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Lasers without inversion: Single-atom transient response

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We discuss the effect of the transient response on the dynamics of lifetime broadened lasers that operate without the need for population inversion. A relationship between the steady-state absorptive transition probability rate and the transient gain and loss is given.

It has recently been shown that a population inversion is not a prerequisite for obtaining laser amplification and oscillation.[1-3] The essential idea is to utilize a system which causes a destructive interference in the absorption profile of lower-level atoms but not in the emission profile of upper-level atoms. An example of such a system is shown in the inset of Fig. 1(b). Level $|1\rangle$ is the lower laser level. Levels $|2\rangle$ and $|3\rangle$ are upper levels which are lifetime broadened with decay rates Γ_2 and Γ_3. This decay may result from autoionization, photoionization, tunneling, or spontaneous emission to a fourth level; but for the ideal case which we discuss here, the decay of both upper levels must occur to a common final continuum. (For example, for autoionization, this continuum is an ion and a free electron of prescribed angular momentum and arbitrary energy.)

In earlier work[1-3] the response of the lower-level atoms has been studied in the steady state, and the initial transient absorption caused by the excitation of the atoms at $t=0$ was ignored. Since this absorption effectively ends within several decay times of the upper levels, and since atoms may remain in level $|1\rangle$ indefinitely, the time-integrated contribution to the total absorption of this transient is vanishingly small, and is neglected when computing the steady-state (Fano-type) interference profiles of level $|1\rangle$ atoms. However, when operating at the zero of the interference profile, this transient absorption is the only absorption and should not be neglected. In fact, we show here, that this absorption determines what is, in effect, a threshold condition for lasers of this type.

When an atom is excited to level $|2\rangle$ or to level $|3\rangle$ (excitation occurs from some level or levels which are not shown), the stimulated response terminates within several decay times, and is, therefore, itself a transient response. For an ensemble of atoms, with rates into both upper and lower levels, the overall gain-loss balance is determined by the (single-atom) transient emission and the combined steady-state–transient absorption. It is often the case, for example in a cw discharge, that an ensemble of atoms is in steady state, while each individual atom has both a transient and a steady-state component. In this Rapid Communication we focus on the individual atom.

We use the equations of Ref. 1. These describe the threefold interference of level-$|1\rangle$ atoms to two upper levels and to the common continuum to which the upper levels decay. Results for a single level and a continuum, or two levels without a direct channel to the continuum, are obtained as special cases. The time-varying amplitudes of a lower level $|1\rangle$ and upper levels $|2\rangle$ and $|3\rangle$ are given by

$$\frac{\partial a_1}{\partial t}+j\Delta\tilde{\omega}_{11}a_1=\kappa_{12}a_2+\kappa_{13}a_3\,, \tag{1a}$$

$$\frac{\partial a_2}{\partial t}+j\Delta\tilde{\omega}_{21}a_2=\kappa_{12}a_1+\kappa_{23}a_3\,, \tag{1b}$$

$$\frac{\partial a_3}{\partial t}+j\Delta\tilde{\omega}_{31}a_3=\kappa_{13}a_1+\kappa_{23}a_2\,. \tag{1c}$$

The quantities in these equations are

$$\begin{aligned}
&\Delta\tilde{\omega}_{11}=-j\frac{W_C}{2}, \quad \kappa_{12}=\tfrac{1}{2}[j\Omega_{12}+(\Gamma_2 W_c)^{1/2}],\\
&\Delta\tilde{\omega}_{21}=\Delta\omega_{21}-j\frac{\Gamma_2}{2}, \quad \kappa_{13}=\tfrac{1}{2}[j\Omega_{13}+(\Gamma_3 W_c)^{1/2}]\,,\\
&\Delta\tilde{\omega}_{31}=\Delta\omega_{31}-j\frac{\Gamma_3}{2}, \quad \kappa_{23}=-\tfrac{1}{2}(\Gamma_2\Gamma_3)^{1/2},
\end{aligned} \tag{2}$$

