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Radiation amplification through autoionizing resonances without population inversion

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We present a quantitative study of radiation amplification without population inversion based on models recently proposed by Harris [Phys. Rev. Lett. **62**, 1033 (1989)]. We discuss both two-level and three-level arrangements in terms of the density matrix. Our calculations, which are fully time dependent, incorporate an incoherent pumping mechanism that is essential for amplification. We discuss in detail the range of and conditions on atomic parameters for which amplification is possible. Its feasibility is demonstrated through the direct calculation of the change of the intensity of a probe beam interacting with the atom.

I. INTRODUCTION

In a very recent paper,[1] Harris has presented a theoretical scheme for the amplification of radiation, and possibly lasing, without population inversion. His arguments, which represent an interesting extension of ideas first proposed by Arkhipkin and Heller,[2] rely on certain properties of autoionizing states (AIS). Although the analysis by Harris does not explicitly include a pumping mechanism, it implicitly assumes its existence, because his calculation of gain employs steady-state conditions. Arkhipkin and Heller, on the other hand, have included an incoherent pumping mechanism in their equations from which they have deduced some conditions for net amplification. They have, however, limited their analysis to a qualitative discussion of the two-level case only.

Our purpose in this paper is to pursue these interesting ideas to a more quantitative level by studying the process of amplification itself in a fully time-dependent context. Since the generation of coherent short-wavelength radiation underlies much of the motivation for such studies, the quantitative understanding of the amplification of a pulse is particularly relevant, given that a standard laser cavity arrangement is not possible for very short wavelengths. Our analysis is, moreover, intended to provide an avenue towards the possible experimental investigation of these ideas in connection with ongoing studies[3] of AIS through multiphoton absorption, a connection that we plan to discuss in a future paper.[4]

In order to provide an analysis which can include pumping as well as an explicit account of the degree of amplification, we formulate the problem in terms of a set of density-matrix equations. Assuming a certain rate of pumping for the AIS and for the repopulation of the ground state, as well as an external, pulsed probe beam of frequency ω, we can obtain the complete evolution of the atomic system in time, from which we can calculate the amplification or attenuation of the probe beam. If amplification is possible without population inversion, what is the minimum required population of the AIS, and how is it related to pumping and the other parameters of the atomic system? How is pumping and the resulting gain related to the autoionization width and the asymmetry of the AIS? These are some of the questions we wish to probe here.

II. MODEL AND FORMULATION

The energy-level diagram of the atomic system under consideration is shown in Fig. 1. It consists of a ground state, $|1\rangle$, and two excited states, $|2\rangle$ and $|3\rangle$, embedded in a continuum, $|c\rangle$. The excited states are coupled to the continuum by configuration interaction and therefore autoionize. This formulation[5] of autoionization is adequate for isolated autoionizing (AI) resonances (well separated from each other), which is the case throughout this work, as well as Harris's paper. The density-matrix equations that govern the time evolution of this system are

$$\dot{\rho}_{11} = -\gamma_1\rho_{11} - 2\,\mathrm{Im}\left[\tilde{\Omega}_2\left(1-\frac{i}{q_2}\right)\rho_{21}\right] - 2\,\mathrm{Im}\left[\tilde{\Omega}_3\left(1-\frac{i}{q_3}\right)\rho_{31}\right] + Q_1 P - Q_2\rho_{11}\,, \tag{1}$$

$$\dot{\rho}_{22} = -\Gamma_2\rho_{22} + 2\,\mathrm{Im}\left[\tilde{\Omega}_2\left(1+\frac{i}{q_2}\right)\rho_{21}\right] - 2\,\mathrm{Im}(\Omega_{32}\rho_{23}) + Q_2\rho_{11}\,, \tag{2}$$

$$\dot{\rho}_{33} = -\Gamma_3\rho_{33} + 2\,\mathrm{Im}\left[\tilde{\Omega}_3\left(1+\frac{i}{q_3}\right)\rho_{31}\right] - 2\,\mathrm{Im}(\Omega_{32}\rho_{32})\,, \tag{3}$$

