechanism called Dicke narrowing occurs for atoms confined to a region smaller than an optical wavelength and substantially decreases the linewidth of the resulting spectral features [7]. One can easily understand the origin of this phenomena by considering the case where an atom in state \(|n\rangle\) sequentially absorbs a photon and spontaneously emits a photon to end up in state \(|m\rangle\). The rate at which this occurs is \(\gamma_{nm} = \gamma_{sc}|M|^2\), where \(M = \langle m | \exp(ik\hat{z}) | n \rangle\) is the transition amplitude for this process. Using the relationship \(\hat{z} = \sqrt{\hbar/2m\omega_{\text{vib}}} (\hat{a} + \hat{a}^\dagger)\), one can write

\[
|M|^2 = \sum_j \left| \langle m | \left[ i \sqrt{\hbar k^2/2m\omega_{\text{vib}}} (\hat{a} + \hat{a}^\dagger) \right]^j \right| n \rangle \right|^2 ,
\]

where \(\hat{a}^\dagger\) and \(\hat{a}\) are the harmonic oscillator raising and lowering operators, respectively.
For an atom bound deeply in the potential well, the quantity \( k \Delta z_n \ll 1 \), where

\[
\Delta z_n = \left( \langle n | \hat{a}^2 | n \rangle \right)^{1/2} = \left[ \frac{(2n + 1)\hbar}{2m\omega_{vib}} \right]^{1/2}
\]

is the root-mean-squared (rms) extent of the atomic wave function. Evaluating Eq. 2.35 in terms of this small quantity, one finds that

\[
|M|^2 = \delta_{m,n} + (k\Delta z_n)^2 \left( -2\delta_{m,n} + \frac{n}{2n+1} \delta_{m,n-1} + \frac{n+1}{2n+1} \delta_{m,n+1} \right) + O[(k\Delta z_n)^4],
\]

where \( \delta_{m,n} \) is the Kronicker delta function.

The probability of remaining in the same state after scattering is nearly unity, while scattering between different levels is reduced by the Lamb-Dicke factor \( \varepsilon = E_r / \hbar\omega_{vib} \). Thus, while the total scattering rate remains unchanged, the probability of elastic scattering becomes greatly enhanced with respect to inelastic scattering. One can alternatively understand the recoil-free scattering of bound atoms in terms of the Mössbauer effect [60], which was originally discovered in the context of nuclear physics. In this case, one can view the absorption-emission cycle as a collective scattering process, where the recoil is taken up by the lattice as a whole. Because the mass of the many-atom lattice is much larger than that of a single atom, the recoil on any given atom is negligible.

### 2.3.2 Multilevel atom in a dissipative optical lattice

Up to this point, I have discussed only the motion of two-level atoms in a conservative optical lattice. If one considers multi-level atoms, new cooling mechanisms arise that enable sub-Doppler temperatures. In general, these mechanisms are based on non-
adiabatic optical pumping between internal atomic states as the atoms move. Because the light shifts of the individual atomic levels in an optical lattice can be different from one another, an atom can gain or lose energy by jumping from one potential surface to the next. Cooling occurs when one chooses a configuration such that the atoms repeatedly jump from a higher to lower potential surface as they move through space. Because the atoms find themselves endlessly climbing potential hills, this sub-Doppler cooling process is known as Sisyphus cooling.

A simple example of Sisyphus cooling occurs in the 1D lattice described in Sec. 2.3 (i.e., co-polarized, counterpropagating fields) if one considers a $J_g = 1/2 \rightarrow J_e = 3/2$ transition (level scheme shown in Fig. 2.3 a) and circularly-polarized optical fields. For concreteness, I choose $\sigma^+$-polarized fields without loss of generality. In this case, one finds two potentials $U_{\pm 1/2}$ that describe the light shifts for the $m_g = \pm 1/2$ states, where $U_{1/2} \geq U_{-1/2}$ because the $m_g = 1/2$ state couples more strongly to the field than the $m_g = -1/2$ due to the transitions’ respective Clebsch-Gordan coefficients (see Fig. 2.3 a). Optical pumping by the $\sigma^+$ beams tends to put the atoms into the $m_g = 1/2$ state. In the limit of low field intensities, the atoms adiabatically follow the $U_{1/2}$ potential as they move through space (see Fig. 2.3 b).

When one includes a transverse magnetic field, though, the atoms no longer simply follow the potential. In the lattice nodes, where the optical intensity is zero, the atoms undergo Larmor precession, which effectively converts an $m_g = 1/2$ state into an $m = -1/2$ state. Because $U_{1/2} = U_{-1/2}$ at the nodes, the atomic energy does not change in this process. As the atoms move into the antinodes, they are preferentially optically pumped back into the $m_g = 1/2$ state, thereby causing them to lose energy. The atoms then climb up the $U_{1/2}$ potential curve until arriving at the next node, where the process repeats. This process is typically referred to as magnetically-induced laser cooling (MILC), due to the requirement of an externally-applied magnetic field.
Figure 2.3: MILC in a $J_g = 1/2 \rightarrow J_e = 3/2$ atom. a) Atomic level structure for a $J_g = 1/2 \rightarrow J_e = 3/2$ transition including along with the square of the Clebsh-Gordan coefficients (defined as a fraction of the reduced dipole moment for the transition). b) Cooling scheme used in MILC. Starting at position 1, an atom in the $m_g = 1/2$ state converts kinetic into potential energy as it climbs to position 2. Larmor precession then transfers the atom to the $m_g = -1/2$ state, the atom continues to position 3, and then undergoes optical pumping back to the $m_g = 1/2$ state in which the atom radiates energy and ends up at position 4 with a reduced total energy. This figure is adapted from Ref. [53].
A method for realizing all-optical sub-Doppler cooling (i.e., without requiring an external magnetic field) involves counterpropagating beams with orthogonal polarizations. In this case, the fields do not interfere, and the intensity is uniform in space. However, the polarization of the total electric field varies in space. For example, linearly cross-polarized, counterpropagating fields (see Fig. 2.4a), give rise to a total electric field

\[ E_\perp(z) = E_0(e^{ikz}\hat{x} - ie^{-ikz}\hat{y}) = \sqrt{2}E_0[\cos(kz)e_- - i\sin(kz)e_+], \] (2.38)

where \( e_\pm = (\hat{x} \pm i\hat{y})/\sqrt{2} \) are unit vectors in the spherical basis. This choice of polarizations is typically referred to as the lin\(_\perp\)lin configuration. As shown in Fig. 2.4b, the field is polarization is \( e^- \) at \( z = 0 \), linear along \( \hat{x} - \hat{y} \) at \( z = \lambda/8 \), \( e^+ \) at \( z = 3\lambda/8 \), linear along \( \hat{x} + \hat{y} \) at \( z = \lambda/4 \). Because of the different Clebsch-Gordan coefficients, the coupling between the individual atomic levels and the optical field on the field’s polarization at a particular location. As an example, I again consider the simple case of a \( J_g = 1/2 \rightarrow J_e = 3/2 \) transition. Like in MILC, one finds two distinct optical potentials \( U_{\pm1/2} \) associated with the two ground state \( m_g = \pm1/2 \) (see Fig. 2.5a). The \( m = 1/2 \) \( (m = -1/2) \) ground state couples most strongly to the optical field and, therefore, experiences the largest light shift at \( z = (2n+1)\lambda/4 \) \( (z = n\lambda/2) \), where the optical field is polarized in the \( \hat{e}_+ \) \( (\hat{e}_-) \) direction. As the atom moves through space, there is a time lag between when the atom encounters a new optical field polarization and when it undergoes optical pumping. This lag is equal to the inverse of the optical pumping rate \( 1/\gamma_p \). In the limit of low saturation, the atoms tend to lose potential energy by repeatedly climbing a potential hill and then undergoing optical pumping that transitions them to another potential surface with lower energy.
For atoms with $J_g > 1/2$, a related mechanism occurs even when the atoms are already confined to a single potential well (i.e., travel over a distance of less than $\lambda/4$, see Fig. 2.5b). While atoms will tend to accumulate in the stretched states (i.e., $|m_g| = J_g$) at the locations where the light is circularly polarized, atoms displaced from this location have a small probability of undergoing optical pumping into a different internal state that has a larger potential energy. As the atoms oscillate in this potential, they will preferentially be pumped back into the stretched state at the location of the potential minima. The atoms lose energy on average because the energy difference between the potential surfaces is largest where the atoms are most likely to be pumped into the stretched state.

This type of Sisyphus cooling is commonly referred to as polarization gradient cooling, and can occur for any configuration of optical fields that give rise to spatially-varying field polarizations (e.g., counterpropagating beams with non-parallel linear polarizations and orthogonal circular polarizations). While it occurs efficiently, Sisyphus cooling does not cool atoms to arbitrarily low temperatures. Like Doppler cooling,
Figure 2.5: Polarization gradient cooling.  a) Polarization gradient cooling in a \textit{lin}_{\perp} \textit{lin} lattice with a $J_g = 1/2 \rightarrow J_e = 3/2$ transition. Starting at position 1 in the $m_g = 1/2$ state, an atom converts kinetic into potential energy as it climbs up the potential. At position 2, the atom is optically pumped into the $m_g = -1/2$ state and loses energy via spontaneous emission as it transitions to position 3. b) Local polarization gradient cooling for $J_g > 1/2$. The atom starts at position 1 with $|m_g| = J_g$, arrives at position 2 by converting kinetic into potential energy, undergoes optical pumping to a state with $|m_g| = J_g - 1$ at position 3, oscillates back toward the center of this well at position 4, and loses energy radiatively by undergoing optical pumping back to position 1. These figures are adapted from Ref. \cite{55}.
Sisyphus cooling relies on spontaneous emission and necessarily involves some degree of diffusive heating due to the random nature of the process. These approaches, therefore, are intrinsically bounded by the recoil temperature \( T_r = E_r/k_B \), which corresponds to the energy imparted to an atom via the absorption or emission of a photon and are typically on the order of a few hundred nK. Nevertheless, one can show that Sisyphus cooling leads to a larger damping coefficient than Doppler cooling (albeit over a smaller range of velocities), resulting in sub-Doppler temperatures on the order of \( T \sim 10T_r \ll T_D \). Due to the vital role that Sisyphus cooling plays in the results I present later in this thesis, I discuss Sisyphus cooling in a more quantitative and in-depth manner in the next section.

2.4 Sisyphus cooling in a linear lattice

As described above, there are a number of different mechanisms and configurations that allow for Sisyphus cooling. In this section, I focus on polarization gradient cooling in an optical lattice formed by counterpropagating beams with linear, orthogonal polarizations. While similar results occur with other field polarizations, this particular choice of polarizations leads to an intuitive and technically straightforward quantitative description of the cooling mechanism. In addition, I work with a \( J_g = 1/2 \rightarrow J_e = 3/2 \) transition. While such an atomic level structure does not allow for all possible phenomena associated with more complicated level schemes to occur, I choose this configuration in order to construct a minimal model that, as I demonstrate in Ch. 5, includes the core, relevant physics discussed in this dissertation.

Several different methods have been developed to model the internal and center-of-mass dynamics of multilevel atoms moving in optical fields. While semiclassical, analytic approaches [61] to the problem give insight into the physical mechanisms
involved, the approximations required to achieve a closed-form solution typically give only qualitative agreement with experimental observations. On the other hand, Monte-Carlo wavefunction approaches [62] show excellent quantitative agreement with experiments, but give relatively little physical insight into the process underlying a given macroscopic effect. As an intermediate approach, semi-classical Monte-Carlo simulations based on the Wigner representation allow one to formulate the problem in terms of a Fokker-Planck-like equation [63]. This approach gives one increased control over the involved macroscopic parameters and allows one to gain substantial understanding of the associated elementary processes since one can track path of individual atoms through phase space. While the Monte-Carlo techniques are necessary for efficiently simulating the atomic dynamics in multi-dimensional optical lattices and including arbitrary level structures, an additional, fully-quantum approach known as the band method enables one to solve directly for the atomic wave function under certain conditions [64].

In the band method, one takes advantage of the connections between optical lattices and periodic solid state physics by employing Bloch’s theorem to develop the atomic wavefunction in the basis of Bloch states [65]. As for the case of electrons in a lattice of ions, one finds bands of allowed and forbidden atomic energies. In this approach, one begins with a master equation describing the evolution of the density matrix in the presence of both a conservative, external potential and dissipation. One solves this problem perturbatively by first calculating the eigenstates and energy spectrum of the system in the absence of dissipation. One then takes the relaxation terms into account by calculating the transfer rates between each level. Finally, one obtains a set of rate equations for the populations in each eigenstate that allows for the calculation of various properties of the system (such as the evolution of the position and momentum distributions). This approach is formally valid when the Hamiltonian evo-
olution dominates the dynamics, known as the secular limit, which corresponds to the regime that I operate in experimentally.

### 2.4.1 Equations of motion for the ground states

One can derive the optical Bloch equations for an atom in a standing wave under very general conditions and in a fully-quantum mechanical way using a master equation formalism \([66, 67]\). Following the approach developed by Castin et al. \([68]\), I make two assumptions that allow me to simplify the Bloch equations for the experimental regime in which I operate. First, because the atoms are relatively cold, I assume that the atomic Doppler shift \(\omega_D = k \tilde{v}\) is smaller than \(\Gamma\), where \(\tilde{v}\) is the average atomic velocity. Thus, I explicitly ignore Doppler cooling. Second, I assume operation in the low saturation regime, where \(s \Delta \ll 1\). This allows me to adiabatically eliminate the excited state populations and coherences in terms of the ground state density matrix, \(\sigma\). The resulting equation of motion for \(\sigma\) is \([68]\)

\[
\dot{\sigma} = \frac{1}{i \hbar} [\hat{H}, \sigma] + \dot{\sigma}_{sp}, \tag{2.39}
\]

where the first term on the right side of this equation represents the conservative, Hamiltonian part of the evolution (as in Eq. \([2.5]\)), and the second term represents the dissipative, non-Hamiltonian part of the evolution due to spontaneous emission.

In the dipole approximation, the Hamiltonian term becomes

\[
\hat{H} = \frac{p^2}{2m} + \hat{V}(r), \tag{2.40}
\]
where $\hat{V} = \hbar \Delta' \hat{A}$ and $\Delta' = \Delta s_\Delta / 2$. The operator

$$\hat{A}(r) = \left[ \hat{d}^- \cdot \epsilon^*(r) \right] \left[ \hat{d}^+ \cdot \epsilon(r) \right]$$  \hspace{1cm} (2.41)

corresponds to the coherent redistribution of photons between the lattice beams. Here, $\epsilon(r)$ is the position-dependent optical field polarization, $\hat{d}^+$ is an operator that promotes an atom from the ground to the excited state, and $\hat{d}^- = (\hat{d}^+)^\dagger$. For a $J_g = 1/2 \rightarrow J_e = 3/2$ transition, one can write the excitation operator as

$$\hat{d}^+ = \epsilon_+ \left( |e_{3/2}^\pm \rangle \langle +| + \frac{1}{\sqrt{3}} |e_{1/2}^\pm \rangle \langle -| \right) + \epsilon_0 \frac{2}{\sqrt{3}} \left( |e_{1/2}^\pm \rangle \langle +| + |e_{-1/2}^- \rangle \langle -| \right) + \epsilon_- \left( \frac{1}{\sqrt{3}} |e_{-1/2}^- \rangle \langle +| + |e_{-3/2}^- \rangle \langle -| \right),$$  \hspace{1cm} (2.42)

where $\epsilon_0 = \hat{z}$. The states $|\pm \rangle \equiv |g_{\pm 1/2}, p \rangle$ and $|e_{\pm m}^\pm \rangle \equiv |e_{\pm m}, p \rangle$ correspond to the ground and excited states with momentum $p$ along $\hat{z}$. For the field given in Eq. 2.38, one can write

$$\hat{A}(r) = \frac{3}{2} \{ [2 - \cos(2kz)] |+\rangle \langle +| + [2 + \cos (2kz)] |−\rangle \langle −| \}$$  \hspace{1cm} (2.43)

Substituting Eq. 2.43 in to Eq. 2.40 and using the electric field given in Eq. 2.38, the Hamiltonian becomes

$$\hat{H} = \frac{\hat{p}^2}{2m} + U_+(z) |+\rangle \langle +| + U_-(z) |−\rangle \langle −|,$$  \hspace{1cm} (2.44)

where

$$U_\pm(z) = \frac{U_0}{2} \left[ -2 \pm \cos(2kz) \right]$$  \hspace{1cm} (2.45)
are the adiabatic potentials for the $|±\rangle$ states and $U_0 = -2/3\hbar\Delta s_\Delta$ is the well depth. Thus, in the absence of dissipation, the atoms simply move in a sinusoidal bi-potential due to the spatially-dependent light shifts of the two ground states.

One can write the dissipation term as

$$
\dot{\sigma}_{sp} = -\frac{\Gamma'}{2}(\dot{A}\sigma + \sigma\dot{A}) + \frac{3\Gamma'}{8\pi} \int d\Omega_k \sum_{\zeta\perp k} \hat{B}^\dagger_{\zeta} e^{-i\mathbf{k} \cdot \mathbf{r}} \sigma e^{i\mathbf{k} \cdot \mathbf{r}} \hat{B}_{\zeta},
$$

(2.46)

where $\Gamma' = \Gamma s_\Delta/2$. The operator

$$
\hat{B}_{\zeta}(r) = \left[ \hat{d}^- \cdot e^\ast(r) \right] \left[ \hat{d}^+ \cdot \zeta \right].
$$

(2.47)

represents the excitation of an atom by the applied optical field followed by the spontaneous emission of a photon with polarization $\zeta$. For the specific case considered here, one can write

$$
\dot{\sigma}_{sp} = -\frac{\Gamma'}{2}(\dot{A}\sigma + \sigma\dot{A}) + \Gamma' \int_{-\hbar k}^{\hbar k} dp' \sum_m N_m(p')\hat{B}^\dagger_{m} e^{-ip'/\hbar}\sigma e^{ip'/\hbar}\hat{B}_{m},
$$

(2.48)

where

$$
B_0 = \cos(kz) |+\rangle \langle -| + \sin(kz) |+\rangle \langle -|
$$

$$
B_1 = \sin(kz)/\sqrt{2}(3|+\rangle \langle +| + |+\rangle \langle 3| -| \langle -|
$$

$$
B_{-1} = \cos(kz)/\sqrt{2}(|+\rangle \langle +| + 3| -\rangle \langle -|
$$

(2.49)
and

\[
N_0 = \frac{3}{4\hbar k} \left[ 1 + \left( \frac{p'}{\hbar k} \right)^2 \right] ,
\]

\[
N_{\pm 1} = \frac{3}{8\hbar k} \left[ 1 - \left( \frac{p'}{\hbar k} \right)^2 \right]
\]  

(2.50)

describe the normalized distribution of spontaneously emitted photons. Physically, the first and second terms in Eq. 2.48 describe the transition from and to the state \(|\pm\rangle\) via an absorption-spontaneous emission event, respectively. Together, Eqs. 2.39, 2.44 and 2.48 completely describe the atomic evolution.

### 2.4.2 The band method

In order to solve Eq. 2.39, I consider the case where the Hamiltonian terms dominate the dissipative terms. This situation occurs if \(\omega_{\text{vib}}/\Gamma' \gg 1\), which corresponds physically to the situation where an atom oscillates many times in a given well before jumping to another one (either due to optical pumping or tunneling). For the potential given in Eq. 2.45, one can write this condition as

\[
\frac{\omega_{\text{vib}}}{\Gamma'} = 6 \frac{\Delta}{\Gamma} \sqrt{\frac{E_r}{U_0}} \gg 1.
\]  

(2.51)

As I discuss later, this regime allows one to make a secular approximation that greatly simplifies the problem. The experiments described in this thesis typically involve potential well depths \(U_0/E_r \leq 100\) and detunings \(\Delta/\Gamma \geq 3\), and therefore satisfy Eq. 2.51.

Making this approximation enables one to proceed by first solving Eq. 2.39 (i.e., the equation of motion for \(\sigma\)) in the absence of dissipation, which corresponds to the
well-known Bloch problem involving a particle moving in a period potential. For a general potential \( V(z) = \sum V_n \exp(\imath nGz) \), Bloch’s theorem states that one can write the wavefunction \( \psi^{(\nu,q)} \) that satisfies the Schrödinger equation

\[
\hat{H}_0 \psi^{(\nu,q)} = \left[ \frac{\hat{p}^2}{2m} + V(z) \right] \psi^{(\nu,q)} = E^{(\nu,q)} \psi^{(\nu,q)}
\]  

(2.52)
as \( \psi^{(\nu,q)} = \langle z | \nu, q \rangle = \exp(\imath qz)u^{(\nu,q)} \) for a function \( u^{(\nu,q)} = \sum c_n^{(\nu,q)} \exp(\imath nGz) \) with the same periodicity as the potential. Here, \( q \in [-k, k) \) is the quasimomentum of the atom in the first Brillouin zone, \( \nu \) represents the energy band, and \( G \) is the reciprocal lattice vector. Substituting this general form of \( \psi^{(\nu,q)} \) into Eq. 2.52, one arrives at the set of linear equations

\[
\left[ \frac{\hbar^2}{2m} (nG + q)^2 - E^{(\nu,q)} \right] c_n^{(\nu,q)} + \sum_l V_l c_{n-1}^{(\nu,q)} = 0,
\]  

(2.53)
where \( c_n^{(\nu,q)} \) and \( E^{(\nu,q)} \) correspond to the eigenvectors and eigenenergies of the system. In general, the solutions to this equation lead to alternative bands of allowed and forbidden energies.

For the case of the \( J_g = 1/2 \rightarrow J_e = 3/2 \) transition that I consider here, no coherences exist between the two ground states. The two ground states are therefore uncoupled from one another (as evidenced by the potential in Eq. 2.44), which allows one to reduce the problem involving two spin states to two independent, scalar problems for the two ground states. For atoms in the \( |\pm\rangle \) state, the potential in Eq. 2.45 gives \( V_0 = -U_0, V_1 = V_{-1} = \pm U_0/4 \) for \( G = 2k \). In this case, Eq. 2.53 becomes

\[
c_n^{(\nu,q,\pm)} = \pm \frac{U_0}{4} \frac{c_{n-1}^{(\nu,q,\pm)} + c_{n+1}^{(\nu,q,\pm)}}{E_r(2n + q/k)^2 - E^{(\nu,q)} - U_0},
\]

(2.54)
where I now use the notation \( c_n^{(\nu,q,m_j)} \) to include the two ground states. As expected,
one finds that the resulting energies and wavefunctions for atoms in the $|−\rangle$ state are identical to those in the $|+\rangle$ state (up to a spatial translation of $\lambda/4$).

In order to end the recursion relation and solve Eq. 2.54 one must choose a maximum value of $n$ such that $c_N^{(\nu,q,\pm)} \approx 0$. In addition, one can impose periodic boundary conditions on the problem by considering a finite lattice of length $L(\lambda/2)$. This discretizes the quasimomentum at an interval of $2k/L$, and thereby reduces the computation necessary. I find that I require $N = 40$ and $L = 6$ in order to get accurate results for the well depths studied in this dissertation, which agrees with the results presented in Ref. [64].

Figure 2.6a shows the resulting probability distribution $\eta_{\text{norm}}^{(1,0,+)} = |\psi^{(1,0,+)}|^2$ for several well depths, where I have normalized $\eta^{(1,0,+)}$ such that the peak densities are equal in order to facilitate comparison between the distributions. The probability distribution repeats with a period $\lambda/2$, is maximum in the vicinity of the potential minima, and becomes highly-modulated even for relatively shallow wells (e.g., $U_0/E_r \sim 10$). Figure 2.6b shows the energy spectrum for the different bands as a function of quasimomentum. For the higher states, the energy curves resemble the parabolic dispersion relation of free atoms given by $E = q^2/2m$. The lower bands demonstrate the emergence of band gaps as the well depth increases. The energy splitting between the lowest bands is $E_{\nu+1} - E_{\nu} = \hbar \omega_{\text{vib}}$ where $\omega_{\text{vib}}/2\pi = 207 \sqrt{\Gamma_0/2\Delta} \text{ kHz}$ for $^{87}\text{Rb}$. The number of bound states increases as $\sqrt{U_0/E_r}$ [64], where typical widths of the bound bands are on the order of $10^{-4}E_r$. Thus, tunneling between wells is negligible for the most tightly-bound states.

Now that I have calculated the eigenstates of $\hat{H}$, I take into account the dissipative term $\sigma_{sp}$ in Eq. 2.39. For the case discussed above, where $\omega_{\text{vib}} \Gamma' \gg 1$, the energy separation between the lowest bands is much larger than the width of the bands. This allows me to make a secular approximation and assume that $\sigma$ is diagonal in the basis
Figure 2.6: Bloch states. a1), b1), c1) Bloch wavefunctions and a2), b2), c2) energy spectrum for $U_0/E_r = 0, 10$ and $100$, respectively. As $U_0$ increases, the states become more highly-localized and band gaps form.
|ν, q, mg⟩. One can understand this approximation by considering the density matrix element between two different energy bands for the same ground state, which is given by Eq. 2.39

$$\dot{\sigma}_{\nu'\nu} = \frac{1}{i\hbar}(E_{\nu'} - E_{\nu})\sigma_{\nu'\nu} + \Gamma' \langle \nu' | \dot{\sigma}_{sp} | \nu \rangle.$$  \hspace{1cm} \text{(2.55)}

In steady-state, one therefore expects that

$$\sigma_{\nu'\nu} = \frac{\Gamma'}{(\nu' - \nu)\omega_{vib}} \langle \nu' | \dot{\sigma}_{sp} | \nu \rangle \sim \delta_{\nu',\nu} \sigma_{\nu'\nu},$$  \hspace{1cm} \text{(2.56)}

and can ignore the off-diagonal matrix elements, as they on the order of $\Gamma'/\omega_{vib} \ll 1$.

One can make an additional justification for the secular approximation in steady-state via an argument based on symmetry. The fact that $\sigma_{sp}$ contains all of the same symmetries as $\hat{H}$ (e.g., both are symmetric under translations by $\lambda/2$, rotations of $\pi$ about the $z$-axis, and transformation of $m_g = \pm 1/2 \rightarrow m_g \mp 1/2$ with a concomitant translation of $\lambda/4$) implies that no coherences will exist between states with different $\nu$, $q$, and $m_g$. Thus, one must only solve for the populations $\Pi_{\nu,q,m_g} \equiv \langle v, q, m_g | \sigma | v, q, m_g \rangle$.

Using the secular approximation in conjunction with Eq. 2.39 one obtains the simple rate equations

$$\dot{\Pi}_{\nu,q,m_g} = -\gamma_{\nu,q,m_g} \Pi_{\nu,q,m_g} + \sum_{\nu', q', m'_g \neq m_g} \gamma(\nu', q', m'_g \rightarrow v, q, m_g) \Pi_{\nu', q', m'_g},$$  \hspace{1cm} \text{(2.57)}

which describe the evolution of the population in state $|v, q, m_g \rangle$. The first term in Eq. 2.57 describes population leaving $|v, q, m_g \rangle$ at a rate

$$\gamma_{\nu,q,m_g} = \Gamma' \langle v, q, m_g | \hat{A} | v, q, m_g \rangle,$$  \hspace{1cm} \text{(2.58)}
and the second term corresponds to population arriving in $|v, q, m_g\rangle$ at a rate

$$\gamma(v', q', m_g' \rightarrow v, q, m_g) = \Gamma' \int_{-\hbar k}^{\hbar k} dp' \sum_m N_m(p') \left| \langle v', q', m_g' | B_m e^{ip'z/\hbar} | v, q, m_g \rangle \right|^2. \tag{2.59}$$

By introducing the closure relation $\sum_{v, q, m_g} |v, q, m_g\rangle \langle v, q, m_g|$, one can show that

$$\gamma_{v, q, m_g} = \sum_{v', q', m_g'} \gamma(v', q', m_g' \rightarrow v, q, m_g), \tag{2.60}$$

which provides a normalization condition for determining $\Pi_{v, q, m_g}$ and enforces conservation of population. In addition, one can prove that detailed balance holds [i.e., $\gamma(v', q', m_g' \rightarrow v, q, m_g) = \gamma(v, q, m_g \rightarrow v', q', m_g')$]. Thus, a steady-state exists for Eq. 2.57. While the time it takes to reach steady-state scales as $1/\Gamma'$, Eq. 2.57 shows that the steady-state populations depend on the well depth $U_0$ alone (i.e., they are independent of $\Gamma'$).

The relaxation terms described in Eq. 2.57 correspond to two main processes. First, the atom undergoes transitions between different spin states at a rate $\gamma(m_g \rightarrow -m_g) = \sum_{v', q'} \gamma(v', q', m_g \rightarrow v, q, -m_g)$. These rates vary with position and are equal to $\Gamma' \cos^2(kz)$ $[\Gamma' \sin^2(kz)]$ for transitions from $|+\rangle \langle -|$ to $|-\rangle \langle +|$. Thus, this spatially-dependent optical pumping corresponds to the mechanism described qualitatively in Sec. 2.3.2. This process is counterbalanced by heating due to transitions between atoms in the same spin state but with different values of $v$ and $q$ due to the random emission of fluorescent photons. Thus, one obtains a nonzero steady-state temperature.
2.4.3 Atomic phase space distribution

Having solved the problem at hand, I now discuss the properties of the resulting solutions. I first consider the steady-state situation, where I solve Eq. 2.57 with $\dot{\Pi}_{\nu,q,m_g} = 0$. Figure 2.7 shows the steady-state distribution of population among the different bands as a function of the dimensionless well depth $U_0/E_r$, where $\Pi_{\nu} = \sum_{q,m_g} \Pi_{\nu,q,m_g}$. The populations increase initially with increasing well depth before decreasing slightly due to increased heating at larger well depths. The ground state population reaches a maximum value of $\Pi_0 = 0.33$ at $U_0/E_r \sim 60$, where approximately 80% of the total population is in a bound state.

The atomic density distribution $\eta(z)$ therefore becomes highly localized around the potential minima. Figure 2.8a shows sample density profiles for several well depths, where I normalize the density such that the average density $\bar{\eta} = \langle \eta(z) \rangle = 1$. Here $\langle A \rangle \equiv \langle 2/\lambda \rangle \int_{-\lambda/4}^{\lambda/4} A \, dz$ corresponds to an average over a lattice period. For sufficiently deep wells, $\eta$ becomes nonsinusoidal and the contrast scales as $C = \max(\eta)/\min(\eta) \simeq (U_0/10E_r)^{1/4}$ [64]. One can alternatively quantify the atomic density modulation in
The value of $|b|$ is small for shallow well depths where the density distribution is nearly uniform, but increases rapidly in the vicinity of $U_0/E_r \sim 50$ and eventually levels off at $|b| \cong 0.61$ at large well depths. As I discuss later, this sub-wavelength confinement of the atoms can give rise to efficient Bragg scattering via the modulation of the index of refraction of the vapor.

One can also calculate the spread of the atomic momentum distribution. Figure 2.9 shows the dependence of the steady-state kinetic energy of the atomic ensemble on the well depth. Here I use two different measures of the average kinetic energy: $E_{rms} = p_{rms}^2 / 2m$ is calculated using rms momentum spread $p_{rms}$ and $E_{HWMeM} = \delta p^2 / 2m$ is based on the half-width at $1/\sqrt{e}$ maximum (HWMeM) momentum spread $\delta p$. Both measures of the kinetic energy decrease initially, reach a minimum value on the order of $10E_r$, then increase linearly for $U_0/E_r \gtrsim 100$. The well depth at which $E_{rms}$
Figure 2.9: Non-Gaussian nature of the atomic momentum distribution. a) Average kinetic energies $E_{\text{rms}}$ and $E_{\text{FWeM}}$ as a function of well depth. $E_{\text{rms}}$ displays a minimum value at the point of décrochage, which occurs here at $U_0/E_r = 95$. b1), b2) Momentum distribution for $U_0/E_r = 30$ and 140, respectively. The momentum is given in terms of the recoil momentum $p_r = \hbar k$ associated with a photon. The solid line is the distribution calculated via the band model, and the dashed line is a double-Gaussian fit given by Eq. 2.62.

attains its minimum value is commonly referred to as the point of décrochage (roughly translated from French as “taking off”), because the average kinetic energy increases rapidly for shallower wells. Roughly speaking, it corresponds to the point at which Sisyphus cooling most efficiently balances diffusive heating due to atomic recoil. The point of décrochage has been shown experimentally to occur for a fixed lattice beam intensity $I_d$ and is independent of $\Delta [\text{? }];$ this is somewhat surprising because it means that the point of décrochage does not correspond to a fixed well depth ($U_0 \sim I_p/\Delta$) or scattering rate ($\Gamma' \sim I_p/\Delta^2$), which are the central parameters in models of Sisyphus cooling. While no theoretical models to date have reproduced this observation (including the band theory model described here), I treat the independence of $I_d$ on $\Delta$ as an empirical fact. As I discuss later in this thesis, the independence of $I_d$ on $\Delta$ plays an important role in realizing large atom-photon coupling strengths.

Another important observation from Fig. 2.9a is that $E_{\text{rms}} \neq E_{\text{FWeM}}$, whereas
for a Gaussian distribution. In contrast to the initial theoretical work on Sisyphus cooling that predicts a Gaussian distribution [61], a more complete treatment predicts a non-Gaussian momentum distribution [68, 69]. While the deviation from a Gaussian is small for lattice intensities \( I_p \gg I_d \) (where \( E_{\text{rms}} \approx E_{H/W/eM} \)), it is quite pronounced in the region around \( I_d \). Figure 2.9b shows explicitly the momentum distribution \( f(p) \) for two different well depths. In general, the momentum distribution has a narrow central peak sitting on top of a broad base. As expected, the distribution appears nearly Gaussian for deep wells but is clearly non-Gaussian for shallow wells near \( I_d \).

There has been considerable discussion concerning the mechanism responsible for this non-thermal momentum distribution. One explanation involves extending the original Sisyphus theory based on a linear damping force [61] to include a maximum velocity beyond which Sisyphus cooling breaks down [69]. In this approach, one solves a Fokker-Planck-type equation with a velocity-dependent friction force and diffusion coefficient. The resulting momentum distribution is described by the so-called Tsallis function [70], which includes a broad class of functions, including a Gaussian, Lorentzian, and inverted parabola. Atoms in this model experience anomalous diffusion in which they undergo Levy flights (i.e., alternating periods involving short and long trajectories). While this approach has been studied in a variety of different contexts and presents a possible explanation for the resulting momentum distribution, the forms of the diffusion and friction terms are arrived at in a somewhat ad hoc manner.

In 2004, Jersblad et al. [71] proposed an alternative and strikingly-simple interpretation based on a bi-modal distribution. By conducting both a careful experimental and theoretical study of Sisyphus cooling around \( I_d \), they found that the momentum distribution is well-described by a double-Gaussian function. The atomic ensemble therefore consists of two distinct components: a cold component composed of atoms
bound in the potential wells, and a hot component consisting of nearly free atoms. The narrow (broad) Gaussian corresponds to the cold (hot) component with temperature $T_c$ ($T_h$). Cooling in this context occurs via the transfer of atoms from the hot to the cold component, where the fraction of atoms in each component is $f_{h,c}$, respectively \[72\]. Interestingly, the phenomenon of décrochage, as defined in terms of a sharp increase in the width of the momentum distribution, is absent in this model because the transfer between the hot and cold fraction occurs continuously. Instead, the lattice beam intensity normally associated with décrochage corresponds to the point at which most of the atoms occupy the cold component.

The theoretical studies focusing on this bi-modal interpretation \[71\] and \[72\] employed a semi-classical Monte-Carlo approach to Sisyphus cooling that included the full atomic level structure (e.g., a $J_g = 4 \rightarrow J_e = 5$ transition for Cs) and allowed for coherences between the atomic levels. While I do not expect the results of the band model described above to afford the same degree of quantitative agreement with experimental results, I do find that the momentum distribution obtained via the band model is relatively well-fit by the double-Gaussian function (see Fig. 2.9b)

$$f(p) = f_c g(T_c) + f_h g(T_h),$$

(2.62)

where $g(T) \equiv (2\pi mk_B T)^{-1/2} \exp(-p^2/2mk_B T)$ is a normalized Gaussian function and $f_h = 1 - f_c$. Using these fits, I determine $T_{c,h}$ as a function of the lattice beam intensity $I_p$ (see Fig. 2.10a). While $T_c \sim 10T_r$ and remains relatively constant in the vicinity of $I_d$, $T_c \sim 100T_r$ and increases rapidly with increasing well depth. For $I_p \ll I_d$, the momentum distribution of the hot component becomes non-normalizable. This explains why $E_{HWem}$, which is primarily sensitive to $T_c$, continues to decrease below $I_d$ whereas $E_{rms}$, which is sensitive to both $T_c$ and $T_h$, increases sharply.

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Figure 2.10: Theoretical scaling of hot and cold components. a) Temperatures of the hot and cold components. I exclude points for which $f_{c,h} \sim 0$ because it is not possible to achieve an accurate fit at these values. b) Fraction of the atoms in the hot and cold components. The data (points) are well-fit by tanh($U_0/45E_r$) (solid line).

I also determine the fraction of atoms in the cold and hot components from the fit obtained from Eq. 2.62. As shown in Fig. 2.10b, $f_c$ initially increases linearly with $I_p$ before beginning to roll over around $I_d$. Approximately 90% of the atoms are in the cold fraction by the décrochage intensity; by $I_p = 2I_d$, almost no atoms remain in the hot component. While no analytic solution for the dependence of $f_c$ on the lattice intensity exists, I find that the dependence is well-fit by the functional form

$$f_c(I_p) \approx \tanh(I_p/I_c),$$

where $I_c$ is the characteristic intensity for the cooling process and defines the rollover point. For the band theory model, I find that the $I_c$ corresponds to a well depth of $U_0/E_r = 45$. The characteristic intensity is intimately related to $I_d$ since, in the bimodal interpretation, both are related to the transfer of atoms from the hot to the cold component. Thus, while their numerical values differ on account of the particular functional form chosen in Eq. 2.63 (with $I_c \sim I_d/2$), $I_c$ shares $I_d$’s independence on the detuning of the lattice fields.
Figure 2.11: Calculated temporal evolution of the atomic density. a) Spatial dependence of $\eta$ as a function of time. The different curves represent the density at $t = 8$, 225, 750 and 1500 $\mu$s, where the arrow indicates the direction in which time increases. b) Temporal dependence of the atomic bunching. The points represent the band theory calculations, and the solid line is an exponential fit $0.46[1 - \exp(-t/143)]$, where $t$ has units of $\mu$s. For both figures, $U_0/E_r = 100$ and $\Delta/\Gamma = 5$.

In order to get a sense of the time scale over which atomic cooling and localization occurs, I consider the time-dependent solution of Eq. 2.57. Figure 2.11a shows the temporal evolution of the atomic density for an initially uniformly-distributed atomic ensemble at a temperature of $T = 30$ $\mu$K and for a well of depth $U_0/E_r = 100$. For these same initial conditions, I calculate $b(t)$ and find a loading time of 143 $\mu$s and a peak bunching of $b = 0.46$ (see Figure 2.11b). In general, I find that the growth rate is typically on the order of 10-1000 $\mu$s for $\Gamma' \sim 1 - 10 \mu$s, implying that the process of cooling and localization occurs quite efficiently (i.e., requires the scattering of only 10's of photons).

2.5 Summary

In this chapter, I have described some of the basic mechanisms that arise when light and matter interact. I looked first at the response of the internal states of a two-level atom when subjected to an externally-applied optical field and derived an expression
for the resulting atomic polarization. I then showed that this polarization gives rise to mechanical forces that act on the center-of-mass of the atom whenever the optical field involves a position-dependent phase or amplitude. Two main types of forces arise: a dissipative radiation pressure force and a conservative dipole force. The dipole force corresponds physically to a spatially-varying light shift that can result in a modulation of the atomic density distribution, such as in the case of the sub-wavelength confinement of atoms in an optical lattice.