where Γ_2 and Γ_3 are the decay rates of levels $|2\rangle$ and $|3\rangle$; $\Delta\omega_{21}=\omega_2-(\omega_1+\omega)$, $\Delta\omega_{31}=\omega_3-(\omega_1+\omega)$, and ω is the angular frequency of the electromagnetic field; Ω_{12} and Ω_{13} are the respective Rabi frequencies ($\mu E/\hbar$), and W_c is the (direct channel) photoionization rate of level $|1\rangle$ to the continuum. We assume that the basis set has been prediagonalized[1] so that Γ_2, Γ_3, and W_c are real. We note the importance of the cross term κ_{23}, which represents the fact that as level $|2\rangle$ decays it drives level $|3\rangle$ and vice versa. This term arises since both levels couple to the same continuum level and therefore to each other.

We begin by examining computer solutions of these equations. The parameters for the computer runs (Fig. 1) are chosen so as to attain a zero in the steady-state absorption. The parameters for the three-level system are $\Omega_{12}=1/\sqrt{10}$, $\Omega_{13}=1$, $\Delta\omega_{21}=-5$, $\Delta\omega_{31}=50$, $\Gamma_2=1$, $\Gamma_3=10$, and $W_c=0$. For comparison, we also show a two-level system with the same parameters but without the interfering level $|3\rangle$.

Figure 1(a) shows the probability for level $|1\rangle$ occupancy, $|a_1(t)|^2$, versus time for the two-level system with the boundary condition $a_1(0)=1$, $a_2(0)=a_3(0)=0$. The absorption consists of the sum of a transient term and of a (golden-rule) steady-state term. Figure 1(b) shows this same quantity for the ideal three-level system. Here the steady-state term is zero (zero slope) and the absorption consists of only the transient term.

Figures 1(c) and 1(d) show the emission process. Here the boundary condition at $t=0$ is $a_2(0)=1$, $a_1(0)$

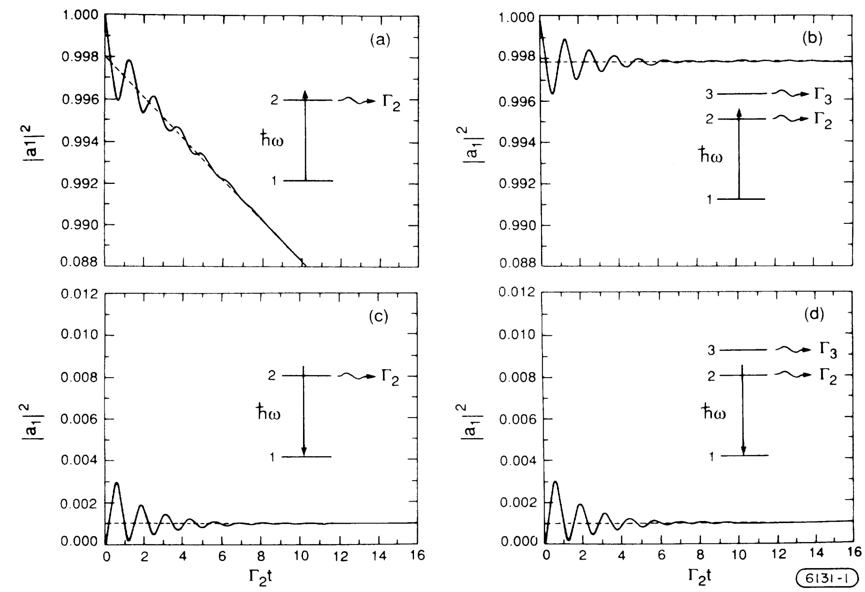

FIG. 1. Single-atom absorption and emission for two and three-level systems. The parameters for the three-level system are chosen so as to attain a perfect Fano-type cancellation. The two-level system has the same parameters, but without the interfering level $|3\rangle$. The solid lines are the computer solutions of Eq. (1) with the boundary condition: (a) and (b), $a_1 = 1$, $a_2 = a_3 = 0$; (c) and (d), $a_2 = 1$, $a_1 = a_3 = 0$. The dashed lines are the analytical solutions given in the text.

