Gain due to level-dependent collisions

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Based on the level dependence of collision cross sections, a mechanism of light amplification without inversion in a Doppler-broadened three-level system is further studied. In the rate equations, we use a collision-kernel model and consider all of the decay and pumping rates. The emission and absorption profiles are derived by solving the coupled mass-velocity equations numerically. Then, the possibility of obtaining positive gain with noninversion for an ensemble of particles is shown.

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I. INTRODUCTION

It was proposed many years ago that population inversion is not a necessary condition for light amplification. Holt [1] reported that lasing without inversion might result from the splitting of emission and absorption spectra due to atomic recoil, but this effect is very small and would be useful in practice only at very high transition frequencies. Arkhipkin and Heller [2] showed that, for a discrete level embedded in a continuum, Fano interference occurs, providing another possible way to achieve noninversion amplification, but this method does not allow many atoms in the discrete levels to acquire higher gain. A few years ago, Harris [3] considered a purely lifetime-broadened three-level system, where the emission and absorption line shapes were different because of Fano-type interference, and where lasing without inversion was believed to be a practical approach. Since then, much research effort has been devoted to this area [4–18].

In Ref. [19] we proposed a new model of light amplification without inversion. We considered a predominantly Doppler-broadened three-level system, where the emission and absorption spectral line shapes were different due to the level dependence of collision cross sections. However, in that paper, we did not account for the influence of pumping and decay rates during the collision process. In this paper we explore this model further. The rate equations include all of these pumping and decay processes, and a collision-kernel model is used to deal with the velocity-changing collision term. Then we obtain coupled mass-velocity equations of excited and ground levels, which have been solved numerically. From the velocity redistribution, it is shown that positive gain can be achieved in some frequency range even if more atoms are in the ground state than in the excited state. Finally, we discuss the conditions that influence this noninversion light amplification effect.

II. THEORETICAL MODEL

We consider a flowing-gas system [shown in Fig. 1(a)]. The target gas emerges from a small hole with mass velocity V_f into a container filled with a buffer gas. The target gas has three Doppler-broadened levels [shown in Fig. 1(b)]. The ground-level atoms are excited to level 3 by electron or optical pumping, and then they relax rapidly to level 2. If we suppose that this process is so quick that the velocity-changing collisions of level 3 can be neglected, i.e., this process just provides the population of level 2, and we can then focus on levels 1 and 2. Since the collision cross sections are level dependent, the velocity-changing collision rates of level 1 or 2 are different, so that the velocity distributions of level 1 or 2 are changed differently when the target gas collides with the buffer gas along the Z axis. Hence there is a displacement of the velocity redistributions between levels 1 and 2, as shown in Fig. 2. The displacement results in different emission and absorption line shapes. By this means, local inversion is made possible, under the condition of noninversion for the ensemble of atoms, and thus—in the corresponding frequency range—light amplification is achieved.

In the above case, the level dependence of the collision cross sections is the physical reason for the noninversion light amplification. To observe this phenomenon, there must be a buffer gas acting as a collision perturber, and the target gas must have nonzero mass velocity relative to the perturber gas; the spectral lines are dominated by Doppler-broadened shapes.

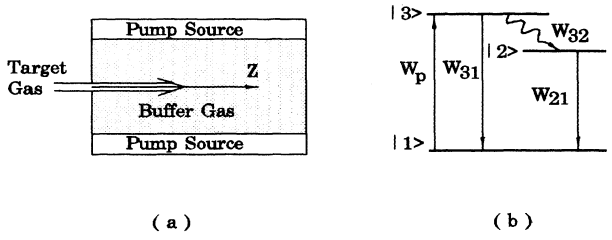

FIG. 1. (a) System scheme. (b) Level scheme. W_p is the pumping rate. W_{ij} is the decay rate from level i to j.

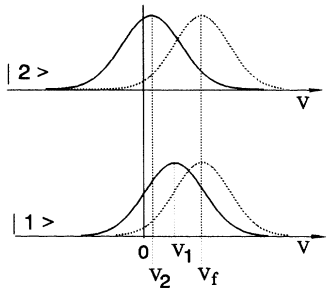

FIG. 2. Dashed lines represent the velocity distributions of ground-state and excited-state atoms $f_1(V,0)$ and $f_2(V,0)$ at $Z=0$, and at this point the mass velocities of the two states atoms are the same, V_f. The solid lines are the velocity distributions $f_1(V,L)$ and $f_2(V,L)$ at $Z=L$; the mass velocities of the lower- and upper-level atoms are V_1 and V_2, respectively.

