Suppression of the line broadening by fast Rabi flopping effects

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We report on a new mechanism that can lead to line narrowing. We show that the line-broadening effects associated with a broadband source can be suppressed by irradiating the system with a coherent pump. We give explicit results for this suppression in spontaneous emission from a two-level system.

One of the important problems of high-resolution spectroscopy is to achieve better and better resolution techniques to suppress the sources of various line-broadening mechanisms. Several dynamical processes are known to lead to line-narrowing phenomena. The well-known example is the collisional narrowing[1] of the spectral lines, which arises due to the mixing of the lines when the pressure increases. The collisional narrowing has been studied extensively in linear as well as in nonlinear spectroscopy. Line narrowing has also been found in several other contexts.[2–5] For example, Agarwal *et al.*[2] reported it in the radiation produced by a Rydberg atom in a cavity driven by incoherent radiation. Lewenstein *et al.*[3] found line narrowing in resonance fluorescence produced by a coherently driven atom contained in an optical cavity. Hanamura[4] and also Agarwal[5] found that the spontaneous emission produced by an atom can be considerably modified when an atom is strongly driven in the presence of certain types of frequency modulations. Carmichael *et al.*[6] have proposed use of squeezed cavities to obtain line narrowing.

Here we report a mechanism that leads to the suppression of the line-broadening effects. We show that a strong competition between the effects of the coherent and incoherent pumps can lead to the narrowing of the spectral profiles, which obviously can have spectroscopic applications, for example, in the study of the otherwise overlapping lines. Consider the interaction of a system (two-level for illustration purposes) with a broadband incoherent pump. It is known that the effects of a broadband pump can be accounted for by changing the relaxation times T_1, and T_2 in the Bloch equations. For example, for the case of radiative relaxation (rate 2γ) we have

$$\frac{1}{T_1}=2(\gamma+\beta)=\frac{2}{T_2}, \quad \beta=\frac{2|x|^2}{\Gamma}, \tag{1}$$

where $|x|$ is the Rabi frequency associated with the broadband pump and Γ is its bandwidth. The spontaneous-emission spectrum from such a system in the presence of a coherent drive (Rabi frequency Ω) is the Mollow spectrum, i.e., it consists of (i) a line at $\omega=\omega_l$ with width $\sim(\gamma+\beta)$ if $\Omega<1/T_1$ and (ii) lines at $\omega=\omega_l$, $\omega_l\pm\Omega$ with widths $(\gamma+\beta)$ and $\frac{3}{2}(\gamma+\beta)$ if $\Omega\gg 1/T_1$. Thus the broadband incoherent pump contributes an amount β to the widths of the observed peaks.

The question arises—is there a way to suppress the effects of β, i.e., how can the spontaneous emission lines be narrowed? We demonstrate that line narrowing is possible if the strength of the coherent pump is such that many Rabi oscillations are possible within the coherence time of the incoherent pump. This is the regime where Bloch equations[7] (or the usual relaxation equations for a multilevel system[8]) cannot be used and thus one has to work with more general relaxation equations and the specific models of the pump field.

We take the atom to be driven by a pump of the form

$$\mathbf{E}(t)=\hat{\epsilon}(\varepsilon_0+\varepsilon_1(t))e^{i\omega_l t+i\mathbf{k}\cdot\mathbf{r}}+\text{c.c.}, \tag{2}$$

where ε_0 is the coherent part and ε_1 is the fluctuating part. We write $\varepsilon_1(t)$ as

$$\varepsilon_1(t)=e^{-i\varphi}x_0(t), \tag{3}$$

where φ is the random phase and $x_0(t)$ is a dichotomic Markov process[9,10] with zero mean and with a correlation time Γ^{-1}, i.e.,

$$\langle x_0(t)x_0(t')\rangle=x_0^2e^{-\Gamma|t-t'|}. \tag{4}$$

Thus the pump spectrum is the sum of a coherent part and a Lorentzian of half-width Γ. It should also be noted that in the limit of $\Gamma\to\infty$, the process $x_0(t)$ is essentially δ-function correlated and the dynamics of the atom is described by optical Bloch equations. The spectrum of the emitted radiation would then be a Mollow spectrum. We will see that the spectral characteristics of the radiation emitted by our system are very sensitive to the relative values of the parameters such as the correlation time and the Rabi frequency associated with the coherent part of the pump.

It should be noted that we have adopted a two-state model for the incoherent part of the pump so that many of the results can be obtained analytically.[9–11] However, the essential physics is expected to be insensitive to the details of the model.

