Amplification of laser beams propagating through a collection of strongly driven, Doppler-broadened two-level atoms

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We investigate theoretically the amplification of a laser beam propagating through a collection of Doppler-broadened two-level atoms driven by an intense counterpropagating laser beam. Large amplification of the beam is predicted when the pump-beam Rabi frequency is comparable to the Doppler width of the atomic transition, even without including the effects of atomic recoil. The microscopic origin of the gain can be attributed to the coherent driving of the atomic dipole moment, suggesting that amplification and lasing due to collective atomic recoil may be influenced by this process. [S1050-2947(97)50203-4]

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The interaction between an intense laser field and a collection of two-level atoms is fundamentally important in the nonlinear and quantum-optics communities because it highlights many aspects of light-matter interactions without being overly complex. One process often ignored in the description of this interaction is the change in the center-of-mass momentum of the atoms that occurs when they absorb or emit radiation; yet this “atomic recoil” effect is paramount in some situations. For example, it is the basis for laser cooling and trapping of atoms [1] and some matter-wave interferometers [2,3]. Also, atomic recoil-induced resonances have been predicted and observed in nonlinear spectroscopy of atoms [3,4].

Recently, Lippi et al. [5] and Hemmer et al. [6] observed amplification of a laser field propagating through a collection of high-temperature (~600 K), Doppler-broadened sodium atoms driven by an intense counterpropagating laser beam. They attribute the gain to an atomic density grating (a periodic bunching of the atoms) formed by the aggregate effect of atomic recoil; a surprising result considering that the modulation depth of the density grating is expected to be quite small at this temperature [7]. Nevertheless, their intriguing results agree qualitatively with recent analyses of this interaction [8].

The purpose of this Rapid Communication is to point out that the interpretation of these results may be richer than originally anticipated; there may be a complex interplay between the amplification process due to atomic recoil and other well-known amplification processes. Specifically, we demonstrate theoretically that large amplification of the laser beam due to coherent driving of the atomic dipole moment is expected even when the effects of atomic recoil are ignored, and that the amplification occurs over a similar spectral region where large atomic-recoil-induced gain is predicted. Furthermore, this process creates a nonlinear polarization grating in the medium that may enhance or interfere with the grating formed by bunched atoms.

Amplification of a laser beam propagating through a collection of high-temperature, Doppler-broadened, laser-driven two-level atoms was pointed out in theoretical studies of saturated absorption in the early 1970s. Haroche and Hartmann [9], for example, find that the medium can amplify the laser beam when the pump beam is sufficiently intense for degenerate-frequency beams (as in the experiment of Lippi et al. [5]) and for nondegenerate frequencies (as in the experiment of Hemmer et al. [6]). They speculate that this process could permit laser oscillation. Using a simplified theory, our analysis shows that probe amplification due to the coherent effects considered by Haroche and Hartmann [9] may occur under the experimental conditions of Lippi et al. [5] and Hemmer et al. [6].

We begin by considering the interaction between a continuous-wave electromagnetic field of the form

\[ \mathbf{E}(\mathbf{r},t) = \sum_{j=p,d} \mathbf{A}_j(\mathbf{r}) e^{-i(\omega_j t - k_j \cdot \mathbf{r}) + c.c.}, \tag{1} \]

and a collection of two-level atoms with resonance frequency \( \omega_{eg} \), electric dipole matrix element \( \mu_{eg} \), atomic mass \( m \), number density \( N \), contained in a cell of length \( L \). \( \mathbf{A}_d \) represents the plane-wave amplitude of the intense pump field of frequency \( \omega_d \) with wave vector \( \mathbf{k}_d \), and \( \mathbf{A}_p \) is the amplitude of the weak probe beam of frequency \( \omega_p \) with wave vector \( \mathbf{k}_p \). The excited state \( |e\rangle \) decays to the ground state \( |g\rangle \) in a time \( T_1 \) and the atomic dipole moment between \( |e\rangle \) and \( |g\rangle \) dephases in a time \( T_2 \). The field induces a polarization \( \mathbf{P}(\mathbf{r},t) \) in the medium which is the source term in the wave equation describing the propagation of the field (1) through the atomic vapor.

