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Lasers without inversion in a Doppler-broadened medium

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Three-level models of lasers without inversion in V and Λ configurations are studied in a realistic Doppler-broadened gas medium. The intensity of a generated coherent radiation is numerically calculated for a wide range of experimental parameters and the role of the laser cavity is fully taken into account. Different geometrical arrangements of the experiment are compared. Significant differences in lasing properties of the studied models are shown and discussed. An amplification mechanism based on the Doppler shift tuning to dressed-state inverted transitions is discussed.

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I. INTRODUCTION

The possible applications of systems exhibiting gain without population inversion between levels involved in a laser action have stimulated intensive studies in the past few years. The process requires asymmetry between absorption and emission properties of the active medium. Theoretical models involving for this purpose autoionizing states [1,2] as well as an additional coherent driving field in two- [3,4], three- [5–8], and four-level models [9–11] have been extensively studied theoretically. While a common term—lasing without inversion (LWI)—is used to describe the properties of all such models, the origin of gain may be different in distinct cases.

Two main mechanisms of gain creation have been found. For most schemes, although the lasing appears to be without inversion in the atomic basis, a clear picture emerges once the system is represented in the so-called "dressed-state" basis obtained after diagonalizing the Hamiltonian including an atom-coherent driving field interaction. The gain is due to dressed-state inversion. The resulting inversion in the dressed-state basis for four-level schemes [10,11] may also be interpreted in the equivalent language of the population-trapping phenomenon [12].

For other proposed models, in particular those involving the autoionizing states [1,2] and some three-level schemes in resonant conditions [5–8], no simple change of basis may explain the amplification. The process is due to coherence among different ionization channels in the former and an induced coherence between dressed states in the latter systems [13]. Alternatively, the amplification may be explained in the language of coherent multiphoton processes with the asymmetry leading to net gain creation introduced by the incoherent pumping [14].

While extensively explored theoretically, a practical realization of LWI seems to be a difficult experimental task. The dressed-state lasers have been shown to work [3], but only partial success has been obtained with other lasing schemes, mainly using the population trapping mechanism [15,16]. Some experiments used a pulsed scheme and utilized not the steady state but a transient gain [16]. No success has been reported up to now for truly "inversionless" systems (of the second type described above), although a related phenomenon of self-induced transparency has been observed [17].

Most of the theoretical treatments have been restricted to linear gain study with the notable exceptions [2,4,6,11] limited, however, to the simplest situations. The aim of this paper is to explore one possible source of experimental difficulties in a realistic situation, namely, the influence of the inhomogeneous broadening due to the Doppler effect. It produces detunings of the pump laser beam and the resonant cavity frequencies from the atomic transitions, which may affect the coherent recycling in atoms and damp the intensity of the laser radiation.

Despite its importance, the effect of inhomogeneous, Doppler-effect-induced broadening has received little attention in the present context. A notable exception available in the literature is a detailed study of an enhancement of the index of refraction in the presence of vanishing absorption in atomic models which are similar to those studed here [18]. That work [18] considers a passive optical system; thus the inhomogeneous broadening may be taken into account in quite a straightforward way by convoluting the final results obtained for a Doppler-free system with the assumed atomic velocity distribution. On the contrary, in the active optical medium considered here, all atoms contribute to the dynamically evolving (and coupled to the cavity mode) macroscopic polarization. The created field influences, in turn, the atomic evolution. Therefore, a full nonlinear laser theory must include the effect of the inhomogeneous broadening from the very beginning, which makes the computation more difficult. To our knowledge, such an analysis for LWI models has not been presented up until now [19].