I then considered the motion of multilevel atoms in an optical lattice and showed that new, sub-Doppler cooling mechanisms arise. In particular, I focused on polarization gradient cooling of atoms with a $J_g = 1/2 \rightarrow J_e = 3/2$ level scheme in a lin⊥lin lattice. To describe the quantum motion of atoms in such an optical lattice, I solved the generalized Bloch equations by using the band method in the secular regime. I found that the steady-state degree of atomic bunching becomes substantial and that the atomic ensemble transforms into a non-equilibrium system consisting of distinct hot and cold modes, where cooling corresponds to the transfer of atoms from the hot to the cold mode. Importantly, the characteristic intensity associated with this population transfer is independent of the detuning of the optical fields from resonance. Finally, I showed that the evolution of the system occurs on a typical time scale of several hundred $\mu$s, which implies that polarization gradient cooling occurs efficiently.

In the next Chapter, I discuss how the basic building blocks of the interaction of light with single atoms described here can give rise to interesting collective effects when photon-mediated interactions between the atoms occur.
Chapter 3

Collective light-matter interactions

In the previous chapter, I introduced several fundamental mechanisms that enable light to interact with an atom. I showed that light can affect both the internal and external (i.e., center-of-mass) dynamics of an atom, and leads to a broad range of possible phenomena. Nevertheless, each of the discussed mechanisms are single-atom effects in which atom-atom interactions play no role. In addition, I assumed that the resulting atomic dynamics arose solely through the atoms’ response to static, externally-imposed optical fields.

In this chapter, I consider the collective regime, wherein atomic couplings are crucial to the behavior of the light-matter system. In order to motivate my own research, I focus on the case of photon-mediated atomic interactions in which the light scattered by an atom at one location can influence the dynamics of another atom at a different location. In particular, I develop a basic theoretical framework for two key processes: mirrorless parametric self-oscillation (MPSO) and superradiance. While MPSO is typically discussed in terms of a NLO wave-mixing instability and superradiance is often considered as a many-body synchronization transition, I show that these two mechanisms are intimately related. After introducing both phenomena for the case of stationary atoms, I show that including atomic motion can lead to MPSO at a reduced pump intensities as well as an altogether new type of superradiance.
3.1 Introduction to MPSO

Since the invention of the laser and the subsequent demonstration of second harmonic generation (SHG), the field of nonlinear optics has played an important role both in fundamental science and technological applications. In general, nonlinear optics refers to the situation where the response of a material is not simply linearly proportional to the incident optical fields. This nonlinear response effectively enables optical fields to interact with one another via the nonlinear optical (NLO) medium, which can give rise to phenomena that are not possible in the linear regime. For example, in the case of SHG, two incident photons simultaneously drive a polarization such that a photon with twice the frequency is emitted. This type of process, in which new optical fields are generated, is an important result in NLO that is not possible in a linear optical material.

Another NLO process, known as parametric self-oscillation (PSO), occurs when a NLO material driven by optical fields gives rise to new, spontaneously-generated fields without changing the state of the system. In contrast to the conventional laser, which relies on stimulated emission, PSO involves a NLO wave mixing process to provide the gain required to amplify vacuum fluctuations to a macroscopic optical field. While one must place the NLO material in an optical resonator for the case of low gain, PSO can occur in free space for sufficiently large gains. I refer to this case as mirrorless parametric self-oscillation (MPSO).

As one approaches the threshold for PSO, the system transitions from a state in which the atoms scatter light individually to one where the radiation from different atoms is phase-locked and adds coherently. As I discuss in this section, the onset of PSO is accompanied by the formation of a spatial modulation of the NLO material's polarization (i.e., a grating forms), which leads to the emission of additional optical
fields through the scattering of the incident beams off this grating. Thus, PSO represents a type of collective scattering due to self-generated organization.

To describe quantitatively MPSO, I begin with Maxwell’s wave equation for the electric field \( \tilde{E} \):

\[
\nabla^2 \tilde{E} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \tilde{E} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2}{\partial t^2} \tilde{P}. 
\]

(3.1)

In general, one can write the polarization (i.e., dipole moment per unit volume) of the material as

\[
\tilde{P} = \varepsilon_0 \tilde{\chi}(\tilde{E}) \tilde{E},
\]

(3.2)

where the susceptibility \( \tilde{\chi}(\tilde{E}) \) is a tensor and can depend on the electric field. In many cases, one can expand the polarization in a power series of the form

\[
\tilde{P} = \varepsilon_0 \left[ \tilde{\chi}^{(1)} : \tilde{E} + \tilde{\chi}^{(2)} : \tilde{E} \tilde{E} + \tilde{\chi}^{(3)} : \tilde{E} \tilde{E} \tilde{E} + \ldots \right] = \tilde{P}^{(1)} + \tilde{P}^{(2)} + \tilde{P}^{(3)} + \ldots = \tilde{P}^{(1)} + \tilde{P}^{(NL)},
\]

(3.3)

where \( \tilde{\chi}^{(n)} \) and \( \tilde{P}^{(n)} \) represent the n-th order susceptibility (which is independent of \( \tilde{E} \)) and polarization, respectively.

From the form of Eq. (3.2) one sees that the polarization acts as a source term that drives the wave equation. For a linear material (i.e., \( \tilde{P} = \tilde{P}^{(1)} \)), the polarization contains only the frequencies of the incident fields. In contrast, the higher-order dependence of the polarization on \( \tilde{E} \) for a NLO material allows for the generation of fields with properties distinct from the incident fields. In this way, the nonlinear polarization \( \tilde{P}^{NL} \) couples the involved optical fields via the material response. Thus, in order to predict the behavior of the coupled light-matter system, one must self-consistently determine both how the fields drive the material and how the material’s response affects the fields. In the case of MPSO, the polarization gives rise to a net flow of power
from the incident optical fields to certain electromagnetic vacuum modes. In order to describe how this can occur, I consider the case of a material with a nonzero value of $\chi^{(3)}$ in the following sections, which is the first order nonlinear response in isotropic media.

### 3.1.1 Wave mixing in a Kerr medium

To motivate and help develop a model for my own experimental results, I follow the approach taken by Refs. [73], [25], and [74], I consider in this section the interaction of monochromatic plane waves

$$\tilde{E} = E(t)e^{-i\omega t} + c.c. \quad (3.4)$$

in a lossless Kerr (i.e., $\chi^{(3)}$) material. Here I take

$$E = \sum_n \hat{e}_n E_n e^{ik_n r + i\delta_n t}, \quad (3.5)$$

where $\hat{e}_n$, $k_n$, $\delta_n$, and $E_n$ are the polarization direction, wave vector, detuning (relative to $\omega$), and complex amplitude of the n-th electric field. I look specifically at the case of near-degenerate interactions, where $\delta_n/\omega \ll 1$. From Eq. (3.3) one expects the polarization to include terms that vary temporally with frequencies near $\pm \omega$ and $\pm 3\omega$; nevertheless, the polarization only efficiently drives fields that oscillate with identical frequencies (i.e., energy conservation must hold). Thus, I make the so-called rotating wave approximation and average over a time long compared to $2\pi/\omega$. This selects out only the terms of the polarization with a frequency close to $\pm \omega$. To allow for temporal changes in the field, I define a slowly-varying amplitude $P$ such that $\tilde{P} = P(t)\exp(-i\omega t) + c.c.$.
If the material is isotropic (as in the case of a gas), one can write the component of $P$ along the $\hat{r}_i$-direction as $P_i = \sum_j \epsilon_0 \chi^{(eff)}_{ij} E_j$, where $\chi^{(eff)}$ is the effective, field-dependent susceptibility tensor $[25]$. Here I focus only on the nonlinear response of the material (i.e., ignore linear dispersion). In addition, I model the finite temporal response of the NLO medium by assuming that $\chi^{(eff)}$ obeys a diffusive Debye relaxation equation of the form $[25]$

$$
\tau \frac{d\chi^{(eff)}_{ij}}{dt} + \chi^{(eff)}_{ij} = \left( A - \frac{1}{2} B \right) \left[ (E \cdot E^*) \delta_{ij} + \frac{1}{2} B (E_i E^*_j + E^*_i E_j) \right] \tag{3.6}
$$

where $A$ and $B$ are real constants that depend on the particular physical processes responsible for the NLO response, $\tau$ is the material response time, and $\delta_{ij}$ is the Kronecker delta function. With the Kerr medium thus defined, I consider several different beam configurations that provide insight into the types of physical processes that occur as well as introduce some of the approximations and mathematical techniques used in the analysis of wave mixing phenomena.

I first analyze the case of degenerate, co-polarized, counterpropagating fields of the form $E = \hat{y} [E_1 \exp(ik_p z') + E_2 \exp(ik_p z')]$ interacting with a Kerr medium of length $L$ (see Fig 3.1 a). From Eq. 3.6 I find that the resulting steady-state susceptibility is

$$
\chi^{(eff)}_{yy} = (A + B/2) \left( |E_1|^2 + |E_2|^2 + E_1^* E_2 e^{i2k_p z'} + E_1^* E_2 e^{-i2k_p z'} \right) = \chi_0 + \chi_+ e^{i2k_p z'} + \chi_- e^{-i2k_p z'}. \tag{3.7}
$$

The interference of the pump fields gives rise to a spatially-varying modulation of the susceptibility with amplitudes $\chi_\pm$, which physically corresponds to a material grating in the internal atomic states. This grating acts to couple the involved fields through enabling one field to scatter off of it into the direction of the other field.
While Eq. 3.7 includes the effect of a finite material response via Eq. 3.6, it assumes that the polarized material is spatially stationary (i.e., does not move). By including the possibility that the polarized material can move, one finds that this motion washes out the grating and effectively reduces the NLO response. One can take this effect into account by replacing Eq. 3.7 with

$$\chi_{yy}^{(eff)} = \chi_0 + h(\chi_+ e^{i2k_pz'} + \chi_- e^{-i2k_pz'})$$, (3.8)

where $h \in [0, 1]$ represents the degree of grating washout due to motion of the excitation. For an atomic gas, thermal diffusive atomic motion plays an important role in determining the contrast of the generated grating. To determine roughly how big this effect is, one must consider the ratio $t_r = \tau / \tau_c$, where $\tau_c$ is the effective decoherence time of the sample (e.g., the time it takes for an atom to move a distance equal to the grating spacing for the case of thermal motion). For $t_r \ll 1$ ($t_r \gg 1$), motion has a small (large) negative impact on the NLO response. Because $\tau_c$ can be much longer in cold (slow-moving) atoms than warm (fast-moving) atoms, cold atoms present the possibility of realizing larger nonlinearities.
The resulting polarization \( P_y = \chi_{y} E_y \) includes terms with spatial frequencies \( \pm k_p \) and \( \pm 3k_p \). A given field is only driven efficiently by the components of the polarization that share the same spatial frequency. This condition, known as phase-matching, corresponds physically to the conservation of linear momentum between the fields. By considering only the terms in the polarization that are phase-matched to the incident fields, one finds that the coupled wave equations for \( E_{1,2} \) are

\[
\frac{\partial E_{1,2}}{\partial z'} = \frac{i k_p}{2\epsilon_0} (A + B/2)(|E_{1,2}|^2 + (1 + h)|E_{2,1}|^2)E_{1,2},
\]  

(3.9)

where I have made the slowly-varying amplitude approximation (i.e., assumed that \(|\partial^2 E / \partial t^2| \ll |\omega \partial E / \partial t| \) and \(|\partial^2 E / \partial z'^2| \ll |k_p \partial E / \partial z'| \)). The solution to Eq. (3.9) is

\[
E_1(z') = E_1(z' = 0)e^{i\beta(|E_1|^2 + (1 + h)|E_2|^2)z'}
\]

\[
E_2(z') = E_2(z' = L)e^{-i\beta(|E_2|^2 + (1 + h)|E_1|^2)z'},
\]  

(3.10)

where \( \beta = k(A + B/2)/2\epsilon_0 \). While no energy is transferred between the fields (i.e., \( I_{1,2} \) does not depend on position), each field experiences a nonlinear phase shift \( \phi_{1,2}^{NL} = \beta(|E_{1,2}|^2 + (1 + h)|E_{2,1}|^2)L \) after propagating across the medium.

### 3.1.2 MPSO in the phase conjugate geometry

I next consider the case where two pairs of co-polarized counterpropagating fields interact with a Kerr medium (see Fig. Fig 3.1 b). I write the total field as

\[
E = \tilde{y} \left( E_1 e^{i k_p \cdot r} + E_2 e^{-i k_p \cdot r} + E_3 e^{i(k_s \cdot r - \delta t)} + E_4 e^{-i(k_s \cdot r - \delta t)} \right),
\]  

(3.11)
Figure 3.2: FWM phase matching via gratings. a) The wave vector associated with the grating formed by a nearly-counterpropagating pump and probe beam [along \(- (k_1 + k_s)\)] is phase-matched for backscattering of a pump photon into the probe direction. b) Here, I show how $E_2$ amplifies $E_s$ by scattering off the grating formed by the interference of $E_1$ and $E_i$.

where $k_p = k_p \hat{z}'$, $k_s = k_s \hat{z}$, and I take $|k_s| = |k_p| = k$ for $\delta/\omega \ll 1$. I refer to $E_{1,2}$ as pump beams and $E_{s,i}$ (i.e., the signal and idler beams, respectively) as probe beams.

I proceed as above in the case of only two beams by first substituting the fields into Eq. 3.6 and solving for the steady-state susceptibility. In the limit that $|E_{s,i}| \ll |E_{1,2}|$, I consider contributions only to first order in $E_{s,i}$. The interference of the fields gives rise to a susceptibility with a spatial modulation at frequencies 0, $2k_p$, and $k_p \pm k_s$. The grating along $k_p \pm k_s$ is phase-matched for coupling the pump and probe beams (see Fig. 3.2a), and therefore leads to gain or absorption.

I find that the solutions for $E_{1,2}$ given in Eq. 3.10 remain unchanged to first order in $E_{s,i}$. Therefore, the pump intensities remain constant throughout the medium. The coupled wave equations for the signal and idler fields become

$$\begin{align*}
\frac{\partial E_s}{\partial z} &= aE_s + bE_i^* \\
\frac{\partial E_i^*}{\partial z} &= aE_i^* + bE_s^*,
\end{align*}$$

(3.12)
where I have again included only phase-matched terms and assumed that $|E_1| = |E_2| = |E_p|^2$. Here $a = i\beta(1+h)|E_p|^2[1+i/(i+\delta \tau)]$ and $b = -2\beta h|E_p|^2/(i+\delta \tau)$. The first term on the right side of Eq. 3.12 corresponds to a modification of a probe beam via two-wave mixing with the pump beams. The second term corresponds to a cross-coupling, where a pump beam scatters into the probe beam direction via the grating formed by the counterpropagating pump and probe beams (see Fig. 3.2b). This situation, where the pump beams backscatter off the gratings, is referred to as backward four-wave mixing (FWM).

Solving Eq. 3.12 for the case where $E_s(z = 0) = E_s(0)$ and $E_i(z = L) = 0$, I find that the output fields are given by

$$E_s(L) = e^{a L} \sec(i b L)E_s(0)$$

$$E_i(0) = i \tan(i b L)E_s^*(0).$$

(3.13)

Because the idler field is simply proportional to the phase conjugate of the incident signal field, this setup is typically referred to as four-wave mixing in the phase conjugate geometry. Using Eq. 3.13 I define the normalized transmission as

$$T = \left| \frac{E_i(z)}{E_i(0)} \right|^2 = |\sec(i b L)|^2$$

(3.14)

and the reflectivity as

$$R = \left| \frac{E_i(z)}{E_i(0)} \right|^2 = |\tan(i b L)|^2.$$  

(3.15)

Figure 3.3 shows the resulting spectra, where the widths of the signals depends on the relaxation time $\tau$. The transmitted signal has a dispersion-shaped gain feature that is anti-symmetric with respect to $\delta = 0$, whereas the idler spectrum has a symmetric profile with a maximum amplitude at $\delta = 0$. 

66
Figure 3.3: FWM spectra. Dependence of a) $T$ and b) $R$ on the normalized two-photon detuning $\delta \tau$ for $h = 1$ and $\beta I_p L = 0.2$.

For the case where $\delta = 0$, Eqs. \[3.14\] and \[3.15\] reduce to the simple forms $T_0 = \sec^2(\phi^{NL})$ and $R_0 = \tan^2(\phi^{NL})$, respectively. Here, the nonlinear phase shift $\phi^{NL} = 2\beta h|E_p|^2 L$. Figure 3.4 shows $T_0$ and $R_0$ as a function of the nonlinear phase shift. The normalized transmission is always larger than the reflectivity, and both increase superlinearly with $\phi^{NL}$. When $\phi^{NL} = m\pi/2$ for integer values of $m$, one sees that both the $T$ and $R$ become infinite. In this case, the amplitudes are unstable in the sense that infinitesimal perturbations from $E_{s,i} = 0$ lead to a runaway growth of the fields. While infinite gain will never occur in real situations due to inevitable depletion of the pump beams, the situation of infinite gain effectively means that vacuum fluctuations can be amplified into macroscopic fields by the wave mixing process. This geometry therefore allows for MPSO; by simply shining sufficiently intense pump beams on the NLO medium, one produces new optical fields (e.g., the probe fields) without the need for an external cavity. Instead, the grating created by the interference of the beams leads to distributed feedback, wherein the growth of $E_s$ amplifies $E_i$ and vice versa.
Figure 3.4: Normalized output intensities for degenerate FWM. Dependence of $T_0$ (solid line) $R_0$ (dashed line) on $\phi^{NL}$ for $h = 1$.

3.1.3 MPSO via FWM in a two-level atom

I carried out the preceding analysis in a general manner that was independent of the physical mechanism responsible for the nonlinearity. In this section, I follow the approach developed by Abrams and Lind [75] to investigate the case of degenerate four-wave mixing via the electronic nonlinearity in a system of stationary two-level atoms. I begin with the susceptibility $\chi(E)$ given in Eq. 2.16 for an ensemble of non-interacting atoms, and take the electric field $E$ to have the form given in Eq. 3.12. Writing the total field in terms of separate contributions due to the pump ($E_0 = E_1 + E_2$) and probe ($\delta E = E_s + E_i$) fields and assuming that $E_0 \gg \delta E$, I perform a Taylor series expansion of $\chi(E)$ about $E_0$. The resulting polarization is

$$P = \epsilon_0 \chi_0 (E_0 + \delta E) - \frac{\epsilon_0 \chi_0}{|E_{sat}|^2} \frac{(E_0 \delta E^* + E_s^* \delta E)E_0}{1 + \tilde{\Delta}^2 + |E_0/E_{sat}|^2},$$

(3.16)

to first order in $\delta E$, where $|E_{sat}|^2 = I_{sat}(0)/2\epsilon_0c$, $\chi_0 = \chi(E_0)$, and $\tilde{\Delta} = (2\Delta/\Gamma)$. Substituting the polarization into Maxwell’s wave equation yields the coupled wave equations
for the probe fields

\[ \frac{\partial E_s}{\partial z} = \bar{a}E_s + \bar{b}^*E_i^* \]
\[ \frac{\partial E_i^*}{\partial z} = \bar{a}^*E_i^* + \bar{b}E_s^*, \]  

(3.17)

where I make the slowly varying amplitude and constant pump approximations. Here

\[ \bar{a} = \frac{a_0(i - \bar{\Delta})(1 + \bar{\Delta}^2)}{2 \left[ 1 + \bar{\Delta}^2 + (|E_1|^2 + |E_2|^2)/|E_{sat}|^2 \right]} \]
\[ = \bar{a}_R + i\bar{a}_I \]  

(3.18)

is the saturated absorption and dispersion for the probe fields (in the presence of the pump fields) and

\[ \bar{b}^* = \frac{-2i\bar{a}E_1E_2}{1 + \bar{\Delta}^2E_{sat}^2} \]  

(3.19)

is the nonlinear coupling coefficient.

Solving Eq. 3.17 and using the boundary conditions \( E_s(z = 0) = E_s(0) \) and \( E_i(L) = 0 \), I find that the normalized output probe intensities are

\[ T_0 = \frac{|\bar{b}\sin(wL)|^2}{|w\cos(wL) + \bar{a}_I\sin(wL)|^2} \]  

(3.20)
\[ R_0 = \frac{|w|^2}{|w\cos(wL) + \bar{a}_I\sin(wL)|^2} \]  

(3.21)

for \( w = (|\bar{b}|^2 - \bar{a}_I^2)^{1/2} \). For the case where \( \bar{a}_I = 0 \) (i.e., a lossless medium), Eq. 3.21 reduces to Eqs. 3.14 and 3.15 and one finds that the condition for MPSO is \( \phi^{NL} = \pi/2 \). In the presence of absorption (i.e., when \( \bar{a}_I > 0 \)), MPSO requires that \( |\bar{b}|^2 > \bar{a}_I^2 \). The oscillation condition in this case becomes \( \tan(wL) = -w/\bar{a}_I \), which implies that \( |\bar{b}|L > \pi/2 \) (see Fig. 3.5). Thus, loss increases the MPSO threshold.

The effects of atomic motion further decrease the NLO response for a given pump
Figure 3.5: Effect of absorption on FWM gain. Dependence of $R_0$ given in Eq. 3.21 for $\bar{a}_i L = 0$ (solid line) and $\bar{a}_i L = 1$ (dashed line).

Intensity, which effectively increases the value of $I_p$ required to achieve MPSO. For example, one finds that $R$ is reduced by roughly a factor $t_{r}^{2}$ \[75\]. Because typical values of the $t_{r}^{hot} \sim 100$ and $t_{r}^{cold} \sim 1$ for hot and cold vapors, $R_{cold}/R_{hot} \sim 10^4$ with all other parameters equal. In addition, studies have shown that the formation of density gratings, in addition to the previously-mentioned polarization gratings, can further enhance the NLO response of cold atoms over warm atoms \[9, 76\] and lead to lower thresholds for MPSO \[11\]. Thus, cold atoms provide the potential for achieving large nonlinearities at low pump powers. Unfortunately, it is often much easier to achieve larger values of $\eta$ and $L$ for warm than cold vapors, which has historically diminished the benefit of using cold atoms for realizing MPSO.

### 3.1.4 Experimental observations of MPSO

Mirrorless parametric self-oscillation has been observed in a variety of different systems over the last 30 years, including hot \[51, 77-83\] and cold \[10, 84\] atomic vapors, solid-state crystals \[85\], and silicon wave guides \[86\], for example. A longer review of previous studies can be found in Ref. \[87\]. Rather than attempting to compile a
comprehensive list, I instead focus on some of the general features and applications of MPSO in atomic vapors.

While the initial studies in atomic vapor cells required pump beam powers on the order of hundreds of milliwatts \[82, 88\], recent studies demonstrate that the threshold in warm atoms can be as low as several hundred microwatts by using improved cells and improved spatial beam profiles \[79\], coherent preparation techniques (e.g., EIT) \[80, 83\], or including an external cavity for the pump beams \[83\].

In a cloud of cold atoms, Schilke et al. \[10\] showed that MPSO can occur for pump beam powers of \(\sim 1\) mW, although this scheme requires additional lattice beams on the order of 1 W. In contrast, I show that MSPO can occur in my system for powers as low as 400 \(\mu\)W for the geometry shown in Ch. 6 and can be reduced to as low as a few \(\mu\)W by using a modified configuration \[84\].

The light generated via MPSO typically shows temporal correlations between the emitted beams \[81, 83\] and reduced quantum noise \[89\]. This makes these systems excellent sources for the generation of “twin beams” \[51, 90\] (i.e., pairs of beams that show entanglement and intensity correlations below the shot noise limit), which are useful for quantum imaging \[24\] and quantum information protocols \[91, 92\]. In addition, Dawes et al. \[27\] observed all-optical switching at low photon numbers using a scheme in which MPSO results in the emission of multiple beams of light in the plane transverse to the pump beams.

### 3.1.5 Quantum four-wave mixing

In the previous discussion, I showed that MPSO gives rise to new beams of light through a NLO process. In the classical description of this process, the amplitudes of the probe fields are initially identically zero. Nevertheless, the idea that one can
amplify nothing and get something is somewhat confusing (even if the amplification is infinite). In addition, I had to postulate the existence of the signal and idler modes in order for them to be included in the theory in the first place. One is actually amplifying vacuum fluctuations of the electromagnetic field, which are present for all possible modes. Thus, a proper treatment of MPSO requires a fully quantum mechanical description of the fields.

While I will not repeat the calculations presented above, I want to briefly discuss the case of quantized optical fields. In the dipole approximation, the part of the Hamiltonian that involves the interaction of light and matter is given by Eq. 2.2. I write the quantized electric field as

\[ \hat{E} = -i \sum_k \varepsilon_k [\hat{a}_k \exp(i\mathbf{k} \cdot \mathbf{r}) - \hat{a}_k^\dagger \exp(-i\mathbf{k} \cdot \mathbf{r})], \]

where \( \varepsilon_k = \sqrt{\hbar c k/2\varepsilon_0 V} \) is the electric field per photon and \( V \) is an arbitrary quantization volume (assumed to be much larger than the atomic sample). The annihilation and creation operators for the electric field with wave vector \( k \), given by \( \hat{a}_k \) and \( \hat{a}_k^\dagger \), respectively, satisfy the commutation relation

\[ [\hat{a}_k, \hat{a}_k^\dagger] = \delta_{k,k'}. \]

With these definitions, the term responsible for FWM is given as \( \hat{H}_{FWM} \propto \hat{P}^{(3)} \cdot \hat{E} \). For the case of degenerate FWM (DWFM) described in Sec. 3.1.3, the relevant terms of the Hamiltonian are of the form

\[ \hat{H}_{DFWM} \propto \sum_{j,k,l,m} \hat{a}_j^\dagger \hat{a}_k^\dagger \hat{a}_l \hat{a}_m. \]

Using Eq. 2.5 to describe the evolution of the fields and replacing the pump field operators by their mean values (i.e., treating them semi-classically), one finds that the coupled wave equations for the quantized probe fields are identical to Eq. 3.17, where \( E_{s,i} \rightarrow \hat{a}_{s,i} \). In the quantum picture, these equations describe the simultaneous destruction (i.e., absorption) of two photons and the creation (i.e., emission) of two photons. For example, the case where the signal beam is amplified via the scattering of a pump beam off the grating formed by the idler and counterpropagating pump
beam corresponds to the term \( \hat{a}_{k_j} \hat{a}_{k_i} \hat{a}_{-k_i} \hat{a}_{k_j} \). Terms of this form, where probe photons are created via the destruction of pump photons, show that power can flow from the pump to the probe beams in the absence of an initial population of probe photons. In addition, one can study higher-order statistical properties of the light, which is not possible in the fully classical picture. The quantum approach therefore naturally describes MPSO as a noise-initiated process that leads to the coherent scattering of pump light into the probe modes.

### 3.2 Superradiance

I now change gears and discuss a related phenomenon that also leads to the coherent amplification of noise via collective scattering. Consider first the case of a single excited emitter undergoing de-excitation, where the radiated electromagnetic field has an amplitude \( E_1 \), intensity \( I_1 \propto |E_1|^2 \), and occurs for a characteristic time \( \tau_1 \) [93]. In “ordinary” fluorescence, a system of \( N \) such emitters radiate independently of one another (i.e., the emitted fields have random phases \( \phi_n \)). This results in an ensemble decay time \( \tau_R = \tau_1 \), a peak intensity \( I_N \propto |\sum_n E_1 \exp(i\phi_n)|^2 = |\sqrt{N}E_1|^2 = NI_1 \), and an essentially isotropic radiation pattern (see Fig. 3.6a). The situation changes dramatically if one allows for photon-mediated interactions between the emitters. This can result in the spontaneous phase-locking of the ensemble, where the emitters instead radiate collectively. With fixed relative phases between the emitters (i.e., \( \phi_n = \phi_1 \)), the emitted intensity becomes \( I_N \propto |\sum_n E_1 \exp(i\phi_1)|^2 = |\sqrt{N}E_1|^2 = N^2 I_1 \). By conservation of energy, this stronger emission leads to a faster decay with duration \( \tau_R = \tau_1/N \). In addition, one typically finds that the radiation is highly directional, with a geometry defined by the spatial distribution of the emitters (see Fig. 3.6b). The case of accelerated, intense emission is referred to in general as either superradiance or super-
Figure 3.6: “Ordinary” and “super” radiance. Spatial radiation distribution and temporal decay profile for a) “ordinary” fluorescence and b) superradiance. The figure is adapted from [93].

fluorescence (depending on the initial conditions of the ensemble).

The transition from “ordinary” fluorescence to collective emission involves the onset of spontaneous global order in the system and corresponds to a quantum phase transition in the thermodynamics limit $N \to \infty$ [94]. Quantum fluctuations initiate the emission and give rise to atom-atom correlations through a spontaneous symmetry-breaking process. The observed macroscopic fields retain some information about the initial fluctuations, which makes superradiance an excellent candidate for studying quantum phenomena. In addition, the light-mediated interactions underlying superradiance lead to other interesting effects, such as quantum chaos [95] and entanglement [96].

In this section, I describe two examples of superradiance. I consider first the case of Dicke superradiance, in which radiative coupling between an ensemble of stationary two-level atoms leads to collective spontaneous emission at an accelerated rate [1]. I then discuss superradiant Rayleigh scattering (SRyS), in which correlated motion of
an atomic ensemble results in coherent optical scattering \[7]. This process is formally equivalent to Dicke superradiance, where the two internal levels in Dicke superradiance are replaced by two different atomic momentum states in SRyS. This matter-wave superradiance represents an example of new physics that can occur in cold atoms systems. In addition, both of these forms of superradiance are closely related to MPSO, which I will discuss in further detail in Sec. 3.3.

3.2.1 Collective spontaneous emission in a two-level atom

Dicke superradiance

In his original proposal [1], Dicke considers \(N \gg 1\) identical, stationary atoms located within a volume whose dimensions are small compared to an optical wavelength \(\lambda\). Each atom has a ground and excited state separated by an energy \(\hbar \omega_0\) and interacts only with an electric field with frequency \(\omega_k\) and wave vector \(k\). The Hamiltonian for the system is given by

\[
\hat{H} = \hbar \omega \hat{a}^\dagger \hat{a} + \hbar \omega_0 \hat{S}_z + \hbar \frac{g_{ap}}{\sqrt{N}} \left( \hat{a}^\dagger + \hat{a} \right) \left( \hat{S}_+ + \hat{S}_- \right),
\]

(3.23)

where \(g_{ap} = \mu \mathcal{E}/\hbar\) is the atom-photon coupling strength and \(\{\hat{a}^\dagger, \hat{a}\}\) are annihilation and creation operators for the optical field. The collective atomic operators \(\hat{S}_{\pm, z}\) are defined as

\[
\hat{S}_+ = \sum_i^N |e\rangle_i \langle g|_i, \quad \hat{S}_- = \sum_i^N |g\rangle_i \langle e|_i, \quad \text{and} \quad \hat{S}_z = \sum_i^N 1/2 \left( |e\rangle_i \langle e|_i - |g\rangle_i \langle g|_i \right).
\]

These operators satisfy the angular momentum commutation relations \([\hat{S}_+, \hat{S}_-] = 2\hat{S}_z\) and \([\hat{S}_\pm, \hat{S}_z] = \mp \hat{S}_\pm\).

It is therefore convenient to map the problem of the interaction of the \(N\) two-level atoms onto a pseudo-spin system of spin \(N/2\), where the ground and excited states correspond to the spin up and down directions in an abstract space. Using this ap-
proach, Dicke showed that the spontaneous emission rate for the ensemble becomes
\[ W_N = \Gamma(J + M)(J - M + 1), \]
where \( J \) is the total spin of the system and \(|M| \leq J\) is the projection of the spin along the quantization axis. Physically, \( J \) (also called the "co-operation number") is related to the number of interacting atoms and \( M \) corresponds to the population inversion of the system. Here \( \Gamma = 1/\tau_1 \) is the spontaneous emission lifetime of an atom.

For the case where the ensemble starts out in a many-body state that is symmetric with respect to permutation of the atoms (e.g., all atoms in the ground or excited state), the total spin is equal to the maximum possible value \( J = N/2 \) \[\text{[93]}\]. The quantities \( J \pm M \) correspond to the number of atoms in the excited and ground state \( N_{e,g} \), respectively. The energy decay rate (i.e., rate of photon emission) then becomes
\[ W_N = \Gamma N_e(N_g + 1). \] \[\text{(3.24)}\]

One therefore finds that the case where \( M = 0 \) (i.e., \( N_g = N_e \)) gives rise to a maximum decay rate that scales as \( W_N \propto \Gamma N^2 \). Because the rate at which photons are emitted by the ensemble is a factor of \( N \) larger than for independent atoms, this situation is known as superradiance. The physical origin of this collective behavior is the establishment of dipole-dipole correlations between the atoms, which phase-locks the emission from the atoms and creates a macroscopic polarization. This interpretation is emphasized by the fact that the rate of stimulated emission during superradiance remains equal to that of independent atoms.

The system’s transformation to the superradiant state corresponds to a quantum (i.e., zero-temperature), equilibrium phase transition in the thermodynamic limit \( N \to \infty \) \[\text{[94, 97, 98]}\]. The transition occurs when \( g_{ap} = \sqrt{\omega \omega_0}/2 \) and is second order, where both the macroscopic atomic polarization \( \langle S_+ + S_- \rangle \) and scattered field strength
(a) act as order parameters for the transition. This situation is directly analogous to the ferromagnetic phase transition in the mean-field Ising model \[99\]. More recently, it has been shown that first order, nonequilibrium quantum phase transitions can also occur \[100\] and are closely-related to the dynamical Casimir effect \[101\] in the case where \(g_{ap}\) or \(\omega_0\) are time-dependent.

Dicke points out two possible routes to superradiance. One can start with the atoms in the ground state (\(i.e., J = -M = N/2\)) and illuminate the ensemble with a pulse of radiation to excite it directly to the state where \(J = N/2\) and \(M = 0\). This coherently-prepared ensemble then begins to radiate immediately as a single dipole. This situation is completely analogous to the emission from a phased array of classical oscillators. Alternatively, one can prepare the ensemble such that all of the atoms are in the excited state (\(i.e., J = M = N/2\)) such that no macroscopic dipole moment exists initially. The coherently-prepared sample begins to radiate via spontaneous emission, which becomes accelerated as the many-body system decays into the superradiant state (see Fig. 3.7).

Arecchi and Courtens \[102\] extended the model for Dicke superradiance by considering the implications of the finite velocity of light on the system. They found that light from one atom must reach the other atoms before superradiance ends in order for it to participate in the collective emission. This means that, even for atoms within a volume of a single wavelength, the density can be sufficiently high that superradiance terminates in less than a single transit time of light across the sample \(\tau_t\). On the other hand, it is also possible for superradiance to occur in an extended sample (\(i.e.,\) with dimensions larger than \(\lambda\)) for sufficiently high densities. Eberly and Rehler \[103\] studied the case of superradiance in which atoms located in an extended sample with various geometrical shapes are coherently prepared in the excited state. They found that the temporal and spatial properties of the superradiant light depend on both the
number and distribution of atoms as \( N\mu' \), where \( \mu' \) depends purely on the geometry of the sample. For example, \( \mu' = 3\lambda/8L \) for a pencil-shaped distribution satisfying \( L \gg \lambda/2\pi \) and \( F < 1 \), where \( F = A/\lambda L \ll 1 \) is the Fresnel number and \( A \) is the cross-sectional area. The resulting time-dependent intensity of the emitted light has the form \( I(t) \propto (N\mu' + 1)^2 \text{sech}^2[(t - t_D)]/\tau'_R \), where the peak pulse delay is \( \tau_D = \tau'_R \ln(N\mu') \) and the pulse width is \( \tau'_R = \tau_1/(N\mu' + 1) \). For the case of the pencil-shaped geometry, the spatial distribution of the light is sharply concentrated along the sample’s long axis (i.e., the so-called “end-fire” modes).

**Superfluorescence**

In contrast to superradiance, where the sample is initially coherently prepared, Bonifacio and Lugiato [104] consider the case of an ensemble of atoms incoherently prepared in the excited state. In addition, they assume that no initial optical field or macroscopic polarization are present. They refer to this situation as superfluorescence because the
system begins to radiate spontaneously via “ordinary” fluorescence and, after a period of time, develops a macroscopic dipole moment. As in the case of superradiance, superfluorescence leads to accelerated, collective spontaneous emission with a peak intensity $I_N \propto N^2$ and a duration proportional to $1/N$. However, because stimulated emission cannot occur initially, this scenario requires quantum fluctuations to initiate the process. In contrast to the case of a coherently-prepared sample, this situation is therefore inherently quantum in nature. As the system evolves in time, the overlapping radiated fields effectively entrain the neighboring atoms after a time $t_D \sim \tau_R \ln(N)$ such that a macroscopic dipole moment develops. Due to the randomness of the quantum noise that seeds the process, the time delay and amplitude of the peak emitted intensity display random variations from one realization of the system to the next. Thus, the characteristics of the macroscopic field give insight into the nature of the underlying quantum fluctuations.

Using a quantum electrodynamic approach, Bonifacio and Lugliato found that the superfluorescent evolution of the light-matter system is described by the coupled equations:

\[
\begin{align*}
\frac{\partial p}{\partial t} &= Ew \\
\frac{\partial w}{\partial t} &= -Ep \\
\frac{\partial E}{\partial t} + c \frac{\partial E}{\partial z} &= \frac{1}{\tau_R}p,
\end{align*}
\]  
(3.25)

where $p$ and $w$ are the atomic polarization and inversion, respectively. For the case of emission into the end-fire modes of a pencil-shaped sample, Eq. [3.25] reduces to the problem of a damped pendulum. The pendulum, in this case, corresponds to a constant-length vector, known as the Bloch vector, evolving in an abstract space (see Fig. [3.8]) in which the $x$ and $y$ axes correspond to the real and imaginary parts of
Figure 3.8: Superradiance on the Bloch sphere. Bloch sphere representation of superradiance. The evolution of the atomic ensemble corresponds to motion of the Bloch vector $B$ along the sphere, where the radiated field depends on the rate of change of the azimuthal angle $\theta$.

The equation of motion describing the azimuthal angle of the Bloch vector $\theta$ is

$$\frac{\partial^2 \theta}{\partial t^2} + \kappa \frac{\partial \theta}{\partial t} - \frac{\sin(\theta)}{\tau_k^2} = 0,$$

(3.26)

where $\kappa = c/2L$ represents damping due to the loss of photons from the end of the sample. The radiated intensity is related to the Bloch angle as $I_N \propto \dot{\theta}^2$, where $\dot{A} \equiv \partial A/\partial t$.

Starting from an initially-inverted system, one might assume that the correct initial conditions leading to superfluorescence would be $\theta(t = 0) = \dot{\theta}(t = 0) = 0$. Because
Figure 3.9: Effect of damping rate on superfluorescence. Radiated intensity predicted from Eq. 3.26 for $\kappa \tau_R =$ a) 10, b) 0.4 and c) 0.

this corresponds to an unstable equilibrium point, though, the system starting in this state would not radiate. Instead, quantum noise due to the vacuum modes of the electromagnetic field lead to a non-zero initial tipping angle of $\theta(0) = \sqrt{2/N}$. Given this initial condition, the Bloch vector will eventually fall and, in the process, the sample will radiate away its stored energy.

Depending on the value of the quantity $\kappa \tau_R$, Eq. 3.26 describes qualitatively different radiation regimes. In the limit of large damping (where $\kappa \tau_R \gg 1$), one finds that a single burst of radiation of the form $I(t) \propto N^2 \text{sech}^2[(t - t_D)/\tau_R]$ (see Fig. 3.9 a). For the case where $\kappa \tau_R \sim 1$, the sample enters the regime of oscillatory superfluorescence, where it emits multiple pulses (known as ringing) with decreasing amplitudes (see Fig. 3.9 b). This ringing occurs because the emitted photons now have time to act back on the sample, leading to competition between collective spontaneous emission and stimulated scattering. In the extreme case where $\kappa = 0$, a train of identical pulses results, but the height of each pulse is only proportional to $N$ (see Fig. 3.9 c). Stimulated emission dominates the scattering process in this case, resulting in laser-like rather than superradiant operation.

Bonifacio and Lugiato also consider the effects of inhomogeneous broadening in their theory. For an inhomogeneous linewidth $\gamma^* = 1/T_2^*$, the superfluorescent pulse
becomes temporally broadened and reduced in amplitude by a factor of \( \exp(-t/T_2^*) \).

