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Lasers without inversion: Raman transitions using autoionizing resonances

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We consider laser action on a transition involving an autoionizing state $|a\rangle$ and an initially unpopulated state $|f\rangle$, the population of which builds up because of spontaneous emission and Raman processes. The autoionizing state is pumped by a coherent field. We discuss the transient amplification of the probe field on the lasing transition for a range of parameters. The coherent field on the pumping transition leads to minima in the absorption spectra if the atomic population were in the state $|f\rangle$. Such regions of minima can be used for obtaining gain even for long times.

I. INTRODUCTION

Recently Harris[1] has discussed how interference effects can be utilized for obtaining laser action without population inversion.[2-4] He considered two excited states (of the autoionizing type) which were decaying to the same continuum. Other systems have been shown to have similar properties.[3-5] He argued that in the range of parameters, where a Fano minimum occurs, one can have gain without population inversion. Harris introduced a scheme for pumping the population into the exciting state. The pumping in his model was determined intrinsically rather than by external parameters. The pumping mechanism, of course, is very critical in determining laser action.[6] In view of this we consider a realistic model in which we pump population from a third state $|i\rangle$ to the autoionizing state $|a\rangle$. The population then decays either by autoionizing or by spontaneous emission to the level $|f\rangle$. We investigate the amplification of an optical wave acting on the transition $|a\rangle\leftrightarrow|f\rangle$. We discuss the transient gain. We also show regions of gain in the long-time limit if we work in a range of parameters such that the absorption starting from $|f\rangle$ is a minimum. This is because for such times much of the population will reside in $|f\rangle$ but this will not contribute to absorption due to the field-induced minimum. Thus even a small effective population in the excited state will lead to gain. The plan of the paper is as follows. In Sec. II we discuss basic equations for the model and show how the gain on the probe transition can be calculated. In Sec. III we show how the spontaneous emission on the probe transition can be included and how the resulting density matrix equation can be solved. In Sec. IV we present numerical results. We show that the probe can be amplified for a wide range of parameters.

II. MODEL FOR LASER ACTION WITHOUT POPULATION INVERSION

Consider the scheme of the energy levels shown in Fig. 1. Here $|a\rangle$ is the autoionizing state; $|i\rangle$ and $|f\rangle$ are the bound states of the system and can be reached by laser excitation. We assume that the population is initially in the state $|i\rangle$. We pump the population from the state $|i\rangle$ to $|a\rangle$ by a laser of frequency ω_1. We examine whether a probe field of frequency ω_2 applied on the transition $|a\rangle\leftrightarrow|f\rangle$ can be amplified. Let Γ be the rate of autoionization of the state $|a\rangle$. Such a model has been studied quite extensively[7,8] and photoelectron as well as spontaneous emission spectra calculated. Even the absorption spectrum has been studied. In the following we present calculations[9] for the gain, keeping the laser action in mind. The Hamiltonian for the above system can be written as[7] ($\hbar=1$)

$$\begin{aligned}H={}&E_a|a\rangle\langle a|+\int E|E)(E|\,dE+E_f|f\rangle\langle f|+E_i|i\rangle\langle i|+\int[V_{Ea}|E)\langle a|+\text{H.c.}]dE\\&+\int[\tilde{v}_{Ei}|E)\langle i|e^{-i\omega_1 t}+\text{H.c.}]dE+\int[\tilde{v}_{Ef}|E)\langle f|e^{-i\omega_2 t}+\text{H.c.}]dE\\&+(\tilde{v}_{ai}|a\rangle\langle i|e^{-i\omega_1 t}+\text{H.c.})+(\tilde{v}_{af}|a\rangle\langle f|e^{-i\omega_2 t}+\text{H.c.})\,,\end{aligned}\tag{2.1}$$

where V_{Ea} is the configuration interaction, and $\tilde{v}$ gives the interaction with the electromagnetic field. The configuration interaction can be diagonalized by introducing the Fano state $|E\rangle$ whence Eq. (2.1) reduces to

$$\begin{aligned}H={}&\int E|E\rangle\langle E|dE+\int(v_{Ei}|E\rangle\langle i|e^{-i\omega_1 t}+\text{H.c.})dE\\&+\int(v_{Ef}|E\rangle\langle f|e^{-i\omega_2 t}+\text{H.c.})dE\\&+E_f|f\rangle\langle f|+E_i|i\rangle\langle i|\,,\end{aligned}\tag{2.2}$$

