

Comment on “Resonance-fluorescence and absorption spectra of a two-level atom driven by a strong bichromatic field”

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We identify discrepancies between two published predictions of the resonance fluorescence from a two-level atom driven by a strong bichromatic field [J. Opt. Soc. Am. B **8**, 1163 (1991) and Phys. Rev. A **48**, 3092 (1993)]. These discrepancies are shown to arise from subtleties in extracting the time-averaged resonance fluorescence from the expansions used to solve the optical Bloch equations. We construct a solution to these equations that corrects the results of the latter reference and reconciles the two predictions.

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In studying the resonance fluorescence from a driven two-level atom, we discovered that two well-known theories [1,2] give somewhat different predictions of the atomic response to a strong bichromatic field. These discrepancies are very small for the examples given in Refs. [1] and [2] but are more prominent for stronger bichromatic fields; see Fig. 1. It was quite surprising to find these inconsistencies, since both formalisms use the optical Bloch equations to describe the atomic response and solve these equations in similar ways.

In this Comment, we analyze the expansions used to solve the optical Bloch equations for the resonance-fluorescence spectrum and develop a solution that facilitates extracting the time-averaged fluorescence from this formalism. We show that this solution gives predictions identical to those from Ref. [1] and allows us to identify the subtleties that led to the discrepancies in the results of Ref. [2]. References [1] and [2] have been cited extensively, and the results that we identify as being problematic have been used in subsequent research [3], so we believe that a resolution of these discrepancies is of general interest.

I. OPTICAL BLOCH EQUATIONS FOR BICHROMATIC EXCITATION

We first review the mathematical model from Ref. [2] of a two-level atom under bichromatic excitation. A two-level atom, with ground state $|g\rangle$ and excited state $|e\rangle$, is illuminated by a bichromatic electric field

$$\mathbf{E}(t) = [\mathbf{E}_1 e^{-i\omega_1 t} + \mathbf{E}_2 e^{-i\omega_2 t}] + \text{c.c.}, \quad (1.1)$$

with field frequencies ω_j and amplitudes \mathbf{E}_j . The atomic response to the bichromatic field is studied through the dynamics of the transition operators $S^+ = |e\rangle\langle g|$ and $S^- = |g\rangle\langle e|$ and the inversion operator $2S^z = |e\rangle\langle e| - |g\rangle\langle g|$. In a frame that rotates with the average field frequency $\omega_s = (\omega_1 + \omega_2)/2$, the expectation values of the transformed operators $\tilde{S}^- = S^- e^{i\omega_s t}$, $\tilde{S}^+ = S^+ e^{-i\omega_s t}$, and $\tilde{S}^z = S^z$ satisfy the optical Bloch equations

$$\frac{d\mathbf{X}}{dt} = \mathbf{A}(t)\mathbf{X}(t) + \mathbf{v}, \quad (1.2)$$

with Bloch vector

$$\mathbf{X}(t) = \begin{bmatrix} X_1(t) \\ X_2(t) \\ X_3(t) \end{bmatrix} = \begin{bmatrix} \langle \tilde{S}^-(t) \rangle \\ \langle \tilde{S}^+(t) \rangle \\ \langle \tilde{S}^z(t) \rangle \end{bmatrix}, \quad (1.3)$$