In addition, they find that superfluorescence only occurs if \( \tau_R \ll T_2^* \). Physically, this means that collective emission must occur faster than dephasing for superfluorescence to occur.

Before proceeding, I make a quick note on semantics. While there have been many studies of collective spontaneous emission, almost all fall into the category of superfluorescence. Nevertheless, many authors do not make the distinction between superradiance and superfluorescence; instead, they simply refer to the phenomena as superradiance. The general disagreement within the scientific community as to the correct nomenclature is highlighted in the following excerpt from a conversation between scientists at the “Collective effects meeting” at Redstone Arsenal in 1976 [106]:

DeTemple: What used to be called a superradiant laser is now called a superfluorescent laser; can we come to some agreement on the words we use to describe these phenomena?

Bonifacio: The physics involved is different: one can distinguish superfluorescence from superradiance by their second moments.

Eberly: The physics is different? The distinction should be experimentally feasible.

Feld: Let me find a place where we agree.

Bonifacio: Nothing.

The result of this confusion is that, even today, many cases of superfluorescence are erroneously called superradiance. In the interest of clarity and consistency, I use whatever terminology that the authors of the papers that I discuss below use, but I also point out whether a given effect is truly superradiance or superfluorescence.
Propagation effects

To go beyond the mean-field models, MacGillivray and Feld [107] considered the case of an optically thick, extended sample, where propagation effects cannot be ignored. Using a semiclassical approach in which the tipping angle was used as a free parameter, they predicted temporal ringing to occur for all sample lengths. While this agrees with the mean-field theories when the $\tau_t < \tau_R$, these theories predict the emission of a single pulse of radiation in the case where $\tau_t > \tau_R$. In the former case, ringing is due to stimulated absorption and reemission of light between different parts of the sample. In the latter case, nonuniformities in the sample (which are ignored in the mean-field theory) lead to ringing.

By taking a fully quantum mechanical approach and including propagation effects, Polder et al. [108] found good agreement with the semiclassical results. The advantages of this approach, though, are that one can predict from first principles the initial tipping angle of the Bloch vector as well as the statistical properties of the emitted light. One finds that characteristics of the initial quantum fluctuations show up directly as easily-measurable properties of the macroscopic superradiant pulse. For example, the first passage time statistics (i.e., the time at which the emitted intensity first reaches a set value [109, 110]) and the second-order field correlations [111] demonstrate the macroscopic effect of microscopic fluctuations.

Steady-state superradiance

Up to this point, the discussion of superradiance has been limited to the transient regime: atoms initially prepared in the excited state eventually decay to the ground state. While some ringing may occur, the collective emission ends once the system radiates away all of the initial, stored energy. While one could continuously replenish
the excited state population by repumping the system, superradiance typically occurs so rapidly that this is not possible. Recently, Meiser and Holland [3] predicted that one could realize steady-state superradiance (or, more accurately, superfluorescence) using ultracold alkaline-earth-metal atoms placed in a high-finesse cavity. These atoms have long-lived excited states that allow a population inversion to build up more rapidly than collective emission depletes it. In addition, coupling to the cavity mode effectively increases the superradiant decay rate by a factor of $C = g_{ap}^2 / \Gamma \kappa_c$, where $\kappa_c$ is the cavity decay rate. For high-finesse cavities, $\kappa_c$ can become very small, leading to a large enhancement of the light-matter coupling strength. Finally, using ultracold atoms reduces the effective dephasing rate, which allows for prolonged superradiance.

Using this scheme, they predict that one can produce a superradiant laser with a millihertz linewidth, which could improve the stability of optical clocks by two orders of magnitude [112]. By studying theoretically the intensity fluctuations of steady-state superradiance, they show that the generated light is second-order coherent in the limit of large $N$. In addition, they point out that the realization of such a system could play an important role in quantum information applications and the study of highly-correlated many-body physics. During the writing of this thesis, Bohnet et al. [113] demonstrated a superradiant laser with a linewidth of a few Hz, although emission from this laser persists for only a few tens of ms.

**Experimental observations of collective spontaneous emission**

The first observation of collective spontaneous emission was performed in 1973 using a long gas of HF molecules [114]. Since that time, many experimental studies have been carried out in a variety of different systems, including hot [115, 116] and cold atomic vapors, solid state crystals [117], quantum dots [118] and circuit QED systems [119]. The systems all demonstrate the characteristic features of superradiance and
gave results in quantitative agreement with theoretical predictions. Superradiance has been used in a variety of applications, such as for producing narrow bandwidth light sources [112], detecting impurities in the atmosphere at large distances [2], generating single photons on-demand [4, 120], and acting as a quantum memory [5]. I discuss additional, specific applications of superradiance as they pertain to my system in Ch. 6.

3.2.2 Collective emission via atomic recoil

Dicke’s original model for collective emission only considered the internal state dynamics of an ensemble of emitters. In this case, superradiance occurs due to the constructive interference of the probability amplitudes for spontaneous decay of multiple atoms. The ensemble then radiates light as a single ‘superatom’. Generalizing this concept beyond the case of spontaneous emission, one expects an identical phenomena to occur for any equivalent system that supports such interference. With the advent of laser cooling and trapping techniques, one can now control the external (i.e., center-of-mass) atomic states with the same precision as the internal states. By replacing the two electronic levels in Dicke’s model with two atomic momentum states, one finds a new and formally-equivalent type of superradiance called superradiant Rayleigh scattering (SRyS). When viewed in a position-space representation, the interference of the atomic momentum states corresponds to the formation of an atomic density grating. Thus, one can intuitively understand SRyS in terms of Bragg scattering, where the collective nature of the scattering is inherent in the ensemble’s capacity to scatter light as a single radiator (i.e., a grating).
Introduction to SRyS

In order to make clear the physical mechanisms involved, I look first at a a semi-classical description of SRyS. It is convenient to consider the simplest case of a pencil-shaped Bose-Einstein condensate (BEC) containing $N_0$ identical atoms. All atoms are initially stationary and in their electronic ground state. When one illuminates the BEC with a single pump beam with wave vector $k_p$, some of the atoms will absorb a photon and get a recoil kick $\hbar k_p$ (see Fig. 3.10 a). Upon decaying back down to the ground state, those atoms will emit a photon and receive another recoil kick. For emission with wave vector $k_s$, an atom accumulates a net change in momentum of $\hbar q = \hbar (k_p - k_s)$. The interference between these two momentum states results in a density grating with an amplitude of $\eta_q = 2\sqrt{N_0 N_q}/V$, where $N_q$ is the number of atoms scattering along $\hat{q}$ and $V$ is the volume of the condensate \cite{7}. The Bragg scattering efficiency is proportional to $\eta_q^2$, which results in a photon scattering rate $W_q \propto \gamma sc N_0 N_q$. This result is very similar to the superradiant scattering rate given in Eq. 3.24 where the excited and ground state populations are replaced by the number of stationary and recoiling atoms, respectively.

The connection between SRyS and superradiance can be made more explicitly by considering the problem quantum mechanically. To first order, one can describe the absorption and emission of a photon coinciding with an atom recoiling with momentum $\hbar \vec{q}$ in terms of an interaction Hamiltonian of the form

$$\hat{H}_{SRyS} \propto c_q^\dagger a_{k_s}^\dagger c_{0} a_{k_p},$$

(3.27)

where \{c_j^\dagger, c_j\} are creation and annihilation operators for atoms in the ground state with momentum $j$. It is worth noting that $\hat{H}_{SRyS}$ is formally identical to $\hat{H}_{DFWM}$ discussed in Sec. 3.1.5. I will discuss this correspondence in more detail in Sec. 3.3.
Using Fermi’s Golden Rule, one immediately finds that the scattering rate

$$W_q \propto \Gamma |\langle N_0 - 1, n_{k_p} - 1; N_q + 1, n_{k_s} + 1 | H_{\text{SRyS}} | N_0, n_{k_p}; N_q, n_{k_s} \rangle |^2$$

$$= \Gamma N_0 n_{k_p} (N_q + 1)(n_{k_s} + 1), \quad (3.28)$$

where $n_{k_i}$ and $n_{k_p}$ are the number of photons in the pump and superradiant modes. In the limit where $n_{k_i} \ll 1$, Eq. (3.28) reduces to $W_q \propto \gamma_{sc} N_0 (N_q + 1)$, which is identical to the scattering rate obtained for Dicke superradiance.

Because each photon scattered along $\hat{k}_j$ corresponds to an atom recoiling along $\hat{q}$, one finds that $\dot{N}_q = W_q$. For the case where $N_q \sim 0$, $\dot{N}_q \propto \gamma_{sc} N_0$ and one recovers standard Rayleigh scattering at a constant rate. When $N_q$ becomes non-negligible, the number of recoiling atoms increases exponentially as the grating depth and scattered optical field mutually amplify one another. Superradiant Rayleigh scattering in this picture therefore begins via normal Rayleigh scattering and evolves into a superradiant state after sufficient scattering has occurred. A more accurate name for this
process would therefore be superfluorescent Rayleigh scattering, but I continue with the common nomenclature nevertheless.

A formal correspondence between SRyS and Dicke superradiance can be made by using a dressed state approach. For the scenario described above, one can picture the relevant atomic structure in terms of a three-level atom consisting of a stationary electronic ground state, a second ground state with momentum $\hbar q$, and an intermediate electronic excited state (see Fig. 3.10b). The system is driven by the pump beam and scatters photons into the signal beam mode. By considering instead the condensate at rest to be dressed by the pump beam (i.e., viewing the stationary-atom-plus-pump-photon system as a single entity), one obtains an equivalent two-level atom (see Fig. 3.10c). In this picture, the new excited state consists of the stationary condensate and pump field, which spontaneously decays to the ground state by emitting a signal photon and a recoiling atom. The analysis of superradiance proceeds in a fashion identical to that of Dicke superradiance in this dressed picture, resulting in intense, accelerated emission. The main difference now is that both photons and atoms are coherently emitted during the superradiant decay.

**Experimental observation of SRyS**

Inouye et al. [7] performed the first SRyS experiment using far off-resonant pump light (i.e., $\Delta/\Gamma \sim -1000$) with intensity $I_p$ propagating along the direction perpendicular to the condensate’s long axis. For the case where the pump field polarization is orthogonal to the BEC’s long axis, they observed directly the emission of intense light along the endfire modes as well as the directional emission of recoiling atoms from the condensate. In this case, preferential initial dipole emission along the endfire modes seeds scattering, which leads to a macroscopic population of recoiling atoms that interferes with the stationary atoms with high contrast. They confirm their interpretation
of the superradiant mechanism by choosing instead a pump beam polarization along the condensate’s long axis. In this case, no scattering occurs along the endfire modes and they observe nearly isotropic emission of light and atoms corresponding to dipole emission. By monitoring the number of recoiling atoms as a function of time for both polarization cases, they further confirm the onset of superradiance by observing an accelerated rate of decay of atomic decay only for the case of orthogonal pump field polarization.

In addition to looking at the number of recoiling atoms, they also studied the intensity of the emitted light and found that its temporal profile agreed with that of Dicke superradiance. Specifically, they found that the light intensity initially grows exponentially, peaks at a time $\tau_p$ that depends on the pumping strength, and then decreases. In contrast, standard Rayleigh scattering would give a constant scattered intensity as a function of time. For stronger pumping, they observed temporal ringing, which stems from secondary recoil events in a manner analogous to propagation-induced ringing in standard superradiance in a dense medium [121]. They also studied the angular distribution of the generated light and showed that multiple, diffraction limited modes centered around the trap’s long axis fired simultaneously, which is consistent with predictions for the case where the Fresnel number $F > 1$ [122]. In addition, the distribution of the firing modes varied randomly from shot to shot, indicating the role of quantum fluctuations in seeding the process.

Repeating these measurements as a function of the pump intensity, Inouye et al. found a distinct threshold $I_{\text{thresh}}$ below which superradiance does not occur. They account for this threshold by modifying the growth rate equation to include a loss term as $\dot{N}_q = (\gamma_g - \gamma_f)N_q$, where $\gamma_g \propto \gamma_{sc}N_0$ and $\gamma_f$ are the gain and loss rates, respectively. Loss corresponds to the decoherence rate of the system, which occurs primarily due to dephasing of the matter wave grating via atomic motion. They find that the generated
light intensity scales linearly with the $I_p$ above the threshold before leveling off at larger pump intensities. Assuming that $\gamma_f$ is independent of $I_p$, they obtain an estimate for $L$ by extrapolating this curve to $\gamma_g = 0$ ($i.e.$, $I_p = 0$). The obtained value of $1/\gamma_f = 32 \mu s$ that agrees well with the width of the measured Bragg scattering signal of 5 kHz. To better study the system below $I_{thresh}$, they shine an additional, weak probe beam along the endfire mode, allow the system to reach steady-state, and then rapidly extinguish the pump beam [123]. As the density grating decays, the scattered probe power decreases, which allows for a direct measurement of $\gamma_f$. In agreement with their assumptions, they find that $\gamma_f$ is constant in this regime. This approach also allows them to show that the system transitions from non-collective to collective scattering as one tunes the pump intensity across the superradiant threshold.

While the initial observation of SRyS employed a BEC, neither quantum degeneracy [7] nor Bose statistics [124] are necessary conditions. For example, SRyS has been observed in an ultracold, thermal Bosonic vapor ($T < 3 \mu K)$ [16] and a degenerate Fermi gas [125] in free space. In these experiments, the scattered photons leave the interaction volume rapidly, which requires the atomic medium to effectively store the coherence. By placing the atoms instead in an optical cavity, the coherence is stored within the optical field and one can observe SRyS at temperatures of up to $T \sim 40 \mu K$ [43]. Thus, it is clear that SRyS simply requires that the system gain exceed the loss ($i.e.$, the decoherence time exceed the superradiant decay time) in order for the atoms to act in a collective manner.

**Semiclassical model for SRyS**

Many theoretical studies focusing on various aspects of SRyS have been carried out previously (e.g., Refs. [121, 122, 126, 127]). In this section, I follow the semiclassical treatment of Piovella et al. [128] because this it agrees quantitatively with the results
of Inouye et al.\[7\] and provides an intuitive explanation for the underlying physical mechanism, despite the fact that it cannot describe some of the inherently-quantum aspects of SRyS (e.g., the radiation statistics). In this model, one considers a pencil-shaped cloud of atoms with length $L$ and width $W$ oriented such that its long axis lies along $\hat{z}$ (see Fig. 3.11). The gas is illuminated by a pump field propagating along $\hat{y}$ and polarized in the $\hat{x}$-direction, which leads to the emission of superradiant light along $\hat{z}$. The total electric field in this case is

$$\tilde{E} = \hat{x} \left( E_p e^{i(k_y - \omega t)} + E_s(z, t) e^{i(k_z - \omega t)} + E_i(z, t) e^{-i(k_z + \omega t)} + c.c. \right), \quad (3.29)$$

where $E_{p,s,i}$ are the pump, signal, and idler fields and I assume that $E_0$ is real and constant. The resulting single atom polarization induced by these fields is $\tilde{\mathbf{p}} = \hat{x} \mu (\rho_{eg} + c.c.)$. Expanding the off-diagonal density matrix element in terms of its Fourier modes to first order, one finds that

$$\rho_{eg} = S_p(z, t) e^{i(k_y - \omega t)} + S_s(z, t) e^{i(k_z - \omega t)} + S_i(z, t) e^{-i(k_z + \omega t)}, \quad (3.30)$$

where $S_{p,s,i}$ are slowly varying atomic polarization amplitudes. For the case where all of the beam intensities are much smaller than the saturation intensity and $\Delta / \Gamma \gg 1$, one finds that $S_{p,s,i} \simeq \mu E_{p,s,i}/\hbar \Delta$. The total polarization is $\tilde{\mathbf{P}} = \eta(r) \mathbf{p}$, where $\eta(r)$ is the spatially-varying atomic density.

Substituting this polarization into Eq. 3.1, making the rotating wave and slowly varying amplitude approximations, and averaging over a grating period to select only the phase-matched terms, one finds that the signal and idler fields evolve according to

$$\left( \frac{\partial E_{s,i}}{\partial t} \pm c \frac{\partial E_{s,i}}{\partial z} \right) = \frac{i \omega \mu \eta}{2\epsilon_0} \left\{ S_p e^{i(k_y - \pi z)} + S_s + S_i e^{\pi 2ik_z} \right\}. \quad (3.31)$$
Here the angular brackets denote a spatial average over a grating period and $\eta \equiv \langle \eta(r) \rangle$ is the average density.

To proceed in a self-consistent way, one must next solve for the dependence of the atomic momentum distribution on the optical fields. Treating the atomic motion classically (i.e., assigning a specific position and momentum to each atom), the equations of motion for the atoms in the presence of the optical dipole force $F = (\vec{p} \cdot \nabla)\vec{E}$ (see Eq. 2.21) become

\[
\begin{align*}
    m \frac{d^2 y}{dt^2} &= -\frac{i k \mu^2 E_0}{\hbar \Delta} \left( E_1 e^{ik(z-y)} - E_2^* e^{ik(z+y)} - c.c. \right), \\
    m \frac{d^2 z}{dt^2} &= \frac{i k \mu^2 E_0}{\hbar \Delta} \left( E_1 e^{ik(z-y)} + E_2^* e^{ik(z+y)} - c.c. \right) + \frac{i 2k \mu^2}{\hbar \Delta} \left( E_1^* E_2 e^{2i k z} - c.c. \right),
\end{align*}
\]

(3.32)

where $m$ is the atomic mass. The interference of the pump and scattered fields therefore produce two orthogonal periodic potentials of the form $U_{s,i} \propto Re\{E_0 E_{s,i}^* \exp[i k(z \mp y)]\}$ in the $\{\hat{y}, \hat{z}\}$-plane, whereas the interference of the scattered fields creates an additional potential $U_{s,i} \propto Re[E_i^* \exp(i 2k z)]$. These potentials cause spatial bunching of the atoms with an identical periodicity, which phase matched various driving terms on the right hand side of Eq. 3.31. For $E_0 \gg E_{s,i}$, one can ignore $U_{s,i}$ and consider only...
Combining Eqs. 3.32 and 3.31, one finds that the coupled light-matter equations become

\[
\begin{align*}
\frac{d\theta_{s,i}}{dt} &= \frac{p_{s,i}}{m} \\
\frac{dp_{s,i}}{dt} &= \pm \frac{4k\mu^2}{\hbar\Delta} \text{Re} \left[ iE_p E_{s,i} e^{\pm i k \theta_{s,i}} \right] \\
\left( \frac{\partial E_{s,i}}{\partial t} \pm c \frac{\partial E_{s,i}}{\partial z} \right) &= \frac{i\omega \mu^2 \eta}{2\varepsilon_0 \hbar \Delta} \left( E_{s,i} + E_p b_{s,i} \right),
\end{align*}
\]

where \( \theta_{s,i} = z \mp y \), \( p_{s,i} = m \dot{\theta}_{s,i} \), and \( b_{s,i} = \langle \exp(\mp ik \theta_{s,i}) \rangle \) is the degree of atomic bunching along the phase-matched density grating. Equations 3.33-3.35 show the scattered fields drive the atomic motion while the atomic bunching leads to the amplification of the optical fields. In addition, one can see that the scattered fields are not coupled to one another in this geometry. It is worth noting that Eqs. 3.33-3.35 are identical to the equations of motion for the free electron laser (FEL) in the high gain regime [129], where the bunching of electrons is replaced by atomic bunching in SRyS.

Direct integration of the equations give rise to a pulse that is well-approximated by a hyperbolic secant function followed by ringing. One can make additional analytic progress by considering the mean field limit, where the propagation term in Eq. 3.35 describing the escape of photons from the interaction volume is replaced by a constant damping term. In this limit, Eq. 3.35 then becomes

\[
\frac{\partial E_{s,i}}{\partial t} = \frac{i\omega \mu^2 \eta}{2\varepsilon_0 \hbar \Delta} \left( E_{s,i} + E_p b_{s,i} \right) - \kappa E_{s,i},
\]

where \( \kappa = c/2L \). Performing a third-order analysis on the resulting equations of motion in the mean-field limit [?] for the case where the scattered field amplitudes are initially zero the atoms start out randomly distributed in space with zero momentum,
one finds that

\[ I_{s,i} \approx \left( \frac{\gamma N}{\Delta} \right)^2 I_p \text{sech}^2 \left[ (t - t_D)g \right], \quad (3.37) \]

where the initial exponential growth rate

\[ g = \frac{1}{\tau_R} = \frac{3\Gamma}{\Delta} \sqrt{\frac{8I_p N \lambda^2}{m \omega \pi W^2}} \quad (3.38) \]

and the time delay

\[ t_D = \ln(2N)/g. \quad (3.39) \]

The form of this solution is identical to that obtained for Dicke superradiance, and displays the characteristic dependence of the radiated intensity on \( N^2 \).

One can easily include the effects of Doppler broadening into the above approach. For a Gaussian spectral distribution with r.m.s. width \( \sigma = k \sqrt{k_B T/m} \), one finds that \( \sigma \tau_R \ll 1 \) in order for superradiance to occur. This is identical to the condition for superfluorescence in the presence of dephasing, where \( \sigma \) plays the role of the inhomogeneous decay time \( T_{2}^* \) described in Sec. 3.2.1. Physically, the momentum spread of the gas washes out the atomic density grating such that the bunching decreases as \( |b| \propto \exp(-t^2/\sigma^2) \). Numerical simulations show that this results in a peak amplitude of the superradiant pulse that decreases as \( I_{s,i}(t \equiv t_D) = I_{\text{peak}} \propto \exp(-\sigma^2 t_D^2) \) [129]. When the grating washes out before collective scattering can occur, one obtains the case of single-atom gain known as recoil-induced resonance [170].

It is also worthwhile to investigate the dependence of the dynamics on the damping rate \( \kappa \). In analogy with Dicke superradiance, one finds that qualitatively different behaviors result depending on the value of \( \kappa \tau_R \). For the case of free space operation studied above, where \( \kappa \tau_R \gg 1 \), one finds that superradiance occurs. In this regime, the superradiant growth rate increases with decreasing \( \kappa \). Thus, by placing the atoms in
a low-finesse cavity, one can overcome the effects of dephasing due to thermal motion and realize SRyS at higher temperatures. In the opposite limit, where $\kappa \tau_R \ll 1$ (e.g., for atoms placed in an optical cavity), the scaling of the output intensity with atom number changes to $N^{4/3}$ [43, 129]. Thus, superradiance is lost due to the narrow cavity linewidth. This regime is referred to as collective atomic recoil lasing (CARL) and operates as a combination of the standard laser and the free electron laser [12]. Nevertheless, the lasing remains a collective effect and has been shown to correspond to a synchronization transition that is formally equivalent to the Kuramoto model [37].

### 3.2.3 Alternative configurations

Several variations of the SRyS/CARL configuration have been studied recently that make possible novel phenomena. For example, Domokos and Ritsch [13] consider the case of atoms placed in a single-mode, high-finesse, standing-wave cavity that is pumped by a red-detuned, standing wave optical field in the direction perpendicular to the cavity axis (see Fig. 3.12 a). Above a threshold pumping power, the system undergoes a transition whereby the atoms self-organize into one of two checkerboard patterns. These patterns are shifted by $\lambda/2$ with respect to one another, and both maximize scattering of the pump fields into the cavity mode. This spontaneous self-organization corresponds to a quantum phase transition in the thermodynamic limit. The order parameters for the transition are the difference between the populations in the two possible checkerboard patterns (see Fig. 3.12 b), which represents the breaking of discrete translational symmetry, and the cavity mode field strength. The coherent scattered field that builds up in the cavity mode scales quadratically with the number of atoms, and is therefore superradiant.

This system was first experimentally realized by Black et al. [19] in a warm atomic
Figure 3.12: Alternative superradiant configurations. a) Schematic of the orthogonally-pumped cavity configuration proposed by Domokos and Ritsch \cite{130}. b) Diagram of the transition from a normal to superradiant phase as a function of the atom-photon coupling strength $g$. The order parameter is the difference in population between the two equivalent configurations \cite{46}. The figures are adapted from the respective publications.

By monitoring the light exiting the cavity, they observed random $\pi$ phase jumps, which they interpret as evidence for the spontaneous reorganization of the atomic structure from one of the allowed patterns to the other. In addition, they found that the collective scattering into the cavity mode leads to atomic cooling due to the irreversible loss of photons from the cavity. This is in contrast to typical cooling schemes in which spontaneous emission provides the dissipation channel.

Using instead Bose-condensed atoms, Baumann et al. \cite{14} observed directly the onset of atomic spatial organization by imaging the spatial density distribution of the BEC. In this case, the Hamiltonian describing the evolution of the system corresponds exactly to the Dicke Hamiltonian with a tunable atom-photon interaction strength \cite{14,21,45}. This system therefore provides a unique experimental platform for studying symmetry breaking in many-body quantum phase transitions \cite{46}.

Recent theoretical extensions of this configuration demonstrate that novel effects can occur when one allows for multiple spatial modes of the superradiant optical field \cite{15,20,47}. The crucial difference between the single- and multi-mode case is that...
both the shape and location of the resulting atomic organization are emergent in the latter. Such a self-consistently generated structure is referred to as a compliant or emergent lattice, and results from the breaking of a continuous (rather than discrete) translational symmetry. This situation therefore more closely models solid-state crystals (as opposed to optical crystals formed via externally-imposed lattices) and enables dislocations, glassiness, frustration, and supersolidity to occur [47].

3.3 MPSO versus superradiance

Traditionally, MPSO and superradiance have been discussed as separate phenomena by different communities. While superradiance is often analyzed in terms of phase transitions, synchronization, and collective scattering, MPSO is typically discussed in the context of NLO wave mixing instabilities and laser oscillation. One source of the artificial distinction between these two phenomena stems from the fact that MPSO typically involves steady-state driving by multiple optical fields, whereas superradiance usually evolves transiently from a specially-prepared initial state. Nevertheless, the physics behind MPSO and superradiance is identical: both systems involve an ensemble of interacting scatters that convert quantum fluctuations into coherent radiation via collective scattering.

In the preceding sections of this chapter, I have attempted to not only discuss both processes using similar language, but also to present the formal correspondence between them under certain circumstances. I do not want to belabor this point, but I explicitly emphasize here their similarities. For example, I showed in Sec. 3.1.2 that FWM can lead to MPSO beyond a critical pumping strength. Thus, the interpretation of SRyS as a FWM interaction between two optical and two matter fields leads directly to an alternative picture of the process in terms of MPSO. Along the same vein, Raymer
et al. showed that stimulated Raman scattering in a three-level atomic system is identical to superfluorescence in a two-level atom.

On the other hand, the collective nature of the scattering in MPSO is often overlooked. Although the system is typically driven by a coherent optical fields in MPSO, the atoms are not properly phased by these fields alone to radiate coherently into the initially-unoccupied optical modes. Oscillation therefore does not occur below a given driving pump power because these fields must first cause the atoms to be sufficiently coupled to one another. This is directly analogous to superradiance, which requires the atoms to interact strongly with the radiated fields of their neighbors, even for an initially coherently prepared ensemble cite [103]. Thus, just as in the case of superradiance, one can analyze MPSO in terms of a many-body phase transition of the system from a disorganized to an organized state.

The observation of this type of transition in a cloud of cold atoms driven by optical fields is at the heart of this dissertation. In the following chapters, I primarily discuss the light-matter interaction in terms of NLO wave mixing when the atoms are weakly driven by external pump and probe fields (i.e., below the transition point, see Chs. 4 and 5). Beyond the transition threshold, where I observe the generation of new, coherent optical fields (see Ch. 6), I compare my results with previous work under the guise of both MPSO and superradiance as appropriate.

### 3.4 Summary

In this chapter, I discussed previously-observed examples of photon-mediated atom-atom interactions that lead to collective scattering. After first introducing the notion that optical fields with different characteristics can couple to one another via the NLO polarization, I looked in detail at the case of FWM in a material with a third-order NLO
response. When a pair of counterpropagating strong pump and weak probe beams are incident on such a material, interference between the beams produces gratings that facilitate the transfer of energy between the beams. This results in an anti-symmetric and symmetric gain spectrum for the transmitted and reflected probe beams, respectively, as a function of the frequency difference between the pump and probe beams. For sufficiently strong driving in the presence of vacuum fluctuations, these gratings can form spontaneously and give rise to the coherent emission of new optical fields. When studied from this point of view, this phenomena is typically referred to as MPSO. I looked specifically at the case of an ensemble of two-level atoms and found that MPSO occurs when the nonlinear phase shift $\phi_{NL} = \pi/2$. Also, when all other parameters are equal, MPSO occurs at lower pump powers in cold than warm vapors.

I also reviewed the process of rapid emission from a system of interacting scatters, known as superradiance. I showed that superradiance leads to the directional emission of a bell-shaped pulse with a peak intensity and duration that scale as $N^2$ and $1/N$, respectively. The onset of superradiance coincides with the spontaneous breaking of a symmetry of the system’s Hamiltonian and represents a quantum phase transition. For the case when the ensemble is initially incoherently prepared (known as superfluorescence), the emitted intensity grows exponentially at first and peaks after a finite time delay $\tau_D \propto \ln(N)/N$. By measuring the statistical properties of the emitted light, one can study indirectly the quantum fluctuations that initiate the process. I then discussed a variety of different configurations in which superradiance arises, such as in free space or in a cavity, in warm or cold vapors, and via spontaneous emission in two level atoms or via recoil in cold atoms.

Regardless of the particular system studied, though, the process of collective amplification of quantum fluctuations by an ensemble of scatters is common to both superradiance and MPSO. Thus, one should consider them to be merely alternate descriptions
of the same phenomena. In the next chapters, I discuss the novel system that I developed that leads to a collective instability in a cloud of cold atoms. Specifically, I discuss my experimental characterization of the system’s NLO response below the instability threshold.
Chapter 4

Bunching-induced optical nonlinearity: experiment

In Ch. 3, I reviewed several relevant situations in which collective scattering arises in an ensemble of atoms. In addition, I showed that the manipulation of the spatial atomic ordering in a cold gas provides advantages over schemes involving only the atomic internal degrees of freedom. In this chapter, I discuss my experimental observation of a novel nonlinear wave mixing process that occurs in cold atoms. Like the previously-discussed schemes, atomic bunching in sub-wavelength gratings plays a crucial role in the nonlinearity that I describe; in contrast to those processes, which operate in the conservative regime, I employ a dissipative optical lattice to combine atomic bunching with Sisyphus cooling. I find that dissipation enhances the nonlinear response, resulting in an extraordinarily large nonlinearity at low light levels that displays unique scalings with material parameters. For sufficiently large nonlinear coupling strengths, the scattered optical fields act back on the atoms resulting in the light-mediated atom-atom interactions that give rise to collective behavior.

This chapter is based partly on Refs. [84] and [131].

4.1 Introduction

Optical instabilities, such as superradiance and mirrorless parametric self-oscillation (MPSO), involve the growth of initial fluctuations into a macroscopic signal. Such processes require a gain mechanism to overcome intrinsic system loss and produce net
amplification. As I discussed in Ch. 3, NLO wave-mixing processes can provide this gain. One can therefore gain insight into the mechanisms leading to the superradiant dynamics that occur above the transition threshold by studying the nonlinear response of the system below the instability threshold.

The NLO response of the system depends both on the magnitude of the susceptibility and the amplitude of the incident fields (see Eq. 3.3). By increasing the susceptibility, one can achieve a given NLO response with fewer input photons. One ultimate goal is to realize nonlinear interactions at the single-photon level, which is important for quantum information applications and lowers the operating power of NLO devices. Unfortunately, one typically requires intense optical fields to drive optical nonlinearities because most materials have a small NLO response. In addition, the magnitude of the NLO susceptibility typically decreases with increasing order (i.e., \( \chi^{(n+1)} \ll \chi^{(n)} \)). Thus, most low-light-level studies to date have focused on lower-order processes. For example, there have been numerous observations of low-light-level NLO interactions based on third-order (\( \chi^{(3)} \)) processes created via electromagnetically-induced transparency (EIT), where a strong coupling beam creates a quantum interference effect that simultaneously renders the medium nearly transparent while enhancing the nonlinearity. Other recent observations of strong third-order NLO effects include a two-photon absorptive switch actuated using < 20 photons interacting with atoms in a hollow-core photonic bandgap fiber and an optical pattern-based switch using ~600 photons interacting with a warm atomic vapor. In addition, the nonlinearity underlying all experimental realizations of SRyS and CARL to date rely on a \( \chi^{(3)} \) process.

Despite the success of these approaches, some applications require or can benefit from higher-order nonlinearities. One can use materials with a large, fifth-order (\( \chi^{(5)} \)) response in quantum information networks as new sources of correlated pulse...
pairs [132], quantum memories [132, 133], and for performing 3-bit quantum processing [134, 135], for example. By controlling the relative contributions of the $\chi^{(3)}$ and $\chi^{(5)}$ NLO response, one can create novel states of light such as liquid light condensates [136] as well as use the interference of the resulting four- and six-wave mixing signals for high-precision measurements and nonlinear spectroscopy [137]. Quintic media can also improve high-precision measurements [137] and reduce phase noise for enhanced interferometry performance [138], as well as give rise to new types of transverse optical patterns and multi-dimensional solitons [139]. Finally, combining higher-order wave mixing processes with SRyS can significantly enhance the generation of superradiant light [140]. Thus, the realization of efficient $\chi^{(5)}$ materials is important for fundamental studies in quantum nonlinear optics as well as improving the performance of NLO devices.

In this Chapter, I describe my discovery of a dissipation-enhanced NLO process in a cloud of cold atoms that gives rise to the largest fifth-order NLO susceptibility ever reported. At the same time, the system displays high transparency. In the presence of a dissipative optical lattice imposed by weak optical fields, the atoms simultaneously undergo Sisyphus cooling and load into the lattice. The resulting density grating enables efficient Bragg scattering. In contrast to previous studies of wave mixing via atomic bunching that produce a third-order NLO response [7, 9, 17], dissipative effects are crucial to the nonlinearity discussed here and cause the lowest-order nonlinearity to be fifth-order in the applied fields. Surprisingly, the achievable nonlinear interaction strength observed in our experiments from the $\chi^{(5)}$ response is just as large as that obtained in previous experiments dominated by a $\chi^{(3)}$ response. This strong light-matter coupling enables the scattered fields to act back on the atoms, resulting in greatly reduced group velocities, enhanced atomic coherence times, and optical instabilities.
the interaction of incident laser beams with cold atoms. I create the cold vapor using a highly anisotropic magneto-optical trap (MOT, described in Apps. [A] and [B]) to confine $^{87}$Rb atoms in the $5^2S_{1/2}(F = 2)$ state within a cylindrical region of length $L=3$ cm (along $\hat{z}$) and $1/e$ diameter $W=430$ $\mu$m (along $\hat{x}$ and $\hat{y}$) [36]. The resulting cloud has typical atomic temperatures $T = 30$ $\mu$K and densities $\eta = 2 \times 10^{10}$ cm$^{-3}$, which leads to a typical on-resonant optical depth of $OD = 20 - 30$ ($OD \sim 1$) along the $\hat{z}$- ($\hat{x}$- and $\hat{y}$-) direction(s).

I shine a pair of counterpropagating pump beams (wave vectors $\mathbf{k}_{p1} = k\hat{z}' = -\mathbf{k}_{p2}$, frequency $\omega_p$, and polarizations $\hat{e}_{p1,p2}$) on the vapor at an angle of $\theta = 10^\circ$ relative to the $\hat{z}$-axis. The pump beams are detuned from the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ atomic transition by $|\Delta| = 2 - 20\Gamma$, have intensities of $I_p = 0.1 - 30$ mW/cm$^2$, and diameters of 3 mm. In addition, I shine a weak signal beam along the trap’s longitudinal axis (i.e., $k_s = k\hat{z}$) with a polarization $\hat{e}_s$. This signal beam is detuned from the pump beams by $\delta = \omega_s - \omega_p$, has a diameter of 200 $\mu$m, and an intensity of $I_s = 1 - 100$ $\mu$W/cm$^2$ (although $I_s$ is typically $\mu$W/cm$^2$ unless otherwise specified). The wave-mixing interaction generates an idler beam that is counterpropagating with the

Figure 4.1: Pump-probe spectroscopy setup. a) Schematic of the experimental setup. b) Timing scheme used in the experiment.

4.2 Experimental setup

Figure 4.1a shows a schematic of the NLO wave-mixing experiment, which involves the interaction of incident laser beams with cold atoms. I create the cold vapor using a highly anisotropic magneto-optical trap (MOT, described in Apps. [A] and [B]) to confine $^{87}$Rb atoms in the $5^2S_{1/2}(F = 2)$ state within a cylindrical region of length $L=3$ cm (along $\hat{z}$) and $1/e$ diameter $W=430$ $\mu$m (along $\hat{x}$ and $\hat{y}$) [36]. The resulting cloud has typical atomic temperatures $T = 30$ $\mu$K and densities $\eta = 2 \times 10^{10}$ cm$^{-3}$, which leads to a typical on-resonant optical depth of $OD = 20 - 30$ ($OD \sim 1$) along the $\hat{z}$- ($\hat{x}$- and $\hat{y}$-) direction(s).

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signal beam (i.e., \( k_i = -k_s \)), has an intensity \( I_i \), and a frequency \( \omega_i = \omega_p - \delta \). I refer to the signal and idler beams collectively as probe beams, and take \( |k_p| = |k_s| = |k_i| \cong k \).

Also, because all intensities are typically below the off-resonant saturation intensity 
\[ I_{sat}(\Delta) = I_{sat}[1 + (2\Delta/\Gamma)^2] \] for \( I_{sat} = 1.6 \text{ mW/cm}^2 \), I work in the low saturation limit where \( s_\Delta \ll 1 \).

I control the polarization of each beam using separate wave plates and polarizers (see App. B). In this way, I can study the polarization dependence of the wave mixing process in order to better understand how the system transitions to the superradiant state. Table 4.1 shows the different polarizations that I use. Here I use a naming convention where \( \aleph \), \( \Sigma \), \( \Xi \), \( \Theta \) refer to the situation where the pump beams have linear orthogonal, linear parallel, circularly-counterrotating, and circularly co-rotating polarizations, respectively. The symbols \( \parallel \) and \( \perp \) indicate whether the signal beam has the same or orthogonal polarization as the nearly co-propagating pump beam, respectively.

In the experiment, I cycle between a cooling and trapping period and a wave mixing period (see Fig. 4.1b). First, I cool and trap the atoms for 99 ms with only the MOT beams on. I then turn off the MOT beams and conduct the wave mixing experiment during the remaining 1 ms, which consists of turning on the pump (signal) beams at

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**Table 4.1: Wave mixing polarization states.** Polarization configurations for the pump and probe beams.
time $t_p$ ($t_s$) for a duration $\Delta t_p$ ($\Delta t_s$). During this period, I measure the probe beam intensities with a pair of matched photomultiplier tubes (PMTs).

4.3 Pump-probe spectroscopy

One technique for studying a dynamical system is to observe how it responds when driven sinusoidally. In this section, I measure the spectrum of the vapor's nonlinear response by scanning the signal beam frequency around the pump beam frequency. I record the intensity of both the transmitted signal and reflected idler beams for different pump beam intensities ($I_p$), single photon detunings from resonance ($\Delta$), and polarization configurations, and briefly discuss the mechanisms responsible for the observed spectral features. From these spectra, I gain information about the resulting optical lattice, the atomic momentum distribution, and the gain mechanism responsible for the superradiant transition.

