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Interference of radiatively broadened resonances

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We show that a three-state atomic system where the upper two states have the same J and m_J quantum numbers and decay radiatively to the states of a single atomic level is equivalent to the recently proposed inversionless laser system [S. E. Harris, Phys. Rev. Lett. **62**, 1033 (1989)].

In a recent paper, Harris[1] showed that in some atomic systems population inversion is not a necessary condition for obtaining laser action. He considered a three-level system, where the two upper levels are purely lifetime broadened and decay by autoionization to an identical continuum. For this system, stimulated emission and absorption line shapes are different due to the presence of Fano-type interferences.[2,3] At a certain laser frequency, the absorption rate goes to zero, whereas the emission rate remains nonzero. Amplification of a laser field at this frequency is possible even though the number of lower-level atoms in the system is higher than the number of upper-level atoms. A semiclassical approach was used in the analysis of Ref. 1, thus radiatively broadened resonances were not considered.

In this Rapid Communication, we use a full quantum-mechanical approach to show that a three-state atomic system where the upper two states are radiatively broadened is equivalent to the lifetime broadened systems considered by Harris, provided certain selection rules are satisfied. The selection rules require that the two upper states have the same J and m_J quantum numbers and that they decay to the states of a single atomic level.

The system that we consider is outlined in Fig. 1. A lower atomic state $|1\rangle$ is coupled to two upper states $|2\rangle$ and $|3\rangle$ via a probe laser field at frequency $\omega_{\mathbf{k}_L}$. These upper states decay to a single atomic state $|i\rangle$ by spontaneous emission. The emitted photon can have arbitrary direction and energy.[4] For absorption, we consider an atom which has been in the lower state for a long time compared to the lifetimes of the upper states. In the absence of dephasing events, the interaction of such an atom with a weak monochromatic incident field leads only to Raman scattering into state $|i\rangle$. Raman scattering for this system takes place through two intermediate states ($|2\rangle$ and $|3\rangle$), so that the resulting scattering (identically equal to absorption) probability has an interference term. This interference term may yield a zero in the absorption line shape.

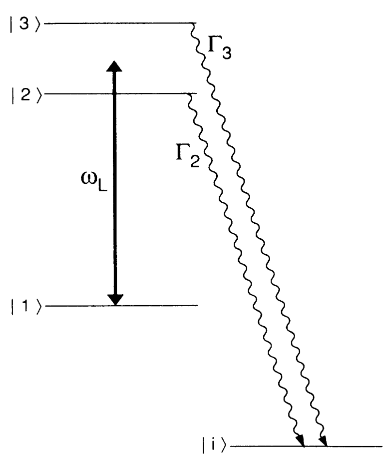

FIG. 1. Radiatively broadened system.

For emission, we consider an atom pumped into state $|2\rangle$ from a reservoir. Due to the coupling of the states $|2\rangle$ and $|3\rangle$ via their decay process, state $|3\rangle$ is also excited, and therefore the stimulated transition to state $|1\rangle$ takes place through two paths. The corresponding interference term, however, is different in this case, giving an emission line shape that is not the same as the absorption.

The basis set that we use in the analysis consists of a number of eigenstates of the noninteracting atom plus radiation field Hamiltonian. Assuming that there are no photons in any but the laser mode of the radiation field initially, we can write the state vector of the total system in the interaction representation[5,6] as

$$|\Psi_I(t)\rangle = a_1(t)\,|1,n_{\mathbf{k}_L}\rangle + a_2(t)\,|2,n_{\mathbf{k}_L}-1\rangle + a_3(t)\,|3,n_{\mathbf{k}_L}-1\rangle + \sum_{\sigma\mathbf{k}} a_{i,\sigma\mathbf{k}}(t)\,|i,n_{\mathbf{k}_L}-1,1_{\sigma\mathbf{k}}\rangle\,, \tag{1}$$

where a_1, a_2, a_3, and $a_{i,\sigma\mathbf{k}}$ are the probability amplitudes; $n_{\mathbf{k}_L}$ is the number of photons in the laser mode; $1_{\sigma\mathbf{k}}$ represents the fact that the number of photons in the radiation mode $\sigma\mathbf{k}$ is one; $\mathbf{k}$ and σ are the wave vector and polarization of the spontaneously emitted (or Raman scattered) photon, respectively. The expansion in (1) assumes that the radiative decay into atomic state $|1\rangle$ is negligible and that only the eigenstates with approximately equal energy are coupled by the interactions. The latter assumption is equivalent to the rotating wave approximation in the semiclassical approach.

