Asymmetric resonance fluorescence spectra in partially coherent fields

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(Received 21 September 1977)

A simple physical model is presented which explains the asymmetry recently predicted in the fluorescence spectra from a two-level atom driven slightly off resonance by a partially coherent field. The asymmetry is due to a continuing reinitiation of the transient response of the atom as its dipole moment tries to settle down to a steady-state phase relation with the fluctuating applied field. A Lorentz-model calculation based on this idea is able to give a qualitative explanation of the spectra at all field strengths, and a quantitative explanation of the low-intensity spectra. The predicted spectra are compared with those recently derived by Kimble and Mandel.

I. INTRODUCTION

Resonance fluorescence from a two-level atom interacting with a coherent radiation field has been the subject of a number of theoretical papers. The subject is now well understood.[1] Current theoretical activity is concentrating on the effects of field coherence on the fluorescence spectrum. A simple model to describe the role of phase and amplitude fluctuations in resonance fluorescence was introduced by Eberly[2] and used by Kimble and Mandel[3] to study the emission spectrum in the case of off-resonance excitation.

The steady-state resonance fluorescence spectrum from a two-level atom driven by a purely coherent laser field at frequency ω_L is symmetric even if the driving field is detuned by a large amount from the resonance frequency ω_0. Kimble and Mandel[3] (KM) have found that the radiated spectrum becomes markedly asymmetric with detuning if the field possesses a bandwidth described by a phase diffusion model. Their asymmetries persist even for low intensities or for large detunings where the Rabi frequency Ω is unimportant. They offer no physical explanation for these asymmetries. Similar asymmetries have been found by others considering the spectra produced by pulsed-laser excitation.[4] In a recent paper, Renaud, Whitley, and Stroud[5] (RWS) find asymmetries in the nonstationary emission spectrum induced by a completely coherent field. Their field is of finite extent and the atoms in an atomic beam have a transient response as they enter the exciting field. These asymmetries die out as the transients die out. Further, they also find that these transient asymmetries are most pronounced at low intensities where the linear Lorentz model approximation is valid. For short times there is not only the usual elastic response at the driving frequency ω_L, but also a sideband at ω_0. For large times the dipole settles down to a fixed phase relationship with the driving field and the elastic response approaches the expected δ-function spectrum. The asymmetry is due wholly to the transient response at ω_0. In this paper we show that the low-intensity KM asymmetries due to partially coherent excitation may be attributed to this transient response even though their spectrum is a steady-state spectrum. In their problem this transient response is continually reinforced by the random changes in the phase of the driving field. These random phase changes force the atom back into the transient regime and generate an asymmetric steady-state response at both ω_L and ω_0.

II. TRANSIENT RESPONSE AND EFFECT OF FIELD FLUCTUATIONS

The dynamics of the driven two-level atom is described by the operator Bloch equations.[6] The positive frequency part of the dipole moment operator $\hat{\sigma}_+(t)$ obeys the equation of motion

$$\dot{\hat{\sigma}}_+(t) = (i\omega_0 - \gamma_N)\hat{\sigma}_+(t) + i(d/\hbar)\hat{\mathcal{E}}^-(t)\hat{\sigma}_3(t)\,, \tag{1}$$

where γ_N is one-half the natural decay width, d is the transition dipole moment, and $\hat{\mathcal{E}}^-(t)$ is the negative frequency electric field operator. The basic physics of our discussion is already contained in the low-intensity limit where $\langle\hat{\sigma}_3(t)\rangle \simeq -1$. We assume the atom remains more or less in its ground state and linearize Eq. (1) to obtain the "operator Lorentz equation,"

$$\dot{\hat{\sigma}}_+(t) = (i\omega_0 - \gamma_N)\hat{\sigma}_+(t) - (id/\hbar)\hat{\mathcal{E}}^-(t)\,. \tag{2}$$