coupling matrix

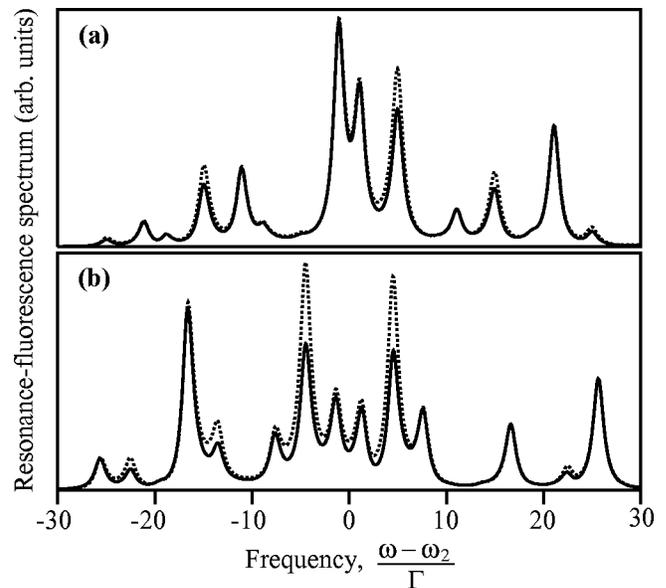


FIG. 1. Discrepancies in the predictions of the resonance-fluorescence spectrum, with (a) $\delta = 5\Gamma$, $\Delta = 0$, $\Omega_1 = 8\Gamma$, and $\Omega_2 = 13.6\Gamma$ [originally investigated in Ref. [2], Fig. 4(c)], and (b) $\delta = 9\Gamma/2$, $\Delta = -11\Gamma/2$, $\Omega_1 = 8\Gamma$, and $\Omega_2 = 16\Gamma$. Solid lines show the predictions from Ref. [1], and dashed lines show those from Ref. [2]. Note that Ref. [1] computes the inelastic fluorescence spectrum and Ref. [2] computes the total fluorescence spectrum; these spectra should agree everywhere except for frequencies $\omega = \omega_2 - 2l\delta$ (for integer l), which correspond to the locations of the spikes in the elastic spectrum. We have omitted these frequencies in these plots.

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$$\mathbf{A}(t) = \begin{bmatrix} -\frac{1}{2}\Gamma - i\Delta & 0 & \Omega_1 e^{i\delta t} + \Omega_2 e^{-i\delta t} \\ 0 & -\frac{1}{2}\Gamma + i\Delta & \Omega_1 e^{-i\delta t} + \Omega_2 e^{i\delta t} \\ -\frac{1}{2}(\Omega_1 e^{-i\delta t} + \Omega_2 e^{i\delta t}) & -\frac{1}{2}(\Omega_1 e^{i\delta t} + \Omega_2 e^{-i\delta t}) & -\Gamma \end{bmatrix}, \quad (1.4)$$

and decay term

$$\mathbf{v} = \begin{bmatrix} 0 \\ 0 \\ -\frac{1}{2}\Gamma \end{bmatrix}. \quad (1.5)$$

In these equations, $2\delta = (\omega_2 - \omega_1)$ is the frequency difference between the two applied fields, $\Delta = \omega_0 - \omega_s$ is the detuning between the atomic transition frequency ω_0 and the average field frequency, Γ is the Einstein A coefficient for spontaneous emission, and $\Omega_j = 2\boldsymbol{\mu} \cdot \mathbf{E}_j / \hbar$ are the Rabi frequencies of the applied fields (where $\boldsymbol{\mu}$ is the atomic transition electric dipole moment). Equation (1.2) assumes an electric dipole interaction between the atom and the field and makes use of the rotating-wave approximation.

II. SOLUTION OF THE BLOCH EQUATIONS

Since the elements of the coupling matrix $\mathbf{A}(t)$ and the decay term \mathbf{v} in the optical Bloch equations (1.2) are either time independent or periodic in time with period $2\pi/\delta$, in the long-time limit, the atomic expectation values will respond at the harmonics of frequency δ . Thus, the optical Bloch equations in this limit are solved by a Floquet expansion,

$$\mathbf{X}(t) = \sum_{l=-\infty}^{\infty} \mathbf{X}^{(l)} e^{il\delta t}. \quad (2.1)$$

The expansion coefficients $\mathbf{X}^{(l)}$ are needed to determine the resonance-fluorescence spectrum, so we provide a prescription for calculating them here. By inserting Eq. (2.1) into Eq. (1.2), it has been shown [1,2] that the inversion components obey a three-term recurrence relation,

$$a_l X_3^{(l)} + b_l X_3^{(l-2)} + d_l X_3^{(l+2)} = g_l, \quad (2.2)$$

where

$$a_l = (il\delta + \Gamma) + \frac{1}{2}\Omega_1^2 \left[\frac{1}{Q_{l-1}} + \frac{1}{P_{l+1}} \right] + \frac{1}{2}\Omega_2^2 \left[\frac{1}{P_{l-1}} + \frac{1}{Q_{l+1}} \right], \quad (2.3a)$$

$$b_l = \frac{1}{2}\Omega_1\Omega_2 \left[\frac{1}{P_{l-1}} + \frac{1}{Q_{l-1}} \right], \quad (2.3b)$$

$$d_l = \frac{1}{2}\Omega_1\Omega_2 \left[\frac{1}{P_{l+1}} + \frac{1}{Q_{l+1}} \right], \quad (2.3c)$$