By substituting (1) in Schrödinger's equation[6]

$$i\hbar\frac{\partial|\Psi_I(t)\rangle}{\partial t} = \hat{H}_I(t)\,|\Psi_I(t)\rangle\,, \tag{2a}$$

where

$$\hat{H}_I(t) = i\hbar \sum_{\substack{\sigma,\mathbf{k}',m,n\\ m\neq n}} g_{nm\sigma\mathbf{k}'}\{|m\rangle\langle n|a_{\mathbf{k}'}\exp[i(\omega_{mn}-\omega_{\mathbf{k}'})t] - a^\dagger_{\mathbf{k}'}|n\rangle\langle m|\exp[-i(\omega_{mn}-\omega_{\mathbf{k}'})t]\} \tag{2b}$$

and setting the energy of the atomic state $|1\rangle$ to zero, we obtain the equations of motion for the probability amplitudes:

$$\frac{\partial a_1(t)}{\partial t} = -g_{12\mathbf{k}_L} n^{1/2}_{\mathbf{k}_L}\exp[i(\omega_2-\omega_{\mathbf{k}_L})t]a_2(t) - g_{13\mathbf{k}_L} n^{1/2}_{\mathbf{k}_L}\exp[-i(\omega_3-\omega_{\mathbf{k}_L})t]a_3(t)\,, \tag{3a}$$

$$\frac{\partial a_2(t)}{\partial t} = g_{12\mathbf{k}_L} n^{1/2}_{\mathbf{k}_L}\exp[i(\omega_2-\omega_{\mathbf{k}_L})t]a_1(t) + \sum_{\sigma,\mathbf{k}} g_{i2\sigma\mathbf{k}}\exp[i(\omega_{2i}-\omega_{\mathbf{k}})t]a_{i,\sigma\mathbf{k}}(t)\,, \tag{3b}$$

$$\frac{\partial a_3(t)}{\partial t} = g_{13\mathbf{k}_L} n^{1/2}_{\mathbf{k}_L}\exp[i(\omega_3-\omega_{\mathbf{k}_L})t]a_1(t) + \sum_{\sigma,\mathbf{k}} g_{i3\sigma\mathbf{k}}\exp[i(\omega_{3i}-\omega_{\mathbf{k}})t]a_{i,\sigma\mathbf{k}}(t)\,, \tag{3c}$$

and an equation for each $\sigma\mathbf{k}$:

$$\frac{\partial a_{i,\sigma\mathbf{k}}(t)}{\partial t} = -g_{i2\sigma\mathbf{k}}\exp[-i(\omega_{2i}-\omega_{\mathbf{k}})t]a_2(t) - g_{13\sigma\mathbf{k}}\exp[-i(\omega_{3i}-\omega_{\mathbf{k}})t]a_3(t)\,, \tag{3d}$$

where

$$\omega_{mn} = \omega_m - \omega_n\,,$$
$$g_{nm\sigma\mathbf{k}} = e\left(\frac{\omega_{\mathbf{k}}}{2\epsilon_0\hbar V}\right)^{1/2}\hat{\epsilon}_{\sigma\mathbf{k}}\cdot\mathbf{D}_{nm}\,, \tag{4}$$
$$\mathbf{D}_{nm} = \langle n|\mathbf{r}|m\rangle\,,$$

where $\hat{\epsilon}_{\sigma\mathbf{k}}$ is the polarization vector of the photon $\sigma\mathbf{k}$, and V is the normalization volume.

Using the initial condition $a_{i,\sigma\mathbf{k}}(t=0)=0$, we integrate[1,5] Eq. (3d) and substitute for $a_{i,\sigma\mathbf{k}}(t)$ in (3b) and (3c):

$$\frac{\partial a_1(t)}{\partial t} = -g_{12\mathbf{k}_L} n^{1/2}_{\mathbf{k}_L}\exp[-i(\omega_2-\omega_{\mathbf{k}_L})t]a_2(t) - g_{13\mathbf{k}_L} n^{1/2}_{\mathbf{k}_L}\exp[-i(\omega_3-\omega_{\mathbf{k}_L})t]a_3(t)\,, \tag{5a}$$

