Two-photon amplification and lasing in laser-driven potassium atoms: Theoretical analysis

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Two-photon amplification and lasing in laser-driven potassium atoms is investigated theoretically using a rigorous model that takes into account the hyperfine level structure of the atoms for arbitrary intensity drive and lasing fields. The model correctly predicts most features of the recent experiments, such as the characteristics of the multiphoton gain resonances and the two-photon laser emission intensity and line shape. In addition, it constitutes a new tool for exploring behaviors that are difficult to address experimentally, such as interference effects between different quantum pathways. In contrast to single-photon lasers, the laser emission line shape is frequency pushed and has the form of a closed curve composed of a stable and an unstable branch.

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I. INTRODUCTION

The two-photon laser is a new type of quantum oscillator based on the two-photon stimulated emission process whereby two incident photons stimulate an excited atom to a lower-energy state and four photons are scattered coherently by the atom. The laser is intrinsically nonlinear due to the nature of the stimulated emission process and is thus expected to display novel quantum optical and nonlinear dynamical behaviors. For example, it is predicted that a broadband two-photon laser will display spatial solitons [1] and it is believed that the single-mode counterpart will produce polarization-entangled twin beams, which may find application in areas such as quantum information and quantum imaging [2,3]. While there has been continued interest in two-photon lasers since the original conception in the early 1960s (see, for instance Refs. [4–8]), significant progress has been hampered by the difficulty in achieving two-photon lasing. After several pioneering experiments performed in the past [9,10], the very recent results of Pfister et al. on efficient two-photon amplification [11] and lasing [12] in a potassium atomic beam are opening a way for the experimental investigation of the properties of two-photon laser systems.

A special feature of the amplification scheme used in the successful two-photon experiments [10–12] is that the atoms are irradiated by an intense “driving” field, where amplification occurs through multiphoton processes involving simultaneous absorption of drive-field photons and stimulated emission of two probe-field photons. By tuning the drive-field frequency close to the atomic resonances, it is possible to selectively enhance the two-photon stimulated emission process and suppress competing nonlinear optical processes. The added flexibility afforded by such a composite atom - field gain medium has moved forward research on two-photon lasers, but complicates accurate theoretical analyses of the laser. In addition, the laser-driven potassium system is exceedingly rich due to degenerate magnetic sublevels participating in the interaction.

The primary goals of this paper are to summarize a semiclassical model of the interaction between two intense electromagnetic fields with fixed states of polarization and a collection of potassium atoms, use it to explain features of the recent two-photon amplification [11] and lasing [12] experiments, and use it to make predictions for future experiments. Our model takes into account the population and coherence effects brought about by the presence of the intense drive and probe (or lasing) fields as well as the details of the hyperfine level manifolds involved in the atom-field interaction. In particular, the model will allow us to: characterize the different multiphoton gain and absorption resonances and determine the degree of overlap or interaction among them; study the interference effects among different quantum paths contributing to a given gain resonance, and calculate the laser emission intensity and its dependence on the different physical parameters, providing a physical interpretation for the peculiar line shape that characterizes such emission.

We consider the interaction of an intense $\hat{\sigma}_-\cdot$-polarized drive field with an intense $\hat{z}$-polarized probe or cavity field with a collection of optically pumped $^{39}\text{K}$ atoms whose relevant energy-level structure is shown in Fig. 1(a). The ground-(excited-) level manifolds are composed of states $|n,F,M\rangle$, with $n=g$ ($n=e$) and the geometry of the fields and atomic beam is shown in Fig. 1(b). Optical pumping of the atoms is accomplished by auxiliary $\hat{\sigma}_+\cdot$-polarized laser fields that selectively populate the $|g,2,2\rangle$ state. The circularly polarized drive field interacts with transitions $|g,F,M\rangle\leftrightarrow|e,F',M-1\rangle$ with Rabi frequencies $2\alpha_{F'M-1} = 2\alpha_{F'M-1}\hbar/(\hbar\chi)$ (with $\chi$ being the corresponding Clebsh-Gordan coefficients) while the linearly polarized probe field interacts with transitions $|e,F,M\rangle\leftrightarrow|g,F',M\rangle$ with Rabi frequencies $2\beta_{g,F'M} = 2\beta_{g,F'M}\hbar/(\hbar\chi)$ for any allowed value of $F, F'$, and $M$. The drive field is detuned with respect to the transition $|e,2,M\rangle\leftrightarrow|g,1,M\rangle$ by an amount $\Delta_p$ and the detuning of probe field $\Delta_p$ is defined as the difference between the probe and drive frequencies.