II. MODEL

We consider a sample of N three-level atoms in a closed cell placed inside a single-mode ring laser cavity of a passive linewidth Γ, a resonant frequency ω_c, and a wave vector of the cavity mode k_c. A thermal velocity distribution of the atomic sample is a Maxwellian function characterized by a Doppler broadening D, which is related to a temperature of the sample T by $D \sim \sqrt{1/T}$. The formalism developed is also applicable to atomic beam arrangements; then D is related to the residual Doppler width. We study two different three-level models, shown in Fig. 1, i.e., the Λ-level configuration [5] and the V-level scheme [8]. In both cases, one of the optical transitions is coupled by a strong, coherent pump beam of frequency ω_p and wave vector k_p (transition $2 \to 3$ in the Λ model and $1 \to 2$ in the V model), while the laser action occurs on the remaining transition ($3 \to 1$ for both models) in the vicinity of which the cavity resonant frequency is tuned. Additionally, an incoherent pumping on transition $1 \to 3$ is assumed (with pump rate Λ). Each of the excited states n decays radiatively to a ground state m with a transition probability $2\gamma_{mn}$. In our model we neglect processes leading to the depopulation of the ground states such as collisions or escape out of the interaction region.

The time evolution of the density matrix is described by the Liouville–von Neumann equations ($\hbar = 1$)

$$\dot{\rho} = -i\left[\mathcal{H}, \rho\right] + \mathcal{L}_F\rho + \mathcal{L}_S\rho + \mathcal{L}_P\rho. \quad (1)$$

The Hamiltonian part of the evolution reads

$$\mathcal{H} = \sum_{\mu}^{N}\left[\sum_{i=1}^{3} E_i\tilde{\sigma}_{ii}^{\mu} + \left(\Omega e^{i[\omega_p t - \vec{k}_p\cdot(\vec{r}_0^{\mu} + \vec{v}_\mu t)]}\tilde{\sigma}_{ji}^{\mu} + g e^{-i\vec{k}_c\cdot(\vec{r}_0^{\mu}+\vec{v}_\mu t)}\tilde{a}^{\dagger}\tilde{\sigma}_{13}^{\mu} + \text{H.c.}\right)\right] + \omega_c\tilde{a}^{\dagger}\tilde{a}, \quad (2)$$

where $j = 2$ and $i = 3$ for the Λ-level model and $j = 1$ and $i = 2$ for the V-level scheme. A μth atom moves with a velocity $\vec{v^{\mu}}$ and its position at t is $\vec{r_0^{\mu}} + \vec{v^{\mu}}t$. The constant g represents a coupling strength between the atomic transition and the ring-cavity mode a and Ω is the Rabi frequency of the driving field.

The Liouville operators $\mathcal{L}_F$, $\mathcal{L}_S$, and $\mathcal{L}_P$ describe the cavity damping, the spontaneous emission, and the incoherent pumping, respectively, and take the standard form [20], e.g.,

$$\mathcal{L}_F\rho = \Gamma\left(2\tilde{a}\rho\tilde{a}^{\dagger} - \tilde{a}^{\dagger}\tilde{a}\rho - \rho\tilde{a}^{\dagger}\tilde{a}\right). \quad (3)$$

To eliminate the time and spatial dependences from the equations of motion we introduce a new set of variables. For the Λ laser

$$\begin{aligned}
a &= \tilde{a}\exp\{i\omega_p t\}, \quad \sigma_i = \tilde{\sigma}_{ii}, \\
\sigma_{12} &= \tilde{\sigma}_{12}\exp\{i(\vec{k}_p - \vec{k}_c)\cdot(\vec{r}_0^{\mu} + \vec{v}^{\mu}t)\}, \\
\sigma_{13} &= \tilde{\sigma}_{13}\exp\{i[\omega_p t - \vec{k}_c\cdot(\vec{r}_0^{\mu} + \vec{v}^{\mu}t)]\}, \\
\sigma_{23} &= \tilde{\sigma}_{23}\exp\{i[\omega_p t - \vec{k}_p\cdot(\vec{r}_0^{\mu} + \vec{v}^{\mu}t)]\}
\end{aligned} \quad (4)$$

(the transformation for the V model is obtained by an appropriate permutation of indices).