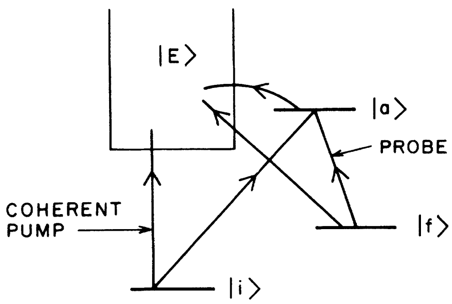

FIG. 1. Schematic diagram for the energy levels involved in the transition.

where v_{Ei} and v_{Ef} refer to the matrix elements of the field interaction between the Fano state $|E\rangle$ and the bound states[7]

$$\begin{aligned} v_{iE} &\approx \tilde{v}_{ia} B_{Ea}, \quad v_{fE} \approx \tilde{v}_{fa} C_{Ea} , \\ B_{Ea} &= b(E,a)\left[1+\frac{2(E-E_a)}{\Gamma q_i}\right] , \\ C_{Ea} &= b(E,a)\left[1+\frac{2(E-E_a)}{\Gamma q_f}\right] , \\ b(E,a) &= \langle a|E\rangle . \end{aligned} \tag{2.3}$$

The fast time dependence in Eq. (2.2) can be removed by transforming to the rotating frame so that

$$\begin{aligned} H = &\int \Delta_E |E\rangle\langle E| dE + \Delta_f |f\rangle\langle f| \\ &+ \int (v_{Ei}|E\rangle\langle i| + \text{H.c.})dE \\ &+ \int (v_{Ef}|E\rangle\langle f| + \text{H.c.})dE . \end{aligned} \tag{2.4}$$

Here we have set

$$\Delta_E=(E-\omega_1), \quad \Delta_f=E_f-(\omega_1-\omega_2), \quad E_i=0 . \tag{2.5}$$

In order to see if the probe field is amplified, we need to calculate the induced polarization $\mathbf{P}(t)$ at the frequency ω_2 which is related to the off-diagonal density matrix element by

$$\begin{aligned} \mathbf{P}(t)\cdot\epsilon_2 &= \int \mathbf{d}^*_{Ef}\cdot\epsilon_2 \rho_{Ef}(t) e^{-i\omega_2 t} dE + \text{c.c.} \\ &= -\int v_{fE}\rho_{Ef}(t) e^{-i\omega_2 t} dE + \text{c.c.} , \end{aligned} \tag{2.6}$$

and hence the *transient susceptibility* χ can be defined by

$$\begin{aligned} \chi(t,\omega_2) &= -\int dE\, v_{fE}\rho_{Ef}(t)/\epsilon_2^2 \\ &= -\int dE\, v_{fE}\psi_E(t)\psi_f^*(t)/\epsilon_2^2 . \end{aligned} \tag{2.7}$$

If $\text{Im}\chi(t,\omega_2)<0$, then there is gain on the transition $|f\rangle \leftrightarrow |a\rangle$. The field ϵ_2 is weak and hence we calculate $\chi(t,\omega_2)$ to all orders in ϵ_1 but to zero order in ϵ_2.

Using Eq. (2.4) we find that

$$\begin{aligned} \dot{\psi}_i &= -i\int dE\, v^*_{Ei}\psi_E , \\ \dot{\psi}_f &= -i\int dE\, v^*_{Ef}\psi_E - i\Delta_f\psi_f , \\ \dot{\psi}_{E_1} &= -i\Delta_{E_1}\psi_{E_1} - iv_{E_1 i}\psi_i - iv_{E_1 f}\psi_f . \end{aligned} \tag{2.8}$$

These are to be solved subject to the initial condition $\psi_i(0)=1$, $\psi_f(0)=0$. Moreover, we need to calculate ψ_f and ψ_E only to first order in v_{Ef}. From Eq. (2.8) it is seen that the Laplace transforms of the amplitudes (with variable z) are given by

$$\begin{aligned} \hat{\psi}_i &= z^{-1} - iz^{-1}\int dE\, v^*_{Ei}\hat{\psi}_E , \\ \hat{\psi}_f &= -i(z+i\Delta_f)^{-1}\int dE\, v^*_{Ef}\hat{\psi}_E , \\ \hat{\psi}_{E_1} &+ \int dE \frac{v_{E_1 i}}{z(z+i\Delta_{E_1})} v^*_{Ei}\hat{\psi}_E + \int dE \frac{v_{E_1 f}}{z+i\Delta_{E_1}} \frac{v^*_{Ef}\hat{\psi}_E}{z+i\Delta_f} \\ &= -i\frac{v_{E_1 i}}{z(z+i\Delta_{E_1})} . \end{aligned} \tag{2.9}$$