Two-photon amplification and lasing is modeled using the semiclassical, density-matrix formalism within the usual rotating-wave, slowly varying envelope, plane-wave and uniform-field approximations. We first summarize our findings.
concerning amplification, followed by a discussion of our results on lasing. Full details of the calculations will be published elsewhere.

II. AMPLIFICATION

From our analysis, we find that amplification of the probe beam can be predicted through a complex “gain” coefficient given by

$$\tilde{G} = \frac{U}{\beta} \left[ \rho_{e22} \hat{u}_{22} + \sum_{i=0,1} \sum_{j,k=1,2} \rho_{eji} \dot{s}_{ik} \right],$$

(1)

where $U$ is the unsaturated gain parameter. Equation (1) is obtained by summation of all the one-photon atomic coherences $\rho_{ul}$ induced on the transitions $u = |e,F,M\rangle \rightarrow l = |g,F',M\rangle$. The imaginary part of Eq. (1), or gain factor $G$, represents the relative increase in the probe-field amplitude per unit of time, and its real part is proportional to the induced change in refractive index experienced by the probe beam. Expression (1) is valid for any intensity of the drive and probe fields and its limitations arise from the fact that: (i) we have neglected the spatial nonuniformity of the fields and atomic beam and the small residual Doppler broadening; (ii) the possible perturbation of the amplification processes by the optical pumping fields has only been taken into account through the atomic relaxation rates (in case they occur simultaneously [11]); and (iii) transitions involving states with $M<0$ have been neglected.

The continuous-line curve in Fig. 1(c) shows the gain $G$ as a function of the probe-pump detuning $\Delta_p$ for the conditions reported in Ref. [11] and with no adjustable parameters. The experimental trace is shown as crosses in the figure (note that the vertical scale is offset for clarity). It is seen that there is very good agreement between the predictions of our model and the experiments, giving us confidence that we can properly assign the origin of the features observed as well as make new predictions. We note that the very small gain features to the low-frequency side of the experimental two-photon gain feature arise from scattering processes originating from states other than $|g,2,2\rangle$ and ending in states with $M<0$ [11] and thus cannot be explained within the context of our current model.

We now proceed to explain the origin of each of the features shown in Fig. 1(c). The absorption dip $A$ corresponds predominantly to the single-photon transitions $|g,2,M\rangle \rightarrow |e,2,M\rangle$. It is seen that the high-frequency side of this feature reduces the size of all the gain resonances and represents one of the main obstacles to achieve efficient two-photon amplification and lasing [11].

The gain peaks in Fig. 1(c) correspond to multiphoton processes of increasing order (nevertheless they could not be described correctly through a perturbative treatment, since the large intensity of the drive and probe fields entails large atomic-level population transfers and ac Stark level shifts that strongly affect the strength and position of the resonances). Peak $B$ corresponds to the Raman process $|g,2,2\rangle \rightarrow |g,1,1\rangle$ involving absorption of one drive-field photon and emission of one probe photon. Peaks $C$ correspond to the processes $|g,2,2\rangle \rightarrow |e,2,0\rangle$ (left peak) and $|g,2,2\rangle \rightarrow |e,1,0\rangle$ (right peak), involving absorption of two drive photons and emission of one probe photon. Thus peaks $B$ and $C$ are not appropriate for two-photon emission. Feature $D$ appearing at $\Delta_p=0$ is brought about by the quantum superposition of the Raman process $|g,2,2\rangle \rightarrow |g,2,1\rangle$ (left peak) entailing absorption of one drive photon and emission of one probe photon, and the higher-order process $|g,2,2\rangle \rightarrow |g,2,1\rangle \rightarrow |g,2,0\rangle$ (right peak) entailing absorption of one further drive photon and one further probe photon. The second process, in principle, could be interesting for two-photon lasing, but its strong interference with the one-photon Raman process makes separation of both channels difficult.

Thus, the most appropriate resonance for two-photon amplification and lasing is the peak labeled as $2ph$, which corresponds to the process $|g,2,2\rangle \rightarrow |g,1,0\rangle$ entailing absorption of two drive photons and emission of two probe photons as shown in Fig. 1(a). Although its strength seems small relatively to the other gain features, it can be controlled in a wide range through its quadratic dependence on the drive intensity.