If the incident field is fully coherent and the atom

is prepared initially in its ground state then

$$\langle\hat{\sigma}_+(t)\rangle = \frac{\frac{1}{2}i\Omega}{i\Delta - \gamma_N}\left(e^{(i\omega_0-\gamma_N)t} - e^{i\omega_L t}\right), \tag{3}$$

where we assume the applied field satisfies

$$\langle\hat{\mathcal{E}}^-(t)\rangle \equiv \mathcal{E}^-(t) = \mathcal{E}_0^-(0)e^{i\omega_L t}, \tag{4}$$

the Rabi frequency is given by $\Omega \equiv 2d\left|\mathcal{E}_0^-\right|/\hbar$ and $\Delta = \omega_0 - \omega_L$. Equation (3) already demonstrates the existence of the transient response at ω_0 as well as the electric response at ω_L. In order to determine the spectrum of the scattered light the dipole-dipole correlation function $\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t+\tau)\rangle$ must be calculated. We find

$$\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t+\tau)\rangle = \frac{\frac{1}{4}\Omega^2}{\Delta^2+\gamma_N^2}\{\exp(-i\omega_L\tau) - \exp[i(\omega_L-\omega_0)t - i\omega_0\tau - \gamma_N(t+\tau)]$$
$$- \exp[-i(\omega_L-\omega_0)t - i\omega_L\tau - \gamma_N t] + \exp[-i\omega_0\tau - \gamma_N(t+\tau) - \gamma_N t]\}. \tag{5}$$

The component at ω_0, and the oscillatory terms due to the sharp turn-on of the field, produce just the transient asymmetries noted by RWS and explicitly shown in their Fig. 6. For $t \gg \gamma_N^{-1}$ we obtain the stationary form

$$\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t+\tau)\rangle = \frac{\frac{1}{4}\Omega^2}{\Delta^2+\gamma_N^2}e^{-i\omega_L\tau}, \quad t \gg \gamma_N^{-1}, \tag{6}$$

and the associated scattered field spectrum is

$$g_s(\omega) = \frac{\frac{1}{2}\pi\Omega^2}{\Delta^2+\gamma_N^2}\delta(\omega-\omega_L), \tag{7}$$

the usual Heitler result.[7]

To include the effects of laser-field fluctuations we will use the phase-diffusion model introduced into this problem by Eberly.[2] The electric field we will describe by a varying amplitude $\mathcal{E}_0(t)$ and phase $\phi(t)$

$$\mathcal{E}^-(t) = \mathcal{E}_0^-(t)e^{i\omega_L t + \phi(t)}, \tag{8}$$

where

$$\langle\langle e^{i[\phi(t)-\phi(t')]}\rangle\rangle = e^{-\gamma_\phi|t-t'|}, \tag{9a}$$
$$\langle\langle\mathcal{E}_0^-(t)\mathcal{E}_0^+(t+\tau)\rangle\rangle = \left|\mathcal{E}_0\right|^2 e^{-\gamma_A|t-t'|}. \tag{9b}$$

We put $\gamma_\phi + \gamma_A = \gamma_L$, the laser bandwidth, and then, straightforwardly, for $t \gg \gamma_N^{-1}$ we obtain the steady-state dipole correlation function

$$\langle\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t+\tau)\rangle\rangle = \frac{\Omega^2}{8\gamma_N}\left(\frac{1}{\gamma_N+\gamma_L+i\Delta} - \frac{1}{\gamma_N+\gamma_L+i\Delta}\right)e^{-(i\omega_0+\gamma_N)\tau}$$
$$+ \frac{\Omega^2 e^{(-i\omega_L-\gamma_L)\tau}}{(\gamma_N-\gamma_L+i\Delta)(\gamma_N+\gamma_L-i\Delta)}, \quad t \gg \gamma_N^{-1}. \tag{10}$$

The important point to note about Eq. (10) is that not only is there a response at ω_L, but there is an additional response at ω_0 even in the steady state. The term which oscillates at ω_L is damped at rate γ_L, and that at ω_0 is damped at rate γ_N. The fluorescence spectrum from Eq. (10) is

$$g_s(\omega) = \frac{\Omega\,\gamma_L/2}{[\Delta^2+(\gamma_N-\gamma_L)^2][\Delta^2+(\gamma_N+\gamma_L)^2]} \times\left(\frac{\Delta^2+2\Delta(\omega-\omega_L)+(\gamma_N^2-\gamma_L^2)}{(\omega-\omega_L)^2+\gamma_L^2} + \frac{\Delta^2-2\Delta(\omega-\omega_0)-(\gamma_N^2-\gamma_L^2)}{(\omega-\omega_0)^2+\gamma_N^2}\right). \tag{11}$$

For large Δ, and γ_L less than γ_N, $g_s(\omega)$ is made up of two resonance curves centered, respectively, at ω_L and ω_0. Figure 1 shows the spectrum for $\Omega = 0.1\gamma_N$, for fixed detuning $\Delta = +5\gamma_N$ and for increasing values of γ_L; all these values were chosen to correspond with those of KM's Fig. 2. The spectrum we have derived agrees with that derived by KM.