and

$$g_l = -\frac{1}{2}\Gamma \delta_{l,0}, \quad (2.3d)$$

where we have defined

$$P_l = il\delta + \frac{1}{2}\Gamma + i\Delta \quad (2.4a)$$

and

$$Q_l = il\delta + \frac{1}{2}\Gamma - i\Delta. \quad (2.4b)$$

The coefficients $X_3^{(l)}$ can be found numerically from this recurrence relation using continued-fraction [1] or matrix [2] methods. Then, the transition-operator components are given by

$$X_1^{(l)} = \frac{1}{P_l} [\Omega_1 X_3^{(l-1)} + \Omega_2 X_3^{(l+1)}] \quad (2.5a)$$

and

$$X_2^{(l)} = \frac{1}{Q_l} [\Omega_1 X_3^{(l+1)} + \Omega_2 X_3^{(l-1)}]. \quad (2.5b)$$

III. RESONANCE-FLUORESCENCE SPECTRUM

The resonance-fluorescence spectrum [2] is given by

$$S(t, \omega) = \Gamma u(\hat{\mathbf{r}}) \operatorname{Re} \int_0^\infty d\tau \langle S^+(t) S^-(t+\tau) \rangle e^{i\omega\tau}, \quad (3.1)$$

where $u(\hat{\mathbf{r}}) = (3/8\pi) \sin^2\theta$, with θ the angle between the observation direction $\hat{\mathbf{r}}$ and the atomic transition dipole moment $\boldsymbol{\mu}$. The correlation function in the integrand of Eq. (3.1) is computed by introducing a two-time correlation vector $\mathbf{Y}(t, \tau)$, with components

$$Y_1(t, \tau) = \langle \tilde{S}^+(t) \tilde{S}^-(t + \tau) \rangle, \quad (3.2a)$$

$$Y_2(t, \tau) = \langle \tilde{S}^+(t) \tilde{S}^+(t + \tau) \rangle, \quad (3.2b)$$

and

$$Y_3(t, \tau) = \langle \tilde{S}^+(t) \tilde{S}^z(t + \tau) \rangle. \quad (3.2c)$$

Applying the quantum regression theorem [4] to the optical Bloch equations, this vector is found to satisfy the partial differential equation

$$\frac{\partial \mathbf{Y}}{\partial \tau} = \mathbf{A}(t + \tau) \mathbf{Y}(t, \tau) + X_2(t) \mathbf{v}. \quad (3.3)$$

The subtleties in solving this equation arise from the fact that the coupling matrix depends on the final time ($t + \tau$),

whereas the decay term depends on the initial time t , of the two-time correlation. To further intertwine the role of these two time quantities, note that the differential equation (3.3) involves a derivative with respect to the time difference τ , but this equation is used to determine the dc resonance fluorescence spectrum by averaging Eq. (3.1) over the time t for a period $2\pi/\delta$.

IV. EXPANSION OF THE CORRELATION VECTOR

The differential equation (3.3) can be solved by an expansion of the form

$$\mathbf{Y}(t, \tau) = \sum_{l=-\infty}^{\infty} \mathbf{Y}^{(l)}(t, \tau) e^{il\delta(t+\tau)}. \quad (4.1)$$

Inserting this expansion into Eq. (3.3), we find that

$$\begin{aligned} \sum_{l=-\infty}^{\infty} \frac{\partial \mathbf{Y}_3^{(l)}}{\partial \tau} e^{il\delta(t+\tau)} = & -\frac{1}{2} \Gamma X_2(t) - \sum_{l=-\infty}^{\infty} (\Gamma + il\delta) Y_3^{(l)} e^{il\delta(t+\tau)} \\ & - \frac{1}{2} \sum_{l=-\infty}^{\infty} [\Omega_1 Y_1^{(l+1)} + \Omega_2 Y_1^{(l-1)} + \Omega_1 Y_2^{(l-1)} + \Omega_2 Y_2^{(l+1)}] e^{il\delta(t+\tau)}, \end{aligned} \quad (4.2)$$

where we show only the third component of the vector expansion for brevity. Equation (4.1) expresses an unknown function $\mathbf{Y}(t, \tau)$ in terms of a set of functions $\{\mathbf{Y}^{(l)}(t, \tau)\}$, and there are an infinite number of ways to partition the unknown function in this way. The expansion functions $\mathbf{Y}^{(l)}(t, \tau)$ are only specified uniquely when additional constraints are imposed on them; once such constraints have been stipulated, we can extract differential equations that are satisfied by the functions $\mathbf{Y}^{(l)}(t, \tau)$ individually from the infinite sums in Eq. (4.2).

