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Comment on absorption and fluorescence in strong frequency-modulated and amplitude-modulated fields

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It is shown that the interaction of frequency- and amplitude-modulated optical fields with a two-level atom, as treated by Nayak and Agarwal [Phys. Rev. A **31**, 3175 (1985)] and Agarwal and Nayak [J. Phys. B **19**, 3385 (1986)], respectively, can be described by the same Bloch equations, and that both have close counterparts in magnetic resonance. The matrix continued-fraction solution for the system is refined, and fluorescence and absorption spectra are explicitly related.

In two recent papers, to be referred to as I and II, respectively, Nayak and Agarwal[1] and Agarwal and Nayak[2] have treated the response of a strongly driven two-level atom to frequency-modulated (I) and amplitude-modulated (II) optical fields. Solutions of the Bloch equations are obtained in terms of two-dimensional matrix continued fractions, valid for an arbitrary index of modulation. In this Comment (Sec. I) we show that, by using a different choice of polarization components, both cases can be described by the same equations. The great similarities as well as the differences between the two cases are pointed out, and a comparison is made with magnetic resonance experiments. In Sec. II it is shown that the matrix continued-fraction solution in our alternative formulation takes a simpler form, especially for the amplitude-modulated (AM) case. In Sec. III some discussion on the resonance behavior of the system is given, and, finally, in Sec. IV, it is shown that the absorption and fluorescence spectra are intimately and simply related, a conclusion that has not been reached fully in the original work.

I. BLOCH EQUATIONS

Following largely the notation of I and II, let a two-level atom be subjected to a frequency-modulated (FM) or AM electric field

$$\mathbf{E}_{\mathrm{FM}}(t)=\mathcal{E}\exp(-i\omega_l t-iM\sin\Omega t)+\text{c.c.}\ , \tag{1}$$

$$\mathbf{E}_{\mathrm{AM}}(t)=\mathcal{E}(1+M\cos\Omega t)\exp(-i\omega_l t)+\text{c.c.}\ , \tag{2}$$

which is detuned from resonance by an amount $\Delta=\omega_l-\omega_0$ and with associated Rabi frequency $g=-\mathbf{d}\cdot\mathcal{E}/\hbar$. As is customary, the dynamics of the interaction are described in a frame rotating at the instantaneous frequency of the field, and counter-rotating terms are dropped. However, rather than working with the off-diagonal elements of the density matrix themselves, as was done in I and II, we follow Allen and Eberly[3] and use the real and imaginary parts U and V of the polarization, along with the atomic inversion W. Both interactions can now be represented by the semiclassical Bloch equations in the form

$$d\underline{\Phi}/dt=\begin{vmatrix}-\gamma_a & \omega_1 & 0\\ -\omega_1 & -\gamma_b & \omega_2+\omega_3\cos\Omega t\\ 0 & -\omega_2-\omega_3\cos\Omega t & -\gamma_c\end{vmatrix}\underline{\Phi}+\underline{C}\ . \tag{3}$$

In Table I the quantities from Eq. (3) are given explicitly for the two cases. From Eq. (3) it is evident that the FM case is remarkably similar to the AM case. The notable difference is the interchange of the roles of the real part

TABLE I. Quantities from Eq. (3). T_1 and T_2 are longitudinal and transverse relaxation times, W^0 is the equilibrium inversion scaled by T_1, and $\lambda_\parallel$ and $\lambda_\perp$ are longitudinal and transverse pumping rates, respectively.

	FM optical	AM optical	rf magnetic
$\underline{\Phi}$	$(W,V,U)^T$	$(U,V,W)^T$	$(U,V,W)^T$
$\omega_1;\omega_2;\omega_3$	$-2g;-\Delta;M\Omega$	$\Delta;2g;2gM$	$\omega_1;\omega_\parallel;\omega$
$\gamma_a;\gamma_b;\gamma_c$	$\frac{1}{T_1};\frac{1}{T_2};\frac{1}{T_2}$	$\frac{1}{T_2};\frac{1}{T_2};\frac{1}{T_1}$	$\frac{1}{T_2};\frac{1}{T_2};\frac{1}{T_1}$
$\underline{C}$	$(W^0,0,0)^T$	$(0,0,W^0)^T$	$\begin{cases}(0,0,\lambda_\parallel)^T\\(\lambda_\perp,0,0)^T\end{cases}$

of the polarization U and the atomic inversion W of the Bloch vector $\underline{\Phi}$, along with its associated damping terms and inhomogeneous term. Also, the roles of the Rabi frequency $2g$ of the carrier field and the detuning Δ (which, in the FM case, can be interpreted as the carrier *phase* of the field) are reversed. From a purely mathematical point of view, i.e., without regard to the roles of the atomic variables, the only difference between the FM and AM cases lies in the nonvanishing component of the inhomogeneous term $\underline{C}$, resulting in different resonance behavior of the system, as discussed below. Before addressing this issue, it should be recognized that Eq. (3) is a very familiar one in the study of magnetic resonance phenomena. Indeed, consider a magnetic dipole under the influence of a combination of static and oscillating magnetic rf fields