From Eq. (2.9) we see that $\hat{\psi}_E$ is at least of order ϵ_2^2 and thus for calculating χ, we solve for ψ_E by ignoring v_{Ef}. Let us define Φ_f by

$$\hat{\Phi}_f = \int dE\, v^*_{Ef}\hat{\psi}_E , \tag{2.10}$$

then Eq. (2.7) shows that

$$\chi(t,\omega_2) = -\Phi_f(t)\psi_f^*(t)/\epsilon_2^2 , \tag{2.11}$$

and Eq. (2.9) leads to

$$\begin{aligned} \hat{\psi}_f &= -i(z+i\Delta_f)\hat{\Phi}_f , \\ \hat{\Phi}_f &= -im_{21}(1+m_{11})^{-1} . \end{aligned} \tag{2.12}$$

In Eq. (2.12) the parameters m are defined by[7]

$$m_{11} = \int \frac{|v_{Ei}|^2}{z(z+i\Delta_E)} dE, \quad m_{21} = \int \frac{v_{Ei}v^*_{Ef}}{z(z+i\Delta_E)} dE . \tag{2.13}$$

A simple calculation shows that for a flat continuum

$$\begin{aligned} m_{11} &= \frac{2}{\Gamma z}|\mathbf{d}_{ia}\cdot\epsilon_1|^2 \left[\frac{(1-i/q_i)^2}{2z/\Gamma+1-i\alpha} + \frac{1}{q_i^2}\right] , \\ m_{21} &= (\mathbf{d}_{ia}\cdot\epsilon_1)(\mathbf{d}_{fa}\cdot\epsilon_2)\frac{2}{\Gamma z}\left[\frac{(1-i/q_i)(1-i/q_f)}{2z/\Gamma+1-i\alpha} + \frac{1}{q_i q_f}\right] , \end{aligned} \tag{2.14}$$

$$\alpha = \frac{2}{\Gamma}(\omega_1 - E_a) .$$

The explicit form of χ can be obtained from Eqs. (2.11)–(2.14).

A relatively simple result can be obtained if we assume that the pumping from the level $|i\rangle$ is weak or if we assume that α is large. It is then sufficient to do a calculation to second order in the field ϵ_1 and thus $\hat{\Phi}_f$ can be approximated by $-im_{21}$, i.e.,

$$\Phi_f(t)=\frac{-2i(\mathbf{d}_{ia}\cdot\epsilon_1)(\mathbf{d}_{fa}\cdot\epsilon_2)}{\Gamma}\left[\left[\frac{(1-i/q_i)(1-i/q_f)}{(1-i\alpha)}+\frac{1}{q_iq_f}\right]+\frac{(1-i/q_i)(1-i/q_f)}{-(\Gamma/2)(1-i\alpha)}e^{-(\Gamma/2)(1-i\alpha)t}\right] \quad (2.15)$$

$$\to \text{const}\times\phi_0 \text{ as } t\to\infty \ . \quad (2.16)$$

On using Eq. (2.16) in Eq. (2.11), we find that

$$\lim_{t\to\infty}\text{Im}\chi(t,\omega_2)=-\frac{\epsilon}{\epsilon^2+\Delta_f^2}|\phi_0|^2 \ , \quad (2.17)$$

where ($\epsilon>0$) is an infinitesimal parameter and is introduced phenomenologically to account for any width of the final state. Equation (2.17) is like in any Raman system where the field on the Stokes transition gets amplified. It should however be borne in mind that the limit $t\to\infty$ and the weak pump field limits are to be taken with great care and these two are not interchangeable. It is thus best to examine the gain in the transient domain.

