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Light amplification without population inversion: Time-dependent study of (1 + 1)-photon emission from autoionizing states

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In a time-dependent dressed-state calculation we have studied the time development of (1 + 1)-photon amplification without population inversion from an autoionizing (AI) state which is coupled to another AI state embedded into the same continuum. We have shown that the (1 + 1)-photon gain evolves with time over the single-photon gain and it can be controlled by changing the relative strength of the coupling of the resonant intermediate state with the ground state and with the two AI states. We have also shown that three-peaked gain on the upper probe field evolves with time and we have demonstrated that the modification of the second peak and the appearance of the third peak are solely due to the presence of the second AI state.

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Recent interest [1–4] in the field of lasing without population inversion has revealed that single-photon lasing from autoionizing (AI) states is possible in the absence of population inversion. In the above cases [1,2] it has been shown that gain occurs in a localized region around the Fano-type absorption minimum and it persists for a much longer time in the presence of another AI state embedded into the same continuum. Moreover, by introducing extra coupling between these two AI states, lasing is obtained in two different spectral ranges [3]. In a semiclassical treatment, which dynamically includes the atom-field coupling in the cavity, the steady-state properties for the inversionless lasing between a bound state and an autoionizing resonance have been studied [5] and the intensity-dependent frequency-pulling and bistability effects were found. Recently, resonantly enhanced two-photon gain has been observed [6] in a driven two-level atom, and it has been shown that two-photon lasing occurs from inverted dressed states in the presence of an intermediate resonant state. In recent experiments [7] the amplification of a probe field and its temporal behavior have been studied in the absence of inversion in atomic systems in a different configuration (i.e., double-lambda scheme). Extensive theoretical study [8] has been done of this type of configuration where two levels are strongly driven by a laser field.

In a previous work [9] we have done a time-independent perturbative calculation (within the limit of the weak-field approximation) in a scheme where two AI states are embedded into different continua (i.e., coupling between two AI states via the continuum is absent). We have shown that the two-photon gain without population inversion is possible and a three-peaked gain for single-photon emission can be obtained due to the interference of near-resonant and resonant AI channels. In the present work we have done a time-dependent nonperturbative calculation for a scheme where two AI states are embedded into the same continuum (i.e., coupled by configuration interaction via the common continuum) and studied the time development of single-photon and two-photon amplification. We have shown that in this case three-peaked gain for a single photon emission can be obtained even when the above-mentioned interference effect is absent, and this switches over to (1 + 1)-photon gain while evolving with time. Furthermore, (1 + 1)-photon amplification can be controlled not only by changing the relative strength of couplings between different bound states (as was shown in our previous calculation) but also by changing the probe pulse durations. We have chosen an AI state as the upper lasing level because of the fact that these high-lying states can cause generations in vacuum ultraviolet and lower wavelength regions, and since these levels are short-lived, it is difficult to create population inversion in these levels. In this scheme, our results give the temporal behavior of amplification on both the probe pulses and further show that amplification can be obtained where the conventional lasing is difficult to achieve. Actually, this type of configuration exists in small diatomic molecules and can be used to test the possibility of lasing in these systems. Since in recent experiments [7] the temporal behavior of single-photon amplification is being studied, the present study on the time evolution of single-photon and two-photon amplification, although in a different configuration, can be considered to be timely.

For the gain on the upper probe field connecting the intermediate resonant state with the AI states our findings are significantly different from the previous results on single-photon lasing from AI states. The advantages over the previous schemes are (i) the gain extends over a wide range of frequencies covering the resonant region (even when both the Fano q parameters are nonzero); (ii) the gain is peaked at three spectral ranges (even in the absence of any extra coupling between two AI states); and (iii) the gain persists for a much longer time (more than an order of magnitude greater) than that obtained in previous calculations. This is because of the fact that the first resonant step in the present scheme can create a bottleneck to the process of absorption, resulting in a slower rise in absorption probability with time. The persistence of gain for a much longer time than that obtained in previous calculations and the possibility of obtaining amplification in a wide range of frequencies make the present scheme more feasible than that for single-photon lasing from AI states. It is to be mentioned here that in this model we have not considered the effect of cavity damping on the amplification process.

