Lasers without Inversion: Interference of Lifetime-Broadened Resonances

S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 23 September 1988)

We show that if two upper levels of a four-level laser system are purely lifetime broadened, and decay to an identical continuum, then there will be an interference in the absorption profile of lower-level atoms, and that this interference is absent from the stimulated emission profile of the upper-level atoms. Laser amplification may then be obtained without inversion. Examples include interfering autoionizing levels, and tunneling systems.

PACS numbers: 42.55.Bi, 32.70.Jz, 32.80.Dz

While it is commonly believed that population inversion is a requirement for obtaining laser amplification, this is not so. In this Letter we show that if two upper levels of a four-level laser system are purely lifetime broadened, and decay to an identical continuum, then there will be an interference in the absorption profile of lower-level atoms, and this interference will be absent from the stimulated emission profile of the upper-level atoms. It is therefore possible to have a substantial gain cross section at frequencies where the absorption cross section is zero and to obtain laser amplification under conditions where the lower-level population greatly exceeds the upper-level population.

Figure 1 is an energy-level diagram for the analysis. Level $|1\rangle$ is the lower laser level. Levels $|2\rangle$ and $|3\rangle$ are upper levels which are assumed to be lifetime broadened and decay to the same continuum with decay rates Γ_2 and Γ_3. This decay may result, for example, from an Auger or autoionizing process. Level $|0\rangle$ is a reservoir from which the other levels may be excited by, for example, electron[1] or optical pumping. We consider two initial conditions: (a) atoms which at $t=0$ are in level $|1\rangle$, and (b) atoms which at $t=0$ are in level $|2\rangle$.

Those atoms which are initially in level $|1\rangle$ experience Fano-type interferences[2] in their absorption profile; i.e., there are several quantum-mechanical paths to the same final continuum level. Atoms which are initially in level $|2\rangle$ do not exhibit this type of destructive interference, but do display unusual properties. For example, even if the oscillator strength from level $|2\rangle$ to level $|1\rangle$ is zero, there will be gain on the $|2\rangle \rightarrow |1\rangle$ transition.

There is a precedent to this idea: Several years ago Arkhipkin and Heller[3] showed that a discrete level, which is embedded in a continuum and exhibits a Fano interference between photoionization to the continuum and (virtual) excitation of the discrete level, exhibits no such interference in emission. Here we show that the interference of two lifetime-broadened levels will create the same type of nonreciprocal interference. Though for generality we include a direct photoionization channel to the continuum, it is not an important or essential ingredient. The consideration of the interference of discrete lines allows much larger gain cross sections at the zero-loss point, and of more importance allows the application of these ideas to systems where the direct channel is absent or too small to be useful. Examples include systems where the lifetime broadening of the two upper levels occurs by autoionization, tunneling, or by radiative decay.[4]

The basis set for this work consists of three discrete levels and a density of continuum levels. We use a method described by Cohen-Tannoudji, Diu, and Laloe[5] and by Lambroupoulos and Zoller[6] to build the continuum into the coupled equations for the discrete levels. This is done by first integrating the continuum equations and then substituting the result back into the equations for the discrete levels. Thereafter, levels $|2\rangle$ and $|3\rangle$ are characterized by their decay rates to the continuum and their oscillator strengths, or equivalently by their Rabi frequencies to level $|1\rangle$. This method imposes a $t=0$, zero boundary condition on the continuum levels, and also requires that the matrix-element-weighted continuum be flat as compared to the spectral bandwidth of the

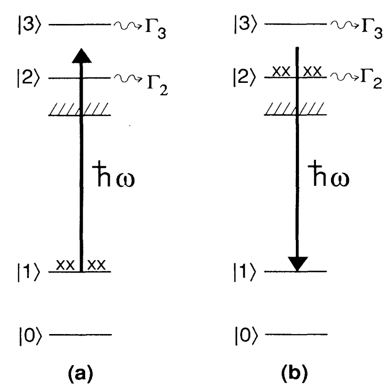

FIG. 1. Energy-level diagram for the analysis. (a) Atoms in level $|1\rangle$ at $t=0$ absorb probe radiation ($\hbar\omega$) and decay to an energy-conserving ion and electron. (b) Atoms in level $|2\rangle$ at $t=0$ both autoionize and are stimulated to level $|1\rangle$, thereby producing gain at the probe frequency. Levels $|1\rangle$, $|2\rangle$, and $|3\rangle$ may be pumped by electrons or photons from level $|0\rangle$.