III. DERIVATION AND DISCUSSION

Since the target gas flows along the Z axis, and the diffusion along other directions is neglected, we just deal with the one-dimensional case.

We define $\sigma_i(Z,V,t)$ as the velocity distribution of state $|i\rangle$ over time t and distance Z, where $i=1,2,3$. Z is the distance from the hole. First, we derive the time evolution of $\sigma_i(Z,V,t)$; the rate equations can then be written as follows:

$$\frac{\partial}{\partial t}\sigma_3(Z,V,t)=W_p\sigma_1(Z,V,t)-W_{31}\sigma_3(Z,V,t)-W_{32}\sigma_3(Z,V,t)-V\frac{\partial}{\partial Z}\sigma_3(Z,V,t)+\mathcal{L}_3(\sigma_3(Z,V,t)) , \quad (1)$$

$$\frac{\partial}{\partial t}\sigma_2(Z,V,t)=W_{32}\sigma_3(Z,V,t)-W_{21}\sigma_2(Z,V,t)-V\frac{\partial}{\partial Z}\sigma_2(Z,V,t)+\mathcal{L}_2(\sigma_2(Z,V,t)) , \quad (2)$$

$$\frac{\partial}{\partial t}\sigma_1(Z,V,t)=W_{21}\sigma_2(Z,V,t)+W_{31}\sigma_3(Z,V,t)-W_p\sigma_1(Z,V,t)-V\frac{\partial}{\partial Z}\sigma_1(Z,V,t)+\mathcal{L}_1(\sigma_1(Z,V,t)) , \quad (3)$$

where W_{ij} is the decay rate from level i to j, and W_p is the pumping rate. $-V\partial/\partial Z$ is a free-flow term and $\mathcal{L}_i$ is the velocity-changing collision operator of state $|i\rangle$. If we assume that W_{32} and W_{31} are much larger than the velocity-changing collision rate and the free-flow rate, so that in the steady state $(W_{31}+W_{32})\sigma_3=W_p\sigma_1$, we then have

$$\frac{W_{32}W_p}{W_{31}+W_{32}}\sigma_1(Z,V)-W_{21}\sigma_2(Z,V)-V\frac{\partial}{\partial Z}\sigma_2(Z,V)+\mathcal{L}_2(\sigma_2(Z,V))=0 , \quad (4)$$

$$W_{21}\sigma_2(Z,V)-\frac{W_{32}W_p}{W_{31}+W_{32}}\sigma_1(Z,V)-V\frac{\partial}{\partial Z}\sigma_1(Z,V)+\mathcal{L}_1(\sigma_1(Z,V))=0 . \quad (5)$$

By definition, we have

$$\sigma_i(Z,V,t)=n_i(Z,t)f_i(Z,V,t) ,$$
$$\int\sigma_i(Z,V,t)dV=n_i(Z,t) .$$

n_i is the density of target atoms in state $|i\rangle$ and f_i is the velocity distribution function. If the target atom is heavy compared to the buffer-gas atom, the velocity change then occurs in small steps; thus in the steady state we can assume local equilibrium, and the velocity distributions then take the following form:

$$f_i(Z,V,t)=\left[\frac{m}{2\pi kT}\right]^{1/2}\exp\left[-\frac{m}{2kT}[V-V_i(Z,t)]^2\right] , \quad (6)$$

where $V_i(Z,t)$ is the mass velocity, and m is the mass of the target atom.

Since

$$\int\mathcal{L}_i[\sigma_i(Z,V,t)]dV=0$$

and

$$\int(V\partial\sigma_i/\partial Z)dV=\partial(n_iV_i)/\partial Z=\partial j_i/\partial Z ,$$

where j_i is the flux density, integrating Eq. (4) or (5) over V, we have

$$\frac{W_{32}W_p}{W_{31}+W_{32}}n_1=W_{21}n_2 . \quad (7)$$

We define $r=n_2/n_1=W_{32}W_p/W_{21}(W_{31}+W_{32})$. If $r\leq 1$, there is no population inversion for the ensemble of atoms.