$$\begin{aligned}\frac{\partial a_2(t)}{\partial t} = {}& g_{12\mathbf{k}_L} n^{1/2}_{\mathbf{k}_L}\exp[i(\omega_2-\omega_{\mathbf{k}_L})t]a_1(t) - \sum_{\sigma,\mathbf{k}}(g_{i2\sigma\mathbf{k}})^2\pi\delta(\omega_{2i}-\omega_{\mathbf{k}})a_2(t)\\ &+ iP\sum_{\sigma,\mathbf{k}}\frac{g^2_{i2\sigma\mathbf{k}}}{\omega_{\mathbf{k}}-\omega_{2i}}a_2(t) + iP\sum_{\sigma,\mathbf{k}}\frac{g_{i2\sigma\mathbf{k}}g_{i3\sigma\mathbf{k}}}{\omega_{\mathbf{k}}-\omega_{3i}}e^{i\omega_{23}t}a_3(t) - \sum_{\sigma,\mathbf{k}} g_{i2\sigma\mathbf{k}}g_{i3\sigma\mathbf{k}}\pi\delta(\omega_{3i}-\omega_{\mathbf{k}})e^{i\omega_{23}t}a_3(t)\,,\end{aligned} \tag{5b}$$

$$\begin{aligned}\frac{\partial a_3(t)}{\partial t} = {}& g_{13\mathbf{k}_L} n^{1/2}_{\mathbf{k}_L}\exp[+i(\omega_3-\omega_{\mathbf{k}_L})t]a_1(t) - \sum_{\sigma,\mathbf{k}}(g_{i3\sigma\mathbf{k}})^2\pi\delta(\omega_{3i}-\omega_{\mathbf{k}})a_3(t)\\ &+ iP\sum_{\sigma,\mathbf{k}}\frac{g^2_{i3\sigma\mathbf{k}}}{\omega_{\mathbf{k}}-\omega_{3i}}a_3(t) + iP\sum_{\sigma,\mathbf{k}} g_{i2\sigma\mathbf{k}}\frac{g_{i2\sigma\mathbf{k}}g_{i3\sigma\mathbf{k}}}{\omega_{\mathbf{k}}-\omega_{2i}}e^{-i\omega_{23}t}a_2(t) - \sum_{\sigma,\mathbf{k}} g_{i2\sigma\mathbf{k}}g_{i3\sigma\mathbf{k}}\pi\delta(\omega_{2i}-\omega_{\mathbf{k}})e^{-i\omega_{23}t}a_2(t)\,.\end{aligned} \tag{5c}$$

Assuming that the modes of the radiation field are closely spaced in frequency, we replace the summation over $\mathbf{k}$ by an integral, in each term. The imaginary principal-value terms (denoted by P) on the right-hand side of Eqs. (5b) and (5c) then give the energy shifts induced by the presence of the vacuum fluctuations. They can be eliminated by a prediagonalization on the original basis set that mixes the "bare" eigenstates so that the energies of the "dressed states" formed with this process are the experimentally observed energies, in the absence of an incident laser field.[7] We proceed by assuming that the atomic states that we have used in our basis set are these prediagonalized dressed states, and set the principal-value terms equal to zero.

The real diagonal terms in Eqs. (5b) and (5c) can be identified as the direct radiative decay rates of the states $|2\rangle$ and $|3\rangle$, respectively.[5,6] The real cross-coupling terms give the finite amplitude for the absorption of a virtual photon emitted from state $|3\rangle$ ($|2\rangle$), by state $|2\rangle$ ($|3\rangle$). These terms lead to observable interference effects only if

$$\omega_{2i} \cong \omega_{3i}\,, \tag{6a}$$

$$\mathbf{D}_{2i} = c\mathbf{D}_{3i} \quad (c \text{ is a scalar})\,. \tag{6b}$$

The assumptions (6a) and (6b) require that the states $|2\rangle$ and $|3\rangle$ be close lying in energy compared to their separation from the state they decay to and that they have the same m_J quantum number, respectively. By a change of variables,

$$a_1(t) = ia_1'(t)\,,$$
$$a_2(t) = a_2'(t)\exp[i(\omega_2-\omega_{\mathbf{k}_L})t]\,, \tag{7}$$
$$a_3(t) = a_3'(t)\exp[i(\omega_3-\omega_{\mathbf{k}_L})t]\,,$$

we arrive at the following equations:

$$\frac{\partial a_1'(t)}{\partial t} = \kappa_{12} a_2'(t) + \kappa_{13} a_3'(t) , \tag{8a}$$

$$\frac{\partial a_2'(t)}{\partial t} + i\Delta\tilde{\omega}_2 a_2'(t) = \kappa_{21} a_1'(t) + \kappa_{23i} a_3'(t) , \tag{8b}$$

$$\frac{\partial a_3'(t)}{\partial t} + i\Delta\tilde{\omega}_3'(t) = \kappa_{31} a_1'(t) + \kappa_{32i} a_2'(t) , \tag{8c}$$

where

$$\Delta\tilde{\omega}_2 = \omega_2 - \omega_{\mathbf{k}_L} - i\frac{\Gamma_{2i}}{2} , \quad \Delta\tilde{\omega}_3 = \omega_3 - \omega_{\mathbf{k}_L} - i\frac{\Gamma_{3i}}{2} , \tag{9a}$$