Recently, in three-level, four-level, and multilevel systems, where two levels are driven by a laser field, the possibility of single-photon lasing with or without population inversion has been studied [10], considering the effect of incoherent pumping from the lower lasing level. In this calculation we have considered that the pumping to the AI state is being done from outside this four-level system by electrical discharge, charge transfer in an AI state, or by a single-photon transition from lower levels other than the lower lasing level, the single-photon transition from the lower lasing level to the AI state being forbidden. We have assumed that these lower levels form a large reservoir [3], and hence the change in population in these levels has been neglected.

The model system considered here is shown schematically in Fig. 1. A two-photon transition from the lower lasing level $|g\rangle$ to the upper lasing level $|a\rangle$, which is an AI state, occurs via a resonant state $|i\rangle$. $|b\rangle$ is another AI state embedded into the continuum $|c\rangle$ adjacent to $|a\rangle$ and is connected to it via configuration interaction through $|c\rangle$. V_{ac} and V_{bc} are the configuration-interaction couplings between $|c\rangle$ and the AI states $|a\rangle$ and $|b\rangle$, respectively.

Starting from the resolvent operator equation [11] $(z-H)G(z)=1$ where $H=H_0+H_{\rm rad}+H_{\rm int}$ and where H_0 is the Hamiltonian for the free atomic or molecular system, $H_{\rm rad}$ is the Hamiltonian for the free radiation field, and $H_{\rm int}$ is the interaction between the atomic or molecular system and the radiation, one can derive a set of equations involving the matrix elements of the resolvent operators: $G_{pq}(z)=\langle p|1/(z-H)|q\rangle$, where $|p\rangle$ and $|q\rangle$ are the product states (i.e., $|g\rangle|n\rangle$, $|i\rangle|n-1\rangle$, $|a\rangle|n-2\rangle$, $|b\rangle|n-2\rangle$, and $|c\rangle|n-2\rangle$), and the choice of $|q\rangle$ will be different for different boundary conditions [9]. Hence the set of equations for the matrix element of resolvent operators (omitting the second subscript on G_{pq}) can be written as

$$(z-E_g)G_g-D_{gi}G_i=A \ ,$$

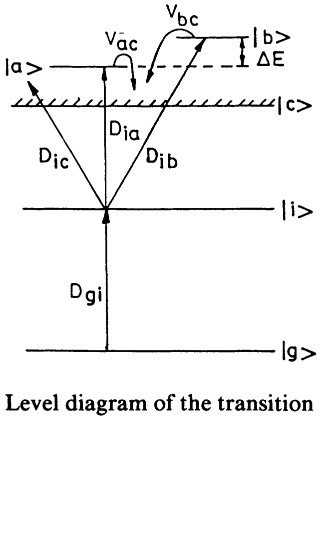

FIG. 1. Level diagram of the transition scheme.

$$(z-E_i+i\gamma_i/2)G_i-D_{ia}(1-i/q_{ia})G_a -D_{ib}(1-i/q_{ib})G_b-D_{ig}G_g=0 \ ,$$

$$(z-E_b+i\Gamma_b/2)G_b-D_{ib}(1-i/q_{ib})G_i +i(\Gamma_a\Gamma_b)^{1/2}G_a/2=0 \ ,$$

$$(z-E_a+i\Gamma_a/2)G_a-D_{ia}(1-i/q_{ia})G_i +i(\Gamma_a\Gamma_b)^{1/2}G_b/2=B \ ,$$