4.3.1 Dependence of gain spectrum on single-photon detuning

I first consider how the spectrum depends on the single-photon detuning of the beams from atomic resonance. Figures 4.2a1 and a2 show the normalized signal beam transmission

\[ T = \frac{I_s(L)}{I_s(0)} \]  

and reflectivity

\[ R = \frac{I_i(0)}{I_s(0)}, \]  

respectively, for the small detuning $\Delta/\Gamma = -2$. Here I take $z = 0$ ($z = L$) to be the location where the signal beam enters (exits) the atomic vapor. In both $T$ and $R$, I observe two distinct resonances: a broad (several MHz) feature near $\Delta = 0$ and
Figure 4.2: Role of wave mixing via electronic nonlinearity

(a1) (b1) Normalized transmission and (a2) (b2) reflectivity for $\Delta/\Gamma = -2$ ($\Delta/\Gamma = -6.5$) and $\alpha_0L = 10(\pm 1)$. For detunings in which the narrow resonance is large, the electronic nonlinearity is negligible.

a narrow (10’s of kHz) feature located around $\omega_s = \omega_p$ (i.e., $\delta = 0$). The broad feature is due to interactions of the optical fields with different electronic levels [i.e., the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ transition, as discussed in Ch. 3.1.1], whereas the narrow feature corresponds to wave mixing processes involving different Zeeman or center-of-mass states. While the amplitudes of the nonlinear responses are comparable for small $\Delta/\Gamma$, the narrow feature dominates the spectrum for larger detunings (see Fig. 4.2b, where $\Delta/\Gamma = -6.5$). As I will show, the components of this narrow feature that are due to transitions between different center-of-mass states are directly related to the system’s transition to superradiance. Therefore, I focus on this feature for the remainder of this thesis.
Figure 4.3: Red and blue spectra. a) Transmission spectrum for $\Delta/\Gamma = -10$ and $I_p = 2.5 \text{ mW/cm}^2$. b) Transmission spectrum for $\Delta/\Gamma = 10$ and $I_p = 26 \text{ mW/cm}^2$. Both spectra were obtained using the $\Theta||$ polarization configuration. Arrows indicate the Rayleigh and Raman transitions.

In addition to the magnitude of the detuning from resonance, one must also consider how the sign of $\Delta$ affects the nonlinearity. Figure 4.3 a (b) shows the narrow transmission gain feature in the $\Theta||$ polarization configuration for $\Delta/\Gamma = -10$ ($\Delta/\Gamma = 10$), where the pump beam intensity used in Fig. 4.3 b is approximately ten times larger than in Fig. 4.3 a. Nevertheless, both spectra show qualitatively similar features; namely, Raman resonances centered at a non-zero two-photon detuning of $\sim 100 \text{ kHz}$ and Rayleigh scattering resonances centered around $\delta = 0$. The Raman features involve processes in which the initial and final states of the system are different (in this case, different vibrational levels of atoms localized in the pump beam lattice), whereas the initial and final states are the same in the Rayleigh processes (see Fig. 2.2). The polarity of the Lorentzian-shaped Raman features is reversed for red ($\Delta < 0$) and blue ($\Delta > 0$) detunings, whereas the polarity of the dispersive-shaped Rayleigh feature is independent of $\Delta$ [172]. This central feature, which is the focus of Secs. 4.5 and 4.4, arises from Bragg scattering of the pump beams off the density grating formed via the interference of a pump and probe beam [55].

Despite the qualitative similarities between the spectra for red and blue detunings,
the amplitude of the signal for $\Delta < 0$ is approximately 100 times larger than that of the $\Delta > 0$ for comparable pump intensities. This is due to the qualitatively different behavior of atoms in red and blue optical lattices (as discussed in Ch. 2). While red lattices lead to a net cooling and atomic localization at the intensity anti-nodes, blue lattices result in atomic heating and localization in the intensity nodes. One therefore expects that a red lattice should give rise to a larger NLO response than a blue lattice for two main reasons. First, the atoms in the red lattice experience a larger average electric field strength, which more efficiently drives the nonlinearity. Second, the colder temperatures in the red lattice lead to an enhanced density grating contrast, which is important for the Bragg scattering process responsible for superradiance. Thus, I consider in detail only the cases where $\Delta < 0$ in the rest of this thesis.

### 4.3.2 Variation of gain spectrum on polarization

Multilevel atoms driven by optical fields exhibit responses that depend on the polarizations of the incident fields. In general, multiphoton processes involving transitions between Zeeman, vibrational, or center-of-mass states can lead to features with sub-MHz linewidths located around $\delta = 0$. Figures 4.4 and 4.4 show $T$ and $R$, respectively, for different polarization configurations and several values of $I_p$. I do not discuss the origin of each feature here; detailed analyses and discussions of the resonances that occur for various atomic structures and optical polarization configurations have been conducted elsewhere [? ? ? ]. Rather, I include each spectrum for completeness, but focus on the particular features that are directly relevant to characterizing the atomic distribution and the onset of superradiance.

In particular, while the spectra display narrow central features for all polarizations, only the $\mathcal{K}$, $\Sigma$ and $\Theta$ configurations give rise to large values of $T$ and $R$ in the immediate
Figure 4.4: Polarization-dependent transmission spectrum. Signal beam transmission for the a) $\Theta \parallel$, b) $\Xi \perp$, c) $\Xi \parallel$, d) $\Sigma \parallel$, e) $\Sigma \perp$, f) $\kappa \perp$, and g) $\kappa \parallel$ polarization configurations. In all figures, $\Delta / \Gamma = -7$, $\alpha_0 L = 14$, and the values of $I_p$ in mW/cm$^2$ used to obtain the spectra are shown. Note the change in span of the horizontal and vertical axes for different figures. h) Definitions for the different polarization configurations.
Figure 4.5: Polarization-dependent reflection spectrum. Reflectivity for the a) $\Theta\parallel$, b) $\Xi\perp$, c) $\Xi\parallel$, d) $\Sigma\parallel$, e) $\Sigma\perp$, f) $\kappa\parallel$, and g) $\kappa\perp$ polarization configuration. The experimental parameters are the same as in Fig. 4.4. Note the change in span of the horizontal and vertical axes for different figures. h) Definitions for the different polarization configurations.
vicinity of $\delta = 0$. One can most clearly see this by looking at the reflectivity, as shown in Fig. 4.5. The common feature of the polarization configurations that yield large nonlinear responses at degenerate two-photon detunings is that they all give rise to strong atomic localization in the lattice formed by the interference of the pump beams, which I refer to as the pump lattice. In addition, they all enable Sisyphus cooling to occur. In contrast, while Sisyphus cooling also occurs for the $\Xi$ configuration, this choice of polarization does not produce a pump lattice (i.e., no density grating forms due to the pump beams). This indicates the importance of atomic bunching in the pump lattice for realizing large nonlinearities.

I compare quantitatively the relative nonlinear response for the different polarization configurations by considering the values of $R_0 = R(\delta = 0)$ in each case (see Fig. 4.6). The $\mathcal{K}_\perp$ and $\Theta_\parallel$ configurations yield the largest nonlinearity (i.e., largest value of $R_0$ for smallest $I_p$). This is most likely because the atoms are optically pumped into the stretched states in the lattice anti-nodes, which maximizes the effective transition dipole moment. The NLO response for the $\mathcal{K}_\parallel$ and $\Sigma_\perp$ configurations are slightly smaller than in the previous cases, but are similar to one another because the spatially-varying optical field polarizations in both lattices are nearly identical. Finally, the $\Sigma_\parallel$ configuration leads to an even smaller NLO response, while the $\Xi_\perp$ and $\Xi_\parallel$ configurations require much larger pump intensities to produce even a weak idler signal. Therefore, in an effort to lower the superradiant threshold and realize the largest possible NLO response, one should choose either the $\mathcal{K}_\perp$ or $\Theta_\parallel$ configuration. I focus primarily on the $\mathcal{K}_\perp$ configuration because the theoretical model for this case has been well-established [? ? ] and is conceptually straightforward. In the following sections, the data corresponds to the $\mathcal{K}_\perp$ configuration, unless specified explicitly otherwise.
Figure 4.6: Variation of reflectivity with polarization. \( R_0 \) as a function of \( I_p \) for the \( \Theta\parallel \) (solid circle), \( \kappa\perp \) (open circle), \( \kappa\parallel \) (open square), \( \Sigma\parallel \) (up-pointing triangle), \( \Sigma\perp \) (down-pointing triangle), \( \Xi\perp \) (solid square), \( \Xi\parallel \) (diamond). For all data, \( \Delta/\Gamma = -7 \) and \( \alpha_0 L = 13.4(\pm 0.5) \). Note that both the vertical and horizontal axes are scaled logarithmically.

4.3.3 Dependence of vibrational resonance on pump beams

While the resonance centered around \( \delta = 0 \) is of particular interest because it leads to the superradiant transition, the vibrational Raman resonances give important information about the optical lattice. As discussed in Sec. 2.3.1, the locations of the Raman resonances correspond to the effective potential well depth \( U_0 \). Figure 4.7 shows the location of the vibrational resonance peaks (\( \omega_{\text{vib}} \)) as a function of \( I_p \) along with a fit from Eq. 2.34 for \( \Delta/\Gamma = -10 \). As expected, \( \omega_{\text{vib}} \propto \sqrt{U_0/E_r} \propto \sqrt{I_p} \). In this way, I measure the effective lattice depth for a given pump beam detuning and intensity. In addition, the widths of the resonances \( \Delta \omega_{\text{vib}} \) reveal information about the degree of atomic localization via the Lamb-Dicke effect (see Sec. 2.3.1). While the scattering rate \( \gamma_{sc} \sim 200 \text{ kHz} \) for the parameters used in Fig. 4.7, the resonance widths are on the order of 80-100 kHz. These widths are larger than the rough estimate \( \Delta \omega_{\text{vib}} = \gamma_{sc} |M|^2 \sim 10 \text{ kHz} \) made using Eq. 2.37, but the reduction nevertheless indicates that the atoms are well-localized in the pump lattice.
Figure 4.7: Location of vibrational Raman peaks. Experimental data (points) and theoretical predictions (via Eq. 2.34, solid line) showing the dependence of $\omega_{\text{vib}}$ on $I_p$ for $\Delta/\Gamma = -10$. The error bar represents typical measurement uncertainty.

4.3.4 Temperature measurement in an optical lattice

I now discuss the technique that I use to measure the temperature of the atoms loaded into the optical lattice. As discussed in Ch. 3, the width of the resonance centered about $\delta = 0$ is directly related to the ground state decay time. For my system, the main decay mechanism is due to atomic motion (i.e., Doppler dephasing), which means that the width of this resonance gives information about the atomic momentum distribution. For the $\mathbf{K}_\perp$ configuration, the central resonance is due almost entirely to the backscattering of a pump beam off the density grating formed by the interference of a pump and nearly counter-propagating probe beam [33]. This allows me to determine the effective “temperature” along the $\hat{z}$-direction according to [?]

$$T_{\text{rms}} \equiv \frac{E_{\text{rms}}}{k_B} = \frac{m\delta_{\text{rms}}^2}{4k_Bk^2\sin[(\pi - \theta)/2]^2},$$

(4.3)

where $\delta_{\text{rms}}$ is half the separation between the gain and absorption peaks of the central resonance and $\theta = 10^\circ$ (i.e., $\theta = 0.175$ rad). See App. B for more details on measuring
the atomic temperature..

I find that $T_{\text{rms}}$ decreases rapidly for small $I_p$ up to $I_p \sim 3 \text{ mW/cm}^2$ and then levels off roughly at $T_{\text{rms}} \cong 2 \mu\text{K}$ (see Fig. 4.8). This trend is consistent with the predictions for cooling in a lin⊥lin lattice in the vicinity of the décrochage intensity $I_d$. From this measurement, I find that $I_d \sim 3 \text{ mW/cm}^2$, which agrees with the value of $I_d$ observed in previous studies [??] using a 3D lin⊥lin lattice to within a factor of 4. I attribute this difference to the fact that I work with a 1D lattice [71] and to the different scattering properties of my anisotropic MOT compared to spherical MOTs [156]. This value of the décrochage intensity implies that the characteristic intensity $I_c \cong I_d/2 = 1.5 \text{ mW/cm}^2$, which is nearly equal to the on-resonant saturation intensity $I_s(0) = 1.6 \text{ mW/cm}^2$ for $^{87}\text{Rb}$. Unlike $I_{\text{sat}}(\Delta)$ [which falls off as $(\Delta/\Gamma)^{-1/2}$], though, $I_c$ is independent of detuning.

Unfortunately, this method of measuring the kinetic energy does not allow me to determine the exact shape of the momentum distribution due to the presence of the Raman resonances (which distort and obscure the tails of the central resonance). Therefore, I use an alternative technique for measuring separately the temperatures of the hot and cold atomic components of the non-thermal momentum distribution (see Sec. 2.4), which I discuss below in Sec. 4.4.2.

### 4.4 Temporal response of the nonlinearity driven with degenerate light

As I showed in Ch. 3, the optical mode with the largest gain typically becomes unstable before other modes with lower gain. Consistent with the observation that the largest nonlinear response occurs at $\delta = 0$ for the laser beam configuration considered in
Figure 4.8: **Longitudinal lattice temperature** Atomic temperature as a function of $I_p$ for $\Delta/\Gamma = -10$. The décrochage intensity corresponds to $I_d \sim 3 \text{ mW/cm}^2$.

Fig. 4.1 I indeed observe superradiant emission at a frequency that is degenerate with that of the pump beams. Therefore, I study in further detail the vapor's behavior when illuminated by degenerate pump and signal beams. In this section, I investigate the transient response of the NLO process when the pump and/or signal beams are rapidly turned on or off during the wave-mixing phase of the experiment. Specifically, I characterize the growth and decay rates of the system, and use this information to gain quantitative information about the hot and cold components.

### 4.4.1 Growth and decay rates

Rather than studying directly the time-dependent probe intensities, it is useful in some circumstances to discuss instead the nonlinear phase shift $\phi^{NL}$ (as defined for FWM in a Kerr medium in Sec. 3.1.1). This is because $\phi^{NL}$ depends only on the material’s optical response according to

$$
\phi^{NL} = |k|L/(4\varepsilon_0c) \left[ \chi^{(3)}I_p + \frac{1}{2\varepsilon_0c} \chi^{(5)}I_p^2 + O(I_p^3) \right],
$$

(4.4)
where $\chi^{(n)}$ is the nth-order susceptibility and I have assumed an isotropic material. This is in contrast to the probe beam intensities measured at the exit of the trap, which also include propagation effects. While I derive in detail an expression for $\phi^{NL}$ for the present experimental setup in Sec. 5.4, the relationship between the reflectivity and NLO phase shift is given simply as

$$\phi^{NL} = \tan^{-1}(\sqrt{R_0})$$

in the limit of small linear absorption.

Using the nonlinear phase shift, I investigate first the growth of the vapor’s NLO response when the pump and probe beams are turned on simultaneously $t_s = t_p = 0$ (see Fig. 4.9). I find that $\phi^{NL}(t)$ is well-described by an exponential growth of the form $\phi^{NL}(t) = \phi_{ss}^{NL} [1 - \exp(-t/\tau_g)]$, where $\phi_{ss}^{NL}$ is the phase shift in steady-state and $\tau_g$ is the growth time. Typical values of $\tau_g$ are on the order of 50-100 $\mu$s, which is comparable to the theoretical predictions for the cooling and loading of atoms in the the pump lattice (see Ch. 3). After several hundred $\mu$s, the system reaches a steady-state, which I study in depth in Sec. 4.5.

While the agreement between the predicted and measured growth time scales indicate that atomic localization in the pump lattice plays an important role in the nonlinearity, the formation of a density grating along the pump lattice (i.e., in the $\hat{k}_p$-direction) is not directly responsible for the wave mixing process. I show this by delaying the time at which I turn on the pump and signal beams such that $t_p = -400 \mu$s and $t_s = 0$ (see Fig. 4.9). In this case, the atoms immediately begin to bunch in the pump lattice, but $R$ and, therefore, $\phi^{NL}$ remain zero. Thus, the reflected idler signal does not arise as a result of scattering of the pump beams off the pump lattice. This is because the pump lattice, with grating period $d_p = \lambda/2$ and reciprocal lattice vector...
Figure 4.9: Growth of the nonlinear response. Time-dependent value of the nonlinear phase shift for $I_p = 2 \text{ mW/cm}^2$, $\Delta/\Gamma = -7$ and $\alpha_L = 13(\pm0.5)$. The signal beam is turned on at $t = 0$ for both curves and $\Delta t_{sp} = 1 \text{ ms}$. The slowly-increasing curve occurs for $t_p = 0$, whereas the rapidly-increasing curve occurs for $t_p = -400 \mu\text{s}$. The dashed lines are fits of the form $\phi_{ss}^N[1 - \exp(-t/\tau_g)]$ used to obtain $\tau_g$.

$G_p = 2k_p$ (see Fig. 4.10a), is not phase-matched for scattering pump photons into the probe beam directions (see Fig. 4.10b).

When I turn the signal beam on at $t = 0$, though, $\phi_{NL}(t)$ jumps up to the same steady-state value as when the pump and signal beams were turned on together, but with a growth time $\tau_g' = 6 \mu\text{s}$. To understand this accelerated growth, one must consider the full 2D optical lattice formed by the pump and probe beams. In the case where $t_s > t_p$, the atoms are already cooled and trapped in the pump lattice by the time I apply the signal beam. When the signal beam turns on, the atoms subsequently organize into the pump-probe lattice with reciprocal wave vector $\mathbf{G} = k_p + k_s$ and lattice period $d = \lambda/2\cos(\theta/2)$ (see Fig. 4.10a). To create a density grating along $\mathbf{G}$, atoms bound in the pump lattice need only to move a distance $d' = d_{\perp}/2$ on average along the direction orthogonal to $\mathbf{G}_p$ ($\mathbf{G}_p^\perp$, see Fig. 4.10a), where $d_{\perp} = \lambda/2\sin(\theta/2)$. Once established, this grating is phase-matched for scattering pump light into the probe direction, and leads to the amplification of the signal beam and creation of the idler beam (see Fig. 4.10b).
Figure 4.10: Grating structure and phase matching conditions. a) Two-dimensional optical lattice formed by the interference of the pump and probe beams, whose wave vectors are indicated by the thick, inward-pointing arrows. The dashed and dotted lines represent the anti-nodes for the pump-probe and pump lattice, respectively. The grating wave vectors and spatial periods are also shown. Phase matching diagram for scattering a pump beam into the idler mode via b) the pump grating and c) the pump-probe grating. The wave vector mismatch is $\Delta k$. 
I look next at the time scale over which the nonlinear response decays. After allowing the system to reach steady-state, I keep the pump beams on but suddenly turn off the signal beam. As the phase-matched density grating along $G$ washes out due to thermal atomic motion, the amount of pump light scattered from the grating will decrease. Thus, recording the decay of the light in the signal and idler mode in the absence of an injected signal beam gives information about the dephasing rate of the system. Figure 4.11 shows the decay of $\phi_{NL}(t)$ after I extinguish the signal beam at $t = 0$, where the decay clearly does not have a simple exponential form. While I study the shape of the decay in detail in Sec. 4.4.2, I define the effective decay time as $\tau_f$ such that $\phi_{NL}(\tau_f) = \phi_{ss}/e$ in order to compare it with $\tau_g$ directly. For the data shown in Fig. 4.11 I find that $\tau_f = 8 \mu$s, which is substantially shorter than $\tau_g$ for similar experimental conditions.

By varying the pump beam parameters, I find that the growth and decay times vary substantially. Figure 4.12 shows the growth and decay rates $\gamma_{g,f} = 1/\tau_{g,f}$, respectively, as a function of $I_p$. The growth rate increases slightly with increasing pump intensity due to the increased optical scattering rate, which leads to more rapid atomic cooling and localization. On the other hand, the decay rate decreases rapidly with increasing
Figure 4.12: Growth and decay rates. Rate at which $\phi_{NL}^N$ grows to $\phi_{ss}^N(1 - 1/e)$ (solid line) or falls to $\phi_{ss}^N/e$ (dashed line) as a function of $I_p$. The rates are equal at $I_p \approx 2$ mW/cm$^2$. Note the logarithmic scale of the vertical axis.

$I_p$ due to a decrease in the atomic temperature (see Sec. 4.4.2). For $I_p \approx 2$ mW/cm$^2$, $\gamma_g = \gamma_f$. Beyond this point, the growth rate exceeds the loss rate, which coincides with a net growth of initial fluctuations in the system and the vapor’s transition to the superradiant state (see Ch. 6 for further discussion).

### 4.4.2 Measurement of hot and cold components

Rather than simply considering the $1/e$ decay time for the decay signal discussed in Sec. 4.4.1, I now study the details of the decay signal’s shape in order to gain information about the underlying atomic momentum distribution. As I show, this approach yields direct evidence regarding the hot and cold component in a dissipative optical lattice. In addition, it enables me to measure the relative contributions to $\phi_{NL}^N$ from the hot and cold components $\phi_{c,h}^{NL}$, where $\phi_{NL}^N = \phi_{c}^{NL} + \phi_{h}^{NL}$.

It is instructive to consider the expected form of the free decay of a 1D density grating with spatial period $\Lambda$. For atoms with a Gaussian momentum distribution $g(p, T) = (2\pi mk_B T)^{-1/2}\exp(-p^2/2mk_B T)$, one finds a Gaussian decay $b(t) =$
\[ b_s \exp(-t^2/\tau^2) \propto \phi^{NL}(t) \] due to motion along the grating \[ \text{(141)} \] (see Ch. 5 for additional discussion). Here, \( b \equiv \langle \eta \rangle_\Lambda \) is the degree of bunching averaged over a grating period,

\[ \tau = \Lambda/\pi u(T), \quad (4.6) \]

and \( u(T) = (2k_B T/m)^{1/2} \).

For atoms in a 1D lattice, the predicted momentum distribution is bimodal and of the form \( f(p) = f_c g(T'_c) + f_h g(T'_h) \), where \( T'_{c,h} \) are the temperatures of the cold and hot components along the lattice direction. One therefore expects to find a double-Gaussian decay with decay times \( \tau_{c,h} = \lambda/2\pi u(T'_{c,h}) \) \[ \text{(84)} \]. For my beam geometry, this bimodal momentum distribution occurs along the \( \hat{G}_p \) direction due to the pump beam lattice. In order to extend this result to take into account the 2D lattice that arises from the inclusion of the probe beams, I must additionally consider the momentum distribution along \( \hat{G}_p \perp \). I write the resulting momentum distribution as

\[ f(p_{\parallel}, p_{\perp}) = g(p_{\perp}, T_{\perp})[f_c g(p_{\parallel}, T'_c) + f_h g(p_{\parallel}, T'_h)], \quad (4.7) \]

where \( T'_{c,h} (p_{\parallel}) \) and \( T_{\perp} (p_{\perp}) \) are the temperature (momentum) along \( \hat{G}_p \) and \( \hat{G}_p \perp \), respectively. It is also convenient to define the temperatures along \( \hat{G} \), which are simply \( T_{c,h} = T'_{c,h} \cos(\theta/2) + T_{\perp} \sin(\theta/2) \approx T'_{c,h} \) for \( T_{\perp}/T'_{c,h} \tan(\theta/2) \ll 1 \). Because thermal motion along any one of these directions leads to grating washout, one expects \( \phi^{NL}(t) \) to display a multi-Gaussian decay.

Figure [4.13] shows the decay of \( \phi^{NL} \) for three different values of \( I_p \). For small \( I_p \), where almost all of the atoms are in the hot, unbound component (i.e., \( f_c \sim 0 \)), \( \phi^{NL} \) decays after a time \( \tau = 1.8 \mu s \) due to diffusion of the hot component along \( \hat{G} \) (see Fig. [4.13 a]). Using Eq. (4.6) with \( \Lambda = d\hat{G} \), I find that \( T_h = 25 \mu K \). This is consistent with the predictions for Sisyphus cooling in a 1D lattice (see Sec. 2.4), and confirms that
the grating along $\hat{G}$ (rather than along $\hat{G}_p^\perp$) is responsible for the observed nonlinearity [33]. In addition, while the $T_h$ is a factor of 10 larger than $T_c$, it is still smaller than the equilibrium temperature of the atoms in the MOT (see Sec. B.2.2).

Increasing $I_p$ increases $f_c$ and gives rise to a second Gaussian decay due to the cold atomic component. This constitutes direct evidence for the existence of a hot and cold component. Because the average atomic kinetic energy of the cold component is much smaller than the well depth in this regime (e.g., $k_B T_c/U_0 = 0.3$ for $U_0/E_r = 50$), grating decay along $\hat{G}_p$ is highly suppressed. Therefore, grating washout predominantly occurs due to atomic motion along $\hat{G}_p^\perp$, which determines the decay time for the cold component. By fitting the decay in Fig. 4.13 b with a double-Gaussian, I find that $\tau_c \sim 20 \mu s$. This corresponds to $T_\perp = 30 \mu K$ for $\Lambda = d' = 11.5d$.

For larger $I_p$, where the optical gain becomes large and $\tau_f \sim \tau_g$, the back-action of the amplified probe fields strongly influences the coupled light-matter dynamics. In this regime, additional cooling along the direction perpendicular to $\hat{G}$ begins to occur via the lattice formed by the pump and nearly-copropagating probe beams and reduces $T_\perp$. The slow motion of atoms cooled in both the $x$- and $z$-directions give rise to a third decay time of $\tau \sim 100 \mu s$, as shown in Fig. 4.13 c. The complicated dynamics due to

Figure 4.13: Multi-Gaussian decay profile Decay of the nonlinear phase shift as a function of time after the signal beam is turned off at $t = 0$ for $\Delta/\Gamma = -5$, $\alpha_0L = 14(\pm 0.5)$ and $I_p = a) 0.45, b) 0.7$ and c) 1.25 mW/cm$^2$. The solid and dashed lines correspond to experimental data and a double-Gaussian fit, respectively (note the change in scale of the horizontal axis).
Figure 4.14: Nonlinear phase shift due to hot and cold components. Dependence of the nonlinear phase shift due to the cold (circle) and hot (square) components on $I_p$. The error bar represents typical measurement uncertainty.

The back-action of the scattered light on atomic motion in this regime do not allow me to directly extract the atomic temperature from the observed decay time. Nevertheless, the strong coupling between the evolution of the atomic and optical fields leads to decay times that are over 50 times larger than in the case of weak pump beams. In addition, because grating washout represents the dominant source of decoherence in the system, one can interpret the decay time $\tau$ as the effective coherence time of the system. Thus, I find that the coherence time of the system can be as large as $\sim 100 \mu s$.

Having identified the hot and cold components via their distinct decay times, I use a multi-Gaussian function to fit $\phi^{NL}(t)$ for various $I_p$ to measure $\phi^{NL}_{h,c}(I_p)$ separately. Figure 4.14 shows the dependence of $\phi^{NL}_{c,h}$ on $I_p$ for $\Delta = -5\Gamma$. The contribution from the cold component increases rapidly with increasing $I_p$ near $I_c$ due to the rise in $f_c$. In contrast, $\phi^{NL}_h$ remains mostly uniform for increasing $I_p$ because the decrease in $f_h$ is somewhat balanced by the larger values of $I_p$. 

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4.5 Steady-state nonlinearity driven with degenerate light

In the previous section, I described how the vapor approaches and leaves its steady-state behavior. In this section, I characterize how the system’s steady-state NLO response depends on various experimental parameters, such as the pump intensity, single-photon detuning from resonance, signal intensity, and atomic density. To quantitatively describe the data, I fit it with simple functional forms motivated by the theoretical work presented in Ch. 5. In addition, I investigate the group velocity of light as it propagates through the vapor.

4.5.1 Scaling with pump beam intensity

The intensity of the pump beams plays an important role in the vapor's nonlinear response, as it strongly influences both the internal and external atomic dynamics. Therefore, I investigate how the nonlinearity scales with $I_p$. Figure 4.15a shows the steady-state reflectivity for degenerate pump and signal frequencies $R_0$ as a function of $I_p$ for different $\Delta$. The reflectivity scales superlinearly with $I_p$ and can approach 1 for weak pump intensities (i.e., $I_p \ll I_s$). Beyond $R \sim 1$, the superradiant instability gives rise to self-generated signal and idler fields in the absence of an injected signal beam [84]. This instability, coupled with the rapid variation of $R$ on $I_p$, ultimately limits the largest values of $R$ that I can measure.

In order to investigate quantitatively the nonlinear susceptibility, I look at how $\phi^{NL}$ depends on $I_p$. Figure 4.15b shows $\phi^{NL}(I_p)$ for different $\Delta$. For small pump intensities (i.e., $I_p \ll I_s$), I find that the nonlinear phase shift is well-fit by a polynomial of the form $\phi^{NL} = AI_p + BI_p^2$ (see inset in Fig. 4.15). The quadratic (linear) components of $\phi^{NL}$
Figure 4.15: Dependence of the nonlinearity on pump intensity. Dependence of a) $R_0$ and b) $\phi^{NL}$ on $I_p$ for $\Delta / \Gamma = -3$ (circles), -5 (squares), -7.3 (diamonds), -12.7 (up-triangle), and -18.8 (down-triangle). For both figures, $\alpha_0 L = 13.4(\pm 0.5)$. The vertical dashed line indicates the value of the characteristic intensity $I_c = 1.5 \text{ mW/cm}^2$. 

Figure 4.16: Quadratic dependence of nonlinear phase shift. The inset shows that, for $I_p \ll I_c$, $\phi^{NL}$ is well-fit ($r^2=0.994$) by a quadratic function. The data is the same as shown in Fig. 4.15b for $\Delta / \Gamma = -3$ and $\alpha_0 L = 13.4(\pm 0.5)$. 

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correspond to a $\chi^{(5)}$ ($\chi^{(3)}$) response according to Eq. 4.4. Using this relationship and taking into account the tensorial nature of the susceptibility for the $\kappa_\perp$ configuration, I find $\chi^{(5)}_{xxxxyy} = 1.9(\pm 0.3) \times 10^{-12}$ (m/V)$^4$ for $\Delta = -3\Gamma$. This $\chi^{(5)}$ response gives rise to six-wave mixing (SWM), where the pump beams simultaneously undergo Bragg scattering from the pump-probe density grating and transfer atoms from the hot to the cold component (i.e., cool the atoms). The value of $\chi^{(5)}$ is the largest ever reported, exceeding that obtained for EIT-based SWM by $10^7$ [136, 137] and in three-photon absorption in a zinc blend semiconductor by $10^{24}$ [?].

For the same detuning ($\Delta = -3\Gamma$), I find $\chi^{(3)}_{xxyy} = 2.0(\pm 2.3) \times 10^{-10}$ (m/V)$^2$. The large experimental uncertainty arises from my inability to accurately measure $\phi^{NL}$ at the smallest $I_p$ due to detector noise and scattered light. This $\chi^{(3)}$ response is only $\sim 100$ times smaller than that observed in state-of-the-art EIT-based systems [142], and leads to four-wave mixing (FWM) in which the pump beams scatter off the weak atomic density grating composed solely of atoms in the hot component [143].

For pump intensities larger than the characteristic intensity (i.e., $I_c < I_p \ll I_{sat}(\Delta)$), $f_c \approx 1$ and the $\chi^{(5)}$ response saturates. Nevertheless, $\phi^{NL}$ continues to increase linearly with $I_p$. This is in contrast to $\chi^{(3)}$ processes that increase only sub-linearly beyond saturation.

### 4.5.2 Dependence on detuning

I next look at the dependence of the NLO response on $\Delta$. Figure 4.15 shows that the nonlinearity decreases with increasing $|\Delta|$ for a fixed value of $I_p$. The straightforward way to determine quantitatively how $\phi^{NL}$ decreases with increasing $|\Delta|$ would be to experimentally vary $\Delta$ with all other experimental parameters fixed. Unfortunately, my measurements of $\phi^{NL}$ are limited above by the instability and below by noise, which
precludes this approach. Instead, I fit all of the data shown in Fig. 4.15 with the same functional dependence on $I_p$, where I give an explicit equation for the relationship between $\phi^{NL}$ and $I_p$ in Eq. 5.24. I find that the data for each detuning collapses to a value that depends only on $\Delta$ and with a spread given by the error bars in Fig. 4.17 a). This intensity-independent NLO phase shift, which I denote $\phi^{NL}/f(I_p)$, is well-fit with a function of the form $A(|\Delta/\Gamma|)^{-B}$, where $B = 2.03(\pm0.05)$ and $A$ simply depends on the choice of $f(I_p)$.

This inverse quadratic scaling represents a much slower decrease in the nonlinear susceptibility than, for example, in a two-level atom, where $\chi_{T,LA}^{(n)} \propto (\Delta/\Gamma)^{-n}$. This enables me to obtain large $R$ (and $\phi^{NL}$) while operating far from resonance (where absorption is minimal). Figure 4.17b shows the signal-beam transmission in the absence of the pump beams, given by $T(I_p = 0) = \exp(-\alpha_\Delta L)$. For all of the detunings shown in Fig. 4.15b, $T(I_p = 0) > 0.65$ and approaches 1 for the larger detunings. Thus, this light-matter interaction allows me to realize large nonlinearities with high transparency for low input intensities.
I compare the observed NLO response to previously-reported nonlinearities based on $\chi^{(3)}$ processes by considering the achievable NLO phase shift for a fixed $I_p$. For example, Lo et al. [142] observe a 0.25 rad phase shift for an intensity of 230 $\mu$W/cm$^2$. I find that $\phi^{NL} = 0.25$ rad for $I_p = 560$ $\mu$W/cm$^2$ and $\Delta/\Gamma = -3$, although our system does not require any auxiliary, strong coupling beams (as in the EIT-based setups). Also, $\phi^{NL}$ continues increasing quadratically with $I_p$ for my $\chi^{(5)}$ process (rather than linearly as in $\chi^{(3)}$ processes), which further aids me in achieving large phase shifts at low-light-levels. My system provides the additional advantage that I can work far from resonance and thereby combine large nonlinearities with high transparency (similar to recent predictions for Rydberg-enhanced EIT [144]).

I quantify the tradeoff between absorptive loss and NLO phase shift using the cross-phase-modulation figure of merit

$$\zeta \equiv \frac{\phi^{NL}}{\alpha \Delta L}, \quad (4.8)$$

which corresponds to the ratio of the NLO phase shift to power loss. This is a useful metric because, in many applications, one wants to maximize the nonlinearity while avoiding absorption (which often contributes to loss or noise in a system). Figure 4.18 shows the dependence of $\zeta$ on $I_p$ for several different values of $\Delta$. While Lo et al. [142] observe a maximum value of $\zeta = 0.35$ for an intensity of 230 $\mu$W/cm$^2$, we exceed this value for $I_p > 300$ $\mu$W/cm$^2$, and obtain a maximum value of $\zeta = 26$ for $I_p = 5.6$ mW/cm$^2$. In addition, the fact that the data for each detuning follow the same curve indicates that $\zeta$ is independent of $\Delta$; this confirms the conclusion that $\phi^{NL}$, like $\alpha \Delta$, scales like $(\Delta/\Gamma)^{-2}$.  

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4.5.3 Dependence on atomic density

For a fixed value of $\Delta$ and $I_p$, Figure 4.19 shows that the nonlinear response increases with increasing atomic density. I find that $\phi^{NL}$ is well-described by a linear function that intersects the origin. This implies that the pump light scatters coherently off the pump-probe density grating, as expected for a driven wave-mixing process where the grating is created by the interference of the incident optical fields in a phase-coherent manner.

4.5.4 Influence of signal beam intensity

Up to this point, I have assumed that the signal beam was sufficiently weak so as to influence but not strongly perturb the vapor. As I show in Fig. 4.19, $\phi^{NL}$ is indeed independent of $I_s$ up to approximately 15 $\mu W/cm^2$. After this point, the nonlinearity saturates and $\phi^{NL}$ falls off exponentially. This effect is less pronounced when I repeat the experiment using the same value of $I_p$ but a smaller atomic density (i.e., when the
optical gain is reduced). Therefore, I conclude that one contribution to this saturation is depletion of the pump beams. In addition, radiation pressure forces due to the imbalance of the signal and idler beams can result in an appreciable net acceleration of the atoms for more intense probe beams, thereby reducing the efficiency with which atoms load into the optical lattice.

### 4.5.5 Slow light effects

In addition to large NLO phase shifts, I also observe a substantial decrease in the group velocity of light $v_g = c/n_g$ due to the creation of the atomic density grating $[145]$. Here $n_g = n + \omega_s d n / d \omega_s$ and $n = \sqrt{1 + \chi}$ are the group and complex indices of refraction of the vapor, respectively. Such slow light effects have important applications to a variety of different fields. One obvious application of slow light arises in delaying the arrival of a pulse of light, which is integral to optical buffering techniques in telecommunication networks $[146]$. Slow light also presents a way to localize and store information held in light pulses for applications to quantum computation $[147]$. Furthermore, slow light has been demonstrated to increase the sensitivity of interferometers $[148]$ and
Figure 4.20: Slow-light effects. Dependence of the slow-light delay and group velocity on the NLO phase shift for $\Delta = -5\Gamma$.

I experimentally measure the slow-light group delay $t_d = (n_g - 1)L/c$ by modulating the amplitude of the signal beam by 10% at a frequency of 15 kHz and recording the transmitted signal beam intensity. The relative phase lag between the transmitted beam for $I_p = 0$ and $I_p \neq 0$ gives the time delay due solely to nonlinear effects (i.e., it excludes the propagation time of the light through vacuum such that $t_d = 0$ for $I_p = 0$). Figure 4.20 shows the slow-light delay and group velocity as a function of $\phi_{NL}$ for $\Delta = -5\Gamma$. For $\phi_{NL} \sim 1$, I observe group velocities as large as $v_g/c \sim 10^{-5}$, which corresponds to slow light delays of several $\mu$s. The medium therefore acts like a high-finesse cavity and increases the photon lifetime in the gas from $\tau_{\text{photon}} = L/c = 100$ ps to several $\mu$s. This dramatic increase in the photon lifetime has important consequences on the atom-photon dynamics. For example, it decreases the effective decoherence rate of the system and removes the need to work with either ultracold atoms or high-finesse cavities in order to realize superradiance. In addition, it makes the time scales of the optical evolution comparable to the time scale associated with the atomic dynamics and enables effective long-range atom-atom interactions.
4.6 Summary

In this chapter, I have characterized the nonlinear response of my cold atomic vapor below the superradiant instability. I showed that weak optical fields tuned several natural linewidths below the atomic resonance give rise to a large NLO response, whereas fields with comparable amplitudes tuned above or very close to the atomic resonance yield small nonlinearities. I tested various polarization configurations for the pump and signal beams and found that each produced narrow resonances centered around the pump beam frequency, which results in significant slow-light effects. Nevertheless, the $\kappa_\perp$ and $\theta_\parallel$ configurations yielded the largest nonlinear response for a given incident pump intensity. In addition, the largest nonlinearity occurred for pump and probe beams with degenerate frequencies. I therefore looked in further detail at the case where $\delta = 0$.

I first studied the transient response of the system when the pump and probe beams were rapidly turned on and off. This demonstrated that, while cooling and bunching of atoms in the pump lattice play an important role in the nonlinearity, the pump-probe grating is directly responsible for the scattering of light from the pump to the probe beams. In addition, by studying the rate at which the scattered signal decays after I extinguish the signal beam, I observe direct evidence for the hot and cold atomic components discussed in Ch. 2 as well as a lengthening of the atomic coherence time by a factor of 50.

I then studied how the system’s steady-state NLO response depended on various parameters and found that the vapor displays the largest fifth-order susceptibility reported to date. The NLO response also displays a relatively slow decrease with detuning that scales as $(\Delta/\Gamma)^{-2}$, which allows me to observe simultaneously a large nonlinearity and high transmission. In addition, I observe large slow light delays (i.e., small
group velocities of light) for large NLO phase shifts. In the next chapter, I develop a theoretical model with no free parameters that describes the experimental results described above in the limit of small gain.
Chapter 5

Bunching-induced optical nonlinearity: theory

In Ch. 4 I showed experimental data for a novel, dissipation-enhanced wave mixing process that occurs when a cold atomic vapor is illuminated by weak optical fields. In this chapter, I develop a theoretical model that is based on the light-matter interactions discussed in Chs. 2 and 3 to explain the core physics involved in my experiment. Specifically, I show that the observed NLO response arises from the simultaneous cooling and localization of the atomic ensemble, which leads to a high-contrast density grating that efficiently scatters light. The model reproduces well the measured data with no free parameters, and therefore confirms the interpretation of the nonlinearity proposed in Ch. 4.

This chapter is based partly on Refs. [84] and [131].