time varying level amplitudes. The equations for the time varying amplitudes of a lower level $|1\rangle$ and upper levels $|2\rangle$ and $|3\rangle$ become the following:

$$\partial a_1/\partial t + j\Delta\tilde{\omega}_{11} a_1 = \kappa_{12} a_2 + \kappa_{13} a_3 , \quad (1a)$$

$$\partial a_2/\partial t + j\Delta\tilde{\omega}_{12} a_2 = \kappa_{12} a_1 + \kappa_{23} a_3 , \quad (1b)$$

$$\partial a_3/\partial t + j\Delta\tilde{\omega}_{31} a_3 = \kappa_{13} a_1 + \kappa_{23} a_2 . \quad (1c)$$

The quantities in these equations are

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

where Γ_2 and Γ_3 are the decay rates of levels $|2\rangle$ and $|3\rangle$; $\Delta\omega_{21} = \omega_2 - (\omega_1 + \omega)$; $\Delta\omega_{31} = \omega_3 - (\omega_1 + \omega)$; 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[2] so that Γ_2, Γ_3, and W_c are real. For brevity, a continuum-electric-field-principal-value term[2] which is additive to Ω_{ij} is neglected. 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.

Equations (1) do not allow for dephasing, and rigorously their solution only exhibits rate equation behavior in the limit of zero E field. However, a numerical solution of these equations shows that the formulas which follow are valid for Rabi frequencies which are much less than the complex detunings $\Delta\tilde{\omega}_{12}$ and $\Delta\tilde{\omega}_{13}$. Atoms which are initially in level $|2\rangle$ and are stimulated to level $|1\rangle$ and thereafter recycle, display the same Fano interference and time-averaged absorption as does an atom which is intially in level $|1\rangle$.

The transition rate for absorption W_{ab} is obtained by using the large detunings $\Delta\tilde{\omega}_{21}$ and $\Delta\tilde{\omega}_{31}$, relative to the derivatives, to neglect these derivatives in Eqs. (1b) and (1c). With $a_1 = 1$, the quantities $a_2(t)$ and $a_3(t)$ are substituted into Eq. (1a). The transition rate $W_{ab} = -(\partial/\partial t)|a_1(t)|^2$ is then

$$\frac{W_{ab}}{W_c} = 2\,\mathrm{Re}\left[\frac{1}{2} + \frac{2\kappa_{12}\kappa_{13}\kappa_{23} + j(\kappa_{12}^2\Delta\tilde{\omega}_{31} + \kappa_{13}^2\Delta\tilde{\omega}_{21})}{W_c(\Delta\tilde{\omega}_{21}\Delta\tilde{\omega}_{31} + \kappa_{23}^2)}\right] . \quad (3)$$

Equation (3) thereby neglects the transient portion of the response of $a_2(t)$ and $a_3(t)$. The assumption is that an atom will remain in level $|1\rangle$ for a time which is long compared to the transient, which is over in several decay times.

The transition rate for emission is obtained by applying the boundary condition $a_2 = 1$, $a_1 = a_3 = 0$ at $t = 0$, taking the E field and therefore κ_{12} and κ_{13} equal to zero, and solving for $a_2(t)$ and $a_3(t)$. This solution is then used, in the presence of a small E field, to find $a_1(t)$. As the coupled upper-level populations decay, the lower-level population $|a_1(t)|^2$ grows toward a final value $|a_1(\infty)|^2$. The transition rate for stimulated emission is defined as

$$W_e \equiv \Gamma |a_1(\infty)|^2 ,$$

where

$$1/\Gamma = \int_0^\infty [|a_2(t)|^2 + |a_3(t)|^2] dt . \quad (4)$$

We picture an ensemble of atoms which are excited to level $|2\rangle$ at a uniform rate which is sufficiently slow so that a single atom is not disturbed during its decay. The level $|1\rangle$ population of a single atom approaches $|a_1(\infty)|^2$, and the population of the ensemble grows linearly with time. The line shape, as a function of frequency, is contained within $a_1(\infty)$. We have defined Γ so that the lower-level population equals the downward transition rate W_e times the sum of the steady-state populations of levels $|2\rangle$ and $|3\rangle$. We mention that the numerical solution of Eq. (1) shows that for the case of a perfect Fano interference, even for Rabi frequencies on the order of the autoionizing time, atoms which are stimulated downward remain in level $|1\rangle$ thereafter. By substituting the solutions for the $a_i(t)$ into Eq. (4), we obtain

$$W_e = \Gamma \left| \frac{\kappa_{13}\kappa_{23} + j\kappa_{12}\Delta\tilde{\omega}_{31}}{\kappa_{23}^2 + \Delta\tilde{\omega}_{21}\Delta\tilde{\omega}_{31}} \right|^2 . \quad (5)$$

The expression for Γ is given in Eq. (7b).