where D_{pq}'s are the dipole transition moments between different product states: Γ_a, Γ_b are the autoionization widths of states $|a\rangle$ and $|b\rangle$; γ_i is the photoionization width of the resonant state $|i\rangle$; and q_{ia},q_{ib} are the Fano q parameters for $|i\rangle$ to $|a\rangle$ and $|b\rangle$ transitions, respectively. After doing a little algebra one can write the above system of equations in terms of detunings δ_i, δ_a, and δ_b from states $|i\rangle$, $|a\rangle$, and $|b\rangle$, respectively, and hence in terms of the respective dimensionless detuning $\epsilon_i=\delta_i/(\gamma_i/2)$, $\epsilon_a=\delta_a/(\Gamma_a/2)$, and $\epsilon_b=\delta_b/(\Gamma_b/2)$. In this case $\epsilon_i=0$. The energy gap between AI states $|a\rangle$ and $|b\rangle$ is given as $\Delta E=\delta_a-\delta_b=n_e\Gamma_a/2$, where n_e is an integer.

The equations are solved for absorption and emission with different boundary conditions; i.e., for absorption, $A=1$, $B=0$, and for emission, $B=1$, $A=0$. From the above equations one can obtain formal expressions for $G_p(Z)$'s as $G_p(Z)=f_p(Z)/F_p(Z)$, where $F_p(Z)$ is a polynomial in Z, and in this case it is a fourth-degree polynomial. By solving exactly for the poles of $G_p(z)$, which correspond to the dressed-state energies [12], one can obtain the corresponding matrix elements of the evolution operator $U_p(t)$ (in terms of dressed-state energies) by the inverse Laplace transform of $G_p(z)$ both for absorption [13] and emission. Hence the populations in respective states are given as $|U_p(t)|^2$, and the absorption and emission probabilities at a particular time t are obtained as

$$P_{\rm abs}(t)=1-|U_g^a(t)|^2-|U_i^a(t)|^2 \ ,$$

$$P_{\rm ems}(t)=|U_g^e(t)|^2+|U_i^e(t)|^2 \ .$$

The superscripts a and e denote that the U_p's are obtained by solving for absorption and emission, respectively.

The gain in the system can be written as

$$G=R_aP_{\rm ems}(t)-R_gP_{\rm abs}(t) \ ,$$

where R_a is the pumping rate to the upper lasing level $|a,n-2\rangle$ and R_g that to the lower lasing level $|g,n\rangle$. To obtain G, a few more parameters have been introduced to denote relative transition strengths:

$$x_{ia}=D_{ia}/\Gamma_a, \quad x_{ib}=D_{ib}/\Gamma_b$$

$$y_{ia}=D_{ig}/D_{ia}, \quad y_{ib}=D_{ig}/D_{ib} \ .$$

The absorption, emission, and gain profiles have been calculated as a function of time in units of the AI lifetime of state $|a\rangle$ at a particular value of ϵ_a and also the corresponding line shapes as functions of ϵ_a at a particular value of $\Gamma_a\tau$, for different values of parameters. We have

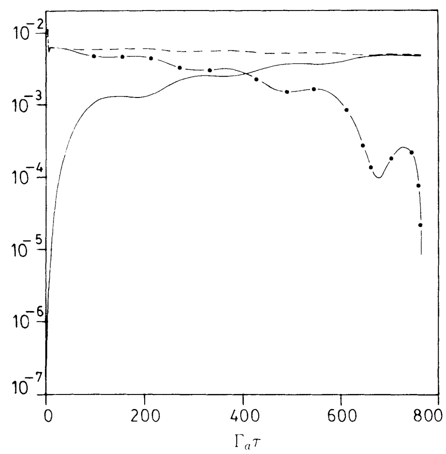

FIG. 2. Time ($\Gamma_a \tau$) dependence of total absorption (P_{abs}), emission (P_{ems}) probabilities, and total gain (G) for the following set of parameters: $\Gamma_a = 1.0$, $\Gamma_b = 0.1$, $q_{ia} = 8.0$, $q_{ib} = 8.0$, $x_{ia} = 0.4$, $y_{ia} = 0.01$, $\epsilon_a = -10.0$, $n_e = 10$, $R_a = R_g$. Full line curve denotes P_{abs}; dashed curve, P_{ems}; dot-dashed curve, gain, all in arbitrary units.

put here $R_a = \Gamma_a$. All the calculations have been done with $R_a = R_g$ unless otherwise stated.