Since in our work we are interested in laser light intensity rather than in noise properties, we follow the semiclassical approach in further calculations. Thus we decorrelate the atomic and field variables and find a set of master equations in the rotating-wave approximation for the quantum averages of atomic and field observables. For the Λ model we obtain explicitly

$$\begin{aligned}
\dot{a} &= -\left[\Gamma + i\left(\omega_c - \omega_p\right)\right]a - ig\sum_{\mu}^{N}\sigma_{13}^{\mu}, \\
\dot{\sigma}_1^{\mu} &= i\left(g^{*}a\sigma_{31}^{\mu} - ga^{\dagger}\sigma_{13}^{\mu}\right) + 2\left(\gamma_{13} + \Lambda\right)\sigma_3^{\mu} - 2\Lambda\sigma_1^{\mu}, \\
\dot{\sigma}_2^{\mu} &= i\Omega\left(\sigma_{32}^{\mu} - \sigma_{23}^{\mu}\right) + 2\gamma_{23}\sigma_3^{\mu}, \\
\dot{\sigma}_3^{\mu} &= -i\Omega\left(\sigma_{32}^{\mu} - \sigma_{23}^{\mu}\right) - i\left(g^{*}a\sigma_{31}^{\mu} - ga^{\dagger}\sigma_{13}^{\mu}\right) \\
&\quad -2\left(\gamma_{23} + \gamma_{13} + \Lambda\right)\sigma_3^{\mu} + 2\Lambda\sigma_1^{\mu}, \\
\dot{\sigma}_{12}^{\mu} &= i[(\vec{k}_p - \vec{k}_c)\cdot\vec{v}_\mu - \omega_{21}]\sigma_{12}^{\mu} - i\Omega\sigma_{13}^{\mu} \\
&\quad + ig^{*}a\sigma_{32}^{\mu} - \Lambda\sigma_{12}^{\mu}, \\
\dot{\sigma}_{13}^{\mu} &= i(\omega_p + \omega_{13} - \vec{k}_c\cdot\vec{v}_\mu)\sigma_{13}^{\mu} - i\Omega\sigma_{12}^{\mu} \\
&\quad + ig^{*}a\left(\sigma_3^{\mu} - \sigma_1^{\mu}\right) - \left(\gamma_{23} + \gamma_{13} + 2\Lambda\right)\sigma_{13}^{\mu}, \\
\dot{\sigma}_{23}^{\mu} &= i(\omega_p - \omega_{32} - \vec{k}_c\cdot\vec{v}_\mu)\sigma_{23}^{\mu} + i\Omega\left(\sigma_3^{\mu} - \sigma_2^{\mu}\right) \\
&\quad - iga\sigma_{21}^{\mu} - \left(\gamma_{23} + \gamma_{13} + \Lambda\right)\sigma_{23}^{\mu},
\end{aligned} \quad (5)$$

with a similar set of equations for the V model. The velocity-dependent terms describe the Doppler shifts of the pump beam frequency and the cavity frequency from the respective atomic transitions. The effect depends (see the equation for σ_{12}^{μ} above) on the mutual orientation of the driving laser beam and the cavity axis.

For further discussion we consider two geometrical realizations of the experiment: (i) the collinear configuration (CC), in which the pump beam is sent along the optical axis of the cavity, and (ii) the orthogonal configuration (OC), in which the pump beam illuminates the cell from the side $\vec{k}_p \perp \vec{k}_c$.

Note that the CC requires the application of wave-

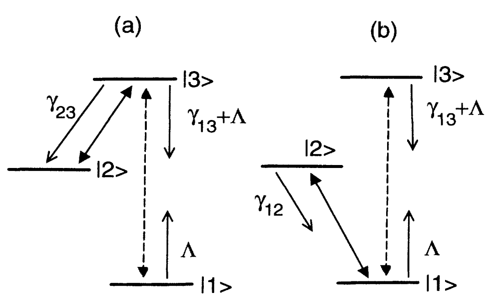

FIG. 1. Two-level schemes studied in our paper: (a) the Λ-level model, and (b) the V-level scheme. One of the optical transitions is coupled by a strong pump beam (solid line, double arrow) and the lasing (dashed line) occurs on the remaining transition to which the cavity resonant frequency is tuned. The remaining arrows indicate pumping and spontaneous decay.

length selective mirrors that are transparent for the pump beam wavelength and highly reflective for the generated coherent radiation. However, for the energy splitting of the two optical transitions larger than several nanometers, such mirrors are commercially available.