Multiplying Eqs. (4) and (5) by V^2 and then integrating them over V, we get

$$\frac{W_{32}W_p}{W_{31}+W_{32}}n_1(Z)\left[\frac{kT}{m}+V_1^2(Z)\right]-W_{21}n_2(Z)\left[\frac{kT}{m}+V_2^2(Z)\right]-\frac{\partial}{\partial Z}\left\{n_2(Z)\left[\frac{3kT}{m}V_2(Z)+V_2^3(Z)\right]\right\}+\int V^2\mathcal{L}_2(\sigma_2(Z,V))dV=0 , \quad (8)$$

$$W_{21}n_2(Z)\left[\frac{kT}{m}+V_2^2(Z)\right]-\frac{W_{32}W_p}{W_{31}+W_{32}}n_1(Z)\left[\frac{kT}{m}+V_1^2(Z)\right]$$
$$-\frac{\partial}{\partial Z}\left\{n_1(Z)\left[\frac{3kT}{m}V_1(Z)+V_1^3(Z)\right]\right\}+\int V^2\mathcal{L}_1(\sigma_1(Z,V))dV=0\ . \quad (9)$$

In the steady state,

$$\frac{\partial n_i}{\partial t}=\frac{-\partial j_i}{\partial Z}=0\ . \quad (10)$$

One can show that

$$\frac{\partial}{\partial Z}\left\{n_i(Z)\left[\frac{3kT}{m}V_i(Z)+V_i^3(z)\right]\right\}$$
$$=2n_i(Z)V_i^2(Z)\frac{\partial}{\partial Z}V_i(Z)\ . \quad (11)$$

The velocity-changing collision term can be treated by using collision-kernel models. When the heavy target atoms are immersed into light buffer-gas atoms and are in the steady state, a microscopic Brownian-motion model can be used. Thus, the velocity-changing collision terms is assumed to be [20]

$$\mathcal{L}_i(\sigma_i(Z,V,t))=\xi_i\frac{\partial}{\partial V}\left[\left[V+\frac{kT}{m}\frac{\partial}{\partial V}\right]\sigma_i(Z,V,t)\right]\ . \quad (12)$$

Upon substituting Eq. (6) into Eq. (12) and after partial integration, we obtain

$$\int V^2\mathcal{L}_i(\sigma_i(Z,V,t))dv=-2\xi_i n_i V_i^2\ , \quad (13)$$

where ξ_i is the velocity-damping coefficient. Here we just consider the collisions between the target gas and the buffer gas. It is true that the real situation involving the collision processes is quite complicated. Collisions in all directions as well as along different lines should be considered. However, since from the latter part of this paper one can estimate that the maximum mass-velocity difference between the target atoms in states $|1\rangle$ and $|2\rangle$ occurs mostly very close to the entrance hole, discussion can be limited to the region not far away from the entrance hole. In this region the Z-component velocity is much larger than the velocity components in the X and Y directions. Hence we can just deal with the one-dimensional case in order to avoid complicated mathematical derivations and to emphasize the physical effect. On the other hand, the relative mass velocity between atoms in states $|1\rangle$ and $|2\rangle$ is much smaller than that between the target atoms and the buffer-gas atoms. For the above two reasons, the damping between the different-state target atoms is neglected. We have [20]

$$\xi_i=\frac{kT}{mD_i},\quad i=1,2 \quad (14)$$

where D_i is the transport coefficient of the $|i\rangle$-state target atoms in the buffer gas; D_i is expressed as [21,22]

$$D_i=\frac{3kT}{4\mu n_b v_r S_i}\quad i=1,2 \quad (15)$$

where n_b is the density of the buffer gas, S_i is the energy-averaged collision cross section, μ is the reduced mass, and v_r is the relative velocity between the two kinds of atoms. Thus we obtain

$$\xi_i=\tfrac{2}{3}an_bS_iv_r\ , \quad (16)$$

where $a=2m_b/(m+m_b)$, and m_b is the mass of the buffer-gas atom. Since the S_i values of different states are generally not the same, the velocity-damping coefficients are level dependent. This is the internal reason for the effect described in this paper. In fact, some other effects are also due to this level dependence of the velocity-damping coefficients, such as light induced drift, for example (see Ref. [20]).