In general the time dependence of the function Φ_f is determined by the roots of $1+m_{11}=0$. These roots are given by

$$z_\pm=-\frac{i\Gamma}{2}(\epsilon_\pm-\alpha) \ ,$$
$$\epsilon^2-[\alpha-i(1+\Omega_i)]\epsilon-\Omega_iq_i^2-i(\alpha-2\Omega_iq_i)=0 \ , \quad (2.18)$$

where

$$\Omega_i=\frac{2\pi}{\Gamma}|\tilde{v}_{Ei}|^2 \ .$$

The roots of Eq. (2.18) are in general complex except at the confluence $\Omega_i=1+\alpha/q_i$ when one of the roots is real, $\epsilon=-q_i$. Thus $\Phi(t)\to 0$ as $t\to\infty$ when $\Omega_i\neq 1+\alpha/q_i$ and when all orders in Ω_i are included. This implies that $\chi\to 0$ as $t\to\infty$ if it is calculated to all orders in the pump field. The situation is slightly different if spontaneous emission is included, as nonzero spontaneous emission leads to buildup of the population of the level $|f\rangle$ which can yield finite absorption.

III. INCLUSION OF SPONTANEOUS EMISSION

Since the transition $|a\rangle\to|f\rangle$ is the lasing transition, it is important to include spontaneous emission in this transition. The effects of spontaneous emission on the pumping transient are generally not important. Let γ_f be the radiative decay coefficient

$$\gamma_f=\tfrac{4}{3}d_{af}^2\frac{\omega_{af}^2}{c^3} \ ; \quad (3.1)$$

then the density matrix for the atomic system can be shown to be given by

$$\frac{\partial\rho}{\partial t}=-i[H,\rho]-\tfrac{1}{2}\gamma_f(A_f^\dagger A_f\rho-2A_f\rho A_f^\dagger+\rho A_f^\dagger A_f) \ , \quad (3.2)$$

where H is given by Eq. (2.4) and A_f is the dipole moment operator connecting the Fano state $|E\rangle$ with $|f\rangle$, i.e.,

$$A_f=\int dE|f\rangle\langle E|C_{Ea} \ . \quad (3.3)$$

The solution of Eq. (3.2) can be constructed from the solution of Eq. (2.8) by modifying the $\dot{\psi}_{E_1}$ equation to include a term $-\gamma_f/2\int dE\, C^*_{E_1a}C_{Ea}\psi_E$.

Let σ be the matrix

$$\sigma_{\alpha\beta}(t)=\psi_\alpha(t)\psi_\beta^*(t) \ . \quad (3.4)$$

Let $\sigma^{(i)}$ (and $\sigma^{(f)}$) be the matrix constructed from Eq. (2.8) under the initial condition $\psi_i\neq 0$ ($\psi_f\neq 0$). Then the solution of Eq. (3.2) can be shown to be given by

$$\hat{\rho}(z)=\hat{\sigma}^{(i)}+\hat{\sigma}^{(f)}\hat{T}^{if}(1-\hat{T}^{ff})^{-1} \ , \quad (3.5)$$

where

$$\hat{T}^{if}=\frac{\gamma_f}{2}\int dE_1\int dE\, C^*_{E_1a}C_{Ea}\hat{\sigma}^{(i)}_{EE_1}+\text{c.c.} \ , \quad (3.6)$$

$$\hat{T}^{ff}=\frac{\gamma_f}{2}\int dE_1\int dE\, C^*_{E_1a}C_{Ea}\hat{\sigma}^{(f)}_{EE_1}+\text{c.c.} \quad (3.7)$$

The induced polarization and hence the gain on the transition $|a\rangle\leftrightarrow|f\rangle$ can be calculated using the solution (3.5). The expressions are rather long[9] and we do not present them here. In Sec. IV we discuss the numerical results.

IV. NUMERICAL RESULTS FOR THE AMPLIFICATION OF THE PROBE FIELD

We have calculated numerically the gain of the probe field for a range of parameters. All gain calculations assume that initially the atom is in the state $|i\rangle$. We show some illustrative results in various figures. The quantity that is plotted is

$$\frac{-q_f^2\Gamma}{2|\langle a|\mathbf{d}|f\rangle|^2}\text{Im}\chi(t,\omega_2) \ .$$

For Fig. 2 we choose ω_1 and ω_2 such that exact Raman resonance occurs, $\Delta_f=E_f-(\omega_1-\omega_2)=0$. For small values of the pump field we find that the probe field is amplified over a very wide range of time, typically running into many autoionization periods. For $\Delta_f\neq 0$ (Fig. 3), a similar behavior occurs. All the gain curves show oscillatory behavior. This is because the atom undergoes a coherent Rabi oscillation between the states $|a\rangle$ and $|i\rangle$, since the pump between the states $|a\rangle$ and $|i\rangle$ is a coherent pump.