Contributions to the polarization include the spatially varying atomic dipole moment (often referred to as the coherence) and variations in the atomic density such as an atomic-recoil grating. Incorporating both effects in the strong-field regime is beyond the scope of the present discussion; our goal is to demonstrate that the spatially varying atomic coherence gives rise to a grating in the polarization that is capable of amplifying the probe beam while ignoring
the effects of atomic recoil. Using a semiclassical theory of this interaction [10], the polarization arising from coherent effects is

$$\mathbf{P}(r,t) = \sum_{j=p,d} \mathbf{P}_j(r) e^{-i(\omega_j t - \mathbf{k}_j \cdot r)} + \text{c.c.,}$$  \hspace{1cm} (2)

where \( \mathbf{P}_j(r) = N \mu_{eg}^2 \langle \sigma_{eg}(\mathbf{r},\omega_j) \rangle_D \), and \( \langle \sigma_{eg}(\mathbf{r},\omega_j) \rangle_D \) is the Doppler-averaged, slowly varying off-diagonal density-matrix element of the driven two-level atom (the "atomic coherence") at frequency \( \omega_j \). Equation (2) represents a moving grating capable of scattering light; its presence may complicate the measurement of atomic number density gratings via Bragg scattering of light as performed by Hemmer et al. [6]. This spatially varying coherence is intimately connected to a spatial variation in the population difference between the atomic states (the inversion) due to their mutual coupling. We note that an alternate, though entirely equivalent, interpretation of amplification due to coherent effects can be formulated in terms of exchange of photons from the pump beam to the probe beam via scattering from the polarization and inversion gratings formed in the atomic medium.

The intensity of the probe beam as it propagates through the medium is determined by solving the coupled Maxwell and density-matrix equations. To simplify our discussion, we consider the case where the probe beam is weak and propagates in the \( +z \) direction (unit vector \( \mathbf{\hat{z}} \)), and the pump beam amplitude remains constant as it propagates through the medium in either along \(-\mathbf{\hat{z}}\) (counterpropagating) or \(+\mathbf{\hat{z}}\) (co-propagating). The constant-pump approximation is reasonable since the intense pump beam saturates significantly the atomic transition thus "burning" its way through the atomic vapor.

The probe beam intensity at the exit of the medium is given by

$$I_p(L) = I_p(0) \exp[-(\alpha(\Delta, \delta))_p L],$$  \hspace{1cm} (3)

where \( I_p(0) \) is the input intensity, \( \langle \alpha(\Delta, \delta) \rangle_p \) is the Doppler-averaged absorption coefficient [proportional to the imaginary part of \( \langle \sigma_{eg}(\mathbf{r},\omega_j) \rangle_D \), \( \Delta = \omega_d - \omega_{eg} \) is the detuning of pump beam frequency from atomic resonance for a stationary atom, and \( \delta = \omega - \omega_d \) is the detuning of the probe and pump frequencies in the laboratory frame. The Doppler-averaged absorption coefficient is related to the moving-atom absorption coefficient \( \alpha(\Delta, \delta, u) \) (with velocity component \( u \), along \( \mathbf{\hat{z}} \)) via

$$\langle \alpha(\Delta, \delta) \rangle_p = \frac{T_z^a}{\sqrt{\pi}} \int_{-\infty}^{\infty} \alpha(\Delta, \delta, u) \exp[-(u T_z^a)^2] du,$$  \hspace{1cm} (4)

where \( T_z^a = (1/k_b) \sqrt{m/2 \hbar^2 T} \) is the Doppler dephasing time, \( k_b \) is Boltzmann’s constant, \( T \) is the temperature, \( u = k v \) is the Doppler shift, and \( k = |\mathbf{k}_p| = |\mathbf{k}_d| \). The absorption coefficient for a moving atom in a strong pump beam with Rabi frequency \( \Omega_d = 2 \mu_{eg} A_d(r)/\hbar \) is given by

$$\alpha(\Delta, \delta, u) = \frac{\alpha_0 w_0 [\beta_T^2(\Delta_T^2 + iT_1)(\Delta_T^2 + iT_2) - \beta_T^2(\Delta_T^2 + iT_2) + |\Omega_d|^2/2(\Delta_T^2 + iT_2)]}{T_z^a(\Delta_T^2 + iT_1)(\Delta_T^2 + iT_2) + |\Omega_d|^2(\Delta_T^2 + iT_2)},$$  \hspace{1cm} (5)