Evolution of absorption, emission, and gain with time for the present four-level scheme is shown in Fig. 2. Comparing this result with that for the case of (1+1)-photon amplification from a single AI state (not shown here), we found that the gain persists for a much longer time on inclusion of the second AI state. We also found that the evolution of absorption in the above cases is much slower than corresponding single-photon transitions (not shown here) because of the fact that the addi-

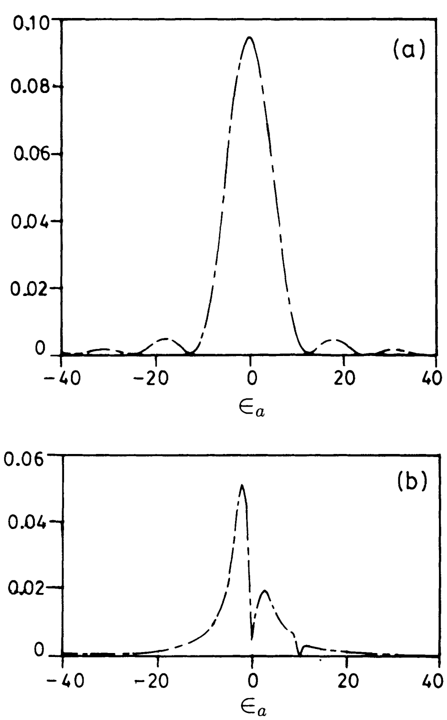

FIG. 3. Total emission and gain as a function of detuning (ϵ_a) from AI state $|a\rangle$ at different values of time: (a) for $\Gamma_a \tau = 1$, (b) for $\Gamma_a \tau = 10$. Here, absorption is negligibly small and hence the emission and gain coincide. The parameters used are $\Gamma_a = 1$, $\Gamma_b = 0.1$, $q_{ia} = 8$, $q_{ib} = 8$, $x_{ia} = 0.4$, $y_{ia} = 0.01$, $n_e = 10$.

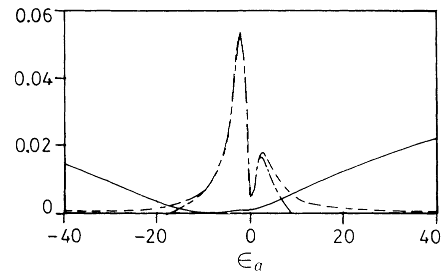

FIG. 4. Profiles for total absorption, emission, and gain in the absence of AI state $|b\rangle$ at $\Gamma_a \tau = 10$. The parameters are $\Gamma_a = 1$, $q_{ia} = 8$, $x_{ia} = 0.4$, and $y_{ia} = 0.01$. Full line curve is absorption; dashed curve is emission; dot-dashed curve is gain.