5.1 Introduction to the problem

Motivated by the experimental results discussed in Ch. 4 I consider the configuration shown in Fig. 5.1a, where optical fields interact with a pencil-shaped cloud of cold atoms with length $L$ and diameter $W$. I assume that the cloud is highly anisotropic ($L/W \sim 100$) and has its long axis aligned along the $\hat{z}$-direction. The atoms are illuminated by a pair of intensity-balanced, counterpropagating pump fields (intensity $I_p$) with orthogonal linear polarizations (lin.$\perp$ lin configuration) that propagate at an angle $\theta = 10^\circ$ relative to the $z$-axis (i.e., the $\hat{z}'$-direction). Weak signal and idler fields (intensities $I_s$ and $I_i$, respectively) counterpropagate along $\hat{z}$ and are orthogonally-
polarized with respect to the nearly-copropagating pump beams. I refer to the signal and idler fields collectively as probe fields.

My goal is to describe self-consistently the coupled evolution of the atoms and the optical fields. To a large degree, previous studies of atomic motion in dissipative optical lattices and nonlinear wave mixing processes have been performed separately. Most of the work on optical lattices assume static, externally-imposed optical fields [55]. In contrast, many nonlinear optical studies in cold atoms ignore the back-action of the generated optical fields on the atomic distribution [150]. While these limiting cases describe well the observed phenomena in many situations, the experimental

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**Figure 5.1: Theoretical model geometry** a) Beam geometry and b) atomic level structure (including the square of the Clebsch-Gordan coefficients the transition) considered in the theoretical model.
findings that I presented in Ch. 4 indicate that these approximations are insufficient for describing my system. Nevertheless, a complete theory describing the motion of multilevel atoms in a dissipative, two-dimensional (2D) optical lattice formed via the interference of the same fields that are simultaneously responsible for wave mixing is challenging to solve. While this can be done numerically, such an approach may obscure the core involved physical mechanisms. Thus, I instead endeavor to construct a minimal model that includes the relevant physics and agrees with the experimental measurements.

In order to guide my approach to the problem, I briefly recall several key results from Ch. 4. First, I find that the intensities of the normalized signal and idler signals displayed a \( \sec^2(\phi^{NL}) \) and \( \tan^2(\phi^{NL}) \) dependence on the nonlinear phase shift \( \phi^{NL} \), respectively. This scaling indicates that propagation effects play an important role in the evolution of the fields, (see the discussion of wave mixing in a Kerr medium presented in Sec. 3.1.2). Second, I observe evidence demonstrating that the atomic momentum distribution transforms into non-thermal distribution consisting of a hot and cold component, which implies that Sisyphus cooling strongly affects the atomic dynamics (as described in Sec 2.4). Third, I show that the largest nonlinear response occurs for degenerate pump and probe beams. Because the instability threshold for this case will be smaller than that of non-degenerate beams, I focus on the case of degenerate fields. Finally, I conduct all of my experiments in the regime where \( I_{s,i} \ll I_p < I_{sat} \), where \( I_{sat}(\Delta) \) is the off-resonant saturation intensity. This suggests that I can treat the probe fields perturbatively and ignore effects due to saturation of the electronic nonlinearity.

With these observations in mind, I briefly outline the main approximations and parameters in my model as well as describe my procedure for solving it. In order to allow for Sisyphus cooling to occur, I take the atoms to have a \( J_g = 1/2 \rightarrow J_e = 3/2 \)
level structure. In addition, I assume that all optical fields have the same frequency $\omega$ and are tuned below the atomic resonance $\omega_0$ by an amount $\Delta = \omega - \omega_0$. I begin by solving for the atomic motion in the 1D lattice formed by the pump beams to all orders in the pump field with the assumption that $I_p$ is spatially and temporally constant (as described in Sec. 2.4). I then solve perturbatively for the modification of the atomic density distribution due to the lattice formed by the interference of a pump and nearly-counterpropagating probe beam. Using this spatially-dependent density, I then determine the atomic polarization to first-order in the probe fields. Substituting the resulting polarization into Maxwell’s wave equation, I derive and solve a set of coupled wave equations that describe the propagation of the probe fields through the vapor.

5.2 Derivation of the coupled wave equations

In this section, I derive a general equation that describes the propagation of the probe fields through the cold atomic cloud. For the optical beam configuration described above, I write the total electric field

$$\tilde{E}(r, t) = E(r, t)e^{-i\omega t} + c.c,$$

(5.1)

where $E = \sum_n E_n \exp(i k_n \cdot r)$ for $n = \{p1, p2, s, i\}$, which corresponds to the two counterpropagating pump, signal, and idler fields, respectively (see Fig. 5.1 a). For my beam geometry, $k_{p1} = -k_{p2} = k_p$ and $k_s = -k_i$ for $|k_p| = |k_i| = k = 2\pi/\lambda$, where $\lambda = 2\pi c/\omega$ and $c$ are the optical wavelength and speed of light in vacuum. Based on my experimental observations of the polarization configuration that leads to the largest NLO interaction strengths, I take $E_n = \tilde{e}_n E_n$ for $\{\tilde{e}_{p1}, \tilde{e}_{p2}, \tilde{e}_s, \tilde{e}_i\} = \{\tilde{x}, \tilde{y}, \tilde{y}, \tilde{x}\}$. 138
Thus, while I show later that the non-zero value of $\theta$ has important implications for phase-matching considerations in the wave-mixing process, I assume that it is small enough to justify the approximation that the field polarizations all lie in the $x$-$y$ plane. I take the pump fields $E_{p1} = E_{p2} = E_p$ to be real and constant with no loss of generality. The signal and idler fields $E_{s,i} = E_{s,i}(z, t)$ have slowly-varying, complex amplitudes, and $|E_{s,i}| \ll |E_p|$. I define the beam intensities as $I_n = 2\varepsilon_0 c |E_n|^2$ and assume $I_{s,i,p} \ll I_{sat}(\Delta)$, where $I_{sat}(\Delta) = I_{sat}[1+(2\Delta/\Gamma)^2]$ is the off-resonance saturation intensity, $I_{sat} \equiv \varepsilon_0 c (\hbar \Gamma/2\mu)^2$ is the resonant saturation intensity, $\Gamma$ is the full width at half maximum natural linewidth of the electronic transition, and $\mu$ is the effective dipole matrix element.

These fields produce an atomic polarization

$$\tilde{P}(r, t) = \sum \chi \tilde{E}(r, t) = P(r, t)e^{-i\omega t} + c.c., \quad (5.2)$$

where $\chi$ is the tensor susceptibility. Substituting this polarization into Maxwell’s equation (i.e., Eq. 3.1) yields

$$\left( \frac{\partial E_{s,i}}{\partial t} \pm c \frac{\partial E_{s,i}}{\partial z} \right) = \frac{i \omega}{2\varepsilon_0} \langle P e^{\mp ik \cdot r} \rangle, \quad (5.3)$$

where I have made the slowly-varying amplitude approximation and the rotating wave approximation (see Sec. 3.1.1). The angular brackets denote a spatial average over a grating period. This effectively enforces phase-matching (i.e., momentum conservation) between the fields and therefore acts to select only the contributions to the polarization that efficiently drive $E_{s,i}$.

As I discussed in Sec. 2.4 one can consider the full multilevel atomic structure to be equivalent to two $V$-type transitions where the dynamics of the $g_{\pm 1/2}$ states (mneu-
monic \( \pm \) are identical (up to a spatial offset of \( \lambda/4 \) discussed later). This situation arises because the applied fields do not drive \( \pi \) transitions and therefore cannot establish coherences between the \( g_{\pm 1/2} \) states, and helps to simplify the model considerably. Thus, I write \( P = P^+ + P^- \) and consider the single-atom polarizations \( p^\pm \) such that

\[
P^\pm = \eta^\pm(r)p^\pm,
\]

(5.4)

where \( \eta^\pm(r) \) is the atomic density at position \( r \) for the \( g_{\pm 1/2} \) states, respectively. By adiabatically eliminating the excited state and solving to first-order in the field strengths for \( (2\Delta/\Gamma)^2 \gg 1 \), I find that

\[
p^\pm = \sum_n p^\pm_n(r)e^{i k_n \cdot r},
\]

(5.5)

where

\[
p^\pm_n(r) = \frac{|\mu^\pm_{\text{eff}}|^2}{\hbar(\Delta + i\Gamma/2)} E_n(r)
\]

are slowly-varying polarization amplitudes such that \( \langle p^\pm \exp(i k_n \cdot r) \rangle \sim p^\pm_n \delta_{nm} \). Here

\[
\mu^\pm_{\text{eff}} = \sum_{j=\epsilon_{\pm 1/2}} \mu_{j,g_{\pm 1/2}} \mu_{g_{\pm 1/2},j},
\]

(5.7)

where \( \mu_{i,j} = \langle j|e_r|i \rangle \) is the dipole matrix element between states. Thus, phase-matching requires a spatial modulation of \( \eta^\pm(r) \) with wave vectors \( k_{s,i} - k_n \).

While the following analysis will display many similarities with the analysis of wave mixing in a nonlinear Kerr medium discussed in Sec. 3.1, the situation described in this section is fundamentally different. For the Kerr medium, I assumed a uniform atomic density distribution and considered the third-order response of the single-atom polarizability, which produced a spatial modulation of the internal atomic state (i.e.,
polarization gratings). In this chapter, I instead consider only the first-order response of the single-atom polarizability, but allow for a spatial modulation of the atomic density. The resulting density gratings phase-match NLO wave mixing in a manner analogous to the polarization gratings, but display novel scalings with material parameters and, as I will show, result in optical instabilities at smaller relative pump intensities for a given optical depth.

### 5.3 Calculation of the spatial density distribution

To calculate the atomic polarization, I first need to determine the spatially-varying atomic density distribution in the \(x-z\)-plane. The field given in Eq. 5.1 produces an optical lattice associated with the ground state potential

\[
U_{\pm}^{2d}(r) = p^\pm \cdot E
\]

\[
\cong U_{\pm} + \delta U_{\pm}
\]  

(5.8)

to first order in the probe field amplitudes. In the limit that \((2\Delta/\Gamma)^2 \gg 1\),

\[
U_\pm = \frac{U_0}{2} \left[ -2 \pm \cos(2k_p \cdot r) \right]
\]  

(5.9)

is the potential due to the pump the pump fields and

\[
\delta U_{\pm}(r) = \delta U_{\pm}^G + \delta U_{\pm}^{G^*}
\]

\[
= \left[ \left( p_{p1}^\pm \cdot E_i^* + (p_{p2}^\pm)^* \cdot E_s \right) e^{iG \cdot r} + c.c. \right] + \left[ \left( p_{p1}^\pm \cdot E_i^* + (p_{p2}^\pm)^* \cdot E_i \right) e^{i(G^* \cdot r)} + c.c. \right]
\]  

(5.10)
is the potential due to the interference of the pump and probe fields. The component of the potential varying along $G = k_p + k_s$ ($G^\perp = k_p - k_s$) is due to the interference of a pump and nearly-counterpropagating (co-propagating) probe beam. This potential differs from those typically considered in studies on atomic motion in dissipative optical lattices in that the lattice is not static: it depends on the probe fields which, in turn, depend on the density distribution via Eq. [5.3]. Thus, solving for the coupled light-matter dynamics, even to first order in the probe fields, requires one to solve simultaneously for the temporally- and spatially-varying optical fields and atomic distribution.

### 5.3.1 Atomic density modulation in the pump lattice

To simplify the problem, I make two additional assumptions. First, I take the light fields to follow adiabatically the evolution of the density grating because the time scale over which the optical fields evolve is typically much shorter than the time scale of the atomic dynamics. Second, because $I_p \gg I_{s,i}$, I assume that the one-dimensional (1D) optical lattice along $k_p$ formed by the pump beams dominates the atomic motion. I therefore solve numerically for the density in the absence of the probe beams using the method described in Sec. [2.4].

For red detunings ($\Delta < 0$), the atoms load into the anti-nodes of the lattice. This leads to an enhanced atomic polarization because the atoms see a higher average field strength than a homogeneously-distributed sample. In addition, the atoms are optically pumped into the stretched states, which have the strongest coupling to the optical field (see Fig. [5.1]b). I write the resulting density due to the pump beam lattice as $\eta_p^+(r) = \eta_p^+(r + q\lambda/2k_p) = \eta_p^-(r + (2q + 1)\lambda/4k_p)$ for integer $q$, which gives rise to long-range anti-ferromagnetic order. One can therefore
write the density in terms of a Fourier series as

$$\eta_p^\pm(r) = \bar{\eta}\sum_m c_m^\pm e^{im2k_p \cdot r \pm \phi_0^\mp},$$

(5.11)

where $\phi_0^+ = 0$ and $\phi_0^- = \lambda/4$ are the relative phase shifts of the ground states. Here $c_m^\pm$ are the amplitudes of the Fourier components chosen such that $c_0^+ + c_0^- \equiv \bar{\eta}^+ + \bar{\eta}^- = \eta$, where $\eta$ is the average atomic density.

As the atoms move in this bright lattice, Sisyphus cooling changes the atomic momentum distribution from a Maxwell-Boltzmann distribution to one that is well-described by a double-Gaussian [71] (as discussed in Sec. 2.4). The narrow (broad) Gaussian distribution component corresponds to a cold (hot), bound (unbound) fraction of atoms, which leads to an interpretation of the cooling mechanism as a transfer of population from the hot to the cold component. While no analytic solution for the fractions of atoms in the hot and cold components $f_c, f_h$ exists, I find that they are well-described by the functional form $f_c \approx \tanh(I_p/I_c) = 1 - f_h$. When the pump intensity equals the characteristic intensity $I_c$, most of the atoms are in the cold component. The characteristic intensity is directly related to the so-called décrochage intensity $I_d$, which has been shown to depend only on the pump intensity (i.e., is independent of $\Delta$) [?].

This leads to an anisotropic momentum distribution $\rho(p)$

$$\rho(p) = g(p_y, T_y)g(p_\perp, T_\perp)[f_c g(p_{||}, T_{c\,||}') + f_h g(p_{||}, T_{h\,||}')],$$

(5.12)

where $g(p, T) = (2\pi mk_B T)^{-1/2}\exp(-p^2/2mk_B T)$, $m$ is the atomic mass, $k_B$ is Boltzmann’s constant, and $T_y (p_y)$, $T_{c\,||} (p_{||})$, and $T_{h\,||} (p_{||})$ correspond to the temperature (momentum) along $\hat{y}$, $\hat{k}_p$, and orthogonal to $\hat{k}_p$, respectively. Typical temperatures in
my experiment are $T_y \sim T_\perp \sim T_{\text{eq}} = 30 \, \mu\text{K}$, $T'_{c} \sim 2 - 3 \, \mu\text{K}$, and $T'_h > 20 \, \mu\text{K}$.

Despite the fact that the atoms can become well-localized along $\hat{k}_p$, this density grating is not phase-matched for scattering pump light into the probe modes and therefore does not directly contribute to the amplification of the probe beams. One can see this explicitly by considering the right-hand-side of Eq. 5.3. For a density modulation due to the pump beams with wave vector $2k_p$, the spatially-averaged polarization becomes

$$\langle p^\pm e^{\pm i k_p \cdot r} \rangle = \langle \eta^\pm p^\pm \rangle = \left\langle \left( \sum_j e^{ij2k_p \cdot r} \right) \left( \sum_n \frac{\mu^\pm}{\hbar(A + i \Gamma/2)} \cdot E_n e^{i k_n \cdot r} \right) \right\rangle = \eta \frac{\mu^\pm}{\hbar(A + i \Gamma/2)} \cdot E_{s,i}. \quad (5.13)$$

This term represents only linear absorption and dispersion of the probe beams as they pass through the vapor.

### 5.3.2 Atomic density modulation in the pump-probe lattice

In contrast, the pump-probe grating is phase-matched for scattering pump light into the probe modes. My experimental results suggest that the scattering of the pump beams off the density grating formed by the interference of a probe and nearly counterpropagating pump field is responsible for the probe beam amplification. I therefore focus on $\delta U_{\pm}^G$ (i.e., the component of the potential with modulation frequency $|G|$) and calculate the resulting perturbation to the atomic density. According to Eq. 5.10, this
potential becomes \( \delta U_G^\pm (r) = \delta U_0^G e^{iG \cdot r} + c.c. \), where

\[
\delta U_{0,\pm}^G = \left[ \frac{\left( \mu_{\pm}^2 \cdot E_{p1} \right) \cdot E_+^i + \left( \mu_{\pm}^2 \cdot E_{p2}^\dagger \right) \cdot E_-}{\hbar \Delta} \right].
\] (5.14)

in the limit that \((2\Delta/\Gamma)^2 \gg 1\). Because the nearly counterpropagating fields are copolarized, Sisyphus cooling due to this lattice does not occur. I therefore assume that the atomic temperature is fixed by the pump lattice.

Before I calculate the resulting modification to the atomic density for my non-thermal, two-component vapor, I consider first the impact of the additional potential \( \delta U_G^\pm \) on a single-component gas with temperature \( T \) along \( \hat{G} \). In this case, the spatially-dependent density becomes

\[
\eta^\pm (r) = \eta_{p,\pm}^\pm + \delta \eta^\pm,
\] (5.15)

where the potential introduces a modulation of the steady-state atomic density distribution given by \([11,141]\)

\[
\delta \eta^\pm = \eta_0^\pm e^{-U_0^G / k_B T},
\] (5.16)

where \( \eta_0^\pm \) is simply a normalization constant that ensures that the total density is properly normalized (i.e., \( \langle \eta^\pm(r) \rangle = \bar{\eta}^\pm \)). I expand \( \delta \eta^\pm \) in terms of its spatial Fourier modes and find

\[
\eta^\pm (r) = \eta_0^\pm \sum_j (-1)^j I_j(\psi^\pm)e^{ijG \cdot r},
\] (5.17)

where \( \psi^\pm = U_{0,\pm}^G / k_B T \) and \( I_j(\psi^\pm) \) is the modified Bessel function of the first kind of order \( j \). In my experiment, \( U_{0,\pm}^G / E_r \sim 1 \) and \( k_B T / E_r \geq 10 \), which implies that \( \psi^\pm \ll 1 \). I therefore Taylor expand Eq. 5.17 about \( \psi^\pm = 0 \) and find

\[
\delta \eta^\pm \sim \eta_0^\pm \left( 1 + b^\pm(T)e^{iG \cdot r} + \left( b^\pm(T) \right)^* e^{-iG \cdot r} \right)
\] (5.18)
to first order in $E_s,i$. Here,

$$b^\pm(T) = -\frac{\langle \mu^2 \cdot E_{p1} \rangle \cdot E_i^* + \langle \mu^2 \cdot E_{p2} \rangle \cdot E_s}{k_B \hbar \Delta}$$

(5.19)

corresponds to the degree of atomic bunching in the density grating along $\hat{G}$. Extending this result to the case of my two-component gas, I find that

$$b^\pm(T) \rightarrow b^\pm(T_c, T_h) = f_c b^\pm(T_c) + f_h b^\pm(T_h),$$

(5.20)

where $T_{c,h} = T_{c,h}' \cos(\theta/2) + T_{\perp} \sin(\theta/2) \sim T_{c,h}'$. Because $T_{\perp}/T_{c,h}' \tan(\theta/2) < 1$, I take $T_{c,h} \sim T_{c,h}'$. 

### 5.4 Solving the coupled wave equations

I next calculate the polarization using Eqs. 5.4 and 5.15. The polarization has two types of contributions that are phase-matched for driving the probe fields. For example, one contribution to the polarization driving the signal field scales as $(E_{p1} E_i^*) E_{p2}$. This corresponds physically to the scattering of a pump field off the grating formed by the interference of the counterpropagating pump field and the idler field and results in a net power transfer between the pump and signal beams (see Fig. 5.2 a). The second contribution has the form $(E_{p2} E_s) E_{p2}$, which corresponds to the scattering of a pump beam off the grating formed by the same pump beam and the signal beam (see Fig. 5.2 b) and results in a phase shift of the signal beam. The waves are therefore coupled via backward FWM, where the pump beams backscatter off the generated gratings.

I investigate the steady-state behavior of the probe fields by substituting the full polarization into Eq. 5.3. Assuming that the optical fields follow adiabatically the evo-
Figure 5.2: Backward four-wave mixing. Scattering of a pump beam into the signal beam direction via the grating formed by the interference of a) $E_{p1}$ and $E_i$ and b) $E_{p2}$ and $E_s$. c) Scattering from the spin-polarized gratings separated by a quarter of an optical wavelength gives rise to backscattered light with a polarization that is identical to the nearly-counterpropagating pump beam.

I find that

$$\frac{\partial E_s}{\partial z} = \frac{i \omega}{2c\varepsilon_0} \sum_{j=\pm} \eta^j \mu^j \frac{\hbar}{\hbar \Delta} \left[ E_s + b^j(T_c, T_h)E_{p2} \right],$$

$$\frac{\partial E_i}{\partial z} = -\frac{i \omega}{2c\varepsilon_0} \sum_{j=\pm} \eta^j \mu^j \frac{\hbar}{\hbar \Delta} \left[ E_i + \left( b^j(T_c, T_h) \right)^* E_{p1} \right],$$

where I keep only phase-matched terms to first order in the probe fields. Carrying out the sum over the contributions from both ground states, Eq. [5.21] reduces to

$$\frac{d E_s}{d z} = i \kappa E_s + i \beta E_i^*,$$

$$\frac{d E_i^*}{d z} = i \kappa^* E_i^* + i \beta E_s.$$  (5.22)

I define the nonlinear coupling coefficient

$$\beta = \beta_c + \beta_h = \frac{\alpha_0 \hbar \Gamma}{64(\Delta/\Gamma)^2 I_{sat}} \left( \frac{f_c}{k_B T_c} + \frac{f_h}{k_B T_h} \right),$$  (5.23)
such that

\[ \phi^{NL} = \beta L \]  

(5.24)
corresponds to the nonlinear phase shift imposed on the weak probe beams by the interaction and \( \phi^{NL}_{c,h} = \beta_{c,h} L \) represents the independent contributions from the cold and hot components. The self-coupling coefficient is given by

\[ \kappa = \beta + \frac{\alpha_0 (-2\Delta/\Gamma + i)}{8(\Delta/\Gamma)^2} = \kappa_R + i\kappa_I. \]  

(5.25)

Here \( \alpha_\Delta \equiv \alpha_0 / [1 + (2\Delta/\Gamma)^2] \) is the absorption coefficient experienced by a single, weak optical field,

\[ \alpha_0 \equiv \frac{\omega}{c} \left[ \frac{2\eta |\mu|^2}{\epsilon_0 \hbar \Gamma} \right] \]  

(5.26)
is the on-resonance absorption coefficient, and the effective dipole moment is defined as

\[ |\mu|^4 = \frac{2\mu_2^4 \epsilon_{1/2} \epsilon_{3/2}}{9} = \left| \sum_{j=\pm} \left[ \left( \vec{\mu}_j \cdot \hat{x} \right) \cdot \hat{x} \right] \vec{\mu}_j \cdot \hat{y} \right|. \]  

(5.27)

It is interesting to note that the tensor nature of the interaction is absent in Eq. 5.21. This is because the scattering of \( \hat{\epsilon}_+ \) light from the \( g_{+1/2} \) state combines with \( \hat{\epsilon}_- \) light from the \( g_{-1/2} \) state, which is spatially offset by a distance \( \lambda/4 \). The combination of these two signals leads to linearly-polarized probe signals with the same polarization as the nearly-counterpropagating pump signals due to coherent backscatter off the spin-polarized density gratings (see Fig. 5.2c).

The coupled wave equations give in Eq. 5.22 have the same general form as those derived by Abrams and Lind for four-wave mixing (FWM) in a two-level atom (see Eq. 3.17). Therefore, imposing the boundary conditions that \( E_i(z = 0) = E_i(0) \) and \( E_i(z = L) = 0 \) (i.e., the same used in Sec. 3.1.3), I immediately find that the
normalized signal transmission and reflectivity are given as

\[ T \equiv \left| \frac{E_s(L)}{E_s(0)} \right|^2 = \frac{|\beta \sin(wL)|^2}{|w \cos(wL) \pm \kappa \sin(wL)|^2} \]  \hspace{1cm} (5.28)

\[ R \equiv \left| \frac{E_i(0)}{E_s(0)} \right|^2 = \frac{|w|^2}{|w \cos(wL) \pm \kappa \sin(wL)|^2}, \]  \hspace{1cm} (5.29)

where \( w = (|\beta|^2 - \kappa_I^2)^{1/2} \). In the limit that \( \kappa_I \to 0 \), these solutions reduce to \( T = \sec^2(\phi_{NL}) \) and \( R = \tan^2(\phi_{NL}) \).

Figure 5.3a show the dependence of \( T \) and \( R \) predicted in Eqs. 5.28 and 5.29 on \( I_p \) for \( \Delta/\Gamma = -4 \). As discussed in Ch. 3, \( T > R \) for all \( I_p \), but both approach infinite gain for certain values of \( \phi_{NL} \). I take as inputs to the model the experimentally-measured values of \( \Delta, I_{p,s,i}, \alpha_{\Delta L}, I_c, \) and \( T_{h,c} \) (as discussed Ch. 4). In general, I find good agreement between the experimental results and theoretical predictions with no free parameters. The fact that the measured signal and idler gain agree with one another for a given value of \( \phi_{NL} \) indicates that I can use measurements of either \( T \) or \( R \) to compare my model with experimental results. Because the experimentally-measured reflectivity has a smaller signal-to-noise ratio, I continue by comparing the predictions of the model with measured values of \( R \).

Figure 5.3b shows the predicted and measured dependence of the reflectivity on \( I_p \) for various values of \( \Delta \). I find that the reduced chi-squared error between the theory and experimental data taken over all measured values of \( \Delta \) and \( I_p \) is \( \chi^2_{\text{red}} = 1.6 \). This excellent agreement indicates the validity of the model approximations discussed above. In addition, it confirms my physical interpretation of the underlying nonlinearity as arising from the cooling and bunching of atoms in a dynamic, dissipative optical lattice. Thus, despite the fact that the form of the coupled wave equations in Eq. 5.22 are the same as that described by Abrams and Lind for FWM in a two-level atom, the nonlinearity studied in this chapter has very different physical origins. Specifi-
cally, while I considered only a linear internal atomic response, I find that including the effects of the incident optical fields on the atomic center-of-mass states results in nonlinear scattering. This has important consequences for the dependence of \( \phi_{NL} \) on material parameters, which I discuss in detail in the next section.

5.5 Dependence of the NLO response on material parameters

In order to determine how the nonlinearity depends on various parameters, I look more closely at the NLO coupling coefficient \( \beta \). In particular, I find that \( \beta \) includes terms that scale roughly as \( \beta \propto \eta(I_p/I_{sat})\tanh(I_p/I_c)(\Delta/\Gamma)^{-2} \). For comparison, Abrams and Lind found that \( \beta_{TLA} \propto \eta(I_p/I_{sat})(\Delta/\Gamma)^{-3} \) for the third-order \( \chi^{(3)} \) nonlinearity due to two-level atoms. In both cases, the linear scaling of \( \beta \) with \( \eta \) means that the light
scattered by the individual atoms adds together coherently. In contrast, $\beta$’s more rapid increase with $I_p$ and slower decrease with $\Delta$ in my system represent a novel result with beneficial consequences. I therefore investigate the origin and implications of this scaling in more detail.

I look first at how the NLO coupling constant scales with the pump intensity in various regimes. In the limit of small pump intensities (i.e., $I_p \ll I_c$), I carry out a Taylor series expansion of $\beta$ and find that

$$\beta \approx \frac{\alpha_0 \hbar \Gamma}{64(\Delta/\Gamma)^2 I_{sat}} \left( \frac{I_p/I_c}{k_B T_c} + \frac{1 - I_p/I_c}{k_B T_h} \right). \tag{5.30}$$

The quadratic (linear) components of $\phi^{NL}$ correspond to a $\chi^{(5)} (\chi^{(3)})$ response according to $\phi^{NL} = |k|L/(4\epsilon_0 c)[\chi^{(3)}_{xxxy} I_p + (2\epsilon_0 c)^{-1}\chi^{(5)}_{xxxxxy} I_p^2]$. Specifically, this gives

$$\chi^{(3)}_{xxxy} = \frac{\alpha_0 \hbar \epsilon_0 c}{16(\Delta/\Gamma)^2 I_{sat} k_B T_h} \frac{1}{k_B T_c} \left( \frac{1}{k_B T_c} - \frac{1}{k_B T_h} \right). \tag{5.31}$$

and

$$\chi^{(5)}_{xxxxxy} = \frac{\alpha_0 \hbar \epsilon_0 c}{8(\Delta/\Gamma)^2 I_{sat} I_c} \left( \frac{1}{k_B T_c} - \frac{1}{k_B T_h} \right). \tag{5.32}$$

From Eq. 5.31, one sees that the $\chi^{(3)}$ response is due solely to atoms in the hot component. This third-order response leads to four-wave mixing in which the pump beams scatter off the weak atomic density grating composed of atoms in the hot component \[143\]. In contrast, the $\chi^{(5)}$ response has a contribution from both the hot and cold components. These contributions appear with opposite signs because the transfer of population from the hot to the cold component (i.e., atomic cooling) increases the $\chi^{(5)}$ response due to the cold component at the expense of the hot component. This fifth-order response gives rise to six-wave mixing (SWM), where the pump beams simultaneously scatter of the atomic density grating and cool the atoms. While both
the hot and cold components contribute to the NLO response, the $\chi^{(5)}$ response is due almost entirely to the cold component because $T_c/T_h \approx 0.1$.

Beyond the region in which the Taylor series expansion given in Eq. 5.30 is valid [i.e., $I_c < I_p \ll I_{sat}(\Delta)$], the population in the cold state saturates (i.e., $f_c \approx 1$). This leads to a corresponding saturation of the $\chi^{(5)}$ response. Nevertheless, $\beta$ continues to increase linearly with $I_p$, which is in contrast to $\chi^{(3)}$ processes that increase only sub-linearly beyond saturation.

Figure 5.4 a shows the predicted and measured values of $\phi^{NL}$ as a function of $I_p$ for different values of $\Delta$. To determine the experimental value of $\phi^{NL}$ for a given measured reflectivity, I invert Eq. 5.29. As expected, $\phi^{NL}$ displays a distinct quadratic dependence on $I_p$ for $I_p \ll I_c$ that rolls over to a linear scaling for $I_p > I_c$ (see Fig. 5.4 b). To go one step farther, I look at the contributions to $\phi^{NL}$ from the hot and cold components separately. Figure 5.5 shows the dependence of $\phi_{c,h}^{NL}$ on $I_p$ for $\Delta = -5\Gamma$. I find that the prediction of Eq. 5.23 agrees reasonably well with the experimental results. The main source of discrepancy between the measured and predicted values stem from the simplified model used to determine $f_{c,h}$ [? ], particularly in the region where $I_p \ll I_c$ and the momentum distribution of the hot component becomes non-normalizable.

I next look at how the NLO response depends on $\Delta$ for fixed $I_p$, and find that $\beta$, $\chi^{(3)}$ and $\chi^{(5)}$ all scale as $(\Delta/\Gamma)^{-2}$. Figure 5.6 a shows the inverse quadratic dependence of $\beta$ on $\Delta$ predicted by Eq. 5.23. To compare with my experimental measurements, I scale the data shown in Fig. 5.4 b by the intensity-dependent portion of $\beta$ given in Eq. 5.23. This allows me to determine an effective, intensity independent nonlinear coupling strength for each detuning, which agrees well with the model predictions.

To put this dependence of $\beta$ on detuning in perspective, I note that the n-th order susceptibility for a two-level atom $\chi_{TLA}^{(n)} \propto (\Delta/\Gamma)^{-n}$. This implies that $\beta_{TLA} \propto (\Delta/\Gamma)^{-3}$.
Figure 5.4: Scaling of the nonlinear phase shift. a) Dependence \( \phi_{NL} \) on \( I_p \) for \( \alpha_0 L = 13.4(\pm 0.5) \) and \( \Delta/\Gamma = -3 \) (circles), -5 (squares), -7.3 (diamonds), -12.7 (up-triangle), and -18.8 (down-triangle). b) Expanded view of the data shown in a) for \( \Delta/\Gamma = -3 \) in the range \( I_p \ll I_c \), where \( \phi_{NL} \) scales approximately quadratically with \( I_p \). The points correspond to experimental data and the solid lines correspond to the prediction from Eq. 5.23.

Figure 5.5: Hot and cold components of the nonlinear phase shift. Dependence of the nonlinear phase shift due to the cold (circle) and hot (square) components on \( I_p \). Points represent experimental data and the solid lines correspond to the predictions of Eq. 5.23. The error bars represent typical measurement uncertainties.
Figure 5.6: Dependence of nonlinearity and figure of merit on detuning. a) Dependence of the nonlinear phase shift on $\Delta/\Gamma$. b) Dependence of the cross-phase-modulation figure of merit on $I_p$ for $\Delta/\Gamma = -3$ (filled circles), -4 (filled squares), -5 (open squares), -7.3 (diamonds), -12.7 (up-triangle), -15.5 (down-triangles), and -18.8 (open circles). Points represent experimental data and the solid lines correspond to the predictions of a) Eq. 5.23 and b) Eq. 5.33. For both panels, $\alpha_0 L = 13.4(\pm 0.5)$.

for FWM and $\beta_{TLA} \propto (\Delta/\Gamma)^{-5}$ for SWM. The relatively slow decrease of $\beta$ with $\Delta$ in my system stems from the independence of $I_c$ on the detuning; since the nonlinearity saturates at a fixed pump intensity for all $\Delta$, one can therefore realize large values of $\phi^{NL}$ for small $I_p$ even at large $\Delta$. Thus, my system enables me to simultaneously minimize the deleterious effects of linear absorption while only reducing slightly the nonlinear phase shift by working several linewidths away from the atomic resonance.

The figure of merit commonly used to quantify the tradeoff between increasing the NLO response and linear absorption of a material is given by $\zeta = \phi^{NL}/\alpha_\Delta L$. In the limit that $(2\Delta/\Gamma)^2 \gg 1$, I find that

$$\zeta = \frac{\alpha_0 \hbar \Gamma}{16} \frac{I_p}{I_{sat}} \left( \frac{f_c}{k_B T_c} + \frac{f_h}{k_B T_h} \right).$$

(5.33)

This result shows that $\zeta$ depends on $I_p$ but is independent of $\Delta$, which implies that one can realize large values of $\zeta$ simply by using larger pump intensities. Figure 5.6b shows that the predicted behavior of $\zeta$ agrees well with the experimental results over
5.6 Instability threshold

As discussed in Sec. 3.1.3, one expects MPSO to occur when the wave mixing gain becomes infinite. I calculate numerically the required threshold pump intensity $I_{\text{thresh}}$ by solving for value of $I_p$ at which $T,R \to \infty$ using Eqs. 5.28 and 5.29. Figure 5.7 a and b show how $I_{\text{thresh}}$ depends on the detuning and optical depth ($OD \equiv a_0L$). I find that the threshold intensity decreases with decreasing detuning and increasing optical depth. One can easily understand this behavior by noting that $\phi^{NL} \propto \psi(I_p)OD/\Delta^2$, where $\psi(I_p)$ describes the functional dependence of the NLO phase shift on pump intensity. If the instability occurs for a fixed value of $\phi^{NL}$ (as in the case of MPSO in a lossless Kerr medium, see Sec. 3.1.2), then one would expect $\psi(I_{\text{thresh}}) \propto \Delta^2/OD$.

This situation is modified slightly because the increase in linear absorption associated with increasing the optical depth effectively increases $I_{\text{thresh}}$, but I find that this effect is relatively small for $|\Delta/\Gamma| > 2$. 

Figure 5.7: Dependence of instability threshold on pump parameters. Dependence of $I_{\text{thresh}}$ on a) $\Delta/\Gamma$ for $OD = 5, 10, 40$ (from top to bottom) and b) $OD$ for $|\Delta/\Gamma| = 2, 3, 15$ (from bottom to top).
I next study how \( I_{\text{thresh}} \) scales with the detuning and optical depth. For \( \phi^{NL} \) as defined via Eqs. 5.23 and 5.24, one expects two different regimes to emerge. When \( I_{\text{thresh}} < I_c \), which typically occurs for large \( OD \) and small \( |\Delta/\Gamma| \), \( \psi(I_p) \propto I_p^2 \) and one expects \( I_{\text{thresh}} \propto \Delta/OD^{1/2} \). In contrast, when \( I_{\text{thresh}} > I_c \), \( \psi(I_p) \propto I_p \) and \( I_{\text{thresh}} \propto \Delta^2/OD \). This case occurs for smaller \( OD \) and larger \( |\Delta/\Gamma| \). To determine the theoretically-predicted scaling of \( I_{\text{thresh}} \), I fit the data shown in Fig. 5.7 a and b with a function of the form \( I_{\text{thresh}} = a(\Delta/\Gamma)^{b_\Delta} \) and \( I_{\text{thresh}} = a/OD^{b_{OD}} \), respectively. Figure 5.8 shows that \( I_{\text{thresh}} \) scales with \( OD \) and \( |\Delta| \) as described above. Namely, \( b_{OD} \approx 0.5 \) for \( |\Delta/\Gamma| < 3 \) and changes rapidly to \( b_{OD} \approx 1 \) for larger detunings, whereas \( b_\Delta \approx 2 \) for \( OD \leq 10 \) and transitions to \( b_\Delta = 1 \) larger values of \( OD \). Thus, the scaling of the instability threshold depends on the choice of system parameters. I compare these predictions to experimental data in Sec. 6.3.3.

Figure 5.8: Scaling of the instability threshold. Dependence of a) \( b_\Delta \) (i.e., scaling of \( I_{\text{thresh}} \) with \( \Delta \)) as a function of \( OD \) and b) \( b_{OD} \) (i.e., scaling of \( I_{\text{thresh}} \) with \( OD \)) as a function of \( |\Delta| \).
5.7 Summary

In this chapter, I developed a theoretical model of the steady-state NLO response of a cloud of cold, multilevel atoms in the presence of a pair of counterpropagating pump and probe beams. The interference of a pump and nearly-counterpropagating probe beam produces a dipole potential that causes the atoms to become localized. This atomic bunching corresponds to the creation of a density grating, which is phase-matched for coupling the pump and probe beams. For orthogonally-polarized pump beams, Sisyphus cooling enhances the loading of atoms into the phase-matched pump-probe lattice, resulting in a larger NLO response.

The combination of atomic cooling and localization also results in a novel scaling of the nonlinear phase shift with various parameters. Specifically, I show that $\phi^{NL} \propto \eta I_p (1 - T_c/T_h) \tanh(I_p/I_c)/\Delta^2 T_c$. The linear dependence of $\phi^{NL}$ on $\eta$ means that the contributions from the atoms add coherently. The dependence of $\phi^{NL}$ on $I_p$ indicates that a SWM process occurs for low pump intensities (where $I_p \ll I_c$ and $\phi^{NL} \propto I_p^2$), which saturates as $\phi^{NL} \propto I_p$ for $I_p > I_c$. Finally, I show that the slow decrease of $\phi^{NL}$ with $\Delta$ occurs because the characteristic intensity is independent of detuning and leads to large values of the cross-phase modulation figure of merit (e.g., $\zeta = 25$).

I verify my model by comparing its predictions to the experimental data shown in Ch. 4. The model, which contains no free parameters, agrees well ($\chi^2_{red} = 1.6$) with the measured data over a range of detunings and pump intensities. This agreement reinforces my interpretation of the NLO response in terms of dissipation-enhanced wave mixing via atomic bunching. In the next chapter, I study experimentally the system's behavior when no probe beams are present and the pump intensities are above the predicted instability threshold.
Chapter 6

The superradiant transition

In Chs. 4 and 5 I investigated experimentally and theoretically the nonlinear optical response of a cold atomic vapor driven by externally-applied optical fields in the limit of small optical gain. In this regime, finite perturbations to the system in the form of a macroscopic, injected signal beam lead to stable, steady-state behavior. In this chapter, I study the situation where the gain is large enough that intrinsic noise is sufficient to cause the system to go unstable. Above a critical pump intensity and optical depth, the vapor spontaneously transforms into a spatially self-organized state: a density grating forms. Scattering of the pump beams off this grating generates a pair of new, intense optical fields that act back on the vapor to enhance the atomic organization. While one can interpret this process in terms of either MPSO or superfluorescence (see Ch. 3), I refer to it as superradiance below in order to be consistent with the literature (see Sec. 3.2.1) and because of its similarities with SRYs.