where \( \Delta_T = \Delta - (\mathbf{k}_d \cdot \mathbf{\hat{z}}) u \), \( \delta_T = \delta - [(\mathbf{k}_d - \mathbf{k}_p) \cdot \mathbf{\hat{z}}] u \), \( \mathbf{k}_p \) and \( \mathbf{k}_d \) are the propagation-direction unit vectors, \( \alpha_0 = A(3\lambda_{eg}^2/2\pi)(T_1/2T_1) \) is the line-center, weak-field absorption coefficient, \( \lambda_{eg} \) is the transition wavelength,

$$w_0 = w^q \left( 1 + \Delta^2_T T_2^2 \over 1 + \Delta^2_T T_2^2 + |\Omega_d|^2 T_1 T_2 \right)$$  \hspace{1cm} (6)

is the population inversion between \( |e\rangle \) and \( |g\rangle \), and \( w^q \) is the absorption in the absence of the laser and \( |e\rangle \). Probe beam amplification due to coherent driving of the atomic dipole moment is determined by evaluating Eqs. (3)–(6) using the appropriate experimental conditions. We first consider the experiment of Hemmer et al. [6], where maximum amplification of \( I_p(L)/I_p(0) = 2 \) is observed when the laser frequencies are tuned near the sodium \(^3P_{3/2} \rightarrow ^3P_{1/2}\) transition with \( \Delta/2\pi = 1.5 \) GHz, \( \Omega_d/2\pi = 2.6 \) GHz, \( T = 340 \) °C, \( N = 6 \times 10^{14} \) atoms \( \text{cm}^{-3} \), and \( L = 1 \) cm [see Fig. 3(b) of Ref. [6]]. For this transition, \( T_1 \approx 32 \) ns, \( T_2 \approx T_1 \) (due to collisional self-broadening of the transition at this temperature [11]), and \( T_1^a = 0.14 \) ns. In our investigation, we take \( \Omega_d/2\pi = 1.3 \) GHz to account for the small self-defocusing present in the experiment [12].

Figure 1 shows the absorptive response for a collection of stationary atoms \( \alpha(\Delta, \delta, u = 0) / \alpha_0 \) and the Doppler-averaged response \( \langle \alpha(\Delta, \delta) \rangle_p / \alpha_0 \) for both counterpropagating and copropagating configurations as a function of \( \delta \) with \( \Delta_T = -151 \), \( \Omega_d T_2 = 131 \), \( T_2/T_1 = 1 \), \( w^q = -1 \), and \( T_1^a/T_2 = 8.75 \times 10^{-3} \). The probe beam experiences loss when \( \alpha > 0 \) and gain otherwise. For stationary atoms [Fig. 1(a)], the spectrum consists of a gain feature (labeled I) that occurs at \( \delta = -\Omega_d' = -\sqrt{\Omega_d^2 + \Delta_T^2} \), where \( \Omega_d' \) is the generalized Rabi frequency, and an absorption feature (III) at \( \delta = \Omega_d' \). These features are easily interpreted using the dressed-state picture; they arise from probe-induced transitions between the dressed states (the composite atom + field states of the strongly driven atom) [10,13]. The dispersion-shaped feature (II) near \( \delta = 0 \) is due to stimulated Rayleigh scattering and can be interpreted in terms of population oscillations between the ground and excited states [10,14]. Spectra similar to Fig. 1(a) have been observed experimentally in a Doppler-free configuration [15].

Inclusion of atomic motion dramatically affects the absorptive response of the driven atoms [9,16]. For the counterpropagating configuration \( \Delta_T = \Delta + u \), \( \delta_T = \delta - 2u \), Fig. 1(b)], the gain feature broadens and its height is significantly reduced, and the Rayleigh feature no longer gives rise to
amplification. Maximum gain no longer occurs at \( \delta = 52 \) V; rather, the Doppler averaging pulls the feature to smaller detunings so it peaks near \( \delta = D \). Interestingly, a small gain feature now exists to the high-frequency side of resonance near \( \delta = T_2 / T_1 = 1, T_2^\ast / T_2 = 8.75 \times 10^{-3} \). For comparison, we show the co-propagating configuration (\( D_2 = D_1 \), \( \delta = 0 \)) in Fig. 1~\( \sim \).

It is seen that the gain feature near \( \delta = 52 \) V is somewhat larger and narrower in comparison to the counterpropagating configuration, and that the Rayleigh feature survives the Doppler averaging. Several research groups have observed experimentally similar spectra \( @17# \) for the copropagating configuration.