$$\kappa_{12} = \kappa_{21} - i g_{12\mathbf{k}_L} n_{\mathbf{k}_L}^{1/2} , \quad \kappa_{13} = \kappa_{31} = i g_{13\mathbf{k}_L} n_{\mathbf{k}_L}^{1/2} , \quad \kappa_{23i} = \kappa_{32i} = -\frac{(\Gamma_{2i}\Gamma_{3i})^{1/2}}{2}\frac{|c|}{c} , \tag{9b}$$

where Γ_{2i} and Γ_{3i} are the radiative decay rates of states $|2\rangle$ and $|3\rangle$, respectively. κ_{23i} and κ_{32i} are the cross-coupling terms, and c is as defined in (6b).

We can see from (8) that the equations of motion for the radiatively broadened system are equivalent to those obtained in Ref. 1 for the interference of autoionizing levels, provided that one sets the photoionization rate W_{c1}, equal to zero in the latter case. The absorption and emission line shapes which are obtained from Eqs. (8a)-(8c) are different from each other.[1,8] When the probe laser is tuned such that $\kappa_{21}(\omega_3 - \omega_{\mathbf{k}_L})\Gamma_{2i}^{1/2} = -\kappa_{31}(\omega_2 - \omega_{\mathbf{k}_L})\Gamma_{3i}^{1/2}$, the absorption rate goes through a zero whereas the emission rate remains nonzero. This fact implies that amplification of the laser field without having a population inversion is possible.

If there is radiative decay into more than one final atomic state, Eqs. (8) and (9) should be modified as

$$\Gamma_{2i} \rightarrow \sum_i \Gamma_{2i} , \quad \Gamma_{3i} \rightarrow \sum_i \Gamma_{3i} , \quad \kappa_{23i} \rightarrow \sum_i \kappa_{23i} , \tag{10}$$

where summation is over all final atomic states. The condition for a "zero" in absorption line shape is then

$$\left[\sum_i \kappa_{23i}\right]^2 = \frac{1}{4}\left[\sum_i \Gamma_{2i}\right]\left[\sum_i \Gamma_{3i}\right] . \tag{11}$$

Of special interest is the case of radiative decay into all m_J states of a single final atomic level. As prescribed by (10), one should first find the decay and cross-coupling rates corresponding to each final m_J state and then add these terms to get the total rates. Using Racah algebra,[9] we show that if $J_2 \neq J_3$, then $\sum \kappa_{23i} = 0$, and if $J_2 = J_3$, then (11) is satisfied. Therefore, only when the two upper states have identical J's, will the interference effects be observable. In other words, the absorption and emission-line shapes given in Refs. 1 and 8 are valid for a system where the two upper states have the same total angular momentum and decay to a single atomic level. The presence of radiative decay into more than one atomic level will, in general, change the zero to a "minimum" in absorption line shape.

An implication of the result stated above is that there is a perfect cancellation point in the Raman scattering profile of a polarized laser field from two close-lying atomic levels, provided that these levels have the same angular momentum.

Interferences in the absorption profile are observable in spite of the fact that there are infinitely many final continuum states given by the direction and polarization of the emitted photon. We explain this by noting that the coupling of states $|2\rangle$ and $|3\rangle$ to the continuum have the same ratio for every channel, where a channel is defined by a given direction and polarization for the emitted photon. When this is the case, scattering probability into every channel as well as the total scattering (absorption) probability, will have the same profile. In other words, as the Raman scattering from the two upper states give identical emission patterns, the interference effects are observable.

Experimental demonstration of these results could be most easily performed in an atomic system where two close-lying upper levels with identical J decay mainly to a single level. Only the states with the same m_J quantum number of the close-lying levels will couple through the continuum to give nonidentical absorption and emission profiles in a transition to some other state to which the two upper states are weakly coupled. One apparent difficulty with this system is that two levels with identical angular momentum generally have energy separations much larger than their radiative decay width, as the energy separations are mainly determined by the electrostatic and spin-orbit interactions and the radiative decay width by the interactions with the vacuum fluctuations. This implies that the gain cross section at the frequency where the absorption goes to zero is smaller by a factor proportional to the square of the ratio of the decay width to the energy separation, compared to the peak gain cross section. Laser systems might be created by using dressed-state ideas to locate two upper states close to each other.

We have extended the previous work on interference of autoionizing levels[1] to show that two close-lying states, having the same J and m_J quantum numbers and decaying radiatively to a single atomic level, will have different absorption and emission line shapes.

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