$$\dot{\rho}_{21} = -\left[i\delta_2 + \left(\frac{\gamma_1+\Gamma_2}{2}\right)\right]\rho_{21} - i\tilde{\Omega}_2\left[\left(1-\frac{i}{q_2}\right)\rho_{11} - \left(1+\frac{i}{q_2}\right)\rho_{22}\right] - i\Omega_{32}^{*}\rho_{31} + i\tilde{\Omega}_3\left(1+\frac{i}{q_3}\right)\rho_{23}\,, \tag{4}$$

$$\dot{\rho}_{31} = -\left[i\delta_3 + \left(\frac{\gamma_1+\Gamma_3}{2}\right)\right]\rho_{31} - i\tilde{\Omega}_3\left[\left(1-\frac{i}{q_3}\right)\rho_{11} - \left(1+\frac{i}{q_3}\right)\rho_{33}\right] - i\Omega_{32}^*\rho_{21} + i\tilde{\Omega}_2\left(1+\frac{i}{q_2}\right)\rho_{32}, \tag{5}$$

$$\dot{\rho}_{32} = -\left[i(\delta_2-\delta_3) + \left(\frac{\Gamma_2+\Gamma_3}{2}\right)\right]\rho_{32} - i(\Omega_{32}^*\rho_{22} - \Omega_{32}\rho_{33}) - i\tilde{\Omega}_3\left(1-\frac{i}{q_3}\right)\rho_{12} + i\tilde{\Omega}_2\left(1+\frac{i}{q_2}\right)\rho_{31}. \tag{6}$$

In the above equations, γ_1 is the direct ionization width of the ground state, while Γ_2 and Γ_3 are the autoionization widths of states $|2\rangle$ and $|3\rangle$, respectively. The detunings are defined as $\delta_1 = \omega - \bar{\omega}_1$ and $\delta_2 = \omega - \bar{\omega}_2$, with ω being the probe frequency and $\bar{\omega}_1, \bar{\omega}_2$ the energies of the AI resonances (*not* of the unperturbed discrete states). The Rabi frequencies are complex;[6] their real parts $\tilde{\Omega}_i$ $(i=2,3)$ being defined as

$$\tilde{\Omega}_i = D_{a_i g} + \mathrm{P}\int dE_c \frac{V_{a_i c} D_{cg}}{E_g + \hbar\omega - E_c},$$

where P denotes the principal part value of the integral, V the configuration interaction, and D the electric dipole coupling. Their imaginary parts involve the q_i $(i=2,3)$ parameters of the two resonances defined in the usual way,[5,6] $q_i = 2\tilde{\Omega}_i/(\gamma_1\Gamma_i)^{1/2}$. Particularly important for the three-level system is the complex quantity Ω_{23} representing the coherent, nonradiative coupling between the two AI states, through the continuum to which they autoionize. This term, defined as

$$\Omega_{23} = \mathrm{P}\int dE_c \frac{V_{a_2 c} V_{c a_3}}{E_g + \hbar\omega - E_c} - \frac{i}{2}(\Gamma_2\Gamma_3)^{1/2},$$

arises from the fact that both levels couple to the same continuum. Finally, Q_1 and Q_2 represent the respective pumping rates, where $P = 1 - \rho_{11} - \rho_{22} - \rho_{33}$. In other words, the replenishment of the ground state takes place through a recombination process, while one (and *not* both) of the excited states is pumped incoherently with a rate Q_2. The inclusion of the incoherent pumping mechanism was one of the reasons that necessitated our use of the density-matrix equations instead of the equations for the amplitudes used by Harris.[1] Apart from the pumping terms, our equations are equivalent to his, in the sense that they can be derived from his set of equations for the amplitudes.

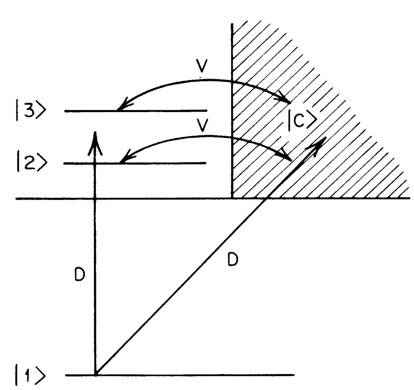

FIG. 1. Energy-level diagram for the atomic system. The pumping mechanism is not shown.