We now focus on the two-photon gain feature and describe one of our primary results concerning interference between quantum pathways arising from the multiple degenerate atomic sublevels in the $F=1$ and 2 states. Figure 2 shows the two-photon gain resonance, for several of different pathways connecting the initial state $|g,2,2\rangle$ with the final
To verify the two-photon nature of the gain peak denoted by $2\phi h$, we determine the probe-beam gain as a function of its intensity. We find that the peak gain increases linearly with probe-beam intensity as expected for an ideal two-photon amplifier, eventually saturating, then decreasing to zero for higher intensities. Figure 3(a) shows the two-photon gain resonance for various probe-beam amplitudes spanning the maximum-gain condition. Simultaneously, the gain feature shifts to lower energy, which we find is due to the probe-field induced ac Stark shift of the final sublevel $|g,1,0\rangle$ to higher energies. As shown below, these features will make a two-photon laser to behave much differently from a normal one-photon laser.

III. LASING

To explore the properties of a two-photon laser based on laser-driven potassium atoms, we consider the case when the atomic beam propagates through an optical cavity with mirrors along the $y$ axis and orthogonal to such axis (to suppress competing wave-mixing processes). If the cavity loss rate is $\kappa$, the “working” point of the two-photon laser can be determined by the intersection of the gain curves of Fig. 3(a) with a horizontal line at height $\kappa$. More precisely, the intersection of the horizontal loss line with the right wing of each gain curve will determine a stable laser emission state, whereas the intersection with the left wing of each gain curve will determine an unstable laser emission state. This leads to the emission profile depicted in Fig. 3(b), which has the form of a closed curve detached from the trivial zero-intensity solution (see inset), and where the upper branch (continuous line) is stable (except for the possible presence of a local bifurcation), and the lower branch (dotted line) is unstable. Thus, there is a dramatic difference between this emission profile and that of a standard single-photon laser, which consists of a symmetric peak whose wings are connected with the zero-intensity branch through a local bifurcation. On the other hand, the two-photon laser can reach a high emission intensity owing to the fact that the maximum gain [shown in Fig. 3(a)] does not occur at zero intensity but at a large value of $\beta$. Figure 3(b) also depicts $-\text{Re} \hat{G}$ (stars correspond to the stable branch and squares to the unstable branch), which directly gives the frequency pushing effect. It is seen that this effect is nearly flat as a function of $\Delta_p$ and is brought about by the proximity of the large absorption dip $A$ shown in Fig. 1(c). This is in sharp contrast to the well-known frequency
and by taking into account that the cavity detuning \( \Delta_c \) is defined by \( \Delta_c = \Delta_p + \text{Re} \ G \). Figure 4 shows a typical profile of the two-photon laser emission obtained in this way in which only the stable branches have been included. The two-photon laser emission line shape is in effect similar to that of the stable branch in Fig. 3(b), except for a global red shift of \( \sim 4-5 \) given by the pushing effect shown in Fig. 3(b). Note also that the two-photon peak intensity is larger than that associated to the other gain features \( B, C, \) and \( D \), and that only the two-photon branch is completely detached from the zero-intensity branch while the other solutions are connected with it by means of unstable branches. Because the zero-intensity branch is stable only in the region of the two-photon emission, the output intensity will display bistability as \( \Delta_p \) is varied. Increasing the cavity losses would lead to shrinking of the two-photon branch from both ends until sudden disappearance of the branch, in contrast to single-photon lasers that would lead to a continuous lowering of the whole curve.

In conclusion, we have established an accurate model that allows one to investigate most aspects of the behavior of the experimental system [11,12] which is the most promising one to achieve very efficient two-photon amplification and lasing and thus to make possible investigation of the variety of properties expected for such a class of phenomena. The model not only explains the main features of the few existing experimental results, but also allows one to predict the gain and laser behavior and its dependence on the different physical parameters, showing phenomena such as interferences between different quantum channels, peculiar two-photon laser emission line shape and potential high-emission efficiency. This should be of great help in designing and interpreting the future experiments. A next step in the model will consist of generalizing it to the case of a laser field with free polarization, which should make possible to explain those recent observations [12] showing polarization instabilities and to make predictions about the possibility of creation of polarization-entangled states.

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