We next investigate an altogether different region of frequency. Note that because of spontaneous emission

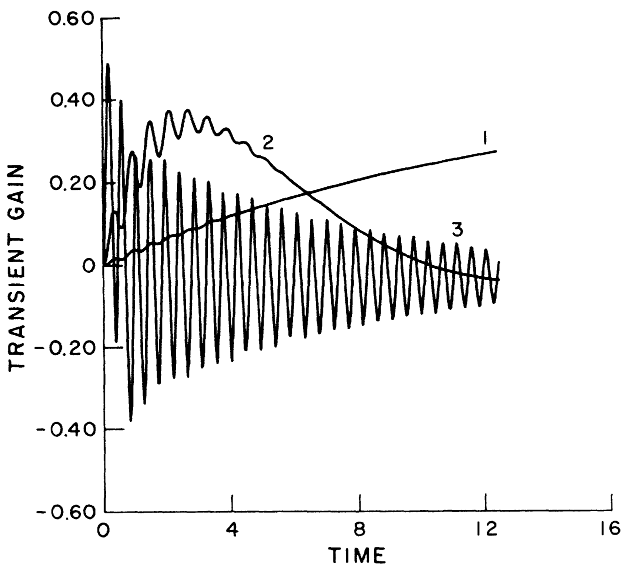

FIG. 2. Transient gain $[-\mathrm{Im}\chi(t,\omega_2)]$ as a function of time $(\Gamma t/2)$ for $\alpha=10$, $\gamma_f/\Gamma=0.1$, $q_i=5$, $q_f=2$, $\Delta_f=2/\Gamma(E_f-\omega_1+\omega_2)=0$, and for the three pump field values [defined by Eq. (2.19)] (1) $\Omega_i=0.01$, (2) $\Omega_i=0.1$, and (3) $\Omega_i=2$. The actual gain is obtained by multiplying the plotted quantities by the factor $\Gamma\Omega_f/2\epsilon_2^2$ [cf. numerical coefficient in Eq. (4.1)].

the population builds up in the state $|f\rangle$. If a large enough population builds up in the state $|f\rangle$, then clearly the probe field will not experience amplification unless for some reason, the absorption is minimal in certain frequency ranges. In an earlier paper[8] the *absorption* of the probe was investigated *assuming the initial population*

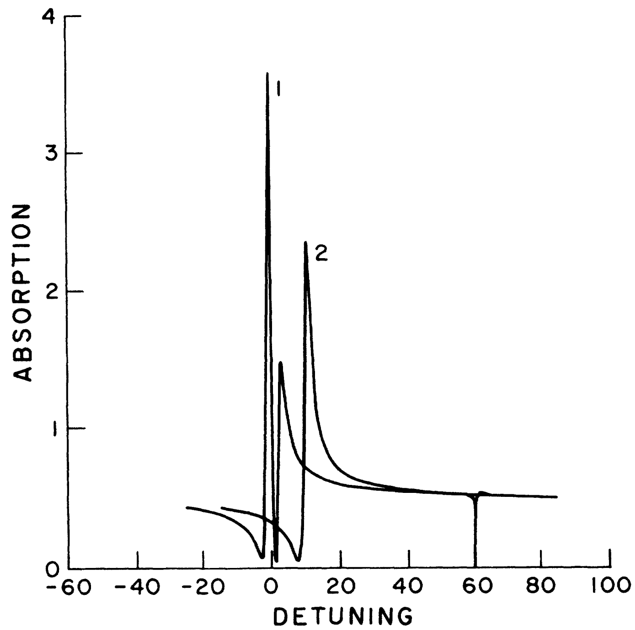

FIG. 4. Steady-state absorption profiles $[\mathrm{Im}\chi(\omega_2)]$ as a function of detuning $\delta=2/\Gamma(E_f+\omega_2-E_a)$ for $q_i=5$, $q_f=2$, $\Omega_i=0.1$, $\gamma_f/\Gamma=0.1$, and for (1) $\alpha=1$ and (2) $\alpha=50$. For clarity the curve for $\alpha=50$ has been shifted to the right by 10 units. This calculation assumes that the atom is initially in the state $|f\rangle$.