III. NUMERICAL RESULTS

To facilitate the numerical integration of the problem we divide the velocity distribution into discrete groups along the relevant direction. The population of those groups is numerically calculated using a Gaussian quadratic procedure and all atoms belonging to a given group are assumed to move with the same mean group velocity.

For the CC ($\vec{k_p} \parallel \vec{k_c}$) the problem is one dimensional since the equations become dependent only on the velocity component parallel to the cavity optical axis. On the other hand, the OC ($\vec{k_p} \perp \vec{k_c}$) requires integration over the two-dimensional velocity space. That significantly increases the number of equations. In calculations the number of velocity groups is typically 30 for the CC and 64 for the OC, corresponding to 272 and 578 real equations, respectively.

We assume natural initial conditions for atomic variables (atoms in the ground state with vanishing initial atomic coherences). As usual in semiclassical laser theory, $a = 0$ gives a trivial solution. We take, therefore, a small nonzero value for the initial field $a(0)$. Equations are numerically integrated using the standard predictor-corrector method. The system of equations reaches a stable solution typically after the time of the order of 300–500 lifetimes of the excited level.

We have studied numerically the behavior of both the V and Λ models in a wide range of parameters [21]. For simplicity let us consider here only the resonant case [$\omega_c = \omega_{31}$ and $\omega_p = \omega_{ij}$, in the index convention of Eq. (2)]. Then the analytic solution for the laser intensity is available for the limiting homogeneous case ($D = 0$) both for the Λ model [6] and for the V system [21]. Both models lead then to LWI in any basis.

The question we now pose is whether this character of the lasing is preserved in the presence of the inhomogeneous Doppler-effect-induced broadening ($D > 0$). For atoms at rest the gain is peaked around the two-photon resonance ($\omega_p - \omega_c = \omega_{ij} - \omega_{13}$). The Doppler effect smears out the resonance so that the intensity decrease with increasing D may be expected. The effect is due to the $(\vec{k}_c - \vec{k}_p) \cdot \vec{v}^{\mu}$ term in Eqs. (5). Note that the influence of the inhomogeneous broadening is sensitive to the mutual orientation of $\vec{k}_c$ and $\vec{k}_p$. We discuss the ring-cavity arrangement in which the driving field and the generated field are copropagating. For counterpropagating fields the Doppler effect will be stronger due to the minus sign above. Let us also note that had we considered the ladder atomic level scheme, the transformation to time-independent frame would have resulted in the $(\vec{k}_c + \vec{k}_p) \cdot \vec{v}^{\mu}$ term in Eqs. (5) and the counterpropagating configuration would have been favored.

For the CC the value of the net detuning is small and may vanish when both transitions are degenerate in frequency. Yet one would prefer a large difference between both transition frequencies to be able to generate radiation in a new frequency range. We assume $|\vec{k_c}| = 1.5|\vec{k_p}|$ so the generated frequency is significantly blueshifted with respect to the driving frequency. For the OC the two-photon resonance condition is not satisfied for most of the active atoms, which should strongly influence lasing capabilities.

