Resonance fluorescence in a weak radiation field with arbitrary spectral distribution

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The authors show that the laser bandwidth effects in resonance fluorescence at low intensities can be treated most readily in terms of simple convolutions, and further that this approach allows one to consider an incident field with an arbitrary spectral distribution, with or without the presence of atomic (line-broadening) collisions.

I. INTRODUCTION

In a recent paper by Knight, Molander, and Stroud[1] (KMS) an interesting pedagogical discussion was given of the effects of laser bandwidth on atomic resonance fluorescence spectra at low intensities. Assuming a Lorentzian-shaped laser spectrum (phase-diffusion model[2,3]) they showed that the finite bandwidth gives rise to an asymmetry in the scattered spectrum when the incident field is detuned from the atomic resonance. They offered a physical explanation for the asymmetry, first found by Kimble and Mandel,[4] in terms of a continuing reinitiation of the transient response of the atom owing to the fluctuations in the laser field. While this explanation is most certainly correct, one point remained unclear, that concerning the role of the convolution in this problem, which is a particular case of linear response theory.

The purpose of this comment is twofold: to show that the laser bandwidth effects in resonance fluorescence at low intensities can be treated most readily in terms of a simple convolution and to show further that this approach allows one to consider an incident field with an arbitrary spectral distribution, with or without the presence of atomic (line-broadening) collisions.

II. EFFECT OF LASER LINE SHAPE

For ease of comparison we will follow KMS and use, where possible, their notation. We start with the low-intensity operator Bloch equation for the positive-frequency dipole operator $\dot{\hat{\sigma}}_+$,

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

where ω_0 is the atomic resonance frequency, γ_N is one-half the natural decay width, d is the transition dipole moment, and $\hat{E}^-(t)$ is the negative-frequency electric field operator for the driving field, which oscillates with frequency ω_L. The scattered spectrum $g(\omega,\omega_L)$ is given in steady state by the Fourier transform of the dipole autocorrelation function $\langle\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t+\tau)\rangle\rangle$, which is obtained by formally solving Eq. (1). This yields

$$g(\omega,\omega_L) = \int_{-\infty}^{\infty} d\tau \exp(i\omega\tau)\frac{d^2}{\hbar^2}\int_{-\infty}^{t} dt' \int_{-\infty}^{t+\tau} dt'' \exp[(i\omega_0-\gamma_N)(t-t')]\exp[(-i\omega_0-\gamma_N)(t+\tau-t'')]\langle\langle\hat{E}^-(t')\hat{E}^+(t'')\rangle\rangle\,, \tag{2}$$

where ω is the frequency of the scattered light. The angular brackets $\langle\langle\;\rangle\rangle$ indicate both a quantum expectation value and an ensemble average over the statistics of the driving field.

For a monochromatic laser the field-correlation function is

$$\langle\langle\hat{E}^-(t')\hat{E}^+(t'')\rangle\rangle = |E_0|^2 \exp[i\omega_L(t'-t'')]\,, \tag{3}$$

giving for the narrow-band spectrum $g_{NB}(\omega,\omega_L)$

$$g_{NB}(\omega,\omega_L) = \int_{-\infty}^{\infty} d\tau \exp(i\omega\tau)\frac{d^2|E_0|^2}{\hbar^2}\int_{-\infty}^{t} dt' \int_{-\infty}^{t+\tau} dt'' \exp[(i\omega_0-\gamma_N)(t-t')]\exp[(-i\omega_0-\gamma_N)(t+\tau-t'')]\exp[i\omega_L(t'-t'')]\,. \tag{4}$$

This result can be evaluated to give

$$g_{NB}(\omega,\omega_L) = \frac{\pi}{2}\frac{\Omega^2}{(\omega_0-\omega_L)^2+\gamma_N^2}\delta(\omega-\omega_L)\,, \tag{5}$$

which shows that only elastic scattering occurs for an isolated atom suffering no collisions.