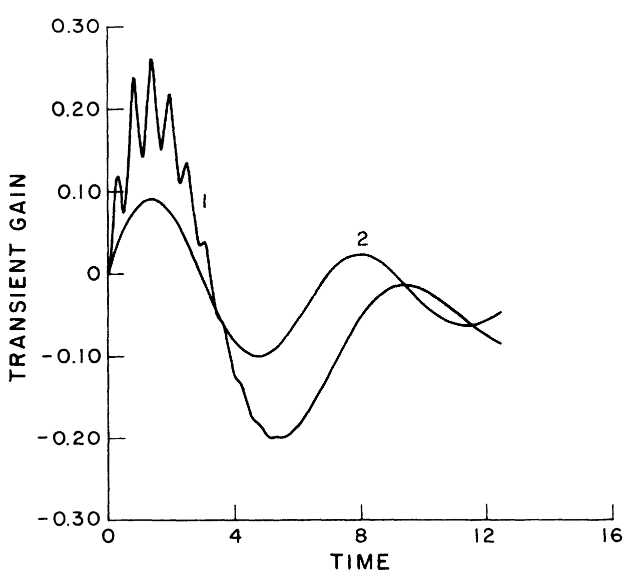

FIG. 3. Transient gain as a function of time for $\Omega_i=0.1$, $\gamma_f/\Gamma=0.1$, $q_i=5$, $q_f=2, \Delta_f=1$, and for pump detuned from the state $|a\rangle$, i.e., for (1) $\alpha=10$ and (2) $\alpha=50$.

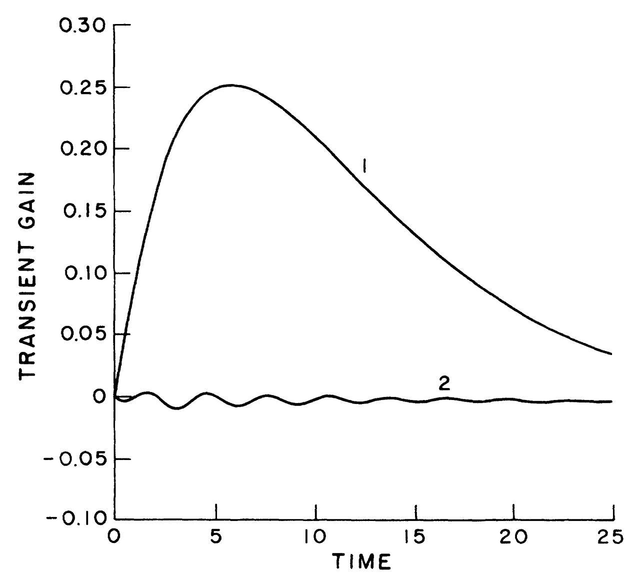

FIG. 5. The variation of transient gain as a function of time for $\alpha=50$ near the pump-field-induced minimum (1) $\delta=50.004 \rightrightarrows \Delta_f=0.004$ and near the Fano minimum (2) $\delta=-2.3 \rightrightarrows \Delta_f=-52.3$, other parameters are chosen as in Fig. 4.

was in the state $|f\rangle$. The steady-state susceptibility for such a case was found to be

$$\chi^{(f)}(\omega_2) = \frac{i\Gamma\Omega_f}{2\epsilon_2^2} \frac{[(\delta-\alpha)(\delta+2q_f+iq_f^2)-q_i^2\Omega_i(q_f/q_i-1)^2]}{(\delta-\epsilon_+)(\delta-\epsilon_-)} \quad (4.1)$$