These formulas may be put into compact form by using the Fano parameters q_i and η_i, defined as $q_2 = \Omega_{12}/(\Gamma_2 W_c)^{1/2}$, $q_3 = \Omega_{13}/(\Gamma_3 W_c)^{1/2}$, $\eta_2 = -\Delta\omega_{21}/(\Gamma_2/2)$, and $\eta_3 = -\Delta\omega_{31}/(\Gamma_3/2)$. To be strictly correct, the q_i should also contain a continuum-electric-field-principal-value term.[2]

The absorption cross section normalized to the continuum cross section σ_c is then

$$\frac{\sigma_{ab}}{\sigma_c} = \frac{(\eta_3 q_2 + \eta_2 q_3 + \eta_2\eta_3)^2}{\eta_2^2\eta_3^2 + (\eta_2 + \eta_3)^2} . \quad (6)$$

This result is implicit in Fano's original paper.[2] Readily obtained special cases include a single line in a continuum, and two interfering lines without a continuum. (Note that to remove a level from this formula, one must set $q = 0$, and $\eta = \infty$, thereby setting both its oscillator strength and its autoionizing time to zero.) Two cases are of special interest: the zero that occurs at the balance of virtual autoionizations for strong lines $|\eta_3 q_2| = |\eta_2 q_3|$, and the zero that occurs at line center ($\eta_2 = 0$) when $q_2 = 0$.

The cross section for stimulated emission by atoms in level $|2\rangle$ at $t = 0$, normalized to the continuum cross section σ_c, is

$$\frac{\sigma_e}{\sigma_c}=\left(\frac{\Gamma}{\Gamma_2}\right)\left[\frac{q_2^2\eta_3^2+[(q_2-q_3)-\eta_3]^2}{\eta_2^2\eta_3^2+(\eta_2+\eta_3)^2}\right], \tag{7a}$$

where

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

The quantity ΔE is the separation of levels $|2\rangle$ and $|3\rangle$. The ratio (Γ/Γ_2) approaches unity as ΔE becomes large. As ΔE becomes small the autoionization rate of level $|2\rangle$ is suppressed. This removes the singularity that would otherwise occur in Eq. (7a) for $\eta_3=0$, as $\eta_2\to 0$.