KMS treat the broad-band laser by assuming the phase-diffusion model[2,3] for the field

$$\langle\langle\hat{E}^-(t')\hat{E}^+(t'')\rangle\rangle = |E_0|^2 \exp[i\omega_L(t'-t'')]\exp(-\gamma_L|t'-t''|)\,, \tag{6}$$

where γ_L is one-half the laser bandwidth. Using this in Eq. (2) leads directly to the main result of KMS[5]:

$$g(\omega,\omega_L)=\frac{\Omega^2\gamma_L/2}{[\Delta^2+(\gamma_N-\gamma_L)^2][\Delta^2+(\gamma_N+\gamma_L)^2]}\times\left(\frac{\Delta^2+2\Delta(\omega-\omega_L)+(\gamma_N^2-\gamma_L^2)}{(\omega-\omega_L)^2+\gamma_L^2}+\frac{\Delta^2-2\Delta(\omega-\omega_0)-(\gamma_N^2-\gamma_L^2)}{(\omega-\omega_0)^2+\gamma_N^2}\right), \quad (7)$$

where the detuning is $\Delta=\omega_0-\omega_L$ and the Rabi frequency is $\Omega=2d|E_0|\hbar$. This spectrum clearly shows two maxima: elastic scattering centered at ω_L and fluorescence centered at ω_0. However, it also seems to contain interesting interference structure, e.g., $(\omega-\omega_L)$ in the numerator. It is stated by KMS that "the spectrum $[g(\omega,\omega_L)]$ is *not* the narrow-band response convolved with the spectral line shape of the incident field" (Lorentzian in this case). This statement is certainly correct for intense, saturating incident fields. However, we will show here that $g(\omega,\omega_L)$ in Eq. (7), valid for low fields, *is* obtainable by a simple convolution. This is as expected in linear response theory.

We will solve the problem in a different, more revealing, way. The power spectrum $P_L(\omega_L')$ of the incident field is generally given by[6]

$$P_L(\omega_L')=\frac{1}{2\pi}\int_{-\infty}^{\infty}d\tau\,\exp(i\omega_L'\tau)\,\langle\langle\hat{E}^-(t)\hat{E}^+(t+\tau)\rangle\rangle, \quad (8)$$

and the inverse can be written as

$$\langle\langle\hat{E}^-(t')\hat{E}^+(t'')\rangle\rangle=\int_{-\infty}^{\infty}d\omega_L'\exp[i\omega_L'(t'-t'')]P_L(\omega_L'). \quad (9)$$

Inserting this form for the correlation function into Eq. (2) for the spectrum $g(\omega,\omega_L)$, and comparing the resulting equation with the narrow-band spectrum $g_{NB}(\omega,\omega_L)$ in Eq. (4), it becomes apparent that the spectrum $g(\omega,\omega_L)$ can be written as

$$g(\omega,\omega_L)=|E_0|^{-2}\int_{-\infty}^{\infty}d\omega_L'g_{NB}(\omega,\omega_L')P_L(\omega_L'). \quad (10)$$

This is the main result of this paper and shows that, for an *arbitrary* laser spectrum $P_L(\omega_L')$, the scattered spectrum $g(\omega,\omega_L)$, at low intensity, is given by the narrow-band scattered spectrum $g_{NB}(\omega,\omega_L)$ convolved with the laser spectrum. Using the δ-function form of $g_{NB}(\omega,\omega_L)$ from Eq. (5) in Eq. (10) results in

$$g(\omega,\omega_L)=|E_0|^{-2}\frac{\pi}{2}\frac{\Omega^2}{(\omega-\omega_0)^2+\gamma_N^2}P_L(\omega), \quad (11)$$

which shows that the scattered light retains a distorted version of the incident light spectrum $P_L(\omega_L')$.