The generality of their treatment requires sophisticated mathematics rather than the intuitively appealing linear Lorentz model which provides more physical insight. In particular, we note the

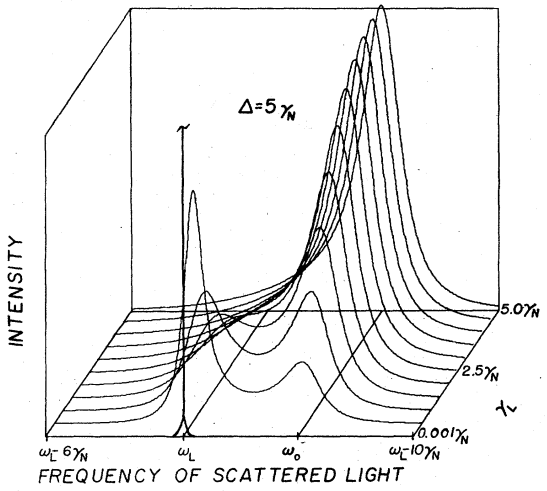

FIG. 1. Emission spectrum of a Lorentz oscillator in a fluctuating field [Eq. (11)]. The values of γ_L, the exciting field bandwidth, and the detuning Δ were chosen to correspond to Fig. 2 of Ref. 3. Provided $\Omega \ll \beta$ the Rabi rate only affects the scale. The sharp peak for $\gamma_L = 0.001\gamma_N$ has been truncated for clarity.

broadening of the elastic component at ω_0, and the marked growth of the inelastic component at the expense of the elastic part. As we predicted above, this spectrum quite closely resembles the transient spectral response of a weakly excited atom given by RWS. One difference is the absence of oscillations on the wings of the resonances in the present case; these occur in the case of transient response because the field is switched on and off suddenly, resulting in a sinc-function behavior which becomes a δ function as $t \to \infty$. In our case, the random phase changes in the incident radiation which reinforce the transient response have an exponential character described by Eq. (8) which suitably "apodizes" the response and eliminates the ringing oscillation.

On resonance ($\Delta = 0$) Eq. (10) simplified to

$$g_s(\omega, \Delta = 0) = \tfrac{1}{2}\Omega^2 \gamma_L / [(\alpha^2 + \gamma_L^2)(\alpha^2 + \gamma_N^2)], \qquad (12)$$

where $\alpha = \omega - \omega_0$. This is a single non-Lorentzian line with a width (FWHM) which tends to $2\gamma_N$ as expected for $\gamma_L \gg \gamma_N$. In the limit of narrow-band excitation with a weak field, the spectrum reduces to that of the excitation. This has been observed experimentally.[8]

III. COMMENTS

We have given a simple intuitive explanation of the asymmetries of the detuned fluorescence spectrum induced by a low-intensity partially coherent field predicted by KM. The emission spectrum is *not* the narrow-band response convolved with the spectral line shape of the incident field, but possesses structure due to the detailed atomic response over the incident frequency distribution. In this se se the spectral distribution we have discussed exhibits features familiar from Raman scattering as "hot luminescence,"[9] where the response at ω_L (the elastic, adiabatic, or Raman response) is supplemented by an additional nonadiabatic response at ω_0 due to the overlap of the excitation spectrum with the absorption line. There has been a great deal of interest in such processes where the incident field is monochromatic but where the radiation process is perturbed by phase-changing collisions.[9] The "fluctuation-induced fluorescence" we have described here has several similarities to such collision-induced fluorescence.

Our analysis applied only in the limit of weak excitation, while Kimble and Mandel's applies more generally for strong resonant saturating fields. However, it is just in the case of weak excitation $\Omega \ll \gamma_N$ or $\Omega \ll |\Delta|$ that the asymmetries are appreciable. The more complex theory must be used for a quantitative understanding of intermediate cases, but the physics underlying the asymmetry is still that demonstrated here. The asymmetries are caused by a continual reinitiation of the transient response of the system.

ACKNOWLEDGMENTS

This research was supported by the Energy Research and Development Administration and the Office of Naval Research. We would like to thank L. Allen, J.H. Eberly, and K. Wodkiewicz for useful discussion. P. L. Knight is grateful to J.H. Eberly for his support, and to Royal Holloway College for the award of a Jubilee Research Fellowship. W.A. Molander is grateful to the Hertz Foundation for fellowship support.

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