In the steady state the damping due to thermodiffusion is neglected and the collision damping is caused only by the directional movement, so $v_r=V_i$:

$$\xi_i=\tfrac{2}{3}an_bS_iV_i\ . \quad (17)$$

Substituting Eqs. (11), (13), and (17) into Eqs. (8) and (9), we obtain coupling equations for V_1 and V_2:

$$2V_2^2(Z)\frac{\partial}{\partial Z}V_2(Z)=\frac{W_{32}W_p}{r(W_{31}+W_{32})}V_1^2(Z)$$
$$-W_{21}V_2^2(Z)-\tfrac{4}{3}an_bS_2V_2^3(Z)\ , \quad (18a)$$

$$2V_1^2(Z)\frac{\partial}{\partial Z}V_1(Z)=rW_{21}V_2^2(Z)-\frac{W_{32}W_p}{W_{31}+W_{32}}V_1^2(Z)$$
$$-\tfrac{4}{3}an_bS_1V_1^3(Z)\ . \quad (18b)$$

We use the Runge-Kutta method to solve Eqs. (18a) and (18b) numerically. Figure 3 shows $\Delta V=V_1-V_2$ as a function of Z. If S_2 is larger than S_1, the excited atoms slow down more quickly than the ground-state atoms, and if S_2 is smaller than S_1, ΔV is negative. Hence, due to the level dependence of the collision cross sections, different velocity redistributions for levels 1 and 2 can be obtained, which leads to the separation of absorption and emission profiles.

Using the formal density-matrix method, one can easily obtain the following formula for the gain of a probe light through this system:

$$G\propto g(\Delta v,r,\Delta V)=r\exp(-x^2)-\exp[-(x-b)^2]\ , \quad (19)$$

where

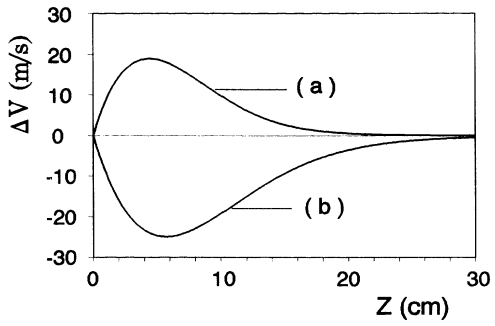

FIG. 3. Relation between ΔV and Z. For curve (a), $\Delta S/S_1=0.35$; for curve (b), $\Delta S/S_1=-0.35$. The other parameters are $V_f=250$ m/s, $n_b=2.1\times10^{20}$ m^{-3}, $W_{21}=10^3$ s^{-1}, $r=1$, $S_1=2.9\times10^{-19}$ m^2, and $a=0.3$.

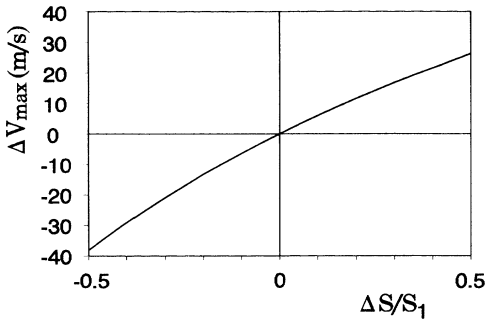

FIG. 4. $\Delta V_{\max}$ as a function of ΔS. Here, $V_f=250$ m/s, $n_b=2.1\times10^{20}$ m^{-3}, $W_{21}=10^3$ s^{-1}, $S_1=2.9\times10^{-19}$ m^2, $r=1$, and $a=0.3$.