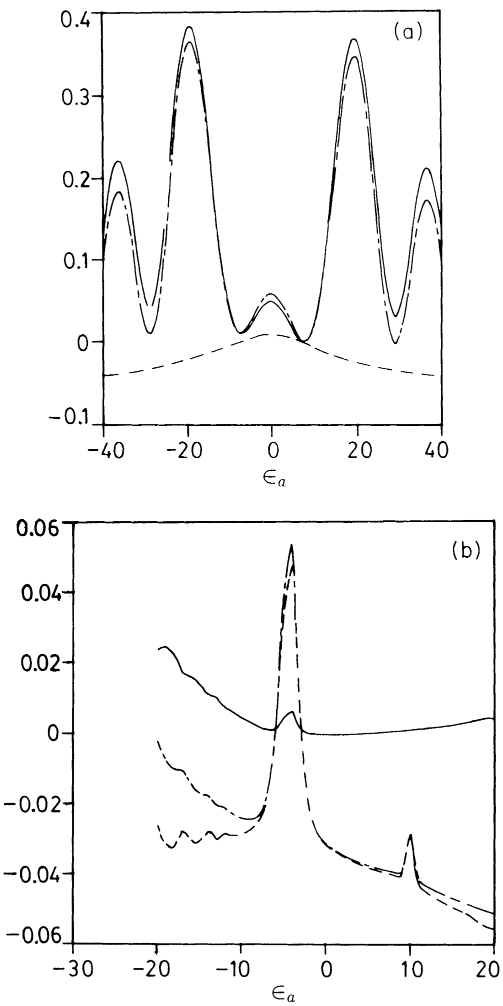

FIG. 5. Profiles for single-photon, (1+1)-photon, and total gain at different values of time: (a) $\Gamma_a \tau = 1$, (b) $\Gamma_a \tau = 20$. Other parameters are $\Gamma_a = 1.0$, $\Gamma_b = 0.1$, $q_{ia} = 96.0$, $q_{ib} = 0.1$, $x_{ia} = 6.0$, $y_{ia} = 0.04$. Dashed curve is (1+1)-photon gain; full line curve is single-photon gain; dot-dashed curve is total gain.

tion of a lower resonant step can create a bottleneck to the net absorption. This is one of the features that makes the present scheme more advantageous over the previous schemes for single-photon lasing from AI states. We have also seen that for smaller values of Γ_a and Γ_b, i.e., for weaker transition strengths (since D_{ia} and D_{ib} are proportional to Γ_a and Γ_b), the gain can be sustained for a time (not shown here) that is greater by more than two orders of magnitude than that shown in Fig. 2, although the pumping rate R_a is 100 times smaller than R_g, i.e., $R_a/R_g=0.01$.

Figure 3 shows the time development of three-peaked gain as a function of detuning of the upper probe field from state $|a\rangle$. Actually, gain at the regions on both sides of $\epsilon_a=0$ and also $\epsilon_b=0$ grows in strength with time, and the two peaks in between $\epsilon_a=0$ and $\epsilon_b=0$ overlap, leading to the second peak. The first and the third peaks arise from the generations on the left sideband of $\epsilon_a=0$ and on the right sideband of $\epsilon_b=0$, respectively. The third peak arises solely due to the presence of the second AI state, which can be demonstrated by comparing this gain profile with one in which it is absent (Fig. 4). It is to be noted (Fig. 4) that the absorption profile obtained in $(1+1)$-photon autoionization exhibits a broad minimum extending over a wide range of frequencies (unlike single-photon autoionization), and hence a two-peaked gain profile is obtained even in the absence of a second AI state. Therefore in the present scheme the gain is possible in a wide range of frequencies, and it is peaked in three spectral ranges, contrary to the previous results on single-photon lasing from AI states.

These gain profiles are further modified by the change of the Fano q parameters. In the above cases single-photon gain (i.e., the gain on the upper probe field) dominates the $(1+1)$-photon gain. But for $q_{ia} \gg q_{ib}$, the $(1+1)$-photon gain can be obtained by increasing the strength of the upper transition, i.e., for large values of x_{ia}. Figure 5 shows the time evolution of $(1+1)$-photon gain over single-photon gain as a function of ϵ_a. It also shows that a strong $(1+1)$-photon amplification is possible around the left sideband of the resonant transition $\epsilon_a=0$. Therefore, in the present scheme, gain can be obtained on both the probe fields, which is also an advantage over the previous schemes on single-photon lasing.

In conclusion, in a time-dependent dressed-state calculation we have shown that the gain for the $(1+1)$-photon emission from the AI states can be controlled by the proper choice of system parameters and probe pulse durations. Moreover, the gain on the two side peaks of the $(1+1)$-photon resonant transitions $\epsilon_a=0$ and $\epsilon_b=0$, i.e., connecting $|g,n\rangle$ to $|a,n-2\rangle$ and $|g,n\rangle$ to $|b,n-2\rangle$, respectively, evolves with time, leading to the three-peaked gain profile.

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