$= a_3(0) = 0$. We see that the response of the two- and three-level systems are quite similar. An atom which is initially in level $|2\rangle$ decays primarily via its lifetime broadening, and during this process has a probability of about 10^{-3} of being stimulated to level $|1\rangle$. Though it is not apparent in these figures, there is an important difference between the emissive processes in the two- and the three-level systems: At large time the lower-level population $[|a_1(t)|^2]$ of the two-level atom exhibits a steady-state decay, while the three-level atom is locked into the lower level.

We now return to the analytical treatment. We make the assumption that the applied electromagnetic field is infinitesimally small, or equivalently that the Rabi frequencies are much less than the smaller of the decay times or detunings. With this assumption, one may show (exactly) that the probability P_{ab} of an atom which at $t=0$ is in level $|1\rangle$, having been excited from this level at time t, *as t becomes very large*, is

$$P_{ab}(t) = 1 - |a_1(t)|^2 = \Delta + W_{ab} t \,, \tag{3a}$$

where Δ and W_{ab} are

$$\Delta = 2\,\mathrm{Re}\left\{ \frac{[\kappa_{23}^2(\kappa_{12}^2+\kappa_{13}^2) - \kappa_{12}^2\Delta\tilde{\omega}_{31}^2 - \kappa_{13}^2\Delta\tilde{\omega}_{21}^2] + 2j\kappa_{12}\kappa_{13}\kappa_{23}(\Delta\tilde{\omega}_{21}+\Delta\tilde{\omega}_{31})}{(\Delta\tilde{\omega}_{21}\Delta\tilde{\omega}_{31}+\kappa_{23}^2)^2} \right\} \tag{3b}$$

$$W_{ab} = 2\,\mathrm{Re}\left\{ j\Delta\tilde{\omega}_{11} + \frac{2\kappa_{12}\kappa_{13}\kappa_{23} + j(\kappa_{12}^2\Delta\tilde{\omega}_{31} + \kappa_{13}^2\Delta\tilde{\omega}_{21})}{\Delta\tilde{\omega}_{21}\Delta\tilde{\omega}_{31}+\kappa_{23}^2} \right\} . \tag{3c}$$

The quantity W_{ab} is the steady-state transition probability rate of atoms to the continuum and is the slope, at large t, of Figs. 1(a) and 1(b). The electrons that are produced as a result of this steady-state absorption have the same energy as the photon which is absorbed and are produced instantaneously, in the same sense as are Rayleigh scattered photons. The quantity Δ is a result of the transient. In general, Δ has a component which is inelastic and leads to real population, followed by subsequent decay, of levels $|2\rangle$ and $|3\rangle$. Though in these figures Δ is positive, it may have arbitrary sign.

Next consider the emission of an atom which at $t=0$ is in level $|2\rangle$. Subject to the small Rabi frequency assumption, the probability G_2 of an atom having made a transition to level $|1\rangle$ at time t, *where t is very long* as compared to the transient response time, is

$$G_2 = \left| \frac{\kappa_{13}\kappa_{23} + j\kappa_{12}\Delta\tilde{\omega}_{31}}{\Delta\tilde{\omega}_{21}\Delta\tilde{\omega}_{31}+\kappa_{23}^2} \right|^2 . \tag{4a}$$

Similarly, the probability G_3 of an atom which at $t=0$ is in level $|3\rangle$ having made a transition to level $|1\rangle$ *at large t* is

$$G_3 = \left| \frac{\kappa_{12}\kappa_{23} + j\kappa_{13}\Delta\tilde{\omega}_{21}}{\Delta\tilde{\omega}_{21}\Delta\tilde{\omega}_{31}+\kappa_{23}^2} \right|^2 . \tag{4b}$$

These expressions for the absorption and emission probabilities are compared to the full numerical solutions in Fig. 1. The dashed lines in Figs. 1(a) and 1(b) show the quantity $1-P_{ab}(t)$ which equals $|a_1(t)|^2$ from Eq. (3a). The transient loss Δ is the intercept and W_{ab} the slope of the straight line. The dashed lines in Figs. 1(c) and 1(d) show the quantity G_2 from Eq. (4a). For large t, the numerical and analytical solutions are in exact agreement.