I find that the superradiant light is nearly coherent, spatially multimode, and displays strong temporal correlations between the various modes. This instability occurs in the absence of an optical cavity and without requiring ultracold atomic temperatures. Under certain conditions, the system emits light continuously, which represents the first realization of steady-state superradiance. By characterizing the system’s response to various parameters, I compare my results with the theoretical models developed in Ch. 3. In addition, I discuss the potential applications of my system.

This chapter is based partly on Ref. [151].
6.1 Experimental setup

To study the system above the superradiant threshold, I use an experimental setup very similar to that described in Sec. 4.2. Briefly, I use a magneto-optical trap (MOT) to produce an anisotropic, pencil-shaped cloud of cold \((T = 30 \mu K)\) \(^{87}\text{Rb}\) atoms with an optical depth of \(OD = 20 – 30\) along the cloud’s long axis \([36]\). The cloud has a length \(L = 3\) cm and \(1/e\) diameter \(W = 430\) \(\mu\)m, which corresponds to a Fresnel number \(F = \pi W^2/(\lambda L) \approx 6\). I illuminate the atomic cloud with a pair of pump beams (intensity \(I_p\) and wave vectors \(\pm k_p\)) that counterpropagate at an angle \(\theta = 10^\circ\) relative to the trap’s long axis and are detuned by an amount \(\Delta\) below the \(5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)\) atomic transition (see Fig. 6.1b). I control the polarization of the pump beams (\(\hat{e}_{p1,p2}\)) using separate wave plates and polarizers (see App. B) and adopt the same naming convention that I used in Ch. 4 where \(\Sigma, \Xi, \Theta\) refer to the situation where the pump beams have linear orthogonal, linear parallel, circularly-counterrotating, and circularly co-rotating polarizations, respectively.

In contrast to the setup used in Ch. 4, I drive the vapor with only the pump beams (\(i.e.,\) I do not apply any probe beams). The interaction generates a pair of new, counterpropagating optical fields oriented along the vapor’s long axis (\(i.e.,\) in the endfire modes), which I refer to as signal and idler fields (intensities \(I_{s,i}\) and wave vectors \(k_{s,i}\),...
respectively). I record the temporal variation of the intensity of the superradiant light with a pair of matched photomultiplier tubes (PMTs). In addition, I image the spatial profile of the signal beam with a charge-coupled device (CCD) camera. I place a 200 mm lens in the path of the signal beam 33 cm beyond the end of the trap and position the CCD camera 35 cm from the lens. In this way, I image the far field such that the images represent the angular distribution of the emitted light.

In the experiment, I cycle between a cooling and trapping period (duration $\Delta t_{\text{MOT}}$ where only the MOT beams are on) and a superradiant period (duration $\Delta t_p$ where the pump beams are on) [151]. During the superradiant period, I either reduce or extinguish the intensity of the MOT beams. When I turn the MOT beams off, I use $\Delta t_{\text{MOT}} = 99$ ms and $\Delta t_p = 1$ ms. When I leave the MOT beams on at a reduced intensity, I use $\Delta t_{\text{MOT}} = 99$ ms and a superradiant period of variable length (up to 5 seconds, limited by electronics). I leave the MOT repump beams and magnetic fields on to ensure that atoms are not pumped into a dark state and to enable continuous cooling and trapping, respectively. I have verified that the MOT magnetic fields do not affect the superradiance when the MOT beams are off [84].

### 6.2 Observation of superradiance

In this section, I discuss the general characteristics of the emitted light for the case where I extinguish completely the MOT beams during the superradiant period. Figure 6.2 shows a typical time series of the measured values of $I_{s,i}(t)$, where I turn on the pump beams at $t = 0$. Initially, the atomic density distribution is uniform and the endfire modes are empty. Nevertheless, fluctuations in the density and electromagnetic vacuum modes act as the impetus that seeds the transition. As the atoms cool and organize in the presence of the pump beams, the signal and idler intensities grow
Figure 6.2: Temporal profile of superradiant emission and the corresponding pump depletion. a) Temporal dependence of $I_{s,i}$ after I turn the pump beams on at $t = 0$. After a dwell time, the fields grow exponentially (as shown in the inset), reach a maximum value of $I_{\text{peak}}$ at time $\tau_D$, and display a high degree of correlation $r_{s,i}(0) = 0.987$. After several additional bursts, oscillation terminates by $\sim 250 \, \mu s$. b) Intensity of the signal and pump beams during superradiance, where the pump depletion corresponds well to the power scattered into signal mode. The data corresponds to $I_p = 4(\pm 0.1) \, \text{mW/cm}^2$, $\Delta / \Gamma = -5(\pm 0.5)$, $OD = 30(\pm 1)$ and the $\kappa$ polarization.

exponentially at a rate $g$ [i.e., $I_{s,i} \propto \exp(g t)$] and reach a maximum value $I_{\text{peak}}$ after a time $\tau_D$. While I study the dependence of these quantities on various parameters in detail below, I find that $g \sim 100 \, \text{kHz}$, $\tau_D$ is typically on the order of 10-100’s of $\mu s$, and $I_{\text{peak}}$ can be as large as $\sim 1 \, \text{mW/cm}^2$ (i.e., 20% of the pump intensity). The generated fields act back on the vapor and strongly affect the ensuing dynamics. The details of the long-term behavior therefore vary from run to run, but typically consist of additional bursts of light with characteristic durations on the order of $10 - 100 \, \mu s$ due to atomic motion in the underlying optical lattice.

Figure 6.2 a also shows that the signal and idler fields display strong temporal correlations due to their mutual coupling. I quantify the degree of correlation using the cross-correlation coefficient

$$r_{s,i}(\tau) = \frac{\langle [I_s(t) - \bar{I}_s][I_i(t+\tau) - \bar{I}_i] \rangle}{\sqrt{\langle [I_s(t) - \bar{I}_s]^2 \rangle \langle [I_i(t) - \bar{I}_i]^2 \rangle}},$$

(6.1)
where $r_{s,i}(\tau) = 1 [r_{s,i}(\tau) = -1]$ indicates that the signal and idler modes are perfectly correlated (anticorrelated). Here, $\langle \ldots \rangle$ represents a time average and $A \equiv \langle A \rangle$. I find that the superradiant modes are highly-correlated, with $r_{s,i}(0) \sim 0.9 - 0.99$. The correlations arise because the generated fields are coupled to one another via the shared, self-generated density grating (see Ch. 5). This is in contrast to experiments carried out using standing-wave cavities, where the mirror boundary conditions necessarily impose correlations. In addition, this effect does not arise in SRyS driven with a single pump beam because the gratings responsible for scattering pump light into the signal and idler modes are separate from one another (see Sec. 3.2.2). While I have measured only the classical correlations between the signal and idler beams, similar wave-mixing schemes have been shown to produce quantum correlations between the emitted photons [? ]. Thus, I anticipate that this system may be useful for twin beam generation [51].

After a few hundred $\mu$s, the emission ends. In contrast to other superradiant systems [7, 43], this transience is not inherent to my configuration. Rather, superradiance terminates in my system because expansion of the cloud along the $\hat{y}$ direction reduces the atomic density and, therefore, the gain. When the gain falls below a threshold value, superradiance terminates. Figure 6.3a shows a simultaneous measurement of the signal beam intensity and the transmission of a weak probe beam through the atomic vapor in the $\hat{x}$-direction. After $\sim 250 \mu$s, the atomic density begins to decrease (i.e., transmission of the probe beam increases), which signals a corresponding decrease in the superradiant intensity. As further evidence for this effect, I find that the duration of the superradiance increases when I start out with more atoms, since the total number of atoms in the interaction volume remains above the threshold atom number for longer in this case (see Fig. 6.3b). This mechanism does not contradict the claim that the atoms are trapped in a 3D lattice, though, because the sequential
firing of the superradiant modes results in only weak average trapping along the $\hat{y}$ axis and ultimately allows the atoms to leave the interaction volume. I discuss my solution to this issue in Sec. 6.4.

I observe directly the transfer of power from the pump to the superradiant fields by measuring the intensity of both the signal beam and one of the pump beams after it has passed through the vapor (see Fig. 6.2 b). As the superradiant intensity grows, the pump intensity decreases (and vice versa). Because the pump beams are significantly larger than the emitted beams, I place an aperture in the pump beam path with a diameter equal to that of the signal beam (see additional discussion on the spatial distribution of the superradiant light below). In this way, I compare quantitatively the intensity of the two beams. One can see that the intensity of the pump beam begins to decrease immediately, whereas the signal beam intensity remains negligible for $\sim 20 \mu s$. During this time, the pump beams are cooling and bunching the atoms in the pump lattice, with most of the light scattered isotropically. Once superradiance begins, nearly all of the light scattered out of the pump beam shows up in the superradiant mode.
I look now at the spatial distribution of the emitted light. As predicted by Eberly and Rehler \[103\], my cylindrically-shaped vapor emits light predominantly along the endfire modes (\textit{i.e.}, I observe almost no light scattered along the $\hat{x}$ and $\hat{y}$ directions). As in MPSO, this counterpropagating geometry provides distributed feedback through the mutual coupling of the fields (see Sec. 3.1). In addition, it balances radiation pressure forces, which is necessary for continuous operation \[18\]. Nevertheless, the fact that the atomic vapor has a Fresnel number greater than unity implies that it supports the propagation of multiple diffraction-limited superradiant modes, where mode competition ultimately leads to emission along the direction in which the superradiant threshold is smallest at a given time.

Figure 6.4 shows the measured transverse intensity distribution of the signal beam $I_s(x, y)$ for three different MOT realizations. The generated light consists of multiple transverse spatial modes, which indicates the existence of atomic self-organization in all three dimensions. The interplay between mode competition and the random initial fluctuations that seed the instability causes $I_{s,i}(x, y)$ to vary from run to run \[122\] and represents the spontaneous breaking of continuous translational symmetry in the system. I therefore do not observe $\pi$ phase jumps in the superradiant field, which are characteristic of discrete symmetry breaking in a cavity-based system \[19, 46\]. My observation of multimode superradiance instead represents the first step toward realizing novel, controllable condensed matter systems involving phase transitions via emergent structure \[15, 20, 21\].

The angular width of each superradiant mode is $\theta_p = 3.1(\pm 0.4)$ mrad, corresponding to a diffraction-limited beam waist ($1/e$ field radius) of $158 \pm 20 \mu$m at the exit of the trap. While multiple modes appear in a single image taken by the CCD camera, this does not necessarily imply that multiple modes fire at once, since each image represents a temporal average over the entire run. In order to learn about the temporal
Figure 6.4: Transverse superradiant mode structure. Images of $I_s(x, y)$ in the far field for three experimental MOT realizations, where the $+$ symbol corresponds to light propagating along the $\hat{z}$-direction. The generated light consists of multiple spatial modes that vary from one run to the next. The angular width of each beam is $\theta_D = 3.1(\pm 0.4)$ mrad, corresponding to a diffraction-limited beam waist ($1/e$ field radius) of $158(\pm 20)$ $\mu$m at the exit of the trap. The dark vertical line in the image corresponds to dust on the surface of the camera.

Firing of the different modes, I insert an aperture along the signal beam path that only allows certain modes to pass and compare the resulting temporal intensity profiles of the signal and idler beams. I find that the temporal correlation between the signal and idler temporal intensity profiles decreases due to the absence of certain peaks in the signal beam time series. I therefore conclude that the mode structure is not fixed for a given run; rather, it changes in time as the atoms move in response to the dynamic optical lattice [152].

Before proceeding to a quantitative study of the superradiant transition, I consider why my system displays superradiance when similar investigations in thermal vapors above $T = 3$ $\mu$K have not. To this end, I look first at situations in which I do not observe superradiance in my system. For example, I find that superradiance does not occur if the pump beams are not on at the same time. I perform this experiment by alternately turning each pump beam off and on with a 500 ns duration. This time scale is much faster than both the superradiance turn on time and the average time it takes an atom
to move one grating period. On average, the atoms see balanced pump intensities
and experience no net radiation pressure force. The modulation therefore acts only
to remove the possibility of scattering involving both pump beams (e.g., multi-wave
mixing, Sisyphus cooling, and bunching in the pump lattice). Because superradiance
does not occur in the absence of these scattering processes, I conclude that they are
crucial to the instability.

On the other hand, modulating the intensity of the pump beams in unison by
$0.05 - 0.1I_p$ does not, in general, affect the system dynamics. I do notice a decrease in
$I_{\text{peak}}$, though, when the frequency of modulation is approximately equal to twice the vi-
brational frequency (i.e., $\omega_{\text{mod}} \sim 2\omega_{\text{vib}}$). In this case, parametric resonance occurs, in
which the atoms gain energy exponentially with time $[153]$. This heating overwhelms
Sisyphus cooling and inhibits the formation of a density grating.

Superradiance also requires that the frequencies of the pump beams be close to one
another. In the case where the pump beam frequencies are no longer degenerate, the
pump lattice is no longer stationary; instead, it moves with a velocity $v_p = \delta_p/k_p$. By
using two separate acousto-optic modulators to create a relative frequency difference
$\delta_p$ between the pump beams, I observe that $I_{\text{peak}}$ decreases monotonically with increas-
ing $|\delta_p|$ (i.e., increasing $|v_p|$). I interpret this reduction in the superradiant amplitude
as due to the fact that atoms load less efficiently into the moving pump lattice because
they have a shorter time in which to do so. Again, this indicates the importance of the
pump lattice in enabling superradiance.

Finally, I only observe superradiance for red-detuned pump beams (i.e., for $\Delta < 0$).
This red-blue asymmetry occurs because red lattices lead to a net cooling and
atomic localization at the intensity anti-nodes, whereas blue lattices result in atomic
heating and localization in the intensity nodes. The atoms in the red lattice therefore
experience a larger average electric field strength, which more efficiently drives the
nonlinearity for $I_p \ll I_{sat}$.[76]. In addition, the lower atomic temperatures in the red
lattice lead to an enhanced density grating contrast and scattering efficiency. Thus, the
situations in which superradiance does not occur are consistent with my interpretation
of the instability as being based on the NLO process described in Chs. 3 and 4. For the
remainder of this chapter, I assume that the pump beams have degenerate frequencies
tuned below the atomic resonance and constant, balanced intensities unless specified
otherwise.

It is therefore clear that the details of the pump beam configuration are paramount
to realizing superradiance. One crucial difference between my system and those used
in previous studies of superradiance is that I use near-resonant counterpropagating
pump beams, whereas previous investigations of SRyS have employed only a single,
far-detuned pump beam [7, 16, 43]. This has several important consequences. For ex-
ample, my beam geometry enables the superradiant modes to couple to one another.
This coupling, combined with the large optical path length of my vapor compared to
standard spherical MOTs (see App. A), leads to increased amplification of the superra-
diant modes as they propagate across the sample. Because the superradiant instability
requires that the gain exceed the loss in the system, this larger resulting gain enables
superradiance to occur in my thermal vapor, despite the fact that the decoherence (i.e.,
loss) rate due to thermal motion is substantially larger than in ultracold samples. In
addition to increasing the gain, proper choice of the pump beam polarizations also
effectively decreases the decoherence rate through providing atomic cooling and local-
ization (as discussed in Sec. 4.4.1). Thus, I overcome the severe temperature restric-
tions typically observed for superradiance in a free space system by simply including
two pump beams and using a dense, anisotropic atomic cloud.

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6.3 Characterization of superradiant emission

In this section, I investigate how the superradiant transition depends on system parameters and compare my results with the theoretical predictions of MPSO and SRyS.

6.3.1 Frequency of the superradiant emission

I look first at the frequency spectrum of the superradiant light. To perform this measurement, I split off some of the light from the same laser used to generate the pump beams and pass it through an acousto-optic modulator (AOM). This additional beam, which I refer to as the local oscillator (LO), is detuned by several MHz from the pump beam frequency. I then interfere the LO with the pump, signal, and idler fields separately on fast detectors. The detector output has an amplitude modulation corresponding to the frequency difference of the respective beams. I then take the Fourier transform of this modulated signal to determine the spectral content of each beam.

Figure 6.5a shows the amplitude spectrum (AS, i.e., the amplitude of the Fourier coefficient corresponding to a given frequency) for the pump, signal, and idler fields (from bottom to top, respectively), where the horizontal axis corresponds to the detuning from the pump beam frequency $\delta$. The superradiant fields are degenerate with the pump field. The FWHM (i.e., -6 dB) bandwidth of the pump beam is 12 kHz, which is due primarily to the finite duration of the measurement, but also includes noise from the AOM. The signal and idler bandwidths are slightly Doppler-broadened due to residual atomic motion, with typical values of $\sim 15 - 25$ kHz. Nevertheless, this is narrower than the central resonance observed via wave mixing (see Ch. 4.3), which indicates that superradiance involves primarily the coldest atoms [127].

By slightly imbalancing the intensities of the pump beams, I find that the frequency of the signal and idler beams shift away from the pump frequency in opposite direc-
Figure 6.5: Superradiant amplitude spectrum. a) Amplitude spectra for the pump, signal, and idler fields (from bottom to top, respectively) as a function of the relative detuning from the pump field. I offset the curves from one another to more easily distinguish them. a) (b) Amplitude spectra for the signal and idler fields when $I_{p1} > I_{p2}$ ($I_{p1} < I_{p2}$), where the signal field is on the left (right) and the idler field is on the right (left). The data was taken using the $\mathcal{K}$ configuration. The frequency resolution is limited to $\sim 6$ kHz by the finite duration of the superradiant signal.

In this case, the radiation pressure force leads to motion of the atoms along the $\hat{z}'$-direction and a corresponding Doppler shift of the scattered light. Figure 6.5 b (c) shows the resulting amplitude spectrum when $I_{p1} > I_{p2}$ ($I_{p1} < I_{p2}$), where the signal beam frequency is larger (smaller) than the pump beam frequency. In this way, one can tune the frequency of the superradiant light over over $\sim 30$ kHz.

If, instead, I misalign the intensity-balanced pump beams so that they are no longer counterpropagating, I find that the degenerate (Rayleigh) superradiance is replaced by non-degenerate (Raman) superradiance. Figure 6.6 shows the AS for Raman superradiance, where almost all of the power occurs at $\omega_{\text{vib}} = 140$ kHz. Gain for this process occurs via Raman scattering between differently-populated vibrational levels of the localized atoms (see Fig. 2.2). The FWHM bandwidth of the emitted light in this case is $\sim 20$ kHz, which is $\sim 10$ times narrower than one would expect for a free atom due to the Lamb-Dicke effect (see Sec. 2.3.1).
Figure 6.6: Raman superradiance. Amplitude spectrum of the signal beam for the case of pump beam misalignment, which leads to Raman superradiance between vibrational levels. The data was taken using the $\mathcal{R}$ configuration.

6.3.2 Superradiant growth rate

In order for superradiance to occur, the gain must exceed the loss in the system. I showed in Sec. 4.4.1 that I can measure the growth and decay rates below the superradiant transition by monitoring the system’s initial approach to (at rate $\gamma_g$) and departure from (at rate $\gamma_f$) steady-state in the presence of an applied probe beam, respectively. I find that the net growth rate $g \equiv \gamma_g - \gamma_f$ increases with increasing pump powers (see Fig. 6.7). Specifically, stronger driving enables atoms to load into the density grating faster, which increases $\gamma_g$. At the same time, the lower atomic temperatures for larger $I_p$ lead to reduced thermal motion-induced decoherence and cause $\gamma_f$ to decrease. This is in contrast to the situation considered by Inouye et al. [7, 123], where the temperature and loss rate were independent of $I_p$. In fact, $\gamma_f$ decreases more rapidly with $I_p$ than $\gamma_g$ increases in my system (see Fig. 4.12), which indicates that the suppression of decoherence plays an important role in my observation of superradiance. This situation is analogous to the case of SRyS in a cavity [43], where increasing the cavity finesse reduces the decoherence and makes superradiance possible.

For a particular value of the pump intensity, which I denote as $I_{\text{thresh}}$, the loss and
Figure 6.7: Dependence of growth rate on pump intensity. a) Dependence of the growth rate on the pump intensity in the vicinity of the superradiant transition. Circles (squares) correspond to the growth rate below (above) the transition, where \( I_{\text{thresh}} = 1.25 \text{ mW/cm}^2 \). b) Growth rate above the transition. Squares correspond to the experimental data and the solid line represents a fit given by \( 0.19(I_p - 1.2)^{0.47} \), where \( I_p \) is in mW/cm\(^2\). The data corresponds to \( \Delta/\Gamma = -5(\pm 0.5) \), \( OD = 30(\pm 1) \) and the K polarization.

I find that \( I_{\text{thresh}} = 1.25(\pm 0.2) \text{ mW/cm}^2 \) for the data shown in Fig. 6.7. Beyond this point, I determine the net growth rate by fitting the initial growth of the measured signal and idler intensities with a function of the form \( I_{s,i} \propto \exp(g t) \) (see the inset in Fig. 6.2a). Figure 6.7a shows that the net growth rate varies continuously through the transition. Above the transition, the growth rate continues increasing with increasing pump intensity. Motivated by the SRyS prediction that \( g \propto \sqrt{N I_p} \) (see Sec. 3.2.2), I fit the data to a function of the form \( a(I_p - I_{\text{thresh}})^b \) and find that \( a = 0.19(\pm 0.15) \mu \text{s}^{-1} \) and \( b = 0.47(\pm 0.08) \). Thus, the scaling with intensity agrees with that predicted in Eq. 3.38 to within the uncertainty of the fit. In addition, the observed amplitude of \( g \) is only a factor of 5 smaller than that predicted by Eq. 3.38 which assumes a sample with no initial momentum spread and therefore overestimates the expected growth rate for my thermal vapor. Thus, despite the differences in the physical systems considered, the underlying mechanism of Bragg scattering via a self-generated density grating yields
Figure 6.8: Dependence of growth rate on atomic number. Dependence of the superradiant growth rate on the \( OD \). The solid line corresponds to a fit given by 0.04(\( OD - 5.2 \))^{0.53}. The data corresponds to \( I_p = 5(\pm 0.1) \) mW/cm\(^2\), \( \Delta / \Gamma = -5(\pm 0.5) \), and the \( \kappa \) polarization.

qualitatively similar results.

I consider next the variation of the superradiant growth rate as a function of the number of atoms. Rather than using directly the atom number, I look at the resonant optical depth \( OD \equiv \alpha(0)L \propto N \) (see App. B.2.1), which provides the same information but is a more directly-measurable and experimentally-relevant parameter. For typical trap parameters, I find that \( OD/N \approx 5 \times 10^6 \). Figure 6.8 shows that superradiance only occurs above a threshold optical depth \( OD_{\text{thresh}} \). While \( OD_{\text{thresh}} = 5.2 \) for the data shown, the value depends on \( I_p \) and \( \Delta \) in general. Beyond the transition, \( g \) increases with increasing \( OD \), which indicates the collective nature of the process. I fit the data above \( OD_{\text{thresh}} \) with a function of the form \( g = a(OD - OD_{\text{thresh}})^b \). I find that \( a = 0.04(\pm 0.03) \) \( \mu s^{-1} \) and \( b = 0.53(\pm 0.28) \), which demonstrates that the scaling of \( g \) with \( N \) is consistent with the predictions of Eq. 3.38.
### 6.3.3 Threshold for superradiance

Having demonstrated that a distinct superradiant threshold exists, I study how $I_{\text{thresh}}$ and $OD_{\text{thresh}}$ depend on various system parameters. Figure 6.9a and b show the dependence of $I_{\text{thresh}}$ on $\Delta$ (for fixed $OD$) and $OD$ (for a fixed $\Delta$), respectively. I observe that $I_{\text{thresh}}$ increases for larger detunings since the atom-photon coupling strength is inversely related to $\Delta$. Alternatively, increasing $OD$ leads to a smaller value of $I_{\text{thresh}}$ due to the larger collective nonlinear response of the system in this case.

I compare the measurements with the predictions of my model by solving for the value of $I_p$ for which $R \to \infty$ in Eq. 5.29 (i.e., where the gain becomes infinite, see Sec. 5.4). The solid lines in Fig. 6.9 show the predicted dependence of $I_{\text{thresh}}$ for the experimental parameters used. In general, the superradiant threshold predicted in this way agrees with the observed value. Specifically, I find that $I_{\text{thresh}}(\Delta) \propto (\Delta/\Gamma)^2$, as expected. On the other hand, the predicted scaling $I_{\text{thresh}}(OD) \propto OD^{-1}$ appears to overestimate the reduction of the threshold intensity with increasing optical depth. This may be due to the increased importance of pump beam absorption at larger optical depths, which is not considered in the model.

I consider next how $I_{\text{thresh}}$ depends on the chosen polarization configuration for a fixed value of $\Delta$ and $OD$. Table 6.1 shows that the $\Lambda$ and $\Theta$ configurations lead to comparable threshold intensities, whereas the superradiance threshold for the $\Sigma$ configuration is approximately twice as large. On the other hand, I do not observe superradiance in the $\Xi$ configuration for any combination of available system parameters (i.e., $I_p < 30 \text{ mW/cm}^2$, $OD < 30$, and $\Delta > 2.5$). This result is consistent with the small observed NLO response of the system below the superradiant transition (see Fig. 4.6).

The lowest threshold that I have measured experimentally is $I_p = 1.1(\pm0.25) \text{ mW/cm}^2$ for $\Delta/\Gamma = -3$ and $OD = 20(\pm1)$, which is similar to that found for SRyS
Figure 6.9: Superradiant threshold. Dependence of the $I_{\text{thresh}}$ on a) $\Delta$ for $OD = 20(\pm 1)$ and b) $OD$ for $\Delta/\Gamma = -7(\pm 0.5)$ in the $\mathcal{K}$ configuration. The points represent experimental data and the solid line is the predicted value via Eq. 5.29 for $T_c = 2.5$ $\mu$K, $T_h = 25$ $\mu$K, and $I_c = 1.5$ mW/cm$^2$. The error bars represent typical measurement uncertainties.

Table 6.1: Polarization dependence of superradiant threshold

<table>
<thead>
<tr>
<th>$I_{\text{thresh}}$ (mW/cm$^2$)</th>
<th>$\mathcal{K}$</th>
<th>$\Sigma$</th>
<th>$\Xi$</th>
<th>$\Theta$</th>
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<tr>
<td>2.3</td>
<td>4.8</td>
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<td>2.7</td>
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in a BEC \[7\] and more than a factor of 10 times smaller than previously observed for MPSO \[10, 27, 80\]. This value of $I_{\text{thresh}}$ corresponds to a total input power of only $P_{\text{thresh}} = 400 \pm 80$ $\mu$W, where I use pump beams with a 5 mm diameter. In contrast to other recently-studied systems that require auxiliary beams (i.e., in addition to pump beams) to initially pre-condition the sample \[10, 80\], $P_{\text{thresh}}$ here represents the total power input to my system. In the ultimate limit, where the pump beams are focused to an optical cross section (i.e., $\lambda^2/2\pi$, where $\lambda = 780$ nm is the optical wavelength), the total number of photons required to observe superradiance is $n \approx I_{\text{thresh}}(\lambda^2/2\pi)/\hbar\omega g$. Because $g \propto \sqrt{I_p - I_{\text{thresh}}}$, I find that $n \propto I_p/\sqrt{I_p - I_{\text{thresh}}}$, which means that the minimum photon number $n_{\text{min}}$ occurs when $I_p = 2I_{\text{thresh}}$. I find that $I_{\text{thresh}} = 2$ mW/cm$^2$ and $g = 130$ kHz for $\Delta/\Gamma = -3$ and $OD = 20(\pm 1)$, which results in $n_{\text{min}} = 60$ photons.
This low threshold suggests that the system may be useful for all-optical switching or quantum information processing applications.

### 6.3.4 Peak superradiant intensity

I next look at how $I_{\text{peak}}$ depends on the pump intensity and optical depth (see Fig. 6.10 a and b, respectively). As expected from the discussion above, I observe no superradiant light when $I_p$ and $OD$ are below the threshold values for a given set of parameters. Immediately beyond $I_{\text{thresh}}$, Fig. 6.10a shows that $I_{\text{peak}}$ increases linearly with $I_p$ and can be as large as $0.2I_p$. For larger pump intensities (i.e., $I_p > 5\, \text{mW/cm}^2$ in the data shown), $I_{\text{peak}}$ levels off to $\sim 1.3\, \text{mW/cm}^2$. This rolloff of the superradiant intensity at larger $I_p$ is similar to that observed for SRyS in a BEC.

I observe direct evidence for the collective nature of the superradiant instability by considering the dependence of $I_{\text{peak}}$ on $OD$ (i.e., the number of atoms). Figure 6.10b shows that the peak intensity of the generated light increases superlinearly with $OD$. 

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**Figure 6.10: Scaling of the peak superradiant intensity.** Dependence of the $I_{\text{peak}}$ on a) $I_p$ for $OD = 20(\pm 1)$ and b) $OD$ for $I_p = 5(\pm 0.1)\, \text{mW/cm}^2$. The dashed lines in a) and b) are given by the equation $I_s = 331(I_p - 1.2)$ and $I_s = 104(OD - 5.2)^2$, respectively, where $I_p$ is in $\text{mW/cm}^2$ and $I_s$ is in $\mu\text{W/cm}^2$. The data corresponds to $\Delta/\Gamma = -5(\pm 0.5)$ for the $K$ polarization, and the error bars represent typical measurement uncertainties.
Figure 6.11: **Superradiant peak delay.** Dependence of $\tau_D$ on a) $I_p$ for $OD = 20(\pm 1)$ and b) $OD$ for $I_p = 5(\pm 0.1)$ mW/cm$^2$. The solid lines in a) and b) are given by the equation $\tau_D = 104(I_p - 1.2)^{-1/2}$ and $\tau_D = 17\log[2 \times 5 \times 10^6(OD - 5.2)]/\sqrt{OD - 5.2}$, respectively, where $I_p$ is in mW/cm$^2$ and $\tau_D$ is in $\mu$s. The data corresponds to $\Delta/\Gamma = -5(\pm 0.5)$ for the $\sigma$ polarization and the error bars represent typical measurement uncertainties.

Beyond $OD_{thresh}$. Specifically, I find that $I_{peak}$ scales quadratically with $OD$, as expected for superradiance. Thus, the observed dependence of the peak intensity on the system parameters are consistent with the SRyS model prediction that $I_{peak} \propto N^2 I_p$ when one takes into account a finite superradiant threshold [? ].

### 6.3.5 Superradiant delay time

The final parameter that I use to characterize the onset of superradiance is the delay time $\tau_D$ (i.e., the time at which the superradiant pulse reaches its first maximum). Figure 6.11a shows that $\tau_D$ is on the order of 100 $\mu$s and decreases with increasing $I_p$. This makes physical sense, as one expects the emission to reach its peak earlier when driven more strongly. I find that the data agrees with the semi-classical SRyS model, which predicts $\tau_D \propto I_p^{-1/2}$ beyond $I_{thresh}$ (see Eq. 3.39).

I consider also how $\tau_D$ scales with the number of atoms via the $OD$. Fig. 6.11b shows that $\tau_D$ decreases with increasing $OD$, where the fact that $\tau_D$ depends on the
number of atoms at all again confirms the collective nature of the emission. I compare the data to the SRyS model prediction that $\tau_D \propto \log(2N)/\sqrt{N}$ (see Eq. 3.39) and find that the observed data is roughly consistent with the expected trend.

### 6.4 Steady-state superradiance

As I discussed in Sec. 6.2, expansion of the cloud along the $\hat{y}$ direction reduces the atomic density and superradiance ends after a few hundred $\mu$s. I overcome this expansion and maintain steady-state superradiance by keeping the radial MOT beams on at reduced intensities during the application of the pump beams. The atoms can therefore continue to interact with the pump beams while remaining free to self-organize. This is directly analogous to a recently-proposed scheme for steady-state superradiance based on the rapid repopulation of a long-lived excited state in alkaline-earth-metals [3]. Rather than employing internal states, I use long-lived center-of-mass states and continuously drive atomic bunching more rapidly than the superradiant emission rate.

Using this scheme, I observe superradiance that persists for up to several seconds (limited only by the timing electronics), where the frequency of the generated light is always degenerate with that of the pump beams. In addition, the amplitude of the superradiant light is smaller than in the absence of the MOT fields (see Fig. 6.12 a and b) and the delay time is longer (typically closer to 1 ms). I understand these changes as due to two main effects. First, the presence of the MOT beams modifies the spatially-dependent polarization of the lattice responsible for superradiance, which affects the degree of atomic cooling and bunching. Second, the damping of the atomic motion caused by the MOT optical molasses reduces the atoms’ ability to respond to the underlying dynamic optical lattice.

After the initial period of exponential growth, the superradiant intensities fluctuate
Figure 6.12: Steady-state superradiance. Typical time series showing a) $I_s(t)$ and b) $I_i(t)$ in steady-state when the MOT beams are left on at a reduced intensity. c) The degree of second order coherence for $I_{s,i}$ as a function of the time delay of the signal $\tau$ for the signal (lower) and idler (upper, shifted for clarity) beams. Here, $I_p = 3(\pm 0.1)$ mW/cm$^2$, $OD = 25(\pm 1)$, and $\Delta/\Gamma = -5(\pm 0.5)$ for the $K$ configuration.

around an average value of $I_{s,i} = 55 \mu$W/cm$^2$ for the data shown in Figs. 6.12a and b. To quantify the intensity fluctuations, I calculate the degree of second-order coherence between the superradiant modes

$$g_j^{(2)}(\tau) = \frac{\langle I_j(t)I_j(t+\tau)\rangle}{I_j(t)^2}, \quad (6.2)$$

where $j = \{s, i\}$. This quantity is similar to the intensity autocorrelation $r_{j,j}$, but is defined in such a way that $g^{(2)}(0) = 1$ for a coherent field and $g^{(2)}(0) > 1 \ [g^{(2)}(0) < 1]$ for a bunched (anti-bunched) field. I therefore find that the light displays slight bunching at zero time-delay but is nearly coherent (see Fig. 3c), which is consistent with recent predictions on steady-state superradiance [? ]. In addition, the coherence time $\tau_c$ [defined as the $1/e$ decay time of $g^{(2)}(\tau)$] is typically several hundred $\mu$s, which is over 100 times larger than that observed for a cloud of unconfined atoms at comparable temperatures [84]. While the intensity coherence time corresponds to the collective decay time, Monte-Carlo simulations for steady-state superradiance shows
Figure 6.13: Correlation between signal and idler modes. a) Temporal variation of $I_{s,i}(t)$ over a short time scale to highlight the correlation of the signals’ amplitude fluctuations. b) Cross-correlation coefficient for the full time series shown in Fig. 6.12. The peak value occurs at $\tau = -15\,\mu s$, where $r_{s,i}(-15) = 0.85$. Here, $I_p = 3(\pm 0.1)\,\text{mW/cm}^2$, $OD = 25(\pm 1)$, and $\Delta/\Gamma = -5(\pm 0.5)$ for the $K$ configuration.

that the amplitude of the electric field is coherent for times on the order of $N\tau_c$ [? ]. This suggests that my system could function as an ultra-stable mirrorless, superradiant laser for optical metrology. This long coherence time, combined with the instability’s low threshold, also indicates its potential as a quantum memory [4] or quantum logic element [52].

As in the case of transient superradiance described in the previous sections, the signal and idler fields show strong correlations for the duration of the steady-state emission. Figure 6.13a shows $I_{s,i}(t)$ together over a short time period, where the relationship between the intensity variations are evident. Again using the cross-correlation coefficient to quantify the similarity between the signals, I find that the peak value of $r_{s,i}$ is typically greater than 0.8. This peak occurs for $|\tau| < 15$, where the lead or lag between the signal and idler fields varies from run to run. Thus, it results from the initial fluctuations responsible for the superradiant transition, rather than any intrinsic asymmetries in the experiment.
6.5 Summary

In this chapter, I showed that a cold, thermal vapor undergoes a transition to a superradiant state under certain conditions. This transition coincides with the breaking of continuous translation symmetry in the system as the initially-uniform vapor evolves into a spatially self-organized state. Simultaneous with the onset of this spatial order, the pencil-shaped gas scatters the incident pump light coherently along the endfire modes. In contrast to previous works, I observe superradiance without using an optical cavity and for initial atomic temperatures that are ten times larger than previously observed.

I find that the superradiant instability demonstrates a finite threshold, below which no organization occurs. I showed that the threshold corresponds to the point at which gain is exactly balanced by loss in the system, and depends on the pump detuning, intensity, and the optical depth (i.e., atom number) of the sample. The magnitude of the superradiant threshold and its scaling with various system parameters are well-described by the predictions in Ch. 5 based on the system’s below-threshold behavior.

Above the threshold, I observe sequential bursts of superradiant light that occupy multiple spatial modes centered around the trap’s long axis. The light is degenerate with the pump beam frequency, but can vary depending on various system asymmetries. The superradiant emission grows exponentially at first and reaches a peak intensity after ~ 100 µs. I characterize the initial period of superradiance by studying the dependence of the temporal growth rate, intensity, and arrival time of the first intensity peak. By comparing the experimental data with the semi-classical model for SRyS developed in Ch. 3, I find that the theoretical predictions describe qualitatively the observed scaling of the measured quantities with various system parameters. This agreement is not completely unexpected, since the majority of the differences between
the below-threshold behavior in my system and that considered in the SRyS model occur for \( I_p < I_c \), whereas superradiance typically occurs for \( I_p > I_c \). In particular, I find that \( g \propto \sqrt{I_pN} \), \( I_{\text{peak}} \propto I_p(N)^2 \), and \( \tau_D \propto \log(N)/g \) above the superradiant transition.

After this initial pulse, emission persists for several hundred microseconds. The counterpropagating superradiant modes demonstrate a high degree of temporal correlation due to their coupling via the atomic density grating. Superradiance ultimately ends due to a reduction of the atomic density in the interaction volume. By using additional beams to weakly confine the atoms in the radial direction, I showed that superradiance can occur continuously (i.e., for several seconds). The light in this steady-state regime is slightly bunched and has a coherence time of several hundred \( \mu s \). This represents the first realization of free-space steady-state superradiance in a cold vapor, and relies on the fact that my beam geometry combines pairs of balanced optical fields (to eliminate radiation pressure effects) and a dissipation channel (to mitigate recoil-induced heating).

The unique properties of my system suggest a variety of possible applications. For example, it could act as a source of entangled or intensity-squeezed light [51]. Also, the combination of a low superradiant threshold and long coherence times imply the possibility of using it to realize a quantum memory [4] or quantum logic element [52]. In addition, the system’s capacity to support multiple spatial modes makes it an excellent candidate for studying continuous-variable quantum information protocols based on spatial multimode entanglement [23, 24], low-light-level all-optical switching [27, 84], and multidimensional optical soliton formation [139]. Finally, the self-phase-matching nature of the collective instability may provide a simple path toward the efficient generation of tunable short-wavelength light [154].
Chapter 7

Conclusions

In this thesis, I describe a novel nonlinear optical process that occurs in a driven cloud of cold atoms. For sufficiently large nonlinear interaction strengths, the system undergoes a phase transition to a spatially-ordered superradiant phase that persists in steady-state. In this chapter, I review the main results presented in each chapter, and discuss future directions and applications of my work.

7.1 Summary

The first three chapters of this thesis serve to motivate my research and provide necessary background for interpreting my results. In Chapter 1, I first introduce the concept of collective light-matter interactions, where interactions between the scatterers cause an entire ensemble to act in concert as a single entity. After reviewing previous work on this topic, I argue that a cloud of cold atoms represents an excellent system in which to study such effects because of the possibility for manipulating the atomic center-of-mass states.

In order to quantitatively describe how one can control these states, I introduce the fundamental physics of light-matter interactions in a non-collective (i.e., single atom) regime in Ch. 2. In this case, atoms interacting with an incident light can become polarized and experience a net force when the optical field varies spatially. Two different types of basic forces result. The radiation pressure force, which is based on absorption-spontaneous emission cycles, is dissipative and occurs when the phase of
the field varies spatially. In contrast, the dipole force is conservative (i.e., described in terms of a potential) and arises when the amplitude of the field varies spatially. The situation in which one creates an array of dipole potentials is known as an optical lattice. By combining optical pumping and spontaneous emission with multilevel atoms in an optical lattice, one finds that atomic cooling and localization can occur. For the case of Sisyphus cooling in a dissipative, one-dimensional lattice, one finds that the r.m.s. value of the atomic kinetic energy temperature decreases rapidly in the vicinity of the so-called décrochage point. More careful studies show that, as the atoms move in this lattice, the gas transforms into a non-thermal system composed of a cold, bound (i.e., spatially localized) and hot, largely unbound component.