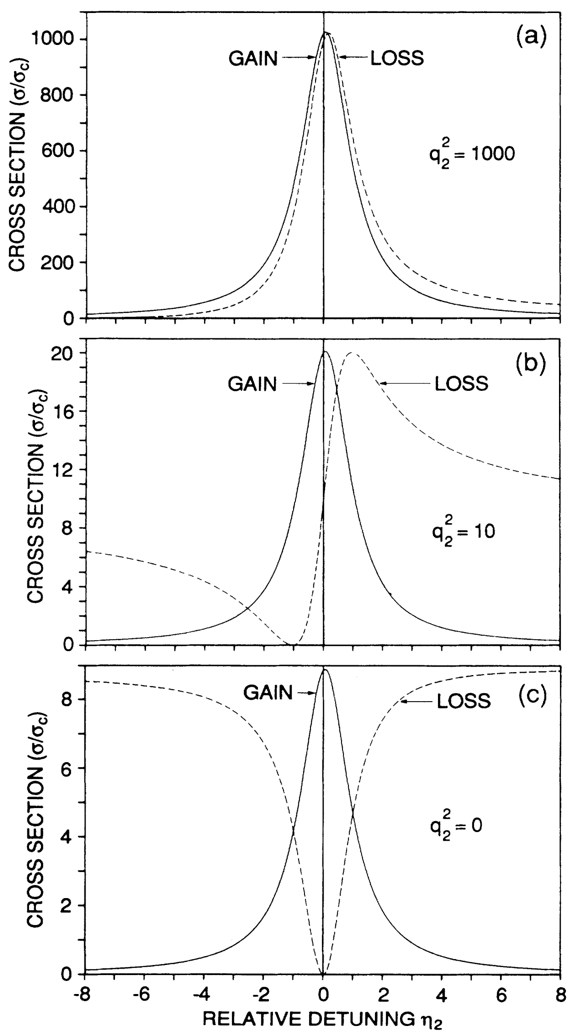

FIG. 2. Gain and loss profiles of interfering levels. For the loss profiles, $a_1=1$, $a_2=a_3=0$ at $t=0$. For the gain profiles, $a_2=1$, $a_1=a_3=0$ at $t=0$. The only parameter which varies between (a)-(c) is q_2^2. The other parameters are $q_3^2=1000$, $\Gamma_2=1$, and $\Gamma_3=250$. Levels $|2\rangle$ and $|3\rangle$ are separated by 2000 linewidths of level $|2\rangle$.

Perhaps the most interesting special case of these formulas is obtained as q_2 approaches zero. We find Lorentzian-shaped gain and loss profiles centered on a line which has zero oscillator strength. Figure 2 shows this result. Both gain and loss are plotted as functions of the normalized detuning η_2. The only parameter which varies between plots is q_2^2. For lines of equal strength the normalized gain is about 1000, and though there is a region where gain exceeds loss, the fractional difference is small. For $q_2^2=10$, a point of zero loss occurs near the line center at a gain of about ten units. For $q_2^2=0$ one obtains nearly symmetrical Lorentzian proflies, with zero loss at the point of maximum gain.

At first glance, one wonders why results of this type do not violate detailed balance. Detailed balance applies between the initial level $|1\rangle$ and the final level, which is an ion and a free electron. A recombination laser, i.e., starting with electrons and ions, could not be made by this method. For a single line in a continuum, recombination exhibits a Fano profile[7]; two lines in a continuum exhibit an interference of their (virtual) dielectronic recombinations.

We note again the neglect of the transient absorptive response in Eq. (3). Though this is always done when computing Fano profiles, it has an important implication for laser processes. This transient response will be initiated whenever a level $|1\rangle$ atom is dephased by a collision, or is produced at $t=0$. Therefore, if level $|1\rangle$ atoms are produced at a rate which equals or exceeds that of level $|2\rangle$ atoms, there will be no net gain. The Fano interference cancels loss which is caused by the lifetime broadening of levels $|2\rangle$ and $|3\rangle$ and, in effect, reduces or prevents the integration or buildup of level $|1\rangle$ population.

The above formulas have also assumed that all processes, i.e., the excitation of the continuum and of levels $|2\rangle$ and $|3\rangle$, produce the same final continuum level. For one-dimensional tunneling this is the case. For certain special cases in atoms, for example, the interference of Rydberg levels which have the same angular momentum, this is also the case, and the interferences and gain cross sections are as given. But in general, different final electrons are produced and only identical channels will interfere.[2,8]

The principal result of this Letter is that interferences between lifetime-broadened discrete levels which decay to the same continuum are sufficient to produce nonreciprocal gain-loss profiles. These effects may occur naturally in extreme-ultraviolet and x-ray laser systems or may be synthesized using artificially layered materials.

The author thanks Roberto Buffa for participating in the development of the type of equations used here, and thanks C. P. J. Barty, M. M. Fejer, W. A. Harrison, A. Imamoglu, J. J. Macklin, A. E. Siegman, and J. F. Young for helpful discussions. I thank J. D. Kmetec for doing numerical work on the large signal behavior of the coupled equations. This work was supported by the U.S. Air Force Office of Scientific Research and the U.S. Army Research Office.

[1]J. C. Wang, R. G. Caro, and S. E. Harris, Phys. Rev. Lett. **51**, 767 (1983).

[2]U. Fano, Phys. Rev. **124**, 1866 (1961).

[3]V. G. Arkhipkin and Yu. I. Heller, Phys. Lett. **98A**, 12 (1983).

[4]A. Imamoglu (to be published).

[5]C. Cohen-Tannoudji, B. Diu, and F. Laloe, *Quantum Mechanics* (Hermann, Paris, France, 1977), Vol. 2, pp. 1343–1347.

[6]P. Lambropoulos and P. Zoller, Phys. Rev. A **24**, 379 (1981).

[7]B. W. Shore, Phys. Rev. **171**, 43 (1968).

[8]U. Fano and J. W. Cooper, Phys. Rev. **137**, 1364 (1965).