For the special case of the phase-diffusion model, used by KMS and described in Eq. (6), the laser spectrum becomes [by inserting Eq. (6) into Eq. (8)]

$$P_L(\omega_L')=|E_0|^2\frac{\gamma_L/\pi}{(\omega_L'-\omega_L)^2+\gamma_L^2}, \quad (12)$$

a Lorentzian, as stated earlier. This then gives, from Eq. (11), the scattered spectrum as

$$g(\omega,\omega_L)=\frac{\pi}{2}\frac{\Omega^2}{[(\omega-\omega_0)^2+\gamma_N^2]}\frac{\gamma_L/\pi}{[(\omega-\omega_L)^2+\gamma_L^2]}, \quad (13)$$

a result which is simple in form, but in fact (after some algebra) is identical to the more complex-looking expression given by KMS in Eq. (7). This shows that the scattered spectrum is the product of two Lorentzians, one peaked at the laser frequency ω_L and the other at the atomic resonance frequency ω_0.

III. COMMENTS

We have shown [Eq. (10)] that, for low-intensity resonance fluorescence in the absence of collisions, the scattered spectrum is given by that derived by assuming a monochromatic driving laser convolved with the spectrum of the actual driving laser. This result holds for *arbitrary* laser line shapes and, in particular, for the Lorentzian line shape derived from the phase-diffusion model[2,3] and used by KMS. The convolution is no longer valid when an intense, saturating driving laser is considered, as the response of the atom is then nonlinear.[4] The conclusion of KMS, that the inelastic component of the scattered light (centered on ω_0) is due to the continuing reinitiation of the transient response of the atom by fluctuations in the driving field, is strengthened by the present result. The conclusion holds regardless of the form of the power spectrum of the fluctuations.

As a further demonstration of the generality of the convolution result, we consider the effects of atomic line-broadening collisions on the scattered spectrum. These include dephasing (or elastic) collisions and quenching collisions, as well as alignment- and orientation-changing collisions. The effect of collisions has been studied in detail for the case of a low-intensity monochromatic driving field.[7-9] In particular, it is known that collisions redistribute the frequency of the scattered light and thus Eq. (5), for the scattered spectrum, is not appropriate in this case. However, denoting the appropriate narrow-band scattered spectrum[10] by $G_{NB}(\omega,\omega_L)$, the scattered spectrum $G(\omega,\omega_L)$ for broad-band excitation can be shown still to be a simple convolution

$$G(\omega,\omega_L)=|E_0|^{-2}\int_{-\infty}^{\infty}d\omega_L'G_{NB}(\omega,\omega_L')P_L(\omega_L'), \quad (14)$$

where again $P_L(\omega_L')$ is the (arbitrary) spectrum of the low-intensity driving field. This result can be verified by inspecting Eqs. (4) and (5) of Ref. 7, which give the scattered spectrum $G_{NB}(\omega, \omega_L)$ as an integral over several terms including the factor

$$I_1(\hat{\epsilon}_1^* \cdot \vec{\mu})(\hat{\epsilon}_1 \cdot \vec{\mu})\exp[-i\omega_L(t_1 - t_3)],$$

where I_1 is the intensity of the incident laser with polarization vector $\hat{\epsilon}_1$ and $\vec{\mu}$ is the atomic dipole operator. For an arbitrary driving field spectrum this factor should be replaced by

$$\langle\langle(\vec{E}_1^*(t_1) \cdot \vec{\mu})(\vec{E}_1(t_3) \cdot \vec{\mu})\rangle\rangle,$$

where $\vec{E}_1(t)$ is the electric field of the laser, and the field average has been performed. This field-correlation function is analogous to the one appearing in the integrand in Eq. (2) for $g(\omega, \omega_L)$, where collisions were ignored. Thus the derivation of $G(\omega, \omega_L)$ in Eq. (14) follows identically to that of $g(\omega, \omega_L)$ in Eq. (10). The convolution result Eq. (14) for the scattered spectrum $G(\omega, \omega_L)$ holds for arbitrary collisional line shapes, both inside and outside the impact region.

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