I go beyond the situation of scattering by single atoms in Ch. 3 where I introduce in greater detail the physics behind two well-known collective light-matter interactions called as MPSO and superradiance. Using a nonlinear optics approach, I describe how MPSO can occur when a pair of counterpropagating optical fields are incident on a Kerr material. When the nonlinear phase shift reaches a value of $\phi_{NL}^{NL} = \pi/2$, the system’s gain effectively becomes infinite and infinitesimal perturbations due to vacuum field fluctuations result in the emission of new, macroscopic fields. Using cold atoms, one can realize MPSO with significantly smaller pump intensities than in a warm vapor with all other parameters equal due to the reduced role of Doppler broadening.

I then introduce superradiance as a synchronization transition, where an ensemble of emitters becomes entrained with one another via their mutual interactions such that a macroscopic, many-atom dipole moment forms. In this case, the atoms radiate collectively such that the intensity (duration) of the emitted light is $N$ times larger (smaller) than observed for non-collective scattering. This phenomena occurs both for spontaneous emission in a gas of stationary atoms as well as via atomic recoil in a vapor of cold atoms (known as SRys). In the latter case, one can view directly the transition
to the superradiant state in terms of the spontaneous onset of spatial organization of the vapor. To describe this process quantitatively, I review a semi-classical model for SRyS. In addition, I point out that superradiance becomes indistinguishable from MPSO when it is viewed in terms of wave mixing between matter and optical waves. This analogy assists me in the analysis of my work.

In Ch. 4, I present my experimental study of a novel wave mixing process in an optically-thick cloud of cold atoms interacting with a pair of counterpropagating pump and probe beams. I show that atomic bunching in the lattice formed by the interference of the pump and nearly-counterpropagating probe beams leads to Bragg scattering of pump light into probe modes. While bunching-induced wave mixing has been observed before, including dissipation via Sisyphus cooling enhances the strength of the NLO response, leads to a unique dependence of the nonlinearity on material parameters, and effectively increases the coherence time of the system. Using this approach, I observe a fifth-order susceptibility \( \chi^{(5)} = 1.9 \times 10^{-12} \text{ (m/V)}^4 \), which is the largest ever reported by seven orders of magnitude and arises from the simultaneous cooling and bunching of atoms in the optical fields. In addition, I find that the resulting nonlinearity scales as \( (\Delta/\Gamma)^{-2} \), which allows me to realize large nonlinear phase shifts (i.e., on the order of \( \pi \)) with pump intensities of less than \( I_{sat} \) and with small linear absorption. I quantify this result in terms of the cross-phase-modulation figure of merit, and show that the nonlinear phase shift can be 25 times larger than the optical depth, which exceeds previous observations by nearly a factor of 100.

To describe the results presented in Ch. 4, I develop in Ch. 5 a simple model for my system that combines the physics associated with atomic motion in a dissipative lattice and nonlinear wave mixing. In this approach, I consider only the linear internal atomic response to the incident optical fields, but allow for the formation of a spatially-dependent atomic distribution. In the presence of Sisyphus cooling, the
model correctly predicts the lowest-order nonlinearity to be fifth-order in the applied fields. Solving a set of coupled wave equations for the probe fields, I derive an analytic expression for the nonlinear response and intensity of the output fields. Writing this result in terms of experimentally-measurable quantities, I show that the model matches quantitatively my observations with no free parameters. This agreement therefore confirms my interpretation of the nonlinearity as due to scattering from an atomic density grating enhanced by dissipation.

In Ch. 6, I show that my system displays a superradiant transition that leads to the generation of new optical fields when the gas is illuminated only by the pump fields. In contrast to previous studies that required ultracold atomic temperatures (i.e., $T < 3 \mu K$) or high-finesse cavities, I realize superradiance in free-space using “tepid” temperatures of $T \sim 30 \mu K$. By connecting this behavior with the bunching-induced wave mixing process described in Ch. 4, I conclude that the system’s transition to a collective, superradiant phase is associated with the onset of atomic spatial organization (i.e., the spontaneous formation of a density grating). I show that the transition only occurs when the wave-mixing gain exceeds the loss in the system, which requires that the pump intensity exceed a threshold value that depends on the number of atoms and detuning used. The lowest threshold intensity that I observe is 1 mw/cm², which is a factor of 50 smaller than previously observations in other systems $[7, 80, 87]$.

I also characterize the light emitted by the vapor in the superradiant phase. The light propagates primarily along the vapor’s long axis, although multiple, randomly-oriented spatial modes arise over a solid angle of a few mrad about this direction. The intensities of the counterpropagating modes demonstrate temporal correlations as large as $r_{si} = 0.987$, can be as large as 20% of the pump beam intensities, and scale nonlinearly with $N$ (as expected for a collective process). While the superradiant emission typically ends after several hundred $\mu$s, I show that I can stabilize the su-
perradiant phase such that emission persists in steady-state (i.e., for several seconds) by imposing an additional, weak confining potential. In this case, I observe that the light displays only slight bunching (i.e., is nearly coherent) and has a coherence time of several hundred µs.

7.2 Future directions

The main results presented in this thesis are: 1) the observation of a new NLO mechanism that yields the largest fifth-order susceptibility ever measured and 2) the realization of steady-state superradiance in a cloud of cold atoms. To interpret and appreciate these accomplishments, I synthesized phenomena typically associated with a variety of different fields, including atomic and condensed matter physics, nonlinear optics, and nonlinear dynamics. It is therefore not surprising that my work has potential applications in both fundamental science and applied technologies in a number of different areas. In this section, I discuss some of the work that is currently underway to extend my previous research as well as other future research directions.

One possible application of my results is in the area of improving existing models of atomic dynamics in dissipative optical lattices. For example, no model to date has correctly predicted the experimentally-observed independence of the décrochage intensity on the lattice detuning. The large NLO response that I observe and its sensitivity to atomic temperature through the hot and cold components represents a new, in-situ technique for characterizing the atomic ensemble in the presence of such a lattice, which could help inform future models.

In addition, while the theoretical model that I develop in Ch. 5 describes well the observed steady-state experimental results, a complete description of the light-matter dynamics in the collective regime (i.e., where the back-action of the probe fields be-
comes important) requires a non-perturbative approach. Such a model would necessarily combine previous work on atomic motion in dissipative, multi-dimensional optical lattices [71] with models of collective scattering via atomic motion in conservative lattices [128]. One could use this model to study the connection between the statistical properties of the emitted light and the fluctuations responsible for seeding the transition. In addition, it is possible that one can use such a model to find new physics not present in previous investigations and motivate future experimental studies.

It is also possible to use my system to study already-proposed collective, many-body phenomena that have not yet been realized experimentally. For example, one might be able to test the theoretical predictions of Gopalakrishnan et al. [47] concerning emergent crystallinity or look for the nonequilibrium Dicke phase transitions proposed by Bastidas et al. [100]. One can also study various effects associated phase transitions in general, such as symmetry-breaking [46], defect formation [101], and multistability [100, 155].

Another interesting research direction involves using my system to realize all-optical switching at the single-photon level. To this end, graduate student Bonnie Schmittberger and I have demonstrated that modifying slightly the orientation of the pump beams leads to a related but distinct light-matter instability with improved performance [84]. By shining small (150 µm) pump beams along the axis of the anisotropic cloud of cold atoms, we find that the generated light produces patterns in the plane transverse to the pump beams (see Fig. 7.1). The confining dipole potential created along the \( \hat{x} - \hat{y} \)-plane by the pump beams effectively traps the atoms and enables superradiance to persist for over 2 ms in the absence of an additional potential. Thus, it may be possible to realize steady-state superradiance without an external potential by further focusing the pump beams.

In addition, we find that the power required to observe the instability in this geom-
Figure 7.1: Pattern formation in cold atoms. a) Beam geometry for pattern-forming instability, where $\theta' \sim 5$ mrad. b) Transverse optical patterns, including i) a nearly full ring and ii) a six- iii) four- and iv) two-spot pattern, for $I_p = 15$ mW/cm$^2$, $\Delta/\Gamma = -8$. The central spot in the images corresponds to residual pump light.

Geometry can be as low as 6 $\mu$W, which is almost 100 times smaller than I observed using off-axis pump beams. A similar configuration has previously been used to demonstrate all-optical switching with a warm vapor using several hundred photons $[27]$. In this work, Dawes et al. observed that decreasing the threshold of the pattern-forming instability leads to a corresponding reduction in the number of photons needed to actuate the switch. Therefore, our observation of a threshold that is 100 times smaller than that observed by Dawes et al. suggests that we could potentially realize switching at the single-photon level.

Beyond studying all-optical switching with few photons, extending the results presented in this thesis to the quantum regime represents another exciting avenue of research. The long coherence times and multimode nature of my system may be useful for optical metrology, as a quantum memory, as a source of entangled photons, or for realizing continuous variable quantum information protocols. To verify the feasibility of these applications, one would need to study the quantum properties of the superradiant light. For example, one could measure the degree of squeezing in a given superradiant mode. Alternatively, one can investigate whether entanglement exists be-
tween photons in different modes. In this way, one could utilize my system as a novel source of quantum light.
Appendix A

Anisotropic MOT physics

The results described in this dissertation require a sample of cold atoms with a large optical path length. While one can achieve this using a large, spherical magneto-optical trap (MOTs), anisotropic MOTs offer several important advantages. For example, effects related to the rescattering of light within the MOT, which result in atomic heating and can compete with the NLO wave mixing processes described in this thesis, are greatly suppressed in such a trap [156]. My choice of a pencil-shaped MOT also enables me to avoid issues related to absorption-induced trapping [36]. In addition, this MOT geometry affects the characteristics of the generated superradiant light and leads to directional emission along the endfire modes, as discussed in Ch. 3. In this chapter, I describe the physical mechanisms underlying atomic cooling and trapping in general, as well as new effects that arise for an anisotropic MOT.

This chapter is based partly on Ref. [36].

A.1 Introduction to MOT physics

Magneto-optical trapping combines optical and inhomogeneous magnetic fields to extract kinetic energy from an atomic ensemble and confine the atoms in a region of space. Over the last twenty years, MOTs have become the most widely-used method for cooling and trapping neutral atoms. The first demonstrations of magneto-optical trapping were performed by initially cooling an atomic beam and then turning on the trap [157]. By trapping atoms directly from a low pressure, room temperature atomic
vapor, Monroe et al. [158] made cold atoms available for almost any laboratory due to the simplicity and robustness of the setup. As discussed above, the standard spherical MOT geometry is not ideal for studying NLO wave mixing; I therefore develop a highly-anisotropic, pencil-shaped MOT based on the work of Vengalatorte et al. [35] that is more suitable for my purposes. While the basic mechanisms underlying the anisotropic MOT are the same as for the case of a spherical MOT, additional phenomena arise due to the large obtainable optical depths along the trap’s long axis that affect its operation.

### A.2 Fundamentals of magneto-optical trapping

The operation of a MOT relies on the scattering (or radiation pressure) force, which results from repeated stimulated absorption and spontaneous emission events [53]. When an optical field with wave vector $k$ is incident on an atom, the net momentum imparted to the atom via $N$ absorption-emission events is (on average) simply $Nhk$; the contribution due to spontaneous emission averages to zero because of the spatial isotropy of the process (see Fig. [A.1]). Atoms thus experience a force in the direction of the incident radiation that depends on the rate of the absorption-emission events. For a two-level atom, one finds that $F_{sp} = \hbar k \Gamma \rho_{ee}$, where $\rho_{ee}$ is the probability that the atom is in the excited state and $\Gamma$ is the natural linewidth (i.e., the excited state population decay rate). For a stationary two-level atom, one can solve for $\rho_{ee}$ in steady state (see Sec. 2.1) and find that

$$F_{sp} = \frac{\hbar k \Gamma}{2} \frac{\Omega^2/2}{(\Gamma/2)^2 + \Delta^2 + \Omega^2/2},$$

(A.1)

where $E = E_0 \cos(\omega_L t)$ and $\Omega = \mu E_0 / \hbar$ are the amplitude and the Rabi frequency of the incident optical field, $\Delta = \omega_L - \omega_0$ is the detuning of the laser ($\omega_L$) from the atomic
The scattering force saturates to a maximum value of \( |F_{sc}| = \hbar|k|\Gamma / 2 \) for larger \( \Omega \).

To describe the process of cooling and trapping in a MOT, I consider the case of an atom moving in the presence of counterpropagating optical fields (with wave vectors \( k_\pm = \pm k\hat{z} \)) and a position-dependent magnetic field that varies linearly along the \( \hat{z} \)-direction as \( B = Az \). The total scattering force in the low intensity limit (\( \Omega^2 / \Gamma^2 \ll 1 \)) is \( F_{MOT} = F_{sc}^+ + F_{sc}^- \), where [53]

\[
F_{sc} = \pm \frac{\hbar k\Gamma}{2} \frac{\Omega^2 / 2}{(\Gamma / 2)^2 + \Delta_\pm^2 + \Omega^2 / 2},
\]

(A.2)

corresponds to the force resulting from each incident beam. The effective detuning seen by the atoms moving with velocity \( v \) at location \( z \) is given by

\[
\Delta_\pm = \Delta \pm \omega_D \pm \omega_Z,
\]

(A.3)

where \( \omega_D = -k \cdot v \) is the Doppler shift, \( v \) is the atomic velocity, and \( \omega_Z = \mu'B \cdot \hat{z} / \hbar \) is the Zeeman shift. Here, \( \mu' = (g_e m_e - g_g m_g)\mu_B \) is the effective magnetic moment for
the transition, \( g_{\epsilon g} \) denotes the Landé g-factor, \( m_{\epsilon g} \) is the magnetic quantum number, and \( \mu_B \) is the Bohr magneton.

In the limit that \( \omega_D, \omega_Z \ll \Delta \), one finds that

\[
F_{\text{MOT}} = -\beta v - \kappa z, \tag{A.4}
\]

where the friction coefficient

\[
\beta = -\frac{\hbar k^2 \Gamma \Delta \Omega^2}{[(\Gamma/2)^2 + \Delta^2 + \Omega^2/2]^2} \tag{A.5}
\]

and the effective spring constant for the trap \( \kappa = \mu' A \beta / \hbar k \). In this regime, \( F_{\text{MOT}} \) leads to damped harmonic motion of the atoms with a damping rate \( \Gamma_{\text{MOT}} = \beta / M \) and oscillation frequency \( \omega_{\text{MOT}} = \sqrt{\kappa / M} \). For typical MOT parameters (i.e., \( A \approx 10 \text{ G/cm} \) and \( \Delta \approx \Gamma \)), the atomic motion is overdamped, with \( \omega_Z \sim \text{few kHz} \) and \( \Gamma_{\text{MOT}} \sim \text{few hundred kHz} \). To better understand the physics underlying the two terms on the right hand side of Eq. [A.4], I consider below the cases of a moving atom in the absence of a magnetic field and a stationary atom in a magnetic field.

In the absence of a magnetic field, Eq. [A.4] reduces to \( F_{\text{MOT}} = -\beta v \), which represents a damping force for red detuned (i.e., \( \Delta < 0 \)) optical fields. This damping arises because a moving atom “sees” the frequency of the counterpropagating (copropagating) beam shifted closer to (further from) resonance due to the Doppler effect (see Fig. [A.2]a). The atom preferentially absorbs light from the field that is closer to the atomic resonance, which results in a net average force that is antiparallel to the atom’s velocity. This process, known as Doppler cooling, effectively extracts kinetic energy from the atoms via spontaneous emission. In addition, this situation is often referred to as optical molasses because the atoms move as if in a viscous fluid. This molasses force,
Figure A.2: Doppler cooling in an optical molasses. a) The effective Doppler shift $\omega_D$ experienced by a moving atom in the presence of counterpropagating laser beams with frequency $\omega_L$ tuned $\Delta$ below an atomic resonance $\omega_0$. The optical frequency viewed by an atom moving toward (away from) a given beam is increased (decreased) relative to the case of a stationary atom. b) The velocity-dependent molasses force given by Eq. A.2. The blue (red) dashed line corresponds to the scattering force imparted by the beam propagating along $\hat{k}$ ($-\hat{k}$), and the solid line corresponds to the total resulting force $F_M$. The dotted line with slope $-\beta$ shows the region of small velocities in which $F_{MOT}$ is approximately linear.

though, only holds on average (i.e., over many scattering events). Random fluctuations in the scattering force result in atomic heating and lead to a non-zero equilibrium temperature on the order of the Doppler temperature $k_B T_D = \hbar \Gamma / 2$, where $T_D = 146 \ \mu K$ for $^{87}Rb$. This optical molasses results in an atomic localization in momentum space only - the atoms are cooled but remain at whatever location they come to rest.

I next consider the case of a stationary atom in a gradient magnetic field, which corresponds to the second term in Eq. A.4. Here, one finds a restoring force $F_{MOT} = -\kappa z$ that pushes atoms toward the location of zero magnetic field. Because the atoms preferentially move to a particular location, this effectively implements spatial trapping. One can understand this trapping force in a manner quite analogous to Doppler cooling: rather than shifting the apparent light frequency via velocity-dependent detuning, the atomic energy levels are altered by the spatially-dependent magnetic field. For an
atom with multiple excited states, the presence of a magnetic field leads to a Zeeman shift that lifts the degeneracy of the magnetic sublevels. By properly choosing the polarization of the counterpropagating optical fields, one can create a force that has a spatial dependence defined by the magnetic field variation.

To demonstrate this effect more explicitly, consider the case of a $J = 0 \rightarrow J = 1$ transition. A linear magnetic field of the form $B = Az$ produces shifts in the $m = \pm 1$ levels as shown in Fig. A.3a, where I assume that the Landé $g$-factors are positive. Because of this shift, an atom is more likely to absorb a photon from a red detuned beam with $\sigma^+$ ($\sigma^-$) polarization for $z < 0$ ($z > 0$). This results in a net force $F_z = -\kappa z$ that guides atoms to the region in which $B = 0$ (see Fig. A.3b). This trapping mechanism is completely commensurate with the formation of optical molasses, so that atoms can be cooled and trapped with the same beams and in the same interaction volume.

Because the scattering force relies on many (typically thousands) absorption and spontaneous emission events to produce effective cooling and trapping, the atoms must be able to continuously interact with the MOT optical fields. To achieve this in a real, multi-level atomic system, I require that atomic species have a cycling transition, where an atom always returns to its original state after an excitation-spontaneous emission event. To this end, I use the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ transition in $^{87}$Rb, where spontaneous transitions to the $5P_{1/2}$ and $5S_{1/2}(F = 1)$ states are forbidden due to parity and momentum conservation, respectively (see Fig. A.4). The requirement of a slight red detuning of the molasses beams opens the absorption-emission cycle by providing a small probability that the atoms will be excited to the $5P_{3/2}(F = 2)$ state, from which they can then decay to the $5S_{1/2}(F = 1)$ state. To protect against atoms getting pumped into this dark state, I include a repump beam that is tuned to the $5S_{1/2}(F = 1) \rightarrow 5P_{3/2}(F = 2)$ state.
While one would expect atomic temperatures on the order of $T_D$ for cooling via optical molasses, an additional mechanism typically plays a role in MOT operation and leads to sub-Doppler temperatures. Sisyphus (or polarization gradient) cooling, as discussed in Ch. 2, arises due to the combination of spatially-varying AC Stark shifts and optical pumping imposed by the incident optical fields on the atoms as they move through the trapping region. This leads to atomic temperatures that are typically on the order of $T \approx 10T_r$, where the recoil temperature $T_r = (\hbar k)^2/2Mk_B$ and $T_r = 361$ nK for $^{87}$Rb.

One can easily extend the description of the 1D MOT operation given above to three dimensions by including additional laser beams and a magnetic field with a gradient in multiple directions. The simplest example of this involves using three pairs of counterpropagating, orthogonally-oriented laser beams and a magnetic field with a
Figure A.4: Rubidium D2 line energy scheme. Energy level scheme for the D$_2$ line in $^{87}\text{Rb}$, including the relative cooling and trapping beam frequency $\omega_{\text{trap}}$ and the repump frequency $\omega_{\text{repump}}$ [159].
constant gradient in all three directions. This results in isotropic cooling and trapping to produce a nearly spherical cloud of atoms. In contrast, I realize an anisotropic MOT by using a quadrupolar magnetic field that varies linearly along the transverse ($\hat{x}$ and $\hat{y}$) directions but is independent of the $\hat{z}$-direction (see App. B for more details). This configuration leads to cooling in 3D, but only traps the atoms in the transverse plane (i.e., the atoms remain free to move along the $\hat{z}$-direction).

A.3 Collective MOT physics

The theory presented above assumes that the interaction of each atom with the magnetic and optical fields is independent of the presence of the other atoms. For magneto-optical traps with sufficiently large optical depths, this assumption breaks down and one can observe additional, collective mechanisms that modify the trap’s operation. One example of this phenomenon results from radiation trapping, where light scatters multiple times from atoms within the trapping volume. This can lead to a repulsive radiation pressure force that causes the atomic cloud to break up into a central ball of atoms surrounded by an orbiting ring when the experimental system contains asymmetries (e.g., in the magnetic field or laser beam profiles) [160]. Even for perfectly aligned beams, vortical radiation pressure results in an irregular atomic density distribution [161]. While these mechanisms play an important role in high-density spherical MOTs, the effects of radiation trapping in my anisotropic MOT are negligible because the geometry enhances the probability that a scattered photon will escape the cloud without interacting with another atom [156]. Instead, a new mechanism, which I refer to as absorption-induced trapping, determines the trap size and location [36].
A.3.1 Introduction to absorption-induced trapping

One can understand absorption-induced trapping in the following way: an atom illuminated by a pair of imbalanced, counterpropagating optical beams preferentially absorbs photons from the more intense beam and therefore sees a net force in the direction of the stronger beam’s wave vector. While a fixed intensity imbalance leads to a constant force and atomic acceleration, stable trapping can occur if the intensity imbalance of the beams is spatially-varying such that the atom’s trajectory is confined to a finite volume \textsuperscript{[162]}. In the case of my anisotropic MOT, the large optical depth along the trap’s long axis causes attenuation of the cooling and trapping beams propagating along the $\hat{z}$-direction. A spatially-dependent intensity imbalance therefore results, which leads to a spatial localization of the atoms that limits the maximum obtainable MOT optical depths.

A.3.2 Theoretical description of absorption-induced trapping

To study this mechanism and its effects on the dynamics of trap formation, I developed a Monte Carlo simulation of the trapping and cooling process. Figure \textsuperscript{[A.5]} shows the magnetic-field and laser beam geometry used for the numerical model as well as the initial (blue) and final (red) positions of the atoms. I consider a magnetic field that is independent of $z$ and has a constant gradient along the $r$-direction. The optical fields consist of one pair of counterpropagating Gaussian beams along the $r$-direction (radial beams) and another along the $z'$-direction (longitudinal beams, which are inclined by an angle $\theta$ relative to the $z$-axis). While I ignore coherences due to optical pumping effects, I explicitly incorporate the polarization-selective absorption necessary for spatial trapping due to the Zeeman effect via the probabilities associated with the absorption of a photon from the four laser beams.
Figure A.5: Absorption-induced trapping model beam geometry. The model beam geometry with radial and longitudinal beams shown. Also, the relative direction and magnitude of the magnetic field are shown. The points and circles represent the initial and final atomic spatial distribution.

I model the dynamics of 500 individual atoms with random initial positions and velocities (below a cutoff of $v = 3.5 \text{ m/s}$), which are weighted by a Maxwell-Boltzmann distribution with $v_{rms} = 350 \text{ m/s}$. At each time step, which is chosen to be long enough that the probability of absorption is nearly unity, each atom absorbs a photon from one of the laser beams and receives a momentum kick along that beam’s wave vector. I calculate the probability of excitation of an atom due to a monochromatic field using the rotating-wave and dipole approximations. The probabilities corresponding to the absorption of a photon from the right/up (+) and left/down (-) radial ($r$) and longitudinal ($l$) beams are given by

$$p_r^{\pm}(r, z) = \frac{1}{4}(\Omega_r^{\pm})^2 \left[ (\Gamma/2)^2 + 1/2(\Omega_r^{\pm})^2 + (\Delta \mp k_r \cdot v \mp \mu_B B / \hbar)^2 \right]$$

(A.6)

$$p_l^{\pm}(r, z) = \frac{1}{4}(\Omega_l^{\pm})^2 \left[ (\Gamma/2)^2 + 1/2(\Omega_l^{\pm})^2 + (\Delta \mp k_l \cdot v)^2 \right]$$

(A.7)

where $v$ is the atom’s velocity and $k_{r,l}$ and $\Omega_{r,l}$ are the radial and longitudinal beams’
wave vectors and Rabi frequencies, respectively. Finally, \( B = Ar \) represents the magnetic field along the direction parallel to the trapping beams, where \( A \) corresponds to the field gradient along the \( r \) direction.

As the beams pass through the gas of atoms, their intensities are exponentially attenuated according to Beer's law as

\[
\frac{I(z')}{I(0)} = \exp \left( -\int_0^{z'} \eta[\zeta] \sigma \hat{k} \cdot d\zeta \right) = \exp(-N[z']\sigma), \tag{A.8}
\]

where \( \eta[\zeta] \) is the density of atoms at \( \zeta \), \( \sigma \) is the effective absorption cross section, \( \hat{k} \cdot d\zeta \) is a small interval along the beam's propagation direction, and \( N[z'] \) corresponds to the total number of atoms in the beam path from the entrance of the trapping region to position \( z' \). The total optical depth of the sample is therefore given by \( OD = N[L]\sigma \).

The Rabi frequency for each beam is defined as

\[
(\Omega^\pm_r)^2 = \frac{2I^\pm_r|\mu|^2}{c\epsilon_0\hbar^2}\exp\left(-z^2/w^2_r\right), \tag{A.9}
\]
\[
(\Omega^\pm_l)^2 = \frac{2I^\pm_l|\mu|^2}{c\epsilon_0\hbar^2}\exp\left(-r'^2/w^2_l\right)\exp\left(\mp N[z']\sigma\right), \tag{A.10}
\]

where \( w_{r,l} \) and \( I_{r,l}^\pm \) are the beam waists and incident intensities (i.e., at the entrance to the interaction volume) of the counterpropagating radial and longitudinal beams, respectively. I define the intensity as \( I = (1/2)\epsilon_0 c|E_0|^2 \), where \( c \) is the speed of light in vacuum and \( \epsilon_0 \) is the permittivity of free space.

I explicitly include the effects of absorption via the dependence of the cooling beam Rabi frequencies on \( N[z'] \). I choose an effective single atom absorption cross section such that the maximum achievable optical depth is on the order of 10 (i.e., comparable to that observed experimentally). It is through the inclusion of this position-dependent attenuation factor that my model provides a mechanism for intensity-dependent spatial...
trapping.

I calculate a normalized probability ($\tilde{P}_i^j$) to determine the likelihood of absorption from each beam as

$$\tilde{P}_i^j = P_i^j / (P_i^+ + P_i^- + P_i^+ + P_i^-),$$  \hspace{1cm} (A.11)$$

where $i = (r, l)$, $j = (+, -)$, and $P_i^j$ represents the probability for the absorption of a photon from any one of the laser beams. With these normalized probabilities, I choose a random number in the range $[0, 1]$ to select which beam is absorbed by each atom. Thus, at each timestep, the positions and velocities of the atoms are updated appropriately, and the integrated density at each point is calculated. I then use this density in the next step to determine the longitudinal beam attenuation (i.e., I ignore absorption along the radial direction because OD < 1 in this plane). If atoms pass out of the trapping region, I consider these atoms to no longer participate in trapping; when this occurs, I randomly place a corresponding number of atoms inside the trapping region.

### A.3.3 Observation of absorption-induced trapping

Using my anisotropic MOT setup, I experimentally investigate the effects of absorption-induced trapping. When the longitudinal beams propagate along the $\hat{z}$-direction (i.e., $\theta = 0$), I find that I am limited to producing a dense, short trap at a well-specified location. The exact location of the trap is reproducible, depends on the frequency and relative intensities of the incident beams, and can occur anywhere along the region of the magnetic-field minimum (i.e., the $z$-axis). By modifying the relative radiation pressure of the counterpropagating longitudinal beams, I quickly ($< 1$ ms) transfer atoms from one end of the trapping region to the other.

Figure A.6 illustrates the effects of an imposed cooling-beam intensity imbalance
Figure A.6: Effect of intensity imbalance on trap formation. The effect of the cooling beam intensity imbalance on trap formation. a1)-a4) Experimental longitudinal atomic density profiles that result when $\theta = 0$, OD = 10, $\Delta = -1.5\Gamma$, $I^+_I^- = 25:4$, 5:3, 3:5, 4:25. b1)-b4) The result of numerical simulations modeling the effects of intensity imbalance when $\theta = 0$, OD = 10, $\Delta = -\Gamma$, $I^+_I^- = 3:2$, 5:4, 4:5, 2:3.

on the resulting trap locations. I show the experimentally measured and numerically simulated longitudinal atomic density distributions in Fig. A.6(a) and (b), respectively, where the ratio $I^+_I^-$ decreases from top to bottom. I find that trapping can occur anywhere along the magnetic-field minimum over a range of several cm and that the trap location is determined by the cooling beam intensity imbalance. This behavior cannot be accounted for by a model that ignores the absorption of the cooling beams, as a beam imbalance simply acts to provide a net acceleration for the atoms, but would not result in a shifted, stable equilibrium position. Alternatively, in the absence of absorption-induced trapping, my model predicts a uniform density over the entire trapping region for perfectly balanced beams. Thus, the excellent agreement between the experimental results and model predictions when I consider absorption-induced trapping confirms its importance to my anisotropic MOT.

I achieve a long, uniform cloud of atoms by altering the orientation of the longitudinal beams. By rotating the longitudinal beams by an angle $\theta = 10^\circ$, I avoid the effects of absorption-induced trapping by exploiting the fact that the trap is optically thin.
Figure A.7: Effect of pump beam rotation on trap formation. a) Experimental longitudinal atomic density profiles of traps obtained for $I_1^+ = I_1^-$ at $\theta = 0^\circ$ and $10^\circ$. b) Numerically modeled atomic density profiles for equal intensity cooling beams at $\theta = 0^\circ$ and $10^\circ$.

along the radial direction (OD<1). The longitudinal beams still produce effectively an optical molasses to cool the atoms along the $z$-axis due to their balanced wave vector components along this direction, but now they only traverse an oblique cross section of the trap and undergo minimal attenuation. Figure A.7 shows the longitudinal atomic density profiles that result when the cooling beams are of equal intensity and oriented at an angle of $\theta = 0^\circ$ and $10^\circ$. While a short trap located in the middle of the trapping region forms when $\theta = 0^\circ$, the trap fills uniformly (i.e., with < 20% variation) the entire trapping region for $\theta = 10^\circ$. Furthermore, I find experimentally that this beam alignment results in a much more robust trap that is more reproducible and less affected by beam misalignments and intensity imbalances. The model results shown in Fig. A.7(b) demonstrate that, despite the presence of absorption-induced trapping forces, rotating the cooling beams results in a trap that is spatially unconfined along the magnetic-field minimum.
A.4 Summary

In this chapter, I showed that the basic mechanism behind magneto-optical trapping is the scattering force. By applying a pair of counterpropagating beams to a cloud of thermal atoms with properly-chosen detunings and polarizations in the presence of a spatially-varying magnetic field, the atoms cool and become trapped at a particular location. In this way, one can realize a dense cloud of cold atoms with a shape specified by the externally-applied magnetic field.

For sufficiently dense clouds, additional, collective effects arise that modify the operation of the MOT. While my choice of an anisotropic, pencil-shaped MOT geometry avoids such effects due to radiation trapping, I find that another mechanism, known as absorption-induced trapping, occurs and can reduce the achievable optical depth of the cold vapor. Because of this effect, I observe a maximum trap length of only a few mm. To improve the trap, I perform an experimental and numerical study of absorption-induced trapping and demonstrate that a simple rotation of the MOT optical fields can result in a uniform (< 20% variation) trap over several cm. In App. B I discuss in detail the experimental realization and characterization of my MOT.
Appendix B

Experimental apparatus

The experimental results presented in this dissertation make use of a highly-anisotropic cloud of cold Rubidium atoms that I developed. In this chapter, I discuss the experimental components required to create the anisotropic MOT as well as the techniques used to characterize the resulting system.

This chapter is based partly on Ref. [36].

B.1 Experimental setup

The last couple of decades have lead to a dramatic simplification in terms of the size and complexity of MOT systems. Today, MOT setups can be found everywhere from sophisticated, cutting-edge experimental setups to undergraduate labs and include free-space and integrated, on-chip architectures [163]. The basic components for building a MOT consist of lasers, magnets, a source of atoms, and a vacuum system. In addition, I require a timing and detection system for conducting NLO studies.

B.1.1 Optical System

A Titanium:sapphire (Ti:saph) crystal laser provides the light for the MOT cooling and trapping beams. The Ti:saph laser produces approximately 600 mW of linearly polarized light with a linewidth of \( \sim 200 \text{ kHz} \). Figure B.1 shows a schematic of the experimental laser system. I use a thick glass plate to split off \( \sim 10\% \) of the light and send it to a saturated absorption spectroscopy setup so that I can set absolutely
the frequency of the laser relative to an atomic resonance line. I typically tune the
frequency $50 - 60$ MHz below the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 3)$ transition in $^{87}$Rb
($\lambda = 780.24$ nm). I then use another thick glass plate to split off an additional $\sim 10\%$
of the light and send it to the pump-probe spectroscopy setup (described later). The
light then passes through a fixed-frequency 40 MHz acousto-optic modulator (AOM, IntraAction AOM-403) so that I can quickly ($\sim$ few hundred ns response time) turn the
MOT beams off and on. I send the first-order diffracted beam, which is upshifted by
40 MHz, through a 50/50 beam splitter (BS) to separate the light into distinct radial
and longitudinal MOT beams.

In general, the spatial uniformity of the intensity profiles of these MOT beams
is critical to the trap’s operation because intensity imbalances in the trapping region
impair the MOT’s performance (see App. A). I create large, spatially-uniform beams
combining a telescope to expand the beams beyond the required size and an aperture
to block the unwanted light. For the longitudinal beam, I use this technique to first
increase its diameters to $\sim 3$ cm and then pass it through an iris, which reduces the
beam size to $\sim 1$ cm. This beam then passes through the trapping region, after which I
use a mirror and quarter wave plate to retro-reflect the beam and rotate its polarization
by 90°. In this way, I achieve sub-Doppler cooling via the resulting 1D lin⊥lin lattice.

I split the radial beam into a pair of beams via a 50/50 BS. I use a telescope to
increase the beams’ diameters to $\sim 6$ cm, then pass them through an aperture so that
the outgoing beam has an elliptical intensity profile with a semimajor and semiminor
axis of 1.5 cm and 1 cm, respectively (defined in the plane perpendicular to the beam’s
wave vector, with the semimajor axis along the $z$-axis). The beams then pass through
quarter wave plates such that the radial beams are oppositely circularly polarized upon
entering the trapping region.

Figure B.1 shows the beam geometry in the MOT interaction region. The longitu-
Figure B.1: Schematic of the MOT optical system. Diagram showing the optical system involved in creating the MOT. The inset shows the beam geometry of the radial and longitudinal MOT beams in the interaction region of the optical cell, where the polarization of each beam is indicated.
dinal beams counterpropagate in the \(x-z\)-plane at an angle of 10° from the \(z\)-axis to produce an optical molasses that primarily cools the atoms along the \(\hat{z}\)-direction. The radial beams counterpropagate in the \(x-y\)-plane and enter the trapping region at an angle of 45° relative to the \(y\)-axis. They then reflect off the mirror base of the optical cell to enable both cooling and trapping in the \(x-y\)-plane.

I generate a repump beam using a distributed feedback (DFB) laser (Eagleyard, EYP-DFB-0780-00080-1500-TOC03-0000), which has a linewidth of \(\sim 10\) MHz and a maximum power of 80 mW. I send several mW to a saturated absorption spectroscopy setup, which I use to help tune the laser frequency to the \(5S_{1/2}(F = 1) \rightarrow 5P_{3/2}(F = 2)\) transition. The rest of the power from the DFB laser enters the MOT setup via the BS used to split the Ti:saph light into the longitudinal and radial beams, as shown in Fig. B.1.

I use additional pump and probe beams to perform NLO experiments. Because I endeavor to study multi-photon resonances that are typically only 10’s of kHz wide (i.e., narrower than the linewidths of the available lasers), I require that the pump and probe beams come from the same laser. I accomplish this by using AOMs to control the frequency and amplitude of the light originating from a separate, external cavity diode laser (ECDL, Toptica DLpro) (see Fig. B.2h). The ECDL has a maximum power of \(\sim 80\) mW, a linewidth of a few hundred kHz, and is actively locked to an atomic resonance line via a saturated absorption spectroscopy setup. I split the output of the ECDL into two paths with a BS and use a pair of matched AOMs (IntraAction, tunable 230 MHz) to separately control the pump and probe beams. This allows me to scan independently the frequencies of the beams over 80 MHz as well as turn them off and on with a response time of \(< 100\) ns. The phase noise between the two AOMs is sufficiently small (few kHz) over a short (\(\sim 100\) \(\mu\)s) time period that I can obtain pump-probe spectra by scanning the frequency of the probe AOM directly. For experiments requiring a fixed
frequency difference between the pump and probe beams for longer times ($\geq 1$ ms),
the phase noise in the AOMs leads to a frequency jitter of $> 50$ kHz, which is broader
than the resonance features that I study. Thus, I use two alternative techniques to
enhance the relative frequency stability.

The main contribution of this frequency jitter comes from the RF electronics used
to drive the AOMs (i.e., not from thermal noise within the acousto-optic crystal itself).
Thus, when I require a fixed, nonzero frequency detuning between the beams, I
amplify (using Mini-Circuits ZHL-1-2W) the output of a high-precision frequency syn-
thesizer (Novatech 2940, stability of 2 ppm and step size of 1 Hz) to drive the AOMs.

Figure B.2: Schematic of optical system for NLO studies. a) Schematic of the optical
system used to generate the NLO pump and probe beams via the DLpro laser and
matched AOMs. b) Beam geometry used to conduct wave mixing experiments.
Alternatively, when I require stable, frequency degenerate pump and probe beams with independently controlled amplitudes, I split off part of the pump beam and double pass it through an AOM. By nearly retroreflecting the beam back into the AOM and choosing the diffracted beam with no net frequency shift, the phase noise of the AOM cancels while enabling me to modulate the probe amplitude separately from that of the pump beam.

I then couple the pump and probe beams into single mode optical fibers (Thorlabs P3-830A-FC-2) using free space fiber couplers (Thorlabs PAF-X-11-B), with typical coupling efficiencies of 60-80%. The beams emerge near the MOT setup, as shown in Fig. B.2b. At the output fiber coupler, I use a polarizer and retarder (i.e., either a quarter or half wave plate) to set the beams’ polarizations. I use a 300 mm lens to focus the probe beam to a $1/e$ field radius of $150(\pm 15) \mu m$ at the location of the atomic sample (corresponding to a Rayleigh range of 9 cm). The probe beam propagates along the $z$-axis. I use a polarizing beam splitter (PBS) to split the pump beam into a pair of counterpropagating beams that intersect the probe beam at the location of the trap and propagate at an angle of $10^\circ$ relative to the $y-z$-plane. The pump beams are collimated and have beam waists of 3-5 mm, depending on the particular output fiber coupler.

B.1.2 UHV system

Creating a MOT by capturing atoms from the low-velocity tail of a thermal vapor requires that one operates at low pressures, since residual thermal atoms present in the cold atom trapping region can negatively impact trap performance by heating the atoms and forcing them out of the trapping volume via atom-atom collisions. Advances in ultra-high vacuum (UHV) systems, which are defined as having pressures on the or-
der of 10\(^{-9}\) Torr, have enabled the realization of simple and robust MOT setups. The UHV system that I use consists of vacuum pumps, metal parts (e.g., flanges, gaskets, valves, etc.), a glass optical cell, and an electrical feedthrough connected to an alkali metal dispenser. I discuss the latter two components in detail below; here, I focus on the techniques used to achieve UHV, and the resulting system performance.