$$\mathbf{H}=H_{\perp}\hat{\mathbf{e}}_{\perp}-(H_{\parallel}+H\cos\Omega t)\hat{\mathbf{e}}_{\parallel}\ , \tag{4}$$

with associated Rabi frequencies $\omega_{\perp}$, $\omega_{\parallel}$, and ω. For this system, the Bloch equations (without making the rotating wave approximation) again take the form of Eq. (3), with the quantities given explicitly in Table I (see, e.g., Stenholm *et al.*[4,5]). Comparison shows that the roles of the atomic variables are as in the optical AM case, as noted by Thomann.[6] However, the inhomogeneous term in the rf case can be either as in the optical AM case for longitudinal pumping of the magnetic doublet, or as in the optical FM case for transverse pumping. Thus, from a mathematical point of view, *the optical FM interaction is identical in form to the rf case under conditions of transverse pumping.* The tremendous body of literature on magnetic resonance experiments, roughly from the period 1955 to 1975, is thus of great value to work in the optical regime for AM *as well as* FM interactions.

II. SOLUTION OF EQ. (3)

Most of the work on magnetic resonance experiments has dealt with special field geometries, e.g., $\omega_{\perp}=0$ (Ref. 7) or $\omega_{\parallel}=0$ (Refs. 8,9). For the latter, it was Stenholm[4] who first obtained (scalar) continued-fraction solutions following Floquet expansion of the Bloch vector,

$$\underline{\Phi}(t)=\sum_{n=-\infty}^{\infty}\underline{\Phi}^{(n)}\exp(-in\Omega t)\ . \tag{5}$$

The more general geometry has been investigated extensively by Yabuzaki *et al.*[10] and by Thomann.[6] Both reduced Eq. (3) to a single, infinite set of five-term recurrence relations for the Floquet coefficients of the atomic inversion W, and obtained the solution by solving a truncated pentadiagonal matrix equation. By comparison, the matrix continued-fraction solutions from I and II significantly reduce the computational effort in solving the problem. Here, we note that a further simplification results by applying the same procedure as in I and II to the Bloch equations in the form of Eq. (3). Upon Floquet expansion as in Eq. (5), and upon elimination of the coefficients $\Phi_1^{(n)}$, Eq. (3) becomes a two-dimensional infinite set of recurrence relations that can be written in matrix form as

$$\underline{A}_n\tilde{\underline{\Phi}}^{(n)}+\tfrac{1}{2}\omega_3(\tilde{\underline{\Phi}}^{(n-1)}+\tilde{\underline{\Phi}}^{(n+1)})=\tilde{\underline{C}}\delta_{n,0}\ , \tag{6a}$$

where

$$\tilde{\underline{\Phi}}^{(k)}=(\Phi_2^{(k)};\Phi_3^{(k)})^T,\quad \tilde{\underline{C}}=(C_3;C_1\omega_1/\gamma_a-C_2)^T\ , \tag{6b}$$

and

$$\underline{A}_n=\begin{bmatrix} \omega_2 & \gamma_c-in\Omega \\ -\gamma_b+in\Omega-\dfrac{\omega_1^2}{\gamma_a-in\Omega} & \omega_2 \end{bmatrix}\ . \tag{6c}$$

Solution of Eq. (6) follows very closely that of I. In particular, the matrix-continued fraction $\underline{X}_1$ has to be evaluated from the recurrence relation

$$\underline{X}_n=-[\underline{A}_n/(\tfrac{1}{2}\omega_3)+\underline{X}_{n+1}]^{-1},\quad n\geq 1\ . \tag{7}$$

For accurate results, the iteration should be started at a value of n no smaller than ω_3/Ω. Note that at each iteration only three components of the $\underline{X}_n$'s have to be computed, since the diagonal elements are equal. Unlike I and II, a similar continued fraction for negative indices is not needed, essentially because the atomic variables U, V, and W are all real. Also, the number of (matrix) multiplications and inversions at each step in the evaluation of the continued fraction is considerably less than for the solution in II for the AM case (for the FM solution in I there is not much difference). Once the $\underline{X}_n$'s have been obtained, the solution of Eq. (6) is given by

$$\tilde{\underline{\Phi}}^{(0)}=[\underline{A}_0+\omega_3\,\mathrm{Re}\{\underline{X}_1\}]^{-1}\tilde{\underline{C}}\ ; \tag{8}$$

$$\tilde{\underline{\Phi}}^{(n)}=\underline{X}_n\tilde{\underline{\Phi}}^{(n-1)},\quad \tilde{\underline{\Phi}}^{(-n)}=\tilde{\underline{\Phi}}^{(n)*},\quad n\geq 1 \tag{9}$$

and the coefficients $\Phi_1^{(n)}$ that had been eliminated from Eq. (3) are obtained as

$$\Phi_1^{(n)}=(C_1\delta_{n,0}+\omega_1\tilde{\Phi}_1^{(n)})/(\gamma_a-in\Omega)\ . \tag{10}$$

As a check, various figures in I and II were recalculated using the preceding equations. Apart from some scaling factors, perfect agreement was found, except for Fig. 3 of II and for the labeling of the curves in Fig. 2 of I.