We have thus far discussed the equations for the three-level system. It is easy to obtain the corresponding equations for a two-level system (having only the atomic states $|1\rangle$ and $|2\rangle$) by eliminating Eqs. (3), (5), and (6), and removing all terms involving matrix elements of the type ρ_{3i} or ρ_{i3} $(i=1,2)$, from the remaining three equations. The pumping mechanism should be left intact.

In order to directly evaluate the process of amplification, we introduce an equation governing the rate of change of the number of photons n of the probe beam. It reads

$$\dot{n} = 2\sum_{j=2}^{3}[\mathrm{Im}(\tilde{\Omega}_j\rho_{j1})] - \gamma_1\rho_{11}. \tag{7}$$

In arriving at this expression, we started from $\dot{n} \sim -(\dot{\rho}_{22}+\dot{\rho}_{33}-\dot{\rho}_{11})$, where we included only those terms [from Eqs. (1)–(3)] that represent radiative transitions; that is, absorption and emission of photons of the probe frequency. Neither autoionization nor pumping are such processes. The terms involving the Rabi frequencies $\tilde{\Omega}_j$, have to be divided by two in order to remove the double counting of the generated photons. This is because these terms count a photon both by the decrease in the population of the excited state and the increase in the population of the ground state. The corresponding equation for a two-level system is obtained from (10) by setting $\tilde{\Omega}_3 = 0$.

To determine the amplification, or lack of it, we calculate the right-hand side of Eq. (7) as a function of time using the solution of the system of Eqs. (1)–(6). It is obvious that, if $\dot{n} > 0$ throughout the duration of the probe pulse, there will be net amplification of the incoming radiation. If, on the other hand, $\dot{n} < 0$ for the entire time of interaction, there will be attenuation. There is of course the possibility that $\dot{n}$ changes sign during the interaction with the pulse (especially around the peak intensity). In that case, one cannot tell in advance whether there will be a net increase in the energy of the pulse at the end of the interaction, or not. However, Eq. (7) can be transformed (by the proper conversion of units) into an equation for the time evolution of the intensity of the probe pulse. If this equation is coupled to the system of Eqs. (1)–(6) and all seven of them are solved self-consistently, one obtains the change of the energy of the pulse at the end of the interaction time. Although this is not a full pulse-propagation calculation, as it lacks the spatial dependence, we have used it in obtaining a value for the overall amplification efficiency of these processes; especially in those cases in which $\dot{n}$ changes sign during the pulse.

III. RESULTS ON THE TWO-LEVEL SYSTEM

We now present some of the results of our calculations, starting with the two-level system. The choice of the atomic parameters is dictated by the requirement that the AIS have an asymmetric absorption line shape exhibiting a minimum. Its q parameter should therefore be small, say $q<10$. On the other hand, one would prefer a narrow AI width, so that the system does not decay very fast, but also a strong Rabi frequency $\tilde{\Omega}_2$, so that there is substantial stimulated emission contributing to the amplification of the incoming radiation. This set of requirements is constrained by the relation[6] $q_2=2\tilde{\Omega}_2/(\gamma_1\Gamma_2)^{1/2}$ which cannot be violated. As a compromise, we have chosen $q_2=4$ and $\Gamma_2\simeq 20$ cm^{-1} (a relatively narrow AI state). An almost Gaussian pulse of peak intensity 1 W/cm^2 (weak in the sense that even at its peak the radiative couplings $\tilde{\Omega}_2$ and γ_1 were much weaker than the coupling responsible for autoionization) and of 20-ps duration is probing the system for amplification. With these conditions and for $q_2=4$, we have $\gamma_1 \ll \tilde{\Omega}_2 \ll \Gamma_2$. In our calculations, the probe frequency was tuned around the resonance to identify the range of detunings over which amplification occurs. The initial condition is that at $t=0$ all of the atomic population is in the ground state ($\rho_{11}=1$).

Without pumping, the system decays quickly and no amplification is found. Only Rabi oscillations can be observed on a time scale comparable to the autoionization time. However, introducing a pumping and recombination mechanism as in Eqs. (1)–(6), we find that only above a certain threshold value for Q_1,Q_2 is it possible to achieve amplification. Assuming for now $Q_1=Q_2$, we find that an optimum value for the strength of pumping is given by $Q_1 \geqq 0.625\Gamma_2$. This condition also implies that the steady state is achieved very quickly (on a time scale comparable to the autoionization time) and before the intensity of the probe pulse rises substantially.