where

$$\delta=\frac{2}{\Gamma}(E_f+\omega_2-E_a)\ . \quad (4.2)$$

In general, this susceptibility has two absorption minima. One is induced by the pump field and the other is the Fano minimum. Thus if we choose to work in the neighborhood of these absorption minima, then even a small population in the state $|a\rangle$ should lead to gain. In Fig. 4 we show some typical absorption spectra, which clearly show these two absorption minima. At low fields the field-induced minimum corresponds to the Raman transition from $|i\rangle$ to $|f\rangle$ (i.e., $\Delta_f=0$). In Figs. 5 and 6, we show the amplification of the probe beam for parameters corresponding to the field-induced minimum. We also show the corresponding gain profile if the parameters are chosen in the vicinity of the other absorption minimum (which is basically the Fano minimum). Figure 5 shows the remarkable result that the amplification of the probe persists over a wide range of times, with the larger gain occurring in the Raman transition. Figures 5 and 6 also show that the gain near the Fano minimum is significant only if the coherent field pumping the transition $|i\rangle\rightarrow|a\rangle$ is close to the resonance. Clearly Fig. 6 also shows that the gain near the Fano minimum is significant if the turn-on time of the field is less than a few Γ^{-1}.

In conclusion we have presented a laser model which works on the interference effects to produce laser action without population inversion. At the same time, although the interaction between the levels is important in establishing the absorption minimum, it is clear from our discussion that the gain without inversion in this case is closely related to the Raman transition from $|i\rangle$ to $|f\rangle$ (and the gain will cease if the population in $|f\rangle$ exceeds that of $|i\rangle$). We would also expect that, for incoherent pumping from some initial state, laser action would again cease when $|f\rangle$ exceeds $|i\rangle$.

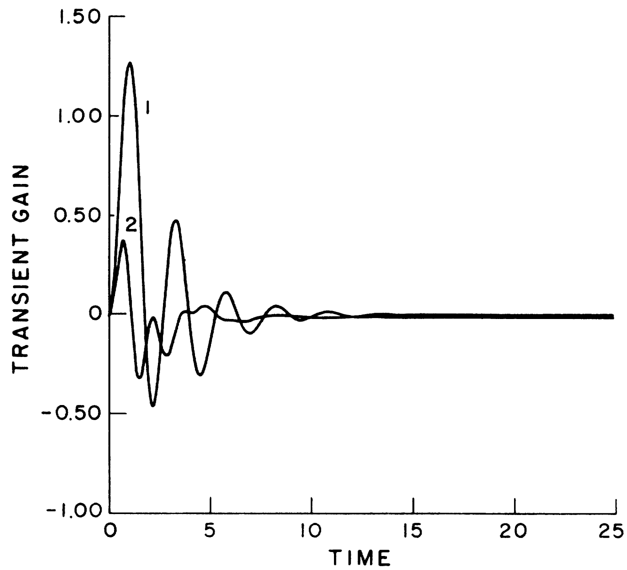

FIG. 6. Same as in Fig. 5 but now $\alpha=1$, (1) $\delta=1.4\Rightarrow\Delta_f=0.4$, and (2) $\delta=-2.7\Rightarrow\Delta_f=-3.7$.

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[3]M. O. Scully, S. Y. Zhu, and A. Gavrielides [Phys. Rev. Lett. **62**, 2813 (1989)] have discussed how ground-state coherence in a Λ system can be used to obtain gain without inversion.

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[6]We would like to comment that the presence of a second state is not absolutely essential. Using Harris's results or more directly using the Fano formulation one can show that the absorption and emission profiles for a single discrete state in the continuum are given by $\Omega_f\Gamma(\delta+q_f)^2/(1+\delta^2)$ for absorption and $\Omega_f\Lambda(1+q_f^2)/(1+\delta^2)$ for emission, where $\delta=(2/\Gamma)(E-E_f-\omega_2)$, and Λ is the rate at which the state $|a\rangle$ is being pumped. The pumping mechanism of Harris gives $\Lambda=\Gamma$. The steady-state population of the state $|a\rangle$ is $\Lambda/\Gamma=p_a$ and, if p_f is the population of the state $|f\rangle$, then the emission exceeds absorption if $p_a(1+q_f^2)>p_f(\delta+q_f)^2$. Thus near the Fano minimum even a small amount of population is $|a\rangle$ may lead to gain.

[7]G. S. Agarwal, S. L. Haan, and J. Cooper, Phys. Rev. A **29**, 2552 (1984); **29**, 2565 (1984). These papers contain an extensive bibliography of the earlier published work on the related problems.

[8]S. Ravi and G. S. Agarwal, Phys. Rev. A **35**, 3354 (1987). The framework for treating the lasers without inversion already exists in this paper, as here the authors deal with the absorption of a probe field for different initial conditions.

[9]For an explicit evaluation of these T matrices, see Appendix B of Ref. 8.