We next discuss the results of our exact calculation valid for arbitrary values of Γ, x, ε_0, etc. Let Ω be the Rabi frequency associated with the coherent part of the pump, i.e., $\Omega=-2\mathbf{d}\cdot\hat{\epsilon}e^{i\mathbf{k}\cdot\mathbf{r}}\varepsilon_0$. The density matrix equation in a frame rotating with the frequency of the pump (which is on resonance with atom) is

$$\frac{\partial \rho}{\partial t} = -\gamma(S^+S^-\rho - 2S^-\rho S^+ + \rho S^+S^-) - i\left[\left[\frac{\Omega}{2} + x(t)\right]S^+ + \left[\frac{\Omega}{2} + x^*(t)\right]S^-, \rho\right] . \quad (5)$$

Introducing the column matrix ψ with components $\langle S^\pm \rangle$ and $\langle S^Z \rangle$, Eq. (5) can be written in the matrix form as

$$\dot{\psi} = \{C_0 - i[x(t)C_+ + x^*(t)C_-]\}\psi + g , \quad (6)$$

where the matrices C_0, $C_\pm$, and g are to be obtained from (5). Calculations show that the ensemble average of ψ over the fluctuations of the pump is

$$\langle \hat{\psi}(z) \rangle = \hat{D}^{-1}(z)[z^{-1}g + \langle \psi(0) \rangle] , \quad (7)$$

$$\hat{D}(z) = z - C_0 + |x|^2 C_+(z + \Gamma - C_0)^{-1}C_- + |x|^2 C_-(z + \Gamma - C_0)^{-1}C_+ , \quad (8)$$

where the carets denote the Laplace transforms.

We have proved that the exact result for the spectrum of the radiation emitted by the two-level atom is obtained from

$$S(\omega) = \mathrm{Re} \lim_{z \to i(\omega - \omega_l)} [\hat{S}_1(z) + \tfrac{1}{2}\hat{D}_{11}^{-1}(z)] , \quad (9)$$

where $\hat{S}_1(z)$ is given by the first element of the column vector

$$\hat{S}(z) = \hat{D}^{-1}(z)\left[\hat{I}_1(z) + \hat{I}_2(z) + M\langle \psi(\infty) \rangle + \frac{g}{z}\psi_2(\infty)\right] , \quad (10)$$

where $\langle \psi(\infty) \rangle$ is the steady-state value of $\langle \psi \rangle$ and

$$\hat{I}_1(z) = -C_+(z + \Gamma - C_0)^{-1}M(\Gamma - C_0)^{-1}C_-|x|^2\langle \psi(\infty) \rangle + \mathcal{A}(C_+ \rightleftarrows C_-) ,$$

$$\hat{I}_2(z) = -C_+(z + \Gamma - C_0)^{-1}N(\Gamma - C_0)^{-1}(z + \Gamma)^{-1} \times C_-\langle \psi(\infty) \rangle |x|^2 + \mathcal{A}(C_+ \rightleftarrows C_-) . \quad (11)$$

Here $\mathcal{A}(C_+ \rightleftarrows C_-)$ denotes terms obtained by interchanging C_+ and C_- and the nonvanishing elements of M and N are $N_{32} = -\gamma$, $M_{13} = -2M_{32} = 1$.

The results (7) and (9) are exact. Thus the spectrum of the emitted radiation can be calculated for a wide range of parameters by using Eqs. (7)–(11). The important parameters are Rabi frequency Ω, correlation time Γ^{-1}, the usual relaxation width $\beta = 2|x|^2/\Gamma$, and detuning Δ.

We show the results for the spectrum of the emitted radiation for some typical cases in Figs. 1 and 2. We only plot the incoherent part of the spectrum for pump on resonance with the atom. Figure 1 shows how the central component narrows with increase in the Rabi frequency. For small Ω the spectrum exhibits only the central peak with width $\gamma + \beta$. However, as Ω increases the central peak exhibits dramatic narrowing approaching asymptotically a value γ for $\Omega \gg \Gamma$. In contrast, the side peaks show only a marginal amount of narrowing. Thus the dramatic reduction of the linewidth occurs predominantly in the region $\omega \sim \omega_l = \omega_0$ when many Rabi floppings during the coherence time of the pump are possible. This in fact is the case when optical Bloch equations fail to characterize the dynamics of the atom. This narrowing is reminiscent of the narrowing noted in related contexts[2–6] and is essentially achieved by effectively decoupling the dipole from the bath degrees of freedom. This can be seen in a qualitative fashion from optical Bloch equations which contain stochastic terms like $x(t)\langle S^z(t) \rangle$, $x(t)\langle S^+(t) \rangle$ etc. If the frequencies associated with $x(t)$ and $\langle S^z(t) \rangle$, for example, are very different, then terms