For simplicity, in all of the calculations reported here, we have assumed $Q_1=Q_2$. This is not as restrictive as it may appear, because relaxation of this condition will only change the time the system needs to reach the steady state and the relative populations of the two levels, without affecting the essence of our conclusions. Closely related to the above is the observation that the AIS population (maintained against autoionization by the pumping) cannot be less than half of the ground-state population (at steady state) if amplification is to be achieved. Crucially important for the amplification is also the central frequency of the incoming radiation. Let us define the reduced (dimensionless) detuning $\epsilon_2=-2\delta_2/\Gamma_2$. As is well known,[5,6] the AI resonance will have a minimum (in the present case, a zero) in its absorption profile for $\epsilon_2=-q_2$ (or $\delta_2=2\Gamma_2$, since here $q_2=4$). Tuning the probe frequency around the region of the minimum, we have found that amplification is possible for $-7<\epsilon_2<-1$. The position of this favorable detuning range, relative to the center of the resonance, will change if the sign of q is reversed. The width of this range depends on the pumping. This dependence is illustrated in Table I.

TABLE I. Two-level system. Probe frequency ranges favorable for amplification vs pumping. The parameters for all examples shown are $q_2=4$; $\Gamma_2=20$ cm^{-1}; $\gamma_1=10^{-10}\Gamma_2$; $\tilde{\Omega}_2=3\times10^{-5}\Gamma_2$.

$Q_1=Q_2$	δ_2	Amplification
$0.76\Gamma_2$	$[0.5\Gamma_2, 3.3\Gamma_2]$	Yes
$0.63\Gamma_2$	$[0.75\Gamma_2, 2.7\Gamma_2]$	Yes
$0.51\Gamma_2$	$\cdots$	No

IV. RESULTS ON THE THREE-LEVEL SYSTEM

We proceed now to the discussion of the three-level system. Many of the conclusions reached for the two-level system are true for the three-level as well, but there are also some differences that make the three-level system more favorable for achieving amplification. Consider first two AIS having exactly the same parameters as the one employed in the two-level system above. The distance between the two resonances is taken as $\Delta E_{32}=5\Gamma_2$ (or 100 cm^{-1}). Tuning the probe frequency around the minimum of the Fano profile of the pumped resonance, we obtain a positive gain coefficient even for pumping rates as low as an order of magnitude smaller than the autoionization rate. This is to be contrasted to the results obtained for

TABLE II. Three-level system. Probe frequency ranges favorable for amplification vs pumping. $\Gamma_2=20$ cm^{-1}; $\Delta E_{32}=100$ cm^{-1}; $\mathrm{Im}(\Omega_{32})=(\Gamma_1\Gamma_2)^{1/2}$ for all examples shown.

q_2	q_3	Γ_3 (cm^{-1})	γ_1	$\tilde{\Omega}_2$	$\tilde{\Omega}_3$	$Q_1=Q_2$	δ_2	Amplification
4	40	20	$2\times10^{-6}\Gamma_2$	$3\times10^{-3}\Gamma_2$	$3\times10^{-2}\Gamma_2$	$0.76\Gamma_2$	$[-0.85\Gamma_2, 0.05\Gamma_2]$	Yes
4	40	20	$10^{-10}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$3\times10^{-4}\Gamma_2$	$0.51\Gamma_2$	$[-0.75\Gamma_2, -0.15\Gamma_2]$	Yes
4	40	20	$10^{-10}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$3\times10^{-4}\Gamma_2$	$0.38\Gamma_2$	$[-0.7\Gamma_2, -0.2\Gamma_2]$	Yes
4	40	20	$10^{-10}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$3\times10^{-4}\Gamma_2$	$0.25\Gamma_2$	$[-0.65\Gamma_2, -0.25\Gamma_2]$	Yes
4	40	20	$10^{-10}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$3\times10^{-4}\Gamma_2$	$0.13\Gamma_2$	$[-0.5\Gamma_2, -0.4\Gamma_2]$	Yes
4	40	20	$10^{-10}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$3\times10^{-4}\Gamma_2$	$\leq 0.1\Gamma_2$	$\cdots$	No
4	40	300	$10^{-10}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$10^{-3}\Gamma_2$	$0.05\Gamma_2$	$[-0.2\Gamma_2, 0]$	Yes
4	10	300	$10^{-10}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$3\times10^{-4}\Gamma_2$	$0.25\Gamma_2$	$[-0.25\Gamma_2, 0]$	Yes
4	10	300	$10^{-10}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$3\times10^{-4}\Gamma_2$	$0.13\Gamma_2$	$[-0.2\Gamma_2, -0.05\Gamma_2]$	Yes
4	10	300	$10^{-10}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$3\times10^{-4}\Gamma_2$	$0.08\Gamma_2$	$[0.2\Gamma_2, -0.1\Gamma_2]$	Yes
40	4	20	$10^{-10}\Gamma_2$	$3\times10^{-4}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$0.25\Gamma_2$	$[-4.7\Gamma_2, -4.35\Gamma_2]$	Yes
<20	4	20	$10^{-10}\Gamma_2$	$<1.4\times10^{-4}\Gamma_2$	$3\times10^{-5}\Gamma_2$	$\leq 0.5\Gamma_2$	$\cdots$	No