We now come to a principal result of this Rapid Communication. It may be shown that the quantities in the preceding equations are related by

$$2(G_2+G_3)=\Delta+W_{ab}\tau\,, \tag{5a}$$

where

$$\tau=\frac{4\Gamma_2\Delta\omega_{31}^2+4\Gamma_3\Delta\omega_{21}^2}{(\Gamma_3\Delta\omega_{21}+\Gamma_2\Delta\omega_{31})^2+4\Delta\omega_{21}^2\Delta\omega_{31}^2}\,. \tag{5b}$$

Thus, as expected, there is a relationship between the parameters which govern stimulated emission and absorption. The relationship does not impose the requirement of inversion to achieve net gain, but does imply a combined inversion–pumping-rate limitation.

First, consider its application to the two-level system of Fig. 1(a). Here, we find $W_{ab}=9.90\times10^{-4}$ sec^{-1}, $\Delta=1.94\times10^{-3}$, $G_2=9.90\times10^{-4}$, and $G_3=0$. The steady-state absorption therefore dominates over the transient absorption after about two autoionizing times of level $|2\rangle$, thereby accounting for its usual neglect.

Now consider the three-level system. In general, for an ensemble of atoms with rates R_i(atoms/cm^{3}sec) into the respective levels, we require for net positive gain

$$R_2G_2+R_3G_3\geq W_{ab}N_1+R_1\Delta\,, \tag{6}$$

where N_1 is the population of level $|1\rangle$. For the case of Figs. 1(b) and 1(d), $W_{ab}=0$, $G_2=1.00\times10^{-3}$, $G_3=1.00\times10^{-4}$, and $\Delta=2.20\times10^{-3}$. Noting $G_3\ll G_2$, from Eqs. (5) and (6) we have the requirement that $R_2G_2>R_1\Delta$, or approximately $R_2>2R_1$. We note though, that there are situations where W_{ab} is finite, $\Delta=0$, and with $N_1>N_2$ there is net gain with no requirement on R_1.

The threshold condition [Eq. (6)] can be rewritten as

$$\frac{R_2}{\Gamma^{(2)}}W_{e2}+\frac{R_3}{\Gamma^{(3)}}W_{e3}\geq W_{ab}N_1+R_1\Delta\,, \tag{7a}$$

where

$$\frac{\Gamma^{(2)}}{\Gamma_2}=\frac{\Gamma^{(3)}}{\Gamma_3}=\frac{(2\Delta E)^2}{(\Gamma_2+\Gamma_3)^2+(2\Delta E)^2}\,, \tag{7b}$$

and ΔE is the separation of levels $|2\rangle$ and $|3\rangle$. The effective stimulated emission probability rates W_{e2} and W_{e3} and the (coupled) decay rates $\Gamma^{(2)}$ and $\Gamma^{(3)}$ of excited level $|2\rangle$ and level $|3\rangle$ atoms are as given in Ref. 1. For $\Delta E\gg\Gamma_2,\Gamma_3$, $R_2/\Gamma^{(2)}=N_2$, and $R_3/\Gamma^{(3)}=N_3$, where N_2 and N_3 are the upper-level steady-state populations.

In this work we have not allowed for noncancellable decay channels from levels $|2\rangle$ and $|3\rangle$. Such channels, and also dephasing collisions, will cause a nonzero steady-state absorption. These topics, as well as the large signal behavior of this type of system are better handled by a density matrix method.[6]

In summary, we have shown a relationship between the quantities which govern stimulated emission and absorption. This relationship often implies that in order to achieve lasing without inversion, the rate into the lower level must be less than that into the upper level. The effect of the Fano cancellation is to render atoms transparent, but only after the termination of a transient response.

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