Substantial amplification of the probe beam due to the coherent driving of the atomic dipole moment is expected for the large atomic number density and path length used in the experiment of Hemmer \( @6# \). We estimate that \( \alpha_0 \sim 5 \times 10^4 \) cm\(^{-1} \) for their experimental conditions resulting in extremely large amplification at optimal probe detuning: \( I_p(L)/I_p(0) \sim \exp(30) \sim 10^{13} \). This amplification is much larger than that observed experimentally.

Several factors reduce the expected value of the amplification. One obvious factor is the multilevel hyperfine structure of the sodium atom; the line-center absorption coefficient should be reduced by a factor of 8 since the laser fields only interact with the \( 3S_{1/2} (F=2, m_F=2) \rightarrow 3P_{3/2} (F=3, m_F=3) \) transition due to the choice of laser beam polarizations. Other factors such as the 1.77-GHz hyperfine splitting of the ground state, optical pumping to the \( 3S_{1/2} (F=1) \) level, velocity-changing collisions, radiation trapping, and self-focusing and/or defocusing effects will reduce the amplification. We find that probe beam amplification similar to that observed by Hemmer \( et al. @6# \) is obtained with \( \alpha_0 = 10^4 \) cm\(^{-1} \), a factor of 50 reduction from
our initial estimate. Figure 2 shows the predicted amplification of the probe beam as a function of $\delta$ for the reduced value of $\alpha_r$. The predicted and observed [see Fig. 3(b) of Ref. [6]] spectra are strikingly similar for $\delta<0$, where they attribute the gain to collective atomic recoil effects. Note that the sense of the frequency scan is reversed for our plots compared to Hemmer et al. [6]. Contrary to our prediction, there is no probe beam amplification for $\delta>0$ in the experiments. This discrepancy could be due to competition between the recoil-induced gain and coherent driving of the dipole moment, self-focusing of the probe beam, or other factors such as those mentioned previously.

We now turn our attention to the experiments of Lippi et al. [5]. Comparing our theoretical predictions with their experiments is complicated since they use a buildup cavity to boost the probe-beam intensity and to enhance the small gains experienced by the probe beam. Ignoring this complexity, we find that our simplified analysis also predicts laser beam amplification under their experimental conditions of degenerate pump and probe frequencies ($\delta=0$), $T=240^\circ C$ (resulting in $N\approx 1.6 \times 10^{13}$ atoms cm$^{-3}$, $T_2=2T_1=32$ ns, $\alpha_0 \approx 1.3 \times 10^4$ cm$^{-1}$, and $T_2^p=0.16$ ns), and $L=0.6$ cm. At the large pump-beam Rabi frequency quoted in the experiment ($\Omega_g/2\pi=16$ GHz, the medium is almost fully saturated and there is little amplification or absorption of the probe beam. We find better agreement when the pump-beam Rabi frequency is comparable to but somewhat larger than the Doppler width.

Figure 3(a) shows the absorptive response for counterpropagating beams as a function of $\Delta$ for the experimental conditions similar to that of Lippi et al. [5] with $\Omega_g/2\pi=2$ GHz ($\Omega_gT_2=402$, solid line) and $\Omega_g/2\pi=4$ GHz ($\Omega_gT_2=804$, dashed line) with $\delta=0$, $T_2=2T_1$, $w^q=-1$, $T_2^p/T_2=5.1 \times 10^{-3}$. It is seen that there are spectrally broad regions in the wings of the atomic resonance that exhibit amplification of the laser beam.

The transmission of the probe beam through the cavity is given approximately by

$$T_{\text{cavity}} = \frac{A}{[1 + F(1-A)/\pi]^2},$$

where $A = \exp[-(\langle \alpha, \delta=0 \rangle)/L]$ and $F$ is the measured cavity finesse. Figure 3(b) shows the predicted cavity transmission for $F=30$, $\alpha_0=267$ cm$^{-1}$ (again reduced by a factor of 50 to partially account for the ground-state structure), and the two different pump-beam Rabi frequencies. It is seen that the maximum value of the probe beam amplification is $\sim 15\%$ for $\Omega_gT_2=402$ and $\sim 3\%$ for $\Omega_gT_2=804$. The former is similar to that observed in the experiment.

Our results suggest that interpretation of these experimental results must be reviewed in light of a possible complex interplay between collective atomic recoil and the coherent driving of the atomic dipole moment. Additional theoretical work that incorporates both effects for a strong driving field and the multilevel structure of sodium is needed to properly identify the mechanisms responsible for the observed amplification.

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[8] See especially the discussion in Sec. 6.