The intensity of the Λ laser as a function of the Doppler width D is presented in Fig. 2. All frequencies and decay rates are expressed in units of γ_{23}. The laser intensity is dimensionless, measured by the corresponding photon number $I = |a|^2$. Indeed the CC setup works better; lasing is suppressed for smaller inhomogeneous broadening for the OC. However, even for the CC the Doppler width permitting lasing is roughly one order of magnitude smaller than the typical inhomogeneous broadening in the atomic cells at the realistic temperatures. On the other hand, much smaller residual widths are possible in the atomic beam experiment. Then, however, the degree of beam collimation is strongly related to the maximum atomic density in the active region. We have verified that for the realistic residual width $D = 8$ the threshold atomic densities for the CC are four *orders of magnitude* smaller than for the OC.

The parameters chosen for the Λ system correspond to LWI in any atomic basis in the linear gain theory [5,13]. As already noted in Ref. [6], for a homogeneously broadened model, a nonlinear theory yields negative inversion only very close to the threshold. Once lasing is established, inversion in the Λ model is created via redistribution of the population due to the generated light. A similar situation occurs for our Doppler-broadened system. By integrating the steady-state populations over the velocity profile we have found that the steady-state laser action is accompanied by atomic inversion (except for the largest D, just at the border of lasing). A lack of

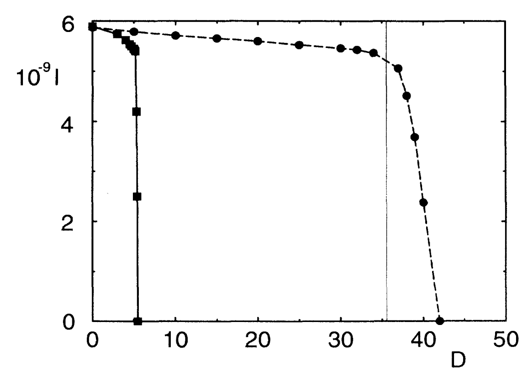

FIG. 2. Λ-level scheme. Intensity of the generated radiation versus the Doppler width D. $N = 10^{10}$, $\Omega = 30$, $\Gamma = 10^{-2}$, $g = 10^{-3}$, $\Lambda = 10^{-1}$, $\gamma_{13} = 0.5$, and $\gamma_{23} = 1$. The squares connected by the solid line correspond to the orthogonal configuration. The circles connected by the dashed line represent the lasing intensity in the collinear configuration. Lasing without inversion appears for the CC on the shoulder of lasing, to the right of thin vertical line.

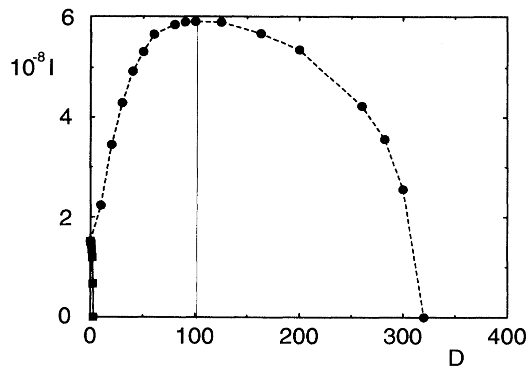

FIG. 3. Same as Fig. 2, but for the V scheme. $N = 10^9$, $\Lambda = 0.75$, and the remaining parameters are the same as in Fig. 2 (but in units of γ_{12}). Lasing without inversion appears in the CC for $D < 105$, to the left of the thin vertical line.

inversion occurs for smaller $N = 5 \times 10^8$; then the lasing D interval is much smaller than in Fig. 2.

Figure 3 presents the calculated laser intensity as a function of the Doppler width for the V model. We keep the values of atomic, cavity, and pump field parameters the same as before, but the incoherent pumping rate Λ value must be significantly larger to achieve lasing. The difference between both considered geometries is even more striking. For the OC the lasing occurs only for very small inhomogeneous broadening. For the CC the surprising growth of the lasing intensity with Doppler width is observed. Strong lasing persists for inhomogeneous broadening values corresponding to typical, realistic temperatures of the gas sample.