III. RESONANCE BEHAVIOR

In I and II, the resonance condition

$$\omega_1^2+\omega_2^2=n^2\Omega^2 \tag{11}$$

is mentioned, corresponding to the vanishing of the determinant of $\underline{A}_n$ in the limit of zero damping. We would like to point out that this condition describes the resonant behavior accurately only in the limit of small ω_3, as is well established in the literature on magnetic resonance. In fact, it has been shown by Yabuzaki *et al.*[10] that the resonance behavior of Eq. (3), i.e., the generalized Bloch-Siegert shift, for arbitrary values of the parameters ω_1, ω_2, and ω_3 is quite complex. In particular, the circles described by Eq. (11) are modified by a pattern of crossings and anticrossings as ω_3 increases. Also, we note that although Eq. (11) is identical for the FM and AM interactions in terms of actual atomic variables, namely, $\Delta^2+4g^2=n^2\Omega^2$, there is an important difference: substituting the values for $\underline{C}$ from Table I, it is seen that

the inhomogeneous term $\bar{C}$ in Eq. (6b) has its nonvanishing component interchanged for the two cases. Thus, the roles of even and odd terms in the Floquet expansion are interchanged. From the literature on magnetic resonance phenomena, this implies an interchange of real and virtual transitions, characterized by odd and even (Haroche[11]) numbers of photons, respectively. Thus we note that the optical FM interaction with zero detuning would constitute an ideal experiment to study Haroche resonances in the optical regime, characterized by elliptical structures in the $(2g;M)$ plane, intersecting the axes at $2g$ equal to an even multiple of Ω, and M equal to a zero of an integer order Bessel function.[9]

IV. ABSORPTION AND FLUORESCENCE SPECTRA

Finally, we emphasize the close relation between fluorescence (in the sense of I and II) and absorption spectra. In I and II, the two are basically treated independently. However, it is clear from the pairs of Figs. 1 and 6, 2 and 9, 3 and 10, 4 and 7, and 5 and 8 in I, and of Figs. 4 and 8, 5 and 10, and 6 and 9 in II, that the two spectra are intimately related, in fact almost identical upon interchange of in-phase and quadrature components. Indeed, from the expression of work done by the fields and from the Bloch equation for the rate of change of the atomic inversion, it is easy to show that for a two-level atom interacting with an *arbitrary* field, the absorption spectrum $\mathcal{P}_a(\omega)$ and fluorescence spectrum $\mathcal{P}_f(\omega)$ are simply related by

$$\mathcal{P}_a(\omega)=(1-i\omega T_1)\mathcal{P}_f(\omega) , \tag{12}$$

where T_1 is the longitudinal relaxation time. For zero frequency, this expresses of course the time-averaged energy balance of the system. For frequencies other than zero it is seen that the magnitude of the absorption spectrum is always greater than that of the fluorescence spectrum, particularly if the longitudinal damping is small.

Let us now denote the rate of fluorescence for both the FM and AM cases by the Floquet series

$$dW_f/dt=(\hbar\omega_l/T_1)\sum_{n=-\infty}^{\infty}\rho_{22}^{(n)}\exp(-in\Omega t) . \tag{13}$$

Also, let us assume that the upper-state population coefficients $\rho_{22}^{(n)}$ have been obtained from the continued-fraction solution. Application of Eq. (12) then yields the rate of absorption, in terms of the *same* coefficients,

$$dW_a/dt=(\hbar\omega_l/T_1)\sum_{n=-\infty}^{\infty}(1-in\Omega T_1)\rho_{22}^{(n)}\exp(-in\Omega t) . \tag{14}$$

This expression can also be obtained directly from the recurrence relations between the Floquet-expansion coefficients. Thus we observe that the relation between absorption and fluorescence is independent of the type of interaction (FM or AM), field strength, modulation index, detuning, and transverse relaxation rates. If the factor $n\Omega T_1$ significantly exceeds unity, which is the case for most of the figures in I and II except near the origins in Ω scans, then the corresponding spectral harmonics are 90° out of phase, and differ in magnitude by that factor. Although the relation between the two spectra in this limit is mentioned for the components at the driving frequency Ω at the conclusion of I for the FM case, we note here that it is quite generally valid. In particular, Eqs. (13) and (14) remain valid for arbitrarily modulated fields as long as both the amplitude and phase are periodic at Ω.

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