$$x=\left[\frac{m}{2kT}\right]^{1/2}\left[\frac{C}{\nu_0}\Delta\nu-V_2(Z)\right],$$

$$b=\left[\frac{m}{2kT}\right]^{1/2}\Delta V,\quad \Delta\nu=\nu-\nu_0\,.$$

ν is the frequency of the probe light, and ν_0 is the center frequency of the transition from $|2\rangle$ to $|1\rangle$. If $g(\Delta\nu,r,\Delta V)>0$, then G has a positive value. It is easy to see that positive gain can be obtained for some frequency range even if $r\leq1$, i.e., amplification without population inversion. The phase "without population inversion" used here relates to the ensemble of all atoms in an inhomogeneous system. It does not have the same meaning as in a homogeneous system. The significance of this kind of noninversion is that it also lowers the demand on pumping power. The gain here is generated from a certain velocity group for which there is inversion, but for the ensemble of atoms there is no population inversion. The inversion of the specific velocity group is nonreciprocal to the pumping source, and it is formed by the different velocity redistributions of the excited and ground states due to the level dependence of collision cross sections between the target atoms and buffer gas under the condition of noninversion for the ensemble of atoms. In a lifetime-broadened noninversion system, the gain is obtained generally based on a quantum-interference effect, which creates different emission and absorption profiles.

In order to discuss the influence of other factors on the gain, we define $\Delta V_{\max}$ as the maximum ΔV along Z, and from Eq. (18) we draw curves of $\Delta V_{\max}$ as functions of $(S_2-S_1)/S_1$ and of W_{21}, respectively; see Figs. 4 and 5. In order to obtain a more noticeable gain, two conditions should be met. One is that the lifetime of $|2\rangle$ should be longer, and the other is that the difference between the collision cross sections of states $|2\rangle$ and $|1\rangle$ should be larger. However, for common atom systems, although the latter condition can usually be met, the lifetime of an electronically excited state is generally very short. For example, we select sodium vapor as the target-atom beam and helium as the perturber gas. The $4S$ state of Na is level 3. Since the coupling of the fine structure $3\,^2P_{1/2}$ and $3\,^2P_{3/2}$ of $3P$ is so strong that atoms in the two states cannot be separated spatially, the fine structure is completely mixed and $3P$ acts as level 2. $3S$ is the ground level. Based on the potentials of Pascal, considering the statistic weight, the average of relevant collision cross-section ratio $(S_2-S_1)/S_1$ is 0.35 [20]. The corresponding b is of the order of the value of the recoil effect [1]. On the other hand, for vibrationally excited molecules, the lifetime of the upper level is long, but the cross-section difference between excited and ground states is generally not large enough. For instance, in the CH_3F/He system, $(S_2-S_1)/S_1$ is about 0.01 [23]. b is about 10^{-3}, three to four orders larger than that of the recoil effect, but still small. However, one can try to select a system fitting the two conditions simultaneously. A metastable state of atoms or an electronically excited state of molecules acting as an upper level may be better choices. One can qualitatively estimate that the effect can become greater in these two cases. Unfortunately, the data for quantitative calculation are not yet obtainable.

In the above, by using a collision kernel, we have obtained the velocity redistributions, in the system considered after solving coupled equations, and, then, from the gain formula, we have shown that positive gain can

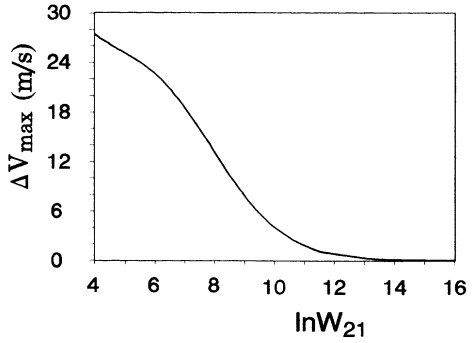

FIG. 5. Relation between $\Delta V_{\max}$ and W_{21}. The parameters are $V_f=250$ m/s, $n_b=2.1\times10^{20}$ m^{-3}, $W_{21}=10^3$ s^{-1}, $S_1=2.9\times10^{-19}$ m^2, $S_2=3.9\times10^{-19}$ m^2, $r=1$, and $a=0.3$.

be obtained even if the ensemble of atoms has no population inversion. The effect of level-dependence collision is to split the Doppler-broadened absorption and emission spectra. Although in some systems this new effect is small, it is generally larger than the recoil effect described in Ref. [1].

ACKNOWLEDGMENTS

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