To explain the striking behavior of the V-laser model we note, following Zhu [8], that for the driving beam sufficiently detuned from the resonance the gain appears due to the population inversion in the dressed-state basis. Moreover, the gain profile is peaked around the shifted frequency value corresponding to the transition from state 3 towards one of the dressed states. Thus, to achieve an efficient amplification, ω_c should be tuned to that frequency. Such is the situation in a homogeneously broadened system.

Consider a single nonzero velocity group. For the moving atoms both ω_p and ω_c are detuned from the resonance in the *same* direction. The moving atom "sees" the cavity shifted in the vicinity of the inverted transition in the (velocity-dependent) dressed-state basis and acts as an amplifier. Note that the effect is not dependent on the velocity sign. A change of the velocity sign changes the sign of the Doppler shift, i.e., the sign of the driving field detuning from the resonance with a given velocity group. Then the relative populations of dressed-states also change [22]. But as the cavity-mode Doppler shift is also dependent on the velocity sign, the cavity remains close to resonance with the inverted transition. The detuning from the inverted dressed-state transition is dependent, via Doppler shifts, on the velocity value for a given velocity group and on a relative magnitude of driving and generated waves' wave vectors. Thus some of the velocity groups contribute via this mechanism much more strongly to the observed gain than the other groups. At the same time, low-velocity groups contribute also to gain due to the LWI mechanism of Ref [8]. That explains the effect observed.

An amplification mechanism (and lasing) based on an inverted dressed-state transition has been studied in detail both theoretically and experimentally [3,4,10,11] for homogeneously broadened systems. An interesting feature of the inhomogeneously broadened system considered here is that the required driving laser detunings appear as Doppler shifts. The discovered velocity-dependent amplification enhancement mechanism discussed above is very sensitive to the mutual directions of the driving coherent field and the ring-cavity mode. Consider that we discuss here the case when $\vec{k}_p$ and $\vec{k}_c$ are parallel (copropagating pumping and generated beams). For antiparallel orientation (counterpropagating beams) ω_p and ω_c would be shifted in opposite directions in the atomic frame (defined for each velocity group separately). Then the similar dressed-state analysis shows that the dressed atoms would act as strong absorbers (since the cavity would be "tuned" to the noninverted dressed-state transition). In a similar way one can convince oneself that for the ladder level scheme the counterpropagating configuration would be favored.

For the OC a small fraction of atoms only is effectively tuned in the vicinity of the dressed transition. The number of atoms contributing to gain becomes very low for higher Doppler widths and the lasing does not occur.

By monitoring the final populations of dressed states and integrating them over the velocity profile we have found that lasing without inversion in the dressed basis occurs for $D < 100$ for the parameters in the figures, while dressed-state inversion dominates lasing for higher values of the Doppler width.

IV. CONCLUSIONS

To summarize, we have presented a theoretical analysis of lasers without inversion in the realistic inhomogeneously broadened medium. We have found that the possibility of observing the lasing depends strongly on the mutual orientation of the pumping laser beam and the ring-cavity axis. Although our theory is not valid for standing-wave cavities, one may expect intuitively that the effect of the inhomogeneous broadening in such cavities will be rather similar to the lasing observed here in the less favorable orthogonal configuration. The waves propagating in both directions have to be considered in such a cavity and the two-photon resonance condition will be strongly suppressed.

Although the V-laser model [8] requires much stronger incoherent pumping in order to lase as compared to the Λ scheme [5], the lasing in the former model may actually be enhanced by the inhomogeneous broadening. This is due to the strong amplification generated in the Doppler-broadened medium. The origin of this mechanism has been traced to gain on inverted dressed-state transitions where the unequal populations of dressed states are due to effective detunings caused by Doppler shifts. For sufficiently strong incoherent pumping and large atomic densities both models may lase efficiently with atomic cell arrangement in a *collinear* geometry only. As far as experimental verification of LWI (in any basis) is concerned, it seems that well collimated atomic beams are necessary (and also the CC) since small Doppler widths are required. For the Λ model, LWI appears only just above the lasing threshold (similarly to the $D = 0$ case [6]).

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