I use a compact UHV system, which allows me to minimize the pumping requirements while still achieving the desired pressures. Before attaching the components, I first inspect the parts to make sure that all knife edges, which are crucial for creating good seals, are sharp. I then clean the metal parts with acetone and methanol in sequential ultrasonic baths. If any part has (or may have) oil on it, I wash it first with a soap (usually Alcanox) and water mixture, rinse it thoroughly with water, then place it in the ultrasonic cleaner with distilled water. I then wrap the parts with aluminum foil until I am ready to use them. I perform all cleaning and assembly while wearing latex gloves.

Once clean, I assemble the UHV system. The pumping station, consisting of a rotary and turbomolecular (Varian, Turbo-V70LP, 70 l/s) pump, attaches to the UHV system via a pair of valves and a flange. An ion pump (Duniway, Varian style, 8 L/S) attaches to the assembly via an additional flange, and remains permanently connected. A metal pipe connects the ion pump to the electrical feedthrough on one end and the optical cell on the other. When I detach the pumping station, the entire MOT UHV system has dimensions of 12 × 20 × 20 in.

I use several pumping and heating stages to achieve UHV pressures. The rotary pump, which is a positive displacement pump that operates by creating a pressure differential to remove the air, can reduce the system's pressure to 10\(^{-4}\) Torr. I connect the rotary pump exhaust to an additional hose to ensure that no oil contaminates the lab environment. Immediately after turning on the rotary pump, I turn on the turbo pump
to prevent the backstreaming of oil into the vacuum system. The turbo pump uses multiple stages of rapidly spinning, angled turbine rotors to give gas molecules successive momentum kicks causing them to leave the pumping region. After pumping for several hours, the pressure typically drops to $10^{-6}$ Torr. To remove residual water and hydrocarbons adsorbed on the internal surfaces of the vacuum components, I bake out the system by wrapping it with heat tape while continuing to pump. I control the heat tape temperature with variable autotransformers (VARIACs) and measure the temperature using K type thermocouples. In particular, I make sure to use that separate heat tapes in the vicinity of the optical cell and glass-to-metal transition to avoid cracking due to the differing thermal expansion properties of the glass, metal, and bonding epoxy (see Sec. B.1.3 for more details). I bake out the ion pump and metal components located far from the cell at temperatures of $\sim 225^\circ$C, and gradually decrease the temperature to $\sim 75^\circ$C at the optical cell.

After baking out the system for approximately 48 hours, I allow the system to cool down. Once the pressure reaches $10^{-7}$ Torr, I turn on the ion pump. This pump uses a strong electric potential to ionize atoms that happen to enter the pump. A magnetic field then accelerates the atoms such that they collide with and remain stuck to a chemically active cathode. The pump may turn on and off several times as it first begins pumping, but the pressure should eventually monotonically decrease such that, after $\sim 12$ hours of pumping, the pressure is $< 10^{-8}$ Torr. I then run the getters to remove additional contaminants (see Sec. B.1.5), which allows me to achieve pressures as low as $9 \times 10^{-10}$ Torr. The pressure typically increases slightly to around $5 \times 10^{-9}$ Torr when I operate the MOT, but decreases quickly once I turn off the getters. In addition, while it is certainly not ideal, I have observed reasonably good MOT operation with background pressures up to $5 \times 10^{-8}$ Torr.
B.1.3 Optical cell

I create and study the cloud of cold atoms in an optical cell that I built specifically for my anisotropic MOT. The key characteristics of this cell are that it allows for good optical access to the trapping region, has a mirror base for realizing a mirror-MOT geometry, and can be placed sufficiently close to the MOT magnets to allow for efficient trapping.

To build the cell, I first clean all components with methanol and acetone in an ultrasonic cleaner for approximately 20 minutes. The cell walls consist of four, anti-reflection-coated BK7 windows (1.25 × 1.25 × 0.125 in., Esco Part #ZQ712125C) arranged in the geometry shown in Fig. B.3. To bond the elements of the cell together, I use TRA-BOND Epoxy BA-2116, which is a space-grade adhesive with minimal outgassing and a room-temperature curing time of approximately 24 hours. While this epoxy is very robust and long-lived, exposing it to acetone causes it to degrade and gives rise to leaks. The bonding procedure involves drying the surface, applying the glue, and applying pressure to the interface for several hours. After gluing the cell walls, I bond a front-surface gold mirror (1.5 × 1.5 × 0.125 in., Esco Part #ZQ715125C) to the bottom of the cell. The top of the cell consists of a thick glass window (1.75 × 1.75 × 0.25 in., Esco Part #Q717250) with a 1 in. diameter circular hole drilled into it. To create this part, I purchased the rectangular window and contracted University Research Glassware (in Chapel Hill, NC) to drill the hole. I glued the top directly to the cell walls. The cell connects to the rest of the UHV system via a glass-to-metal transition that I bond directly to the top surface of the cell.

To further prepare the cell for installation in the UHV system, I attach it directly to the pump station (as described in App. B.1.2), wrap it in heat tape, and bake it out under vacuum to a max temp of 75 °C. After baking for 48 hours, I typically measure
Figure B.3: **Schematic of the optical cell.** Diagram showing the optical cell from the a) top and b) front. c) The cell attaches to the UHV system via a glass-to-metal transition and vacuum flange. d) Photograph of the optical cell along with the MOT magnets immediately below.
pressures as low as $10^{-7}$ Torr, which are limited only by the minimum achievable pressure of the turbopump. Finally, I attach the cell to the rest UHV system.

### B.1.4 Magnets

While the standard MOT utilizes only current-carrying conductors to generate the requisite trapping magnetic fields, I build on the approach taken Vengalattore et al.\cite{35} and use soft-ferromagnetic materials magnetized via externally-applied currents. This approach provides several technical advantages over those based on using current-carrying wires only. It allows one to create larger magnetic fields with smaller currents, which enables for lower power operation (e.g., for integrated MOT devices) and, in my setup, obviates the need to cool the magnets. In addition, one can create larger field gradients farther from the surface of the magnets, which increases the trapping volume and reduces the influence of surface effects on the MOT operation.

The basic element that I use to generate the magnetic field consists of a high-susceptibility ferromagnetic core ($\mu$-metal, CO-NETIC AA Sheet CP025-30-14) wrapped with a solenoid. The cores are rectangular with dimensions $h = 5$ cm, $l = 5$ cm, and $t = 0.64$ mm, and the solenoid consists of $N_{\text{turns}} = 200$ turns of 20 gauge copper transformer wire. Because the aspect ratio of the core is large, I assume that its magnetization is uniform and model it as a pair of current sheets with infinite length, a height $h$, and a separation $w$ (see Fig. B.4a).

The resulting magnetic field is given by \cite{164}

\[
\begin{align*}
B_x &= B_0 \log \left[ \frac{(x + w/2)^2 + y^2 (x - w/2)^2 + (y + h)^2}{(x + w/2)^2 + (y + h)^2 (x - w/2)^2 + y^2} \right] \\
B_y &= 2B_0 \tan^{-1} \left\{ \frac{hw \left[ y(y + h) - x^2 + w^2/4 \right]}{\left[ x^2 + y(y + h) + w^2/4 \right]^2 + h^2(x^2 - w^2/4) - x^2w^2} \right\}
\end{align*}
\] (B.1)
where I take the origin to be at the top center of the magnet. Here

\[ B_0 = \frac{c_0 \mu_0 N_{\text{turns}} I}{4\pi h} \quad \text{(B.2)} \]

is the amplitude of the magnetic field and \( I \) is the current in the wire. The dimensionless constant \( c_0 \) represents the enhancement of the field due to the ferromagnetic core. I experimentally determine \( c_0 \) for my magnet by measuring the magnetic field directly \((\text{i.e., using a Gaussmeter})\) above the center of a magnet and comparing it to Eq. \( \text{B.1} \). For the data shown in Fig. [B.4] I find that \( c_0 = 2300. \)

By placing two such magnets with opposite magnetizations a distance \( s \) from one another, I produce a magnetic field that is independent of the position along the \( z \)-direction and points along the \( \hat{x} \)-direction in the plane of symmetry \( \text{[35]} \). To be useful for magneto-optic trapping, I require a magnetic field with a gradient that changes sign at a specified location \((\text{i.e., has a zero in the desired trapping region})\). I therefore combine two such pairs of magnets (an inner and outer pair) with opposite relative
magnetizations and separations \( s_i \) and \( s_o \) to produce the desired quadrupolar magnetic field in the \( x - y \)-plane (see Fig. B.5a). This field has a magnetic field zero that occurs in the plane of symmetry (which I refer to as the \( x = 0 \) plane) and at a distance \( y_0 \) above the tops of the magnets. In my experiment, I use \( s_i = 1.1 \) cm and \( s_o = 3.2 \) cm and find that the variation of the magnetic field along the \( z \)-direction at \( y_0 \) is \( \Delta B \approx 0.05 \) G. I control the value of \( y_0 \) by adjusting the relative currents provided to the inner (\( I_i \)) and outer (\( I_o \)) pairs of magnets. Since the MOT forms at the location of the magnetic field minimum, I can easily measure the dependence \( y_0 \) on the ratio \( I_o/I_i \) by imaging the location of the resulting atomic cloud. Figure B.5b shows that the height of the trap can vary over approximately 5 mm, which is limited below by the mirror base and above by geometric restrictions (i.e., the top of the cell blocks the radial MOT beams). In addition, I adjust the magnetic field gradient by varying the ratio \( I_o/I_i \), which allows me to vary the depth of the magnetic trap. I typically choose \( I_o = 930 \) mA and \( I_i = 850 \) mA, which results in a MOT that is 8 mm above the top of the magnets (i.e., approximately 3 mm above the top of the mirror base) with a gradient of \( \sim 10 \) G/cm. I use this particular choice of currents in the following data unless specified otherwise.

### B.1.5 Getters

I use alkali metal dispensers (or getters) for two main purposes in my experiment: to supply thermal atomic rubidium for the MOT and to improve and maintain vacuum performance by combining with and removing unwanted gas particles in the UHV system. These dispensers contain an alkali metal chromate contained inside a stainless-steel container with a trapezoidal cross section \([165]\). When resistively heated (using an ac or dc power supply) beyond a threshold temperature (typically 500 to 850 °C),
Figure B.5: Magnetic field characterization. a) MOT magnet geometry consisting of four periodically poled magnets. The solid lines indicate the magnetic field contours (0.6 G/line). The dashed lines show the quadrupolar nature of the magnetic field, and the arrows show the orientation and polarization of the incident radial MOT beams. b) Dependence of the location of the magnetic field zero on $I_o/I_i$, where $I_i = 85$ mA and $y = 0$ at the top of the magnets. The points correspond to data and the solid line represents a fit based on extending Eq. [B.1] to the situation of four magnets shown in a). The error bar represents typical measurement uncertainty.
this mixture undergoes a reduction reaction resulting in the emission of alkali metal vapor via a slit in the front of the dispenser. The rate of emission depends on the temperature of the dispenser, which I control by adjusting the current flowing across it. Most dispensers have a threshold current of $\sim 3.5$ A, below which no metal vapor is emitted.

In the experiment, I use alkali metal dispensers from SAES getters containing natural-abundance rubidium, which I spot-weld to an electrical feedthrough and place inside the vacuum system. To initially activate the dispenser, I slowly ramp up the current at a rate of $\sim 1$ A/hour while running the turbopump to remove any excess particulates that might also be emitted. During this time, I detect the presence of rubidium by illuminating the optical cell with resonant laser light, which leads to an easily-observable fluorescence signal. Once I observe fluorescence from the background rubidium vapor (which typically occurs around 3.5-4 A), I slowly turn the getter off, wait for the fluorescence to disappear, and then ramp up the current again (at a rate of $\sim 1.5$ A/hour) until fluorescence reappears. In this way, I determine the threshold operating current for daily use (which is often slightly lower than the initial threshold). After the initial activation, I typically run a continuous current of 2.8-3.2 A through the getter in order to operate the MOT. The MOT brightness usually decreases substantially within 5 minutes after turning off the current. It is important not to exceed the threshold current by too much, as the rubidium emission rate increases rapidly with increasing current, which therefore greatly reduces the dispenser’s lifetime. By running the getter in this way, I find that each getter typically lasts between six to twelve months.
B.1.6 Detectors

I use several types of light-sensitive detectors to characterize the MOT and study light-matter interactions. I use both a pair of balanced photo-multiplier tubes (PMT, Hamamatsu H-6780) and an avalanche photodiode (APD, Hamamatsu C5460) to measure the temporal dependence of the light generated via wave mixing (both above and below the superradiant threshold). The large detection area of the PMTs and adjustable gain make them useful for measuring spatially multi-mode signals over a range of intensities, although the detector gain and temporal response time are inversely related. On the other hand, the APD has a fixed gain and response time and a small detection area, which enables it to have a fast response time (up to 10 MHz) with high sensitivity (1 V/µW in the near IR). For experiments where I need a less sensitive but faster detector (such as in the beatnote experiments), I use a silicon photodiode (company, part, DC-125 MHz).

For spatial imaging, I use two types of charge-coupled device (CCD) cameras. To image the trap for the purposes of monitoring and characterizing it, I use a standard security camera (Marshall 20III). This camera has a 30 fps refresh rate \( (i.e., \sim 30 \text{ ms/frame}) \) and runs continuously. For applications where I require fast or triggered detection, I use a high-speed camera (Dalsa, CA-D1-0128-STDL). This camera has a maximum frame rate of 490 fps \( (i.e., \sim 2 \text{ ms/frame}) \) and accepts an external trigger to initiate or synchronize image acquisition. I use this camera for conducting expansion imaging of the trap (see Sec. B.2.2) and for recording the spatial intensity distribution of the superradiant light.
B.1.7 Computer control

I use LabVIEW to control the timing and synchronization of the AOMs and triggerable cameras via a digital I/O board (NI PCIe-6536) and connector block (NI CB-2162). One LabVIEW program writes the desired timing scheme to the PCIe card’s on-chip memory, which is limited to $10^6$ points. This limits my total collection time to several seconds for experiments in which I require timing resolution of $\sim 100 \mu s$. I use another LabVIEW program to instruct the card to generate the desired signal, which consists of a configurable 3.3 or 5 V output at a maximum frequency of 25 MHz. While I can use directly the digital signal for some applications (such as triggering the Dalsa camera or the oscilloscope), some components (such as the AOMs) require that I use this output to control secondary, analog devices. For example, I use the digital signal to trigger signal generators (SRS DS345) or as an input to active switches that control external power supplies.

B.2 Characterization of Rb MOT

In this section, I describe the techniques that I use to characterize my anisotropic MOT as well as present the results of my measurements. In particular, I discuss my measurements of the atomic density distribution and temperature.

B.2.1 Atomic Density

I study the atomic density distribution via two methods: directly imaging the MOT fluorescence via a CCD camera and measuring the transmission of a weak probe beam passing through the MOT. In the former method, I illuminate the trapping region with near resonant laser light and use a CCD camera to detect the fluorescent light scat-
tered via absorption-spontaneous emission events. The fluorescent intensity is directly related to the number and distribution of the scatterers. By using one camera to image the $y-z$-plane and another camera to image the $x-z$-plane, I can determine the 3D spatial atomic distribution. Figure B.6 shows sample images of the MOT from two different angles along with corresponding density profiles. I find that the trap has a cylindrically-symmetric Gaussian profile in the radial (i.e. $x-y$) plane with a $1/e$ diameter of $W = 430(\pm 20) \mu m$ and a fairly flat profile (typically $< 20\%$ variation) with a length of $L = 3$ cm along the $z$-direction (which is limited only by the optical cell dimensions, see Fig. B.6). This corresponds to a Fresnel number $F = \pi W^2/(\lambda L) \approx 6$, which implies that the cloud can support multiple transverse optical modes. As mentioned in Sec. B.1.4, I can modify the trap diameter simply by varying the amplitudes of the currents to the magnets. In this way, I can tune the trap diameter between $W = 150 – 500 \mu m$.

I also use the CCD cameras to measure quantitatively the number of trapped atoms. Because the cameras automatically set their contrast, I calibrate absolutely the cameras by simultaneously recording the MOT fluorescence and a separate laser beam of known power. Assuming that the atoms scatter light isotropically (which is the case here because I use multiple beams with various polarizations to illuminate the atoms), I relate the total measured power to the atomic number $N$ according to

$$N = \frac{8\pi \left[ 1 + (2\Delta/\Gamma)^2 + (I_{\text{tot}}/I_{\text{sat}}) \right]}{\Gamma (I_{\text{tot}}/I_{\text{sat}}) t_{\text{exp}} \psi d\Omega N_{\text{counts}}}$$

(B.3)

where $I_{\text{tot}}$ is the total incident optical intensity, $I_{\text{sat}}$ is the saturation intensity of the transition, $t_{\text{exp}}$ is the camera’s exposure time, $\psi$ is the efficiency of the camera (i.e. number of counts per photon), $d\Omega$ is the solid angle of light collected by the imaging system, and $N_{\text{counts}}$ is the number of counts recorded by the camera. Using this
Figure B.6: Images of the anisotropic MOT. CCD images showing the cloud of cold atoms primarily in the a) $-y$- and b) $x-z$-planes, where the dashed arrows indicate the cloud of cold atoms. The normalized, integrated density along the c) $\hat{z}$ (i.e., integrated along the $\hat{y}$-direction) and d) $\hat{y}$ directions (i.e., integrated along the $\hat{s}$-direction) obtained via measuring the fluorescent intensity. I take the origin as the center of the trap in the $\hat{y}$ direction and leftmost side of the trap in the $\hat{s}$-direction.
approach, I typically find a maximum number of trapped atoms \( N = 8 \times 10^8 \), which corresponds to an average atomic density of \( \eta = 3 \times 10^{10} \) atoms/cm\(^3\).

The second method for characterizing the atomic spatial distribution involves shining on the atomic vapor a weak laser beam with dimensions that exceed that of the MOT and measuring the spatial distribution of the transmitted light with a CCD camera. For a beam with input intensity \( I_{\text{in}}(r) \), absorption of the light due to the presence of the atoms yields a transmission signal \( T = I_{\text{out}}(r)/I_{\text{in}}(r) = \exp[-OD(r)] \), where the optical depth (as defined below). Thus, by measuring the spatial distribution of the beam after it passes through the vapor, I can directly determine the integrated atom number along the beam’s direction of propagation. Using this method to determine the radial extent of the MOT, I find that the \( 1/e \) cloud diameter is \( W = 420(\pm 10) \) \( \mu \)m, in agreement with the method described above.

To measure quantitatively the atom number, I choose a linearly-polarized probe beam with a waist that is smaller than the trap dimensions, scan its frequency across the atomic resonance, and record the power of the beam before and after the vapor. By aligning the probe beam along the center of the trap, I effectively measure the peak density and find that \( T = I_{\text{out}}(r = 0)/I_{\text{in}}(r = 0) = \exp[-OD(\Delta)] \). Thus, rather than looking at the spatial dependence of the optical depth, I now consider the detuning dependence of \( OD \). The optical depth is related to the absorption cross section \( \sigma(\Delta) \) according to \( OD(\Delta) = \eta \sigma(\Delta) L \). I take the full atomic level structure of the \(^{87}\text{Rb} \text{ D}_2 \) transition into account to calculate \( \sigma \) by summing the weighted contributions from each possible transition as

\[
\sigma = \sum_{F,F',m,m'} \sigma_{F' m' F m} = \sum_{F,m,m',F'} \rho_{F m} \frac{k \gamma |\mu_{F' m' F m}|^2}{\epsilon_0 \hbar (\Delta_{F' m' F m}^2 + (\gamma)^2)},
\]

(B.4)
where $F, m$ and $F', m'$ denote the ground and excited Zeeman levels associated with the $5S_{1/2}|F, m\rangle \rightarrow 5P_{3/2}|F', m'\rangle$ transition, respectively, $\Delta_{F'Fm}$ is the detuning of the light from a particular transition, $\gamma = 1/2\tau_{sp}$ is half the natural linewidth, and $\tau_{sp} = 26.24$ ns is the spontaneous lifetime $^{[159]}$. Here, $\rho_{Fm}$ corresponds to the probability that an atom is in the ground state $|Fm\rangle$. In thermal equilibrium, for example, $\rho_{Fm} = 1/\sum_{F=1}^{2}(2F + 1) = 1/8$ because all of the levels are equally populated. In the presence of the repump and MOT beams, the atoms are preferentially pumped into the $F = 2$ state such that $\rho_{Fm} = 1/5$. If I take all of the Zeeman sublevels to be degenerate, then $\Delta_{F'Fm} = \Delta_{F'F}$ and Eq. [B.4] reduces to

\[
\sigma = \sum_{F,F'} \sigma_{F'F},
\]

\[
= \sum_{F,F'} \rho_F \frac{k \gamma \sum_{m,m'} |\mu_{F'Fm}|^2}{\Delta_{F'F}^2 + (\gamma)^2}.
\]

I define the component of the dipole matrix element along the $\hat{r}_q$ direction as

\[
\mu_{FmFm'}^q = \langle (J'F')m'|er_q|(JF)m\rangle,
\]

where $J = 1/2$ ($J' = 3/2$) is the total angular momentum of the ground (excited) state, $I = 3/2$ is the nuclear spin, $\hat{r}_q$ is a unit vector in the spherical basis (i.e., $q = \pm 1$ for light with $\sigma^\pm$ polarization, and $q = 0$ for light with $\pi$ polarization), and $e$ is the charge of an electron. I use angular momentum theory $^{[166]}$ for a $F = I \oplus J$ transition.
to describe the matrix element in terms of the reduced matrix elements as

\[
\mu_{F'm'Fm}^q = (-1)^{2F'+1+J+m'} \sqrt{(2F + 1)(2F' + 1)} \times \left( \begin{array}{ccc} F' & 1 & F \\ m' & q & -m \end{array} \right) \left( \begin{array}{ccc} J & J' & 1 \\ F' & F & I \end{array} \right) \langle J', \| \vec{\mu}^{l=1} \| J \rangle
\]

\[
= C_{F'm'Fm} \langle J', \| \vec{\mu}^{l=1} \| J \rangle, \tag{B.8}
\]

where \( \langle J', \| \vec{\mu}^{l=1} \| J \rangle \) is the reduced dipole matrix element and \{...\} and (...) correspond to the Wigner 6-j and 3-j symbols, respectively. I relate the reduced dipole matrix element to the spontaneous decay time according to

\[
\frac{1}{\tau_{sp}} = \frac{(2\pi)^3}{3\pi \epsilon_0 \hbar \lambda^3 (2J' + 1)} \left| \langle J', \| \vec{\mu}^{l=1} \| J \rangle \right|^2, \tag{B.9}
\]

where \( \lambda \) is the wavelength of the atomic transition. Substituting Eqs. B.8 and B.9 into Eq. B.4, I find that

\[
\sigma = \sum_{F',m',F,m} \sigma_0 f_{F'm'Fm} \frac{\gamma^2}{\Delta_{F'}^2 + \gamma^2}, \tag{B.10}
\]

where \( \sigma_0 = 3\lambda^2/2\pi \) is the full atomic cross-section and \( f_{F'm'Fm} = 2C_{F'm'Fm}^2 \rho_F \) describes how this cross section is divided among the various transitions.

I determine the density as

\[
\eta = \frac{OD(\Delta)}{\sigma(\Delta)L}, \tag{B.11}
\]

where I experimentally measure \( OD(\Delta) \) and \( L \) and calculate \( \sigma \). Figure B.7a shows a typical experimentally-measured transmission spectrum \( T \) along the \( \hat{z} \) direction, where the three dips correspond to transitions between the \( F = 2 \rightarrow F' = 1, 2, 3 \) states from left to right. Figure B.7b shows a closeup of the transmission around the \( F = 2 \rightarrow F' = 3 \) transition, which is the spectral region in which I carry out my wave-mixing.

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Figure B.7: MOT transmission spectrum. Transmission of a weak probe beam through the MOT along the $\hat{z}$-axis as I scan its frequency over a) the $F = 2 \rightarrow F' = 1, 2, 3$ and b) $F = 2 \rightarrow F' = 3$ states. The dashed line corresponds to a fit $T = \exp[-OD(\Delta)]$ for $OD = 37$.

experiments. I observe maximum optical depths of $OD_l \sim 60$ and $OD_r \sim 1$ along the longitudinal and radial directions, respectively, although typical values used in this thesis are closer to $OD_l \sim 20$. Combining this result with the theory outlined above and the measurements of $L$ made with the CCD camera, I calculate a maximum atomic density of $\eta = 3(\pm0.3) \times 10^{10}$ cm$^{-3}$, which agrees with the CCD camera-based measurement.

To achieve the largest optical depths, I typically begin with a relatively low getter current such that the number of trapped atoms is small. I then iteratively adjust the alignment of the MOT beams such that the trap appears long and uniform (as imaged by the CCD camera). Next, I monitor the transmission of the weak probe beam propagating along the trap’s long axis, and continue adjusting the MOT beam alignment so that the trap remains long but minimizes the probe transmission (i.e., maximizes the $OD$). Finally, I turn up the getter current to standard working values (see Sec. B.1.5). This leads to an increase in both the total number of trapped atoms as well as the atomic density.
I experimentally control $\eta$ by modifying the frequency of the repump beam. As explained in App. A.2, I require a separate repump laser to keep atoms from being pumped into a dark state via the MOT beams. I use the fact that the cycling transition is not a perfectly closed loop to control the vapor’s optical depth. By keeping the repump power fixed, I vary the repump frequency and therefore more or less efficiently pump atoms out of the dark state so that they can be cooled and trapped. In this way, I can continuously tune the total number of trapped atoms and, therefore, the optical depth.

In addition to measuring the spatial atomic distribution, I study the time scale associated with cooling and trapping in the MOT by monitoring the time-dependent transmission of a weak probe beam propagating along the $\hat{z}$-direction as the MOT forms or decays. I look first at the formation of the atomic cloud by measuring the probe beam transmission immediately after I turn the MOT beams on. To lowest order, one can describe the loading process in terms of the simple rate equation $dN/dt = R - N/\tau$, where $N$ is the number of trapped atoms, $R$ is the loading rate, and $1/\tau$ is the loss rate (e.g., due to collisions between trapped atoms and hot, background atoms). Integrating this equation, one finds that

$$N(t) = N_{ss}(1 - e^{t/\tau}),$$

where $N_{ss}$ is the number of atoms in steady-state. Figure B.8a shows that the normalized atomic density increases in agreement with Eq. B.12 for $\tau = 115$ $\mu$s. This time is shorter than typically observed in MOTs [? ], which may imply that the background pressure at the location of trap formation (which is far from the ion pump) is larger than expected. This may could be due to the fact that I work with natural-abundance rubidium (which is 72% $^{85}$Rb and 25% $^{87}$Rb), but only trap $^{87}$Rb. Thus, the fraction of hot atoms remaining is larger than in the case of a pure $^{87}$Rb sample (in which all atoms
Figure B.8: MOT formation and decay. Normalized MOT density as a function of time when a) the MOT forms immediately after the beams are turned on and b) the MOT decays after the beams are turned off at \( t = 0 \). The dashed line in a) is given by Eq. B.12 with \( \tau = 0.115 \) s. The dashed line in b) is given by Eq. B.14 for \( T_{\text{rad}} = 40 \) µK and \( \sigma_r(0) = 140 \) µm.

are subject to cooling), resulting in a higher loss rate. Alternatively, the fast loading time could indicate additional, pressure-independent loss mechanisms that arise due my choice of magnetic field and beam geometry.

Figure B.8 b shows the case where I first allow the MOT to reach steady state and then turn off the MOT beams. The decay of the signal occurs in only a couple of ms and is due to two main effects: gravity causes the atoms to fall and thermal motion causes the cloud to expand (i.e., the density decreases). Since the probe beam transmission is directly related to the atomic density, one must consider the evolution of the atomic density profile. In steady-state, I write the MOT density in the plane transverse to the probe beam as \( \eta(x, y, t = 0) = N_{ss} \exp\left\{ -\frac{(x(0)^2 + y(0)^2)}{2\sigma_r^2(0)} \right\} / \pi 2\sigma_r^2(0) \), where \( \sigma_r(0) \) is the initial radial width of the MOT. After I extinguish the MOT fields,

\[
\begin{align*}
x & \rightarrow x(t) = x(0), \\
y & \rightarrow y(t) = y(0) - y_c(t), \\
\sigma_r^2 & \rightarrow \sigma_r^2(t) = \sigma_r(0)^2 + \frac{k_B T_{\text{rad}}}{m} t^2, \quad \text{(B.13)}
\end{align*}
\]
where \( y_c(t) = a_g t^2 / 2 \), \( a_g = 9.8 \) m/s\(^2\) is the acceleration due to gravity, and \( T_{rad} \) is the radial temperature. Assuming that the probe beam is aligned along the center of the MOT initially, the resulting time-dependent density experienced by the probe beam becomes \([?]\)

\[
\eta(t) = \frac{N_{ss}}{\pi 2 \sigma_r^2(t)} \exp \left( \frac{-y_c^2(t)}{2 \sigma_r^2(t)} \right).
\]

(B.14)

I find that Eq. (B.14) matches well the observed decay (see Fig. B.8b). This decay time is shorter than for a standard spherical MOT primarily because the radial dimension is nearly an order of magnitude smaller. This rapid decrease in density impedes the realization of long-lived and steady-state behavior. As discussed in Ch. 7, one solution to this problem is to use an additional potential (e.g., a dipole potential in the radial direction) to keep the atoms in the same location after the MOT beams turn off.

### B.2.2 Atomic Temperature

One can measure the temperature of atoms in a MOT in a variety of ways. Most techniques involve turning off the MOT beams, allowing the system to evolve freely, and then performing a measurement after a fixed amount of time. Examples of this technique include the release and recapture method \([167]\), direct expansion imaging \([168]\), and time-of-flight measurements \([169]\). The data in Fig. B.8b, for instance, is an example of a time-of-flight measurement in which the time dependence of the transmitted probe beam depends on the atomic temperature as the trap expands. Using this method, I find that the data corresponds to \( T_{rad} = 40 \pm 10 \) µK. I find that the temperature measured in this way varies between \( T = 30 - 60 \) µK, depending on alignment of the MOT beams.

To supplement this measurement, I also obtain an estimate of the atomic temperature by directly imaging the expansion of the cloud immediately after I turn the MOT
beams off. In this experiment, I use a short pulse of near-resonant light to illuminate the MOT a fixed time after I turn the MOT beams off and record the resulting fluorescence using a fast CCD camera (see Sec. B.1.6). The analysis in this case is similar to that presented in Eqs. B.13 and B.14. The main difference is that, whereas the time-of-flight measurement considers the change in probe transmission due to the effects of gravity and thermal expansion of the gas, imaging the entire cloud allows me to focus only on the effects of thermal expansion because I can account directly for the change in the location of the cloud’s centroid. Figure B.9 shows the width of the cloud after I turn the MOT beams off at \( t = 0 \). I find that the atomic temperature is \( T_{\text{rad}} = 50 \pm 15 \) \( \mu \text{K} \), which is consistent with the time-of-flight measurement. Again, I find that the exact temperature depends on MOT beam alignment and can vary between \( T = 40 – 60 \) \( \mu \text{K} \).

Unfortunately, these approaches have several drawbacks. For example, they require one to make additional assumptions about or measurements of the initial state of the vapor, such as the shape of its steady-state spatial or momentum distribution. Also, the act of measuring the temperature with these techniques necessarily destroys the vapor.
In addition, each approach requires space in which the cloud can fall or expand. Since my anisotropic MOT geometry and optical cell design limit free evolution of the cloud in my setup, it is difficult to make accurate estimations of the temperature in this way. In fact, it is not possible to measure the longitudinal temperature using this technique because the MOT extends to the ends of the cell along the $\hat{z}$-direction. Thus, I use an alternative, non-destructive technique based on a NLO process known as recoil-induced resonances (RIRs) [170,171] to measure the atomic temperature in my setup in-situ.

Recoil-induced resonances arise via stimulated Raman scattering between differently populated atomic momentum states. When one illuminates an atomic sample with a pump (frequency $\omega_1$, wave vector $k_1$) and probe (frequency $\omega_2$, wave vector $k_2$) optical field at an angle of $\theta \neq 0$ relative to one another, the absorption of a photon from one beam and stimulated emission into the other gives the atoms a net momentum kick (see Fig. B.10). This process can occur in two different ways: photons can be scattered from the probe beam into the pump beam or from the pump beam into the probe beam. In the former scenario, scattering transfers an atom with initial momentum $p$ to a final momentum $p + \hbar \Delta k$, where $\Delta k = k_1 - k_2$. In the latter scenario, an atom with initial momentum $p + \hbar \Delta k$ has a final momentum $p$ after scattering.

The simultaneous requirement of momentum and energy conservation imposes constraints on which momentum states the beams interact with for a given two-photon detuning $\delta = \omega_1 - \omega_2$ (i.e., the choice of $\delta$ fixes which two momentum states are coupled, see Fig. B.10 b). Differences in the populations of atoms in the two coupled states, which I denote as $\pi(p)$ and $\pi(p + \Delta k)$, lead to a preferential scattering from one beam into the other and, therefore, a net redistribution of the number of photons in each beam (i.e., gain or absorption). The normalized gain spectrum of the transmitted probe field $g_{RIR}(\delta) = I_{\text{out}}/I_{\text{in}}$ is therefore closely connected to the shape
Figure B.10: Momentum and energy conservation in recoil-induced resonances.

a) Basic geometry for RIR, where the absorption from one beam and stimulated emission from another changes the atom’s momentum by $\Delta \hat{k}$. b) Because of the quadratic dependence of the kinetic energy on the atomic momentum, one can only drive transitions between certain states for a given detuning between the incident fields. Net gain or loss occurs because the states connected in this way in general have different populations (represented by the circles at a given ground state).

of the atomic momentum distribution along $\Delta \hat{k}$. More specifically, $g_{RIR}$ is simply proportional to the derivative of the momentum distribution $[i.e. \partial \pi(p)/\partial p]$ in the limit that $|\Delta \hat{k}| \ll m u$. Here $m$ is the mass of the atom, and $u = (k_B T/m)^{1/2}$ and $T$ are the r.m.s. width of the velocity distribution and temperature along $\Delta \hat{k}$.

For a Maxwell-Boltzmann velocity distribution, one finds that the gain spectrum is given by \[ g_{RIR}(\delta) = -\left(\frac{\pi}{2}\right)^{1/2} \frac{h \delta}{\Delta^2} \frac{\Omega_1^2}{2 m k u^3 \sin(\theta/2)} \exp\left(-\frac{\delta^2}{2 [2 k u \sin(\theta/2)]^2}\right), \] (B.15)

where I take $|k_1| = |k_2| \equiv k$. As shown in Fig. B.11 a, $g_{RIR}$ is anti-symmetric with respect to $\delta$, where gain (absorption) occurs for $\delta < 0$ ($\delta > 0$). The extrema occur at
Figure B.11: Momentum and energy conservation in recoil-induced resonances. a) RIR gain for the case of a pump and probe beam propagating at an angle $\theta = 10^\circ$ relative to one another. The data corresponds to a temperature of $T = 40\pm5$ µK. b) RIR gain for the case where I include an additional, counterpropagating pump beam. The dashed line corresponds to a fit obtained by summing narrow and broad RIR features (with temperatures $T_{rad} = 45\pm5$ and $T_{sz} = 40\pm10$, respectively) with four Raman features (described by Lorentzians centered at $\pm1\omega_{vib}$ and $\pm2\omega_{vib}$ with widths $50/2\pi$ and $90/2\pi$ kHz, respectively). For all data, I use the $\Sigma\parallel$ polarization configuration.

$\delta_e = \pm2k\sin(\theta/2)$, which gives directly the temperature as

$$T = \frac{m\delta_e^2}{4k_B^2}\sin(\theta/2)^2.$$  \hspace{1cm} (B.16)

In addition, since the RIR spectrum’s shape is proportional $\partial\pi(p)/\partial p$, one can recover the entire momentum profile by integrating the measured spectrum.

Extending this result to the case where one uses a pair of counterpropagating pump beams gives rise to two additional benefits. First, one can now simultaneously measure the atomic temperature in two, orthogonal directions. One can understand this by noting that the directions of the atomic recoil due to scattering between the probe and each pump beam are orthogonal to one another. Thus, the gain spectrum consists of two RIR resonances, where the shape of each one is related to the temperature along the direction of atomic recoil (see Fig. B.11b). The narrow central peak (RIR1) is due to recoil between the nearly-copropagating beams, whereas the broad feature
(RIR2) corresponds to recoil between nearly-counterpropagating beams. The analysis in this case is slightly complicated by the presence of additional, Raman resonances between vibrational levels. Nevertheless, one can obtain a fit to the entire spectrum by including the presence of the Raman features as Lorenztians centered at the integer multiples of the vibrational level spacing \( \omega_{\text{vib}} \). For example, I include resonances at \( \pm \omega_{\text{vib}} \) (Ram1) and \( \pm 2\omega_{\text{vib}} \) (Ram2), where \( \omega_{\text{vib}}/2\pi = 170 \text{ kHz} \). Using this method, I find that \( T_{\text{rad}} = 45 \pm 5 \mu\text{K} \) and the temperature along the \( \hat{z} \)-direction is \( T_z = 40 \pm 10 \mu\text{K} \). In general, I find that \( T_z \) and \( T_{\text{rad}} \) can vary between 20-60 \( \mu\text{K} \), depending on the magnetic field gradient and the alignment and polarization of the MOT beams.

### B.3 Summary

In this chapter, I presented the technical details of each system involved in the operation and characterization of my anisotropic MOT, as well as the setup used to perform the wave-mixing experiments described in Chs. 4 and 6. The main systems involved in the creation of the MOT include optical and vacuum systems, a custom-made optical cell, two pairs of magnets constructed using ferromagnetic cores, and getters to provide Rubidium. I also employed a range of different detectors to measure the optical signals used to study the gas. In addition, I developed a computer-controlled system to coordinate the timing of various elements and acquire data.

Using this system, I characterize the atomic density distribution and temperatures of the trapped atoms using several different techniques. I find that the resulting cold atomic cloud is highly anisotropic, with a typical length of \( L \approx 3 \text{ cm} \) and a \( 1/e \) width of \( \sim 430 \mu\text{m} \). The atomic density is typically on the order of \( 10^{10} \text{ cm}^{-3} \), resulting in a longitudinal (radial) optical depth of \( OD \sim 50 \) (\( OD \sim 1 \)). The atomic temperatures along the trap’s radial and longitudinal directions are typically on the order of 30 – 60
$\mu K$, depending on the details of the configuration.
Bibliography


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Biography

Joel Greenberg was born in Bryan, Texas on April 4, 1983. While he moved around the country when he was younger, he attended junior high and high school in Downingtown, Pennsylvania and graduated as valedictorian of his class from Downingtown High school in 2001. He matriculated at Princeton University in August of the same year and completed a B.S.E. degree in Mechanical and Aerospace engineering and a certificate in Engineering Physics. Among his achievements at Princeton, Joel received the “Best Engineering Physics Independent Work Award” and “Donald Janssen Dike Award for Excellence in Undergraduate Research” for his senior thesis work under Dr. Szymon Suckewer. He graduated summa cum laude in 2005.

Joel matriculated at Duke in the summer of 2005 and immediately joined the Quantum Electronics Lab under Dr. Daniel Gauthier. During his time at Duke, Joel was named a James B. Duke Fellow of Physics by the Graduate School, a John T. Chambers Fellow by the Fitzpatrick Institute for Photonics, and a Fritz London Fellow by the Physics Department. In addition, Joel received a Physics Graduate Teaching Fellowship to co-design and teach a course in optics aimed at advanced undergraduate and beginning graduate students with Dr. Gauthier. Joel also participated in the year-long Preparing Future Faculty program, served as the head of the Physics Department graduate mentoring program, and acted as president of the Physics Graduate Student Organization.

Joel received his A.M. in Physics in 2008, a graduate certificate in Photonics in 2012, and his Ph.D. in 2012 for his work on collective light-matter interactions in a cloud of cold atoms. During that same time, Joel married Rachel Gottron in 2006 and welcomed his daughter Madelyn to the world during the writing of his dissertation in 2011.
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