the two-level system, where the necessary minimum pumping rates are considerably higher ($\sim 0.7\Gamma_2$). If, on the other hand, we tune around the resonance that is not pumped, we obtain no gain.

Consider now the case in which $|3\rangle$ (the AIS not pumped) has a large q_3 (>10), a large AI width ($\Gamma_3 \geqq 10\Gamma_2$), and a large Rabi frequency ($\tilde{\Omega}_3 \simeq 10\tilde{\Omega}_2$), while $|2\rangle$ is as described before. Tuning the probe frequency around the assymetric resonance, gain is obtained for a range of detunings lying mostly on the low-energy side of the resonance (see examples in Table II). The width of this range depends on q_3 (the rest being the same) and it increases with increasing q_3. It also depends on the pumping. The stronger the pumping, the broader the frequency range favorable for amplification; however, pumping and q_3 are not independent. The larger q_3 is, the lower the threshold pumping strength one needs to achieve amplification. A summary of typical findings for the three-level system is given in Table II.

The favorable behavior of the three-level system can be attributed to the fact that the coupling represented by Ω_{32} establishes a coherence between the two states $|2\rangle$ and $|3\rangle$, which is sustained because of the pumping and, of course, does not depend on the probe pulse intensity. The imaginary part of this coupling is really instrumental and indispensable to the results presented above. It is, however, rather doubtful that its presence is realistic under most circumstances of two real autoionizing states.

V. CONCLUDING REMARKS AND OUTLOOK

(i) Owing to the pulsed nature of the problem we have considered, and the dependence of amplification on Q_1, Q_2, and δ_2, the resulting gain will vary with the above parameters. It may also vary during the pulse. We can, however, present here some average typical values. We refer to a 50-ps probe and assume $q_2=4$, $q_3=40$, and $\Gamma_2=\Gamma_3=20\ \mathrm{cm}^{-1}$. With pumping rates slower than Γ_2,Γ_3 by about a factor of 2, and an atomic density $10^{17}\ \mathrm{cm}^{-3}$, we obtain net amplification by about a factor of 10 for the two-level system (at $\delta_2=\Gamma_2$) and by a factor of 80 for the three-level system (at $\delta_2=-0.4\Gamma_2$). These numbers are equivalent to gains of 6 and 48 cm^{-1}, respectively.

(ii) Limitations of space have not allowed us to discuss the applicability of rate equations to this problem. Deferring that discussion to a future paper,[4] we will now simply point out that steady state does not imply the automatic validity of rate equations. Additional conditions are required for that to be the case. It turns out that the presence of the continuum and the resulting interferences make this problem significantly different from the standard laser amplifier.

(iii) The results of our analysis demonstrate that relatively short pulses can undergo substantial amplification under rather modest pumping requirements. Since the pumping we have included in our equations is a fairly general incoherent excitation of the resonance, it appears that any form of excitation would provide the conditions for amplification. Spontaneous start-up is a bit more difficult to contemplate at this point because spontaneous photon emission (especially at the minimum) is practically negligible for autoionizing resonances unless one is dealing with highly ionized species. Finally, the description of resonances as discrete states embedded in the same single continuum (inherent in this paper as well as in Refs. 1 and 2) represents a nontrivial limitation whose implications will be discussed in a future paper.[4]

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