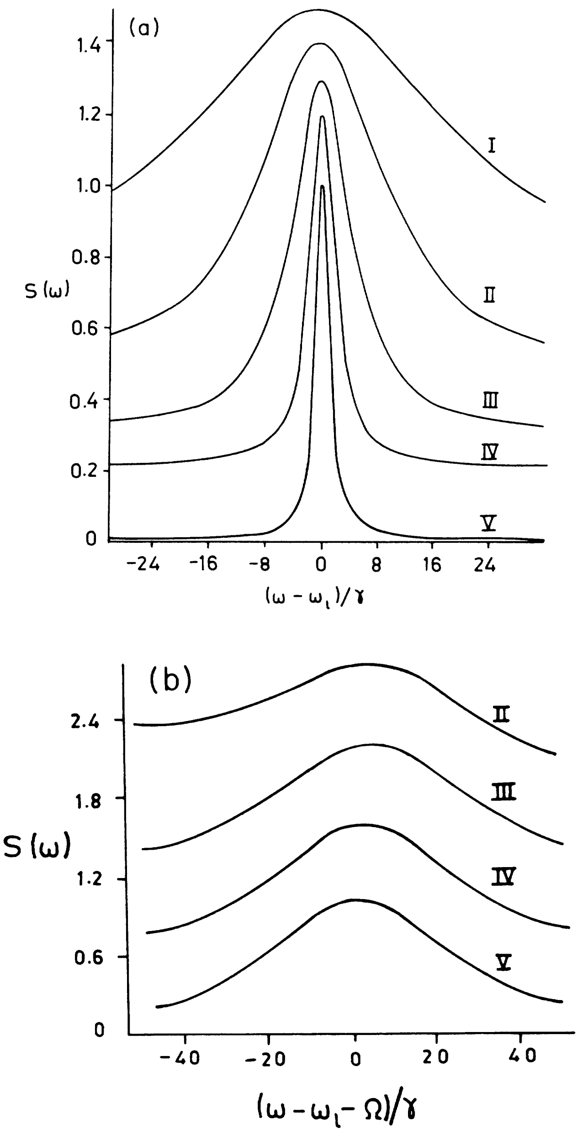

FIG. 1. Spectrum of fluorescence as a function of $(\omega - \omega_1)/\gamma$ for $\beta = 20\gamma$, $\Gamma = 100\gamma$, and for Rabi frequency (Ω/γ), of the coherent pump equal to (I) 40, (II) 200, (IV) 400, and (V) 1000. Part (a) [(b)] gives the behavior of the central (one of the side) peak. The central peak shows dramatic narrowing. For clarity the origin on the y axis is shifted for curves (I), (II), (III), and (IV) in (a) by 0.5, 0.4, 0.3, and 0.2, respectively. In (b) each curve is shifted relative to the next by 0.6.

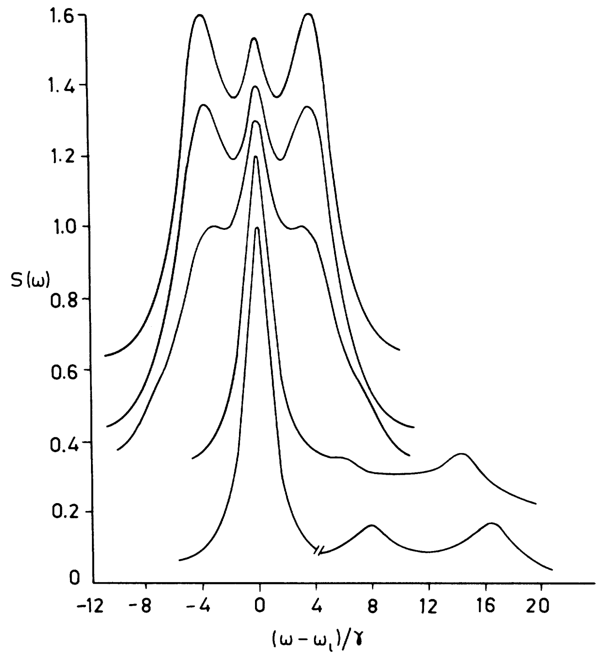

FIG. 2. Same as in Fig. 1 but now the correlation time of the broadband source is comparable to the lifetime of the atom $\Gamma=\gamma$. The curves from top to bottom are in the order of increasing Rabi frequency $\Omega/\gamma=0.2$, 1, 2, 10, and 20. For clarity the maximum in the subsequent curves is shifted relative to the bottom-most curve by 0.2, 0.3, 0.4, and 0.6, respectively. For last two cases only the side bands on one side are shown. The side bands for $\Omega=20\gamma$, are shifted to the left by eight units.

such as $x(t)\langle S^z(t)\rangle$ would average out to zero. Thus the $\langle S^+\rangle$ component effectively gets decoupled.

In Fig. 2 we consider a different regime which is not traditionally considered though experimentally possible. Here the correlation time of the incoherent part of the pump is comparable to the spontaneous-emission time. This is the region in which the non-Markovian effects are most significant. In such a case Mollow-like spectra can be obtained even if the Rabi frequency corresponding to the coherent amplitude of the pump is zero. Figure 2 shows how the characteristics of the spectrum change with changes in Ω. A new feature that we observe now is that the side peaks are split because of the competition between the Rabi flopping effects associated with the parts ε_0 and ε_1 of the pump. For still larger coherence time of the pump our calculations lead to a spectral profile that reflects essentially the coherence time of the incoherent part of the pump. This arises from the pole $z=-\Gamma$ in (11), which in the limit $\Gamma\rightarrow 0$ goes to the elastic component produced by the incoherent field. Note that we have already subtracted the elastic component produced by the pump. The splitting depends on the parameter $|x|$ which essentially is the Rabi frequency associated with the incoherent part of the pump. In fact, peaks occur roughly at the frequencies $\Omega\pm 2|x|$ corresponding to the two fields being in phase or out of phase. The splitting of the side band for the case of long correlation time for the incoherent pump is similar to a recent result[12] obtained while considering the resonance fluorescence in the field of two diffusing modes. This is because in the limit of large coherence time, the second pump also behaves almost like a coherent pump. The changes in the spectral line shapes can be understood in terms of the zeros of the polynomial $\hat{D}(z)$ which depend on the relative values of Γ and Ω for a fixed β.

We next demonstrate on general grounds how the external fields can modify the relaxation dynamics and how this modified dynamics leads to various line-narrowing effects. The modification of the relaxation dynamics is important even if Γ is large but $\Omega\gtrsim\Gamma$. This invalidates the applicability of Eq. (1). To understand this, consider a system with Hamiltonian H_S interacting with a heat bath (which is the source of relaxation) and an external field so that the total Hamiltonian H is equal to $H_S+H_{\text{ext}}(t)+H_R+H_{RS}$. The external interaction is supposed to be coherent in nature. We also assume it to be a resonant interaction. It is then possible to work in a rotating frame so that H becomes $h+H_R+H_{RS}(t)$, where the Hamiltonian h is static in nature. Let ρ_R be the equilibrium density matrix for the heat bath. The degree of freedom associated with the heat bath can be eliminated using the standard master-equation methods.[13] We treat the part h as the unperturbed Hamiltonain. Note that h includes the interaction with the external field and thus the interaction with the field is accounted to all orders. Born and Markov approximations with respect to the system–heat-bath interaction H_{RS} lead to the following master equation for the system:

$$\begin{aligned}&\frac{\partial\rho}{\partial t}+i[h,\rho]+\mathrm{Tr}_R\int_0^{\infty}d\tau[\tilde{H}_{RS}(t),[V(t,t-\tau)\tilde{H}_{RS}(t-\tau)V^{\dagger}(t,t-\tau),\rho_R(0)\rho(t)]]=0\ ,\\&\tilde{H}_{RS}(t)=e^{iH_Rt}H_{RS}(t)e^{-iH_Rt},\quad V(t,\tau)=e^{-ih(t-\tau)}\ .\end{aligned}\tag{12}$$

The usual relaxation dynamics (optical Bloch equations for a two-level system) is obtained if $V\sim 1$. This is possible if $\Omega\tau_c\ll 1$ where Ω is a typical frequency scale associated with h and τ_c is the correlation time for the heat bath. Note that the eigenstates of h are the so-called dressed states and the eigenvalues are the energies of the dressed states. Thus for a two-level system Ω will typically be the Rabi frequency (generalized to take into account the detuning effects). It is clear from (12) that relaxation dynamics will undergo considerable modification if $\Omega\tau_c\gtrsim 1$, as then the evolution of the system described by V, during τ_c, is important. In the context of the system studied in this paper the incoherent part of the pump acts like a stochastic heat bath and we have discussed how the spectrum changes in a dramatic way if $\Omega\tau_c \gtrsim 1$.

It should be evident from our discussion leading to (12) that the effects discussed in this paper occur rather generally. It is only for illustration purposes that we have considered a simple two-level optical transition. The effect reported here can be observed by examining the changes in the spectral profiles produced by a laser with controlled amplitude noise[14] when the intensity of the coherent part is increased substantially.

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