Equation (4.1) is reminiscent of an amplitude-modulation expansion, since the function $\mathbf{Y}(t, \tau)$ is decomposed into a series of carrier signals $e^{il\delta(t+\tau)}$ with varying amplitudes $\mathbf{Y}^{(l)}(t, \tau)$. Indeed, the constraints we impose on the expansion functions $\mathbf{Y}^{(l)}(t, \tau)$ involve their frequency-domain properties. However, it is crucial to emphasize that there are two distinct frequency domains for the function $\mathbf{Y}(t, \tau)$: a “ τ -frequency” domain, suggested by Eq. (3.1), that is defined by a Fourier transform with respect to the time delay τ for fixed time t , and a “ t -frequency domain,” analogously defined by a Fourier transform with respect to the time t for fixed time delay τ .

V. THE SOURCE OF THE DISCREPANCIES: THE τ -FREQUENCY PARTITION

The results of Ref. [2] can be obtained by partitioning the function $\mathbf{Y}(t, \tau)$ in the τ -frequency space. For fixed time t , we demand that the bandwidth of $\mathbf{Y}^{(l)}(t, \tau)$ (defined by a Fourier transform with respect to τ) lies within a frequency window of width δ around the origin for all l . This constraint is equivalent to requiring that each term in Eq. (4.1) covers a

distinct (nonoverlapping) range of the “ τ -frequency” space. In this way, we can partition Eq. (4.2) by these frequency ranges, grouping terms according to the closest carrier frequency $e^{il\delta\tau}$, to find that

$$\begin{aligned} \frac{\partial \mathbf{Y}_3^{(l)}}{\partial \tau} = & -\frac{1}{2} \Gamma X_2(t) \delta_{l,0} - (\Gamma + il\delta) Y_3^{(l)} - \frac{1}{2} [\Omega_1 Y_1^{(l+1)} \\ & + \Omega_2 Y_1^{(l-1)} + \Omega_1 Y_2^{(l-1)} + \Omega_2 Y_2^{(l+1)}]. \end{aligned} \quad (5.1)$$

Recall that the quantity we wish to calculate is the *time-averaged* resonance fluorescence. Although we have constrained the functions $\mathbf{Y}^{(l)}$ according to their τ -frequency ranges, the presence of the raising operator $X_2(t)$ in Eq. (5.1) shows that the functions $\mathbf{Y}^{(l)}$ will not necessarily have a narrow t -frequency range. That is, since Eq. (2.1) establishes that the mean raising operator responds at all harmonics of the frequency δ , we see that the information about the time-averaged resonance fluorescence is distributed over the Fourier transforms of the full set of functions $Y_1^{(l)}$ and can only be extracted by averaging this full set of functions over the time period $2\pi/\delta$. Instead, in Ref. [2], the time-averaged fluorescence was attributed only to the transform of the function $Y_1^{(0)}(t, \tau)$, and this is the source of the discrepancies noted in our Comment.

VI. THE t -FREQUENCY PARTITION

The function $Y_j(t, \tau)$ can also be partitioned based on the frequencies with respect to the time t for fixed delay τ . With this constraint, the differential equation (4.1) is grouped ac-

ording to the nearest carrier frequency $e^{il\delta t}$, which results in the differential equations

$$\frac{\partial}{\partial \tau} Y_1^{(l)} = - \left(\frac{1}{2} \Gamma + il\delta + i\Delta \right) Y_1^{(l)} + \Omega_1 Y_3^{(l-1)} + \Omega_2 Y_3^{(l+1)}, \quad (6.1a)$$

$$\frac{\partial}{\partial \tau} Y_2^{(l)} = - \left(\frac{1}{2} \Gamma + il\delta - i\Delta \right) Y_2^{(l)} + \Omega_1 Y_3^{(l+1)} + \Omega_2 Y_3^{(l-1)}, \quad (6.1b)$$

$$\begin{aligned} \frac{\partial}{\partial \tau} Y_3^{(l)} = & - \frac{1}{2} \Gamma X_2^{(l)} e^{-il\delta\tau} - (\Gamma + il\delta) Y_3^{(l)} \\ & - \frac{1}{2} [\Omega_1 Y_1^{(l+1)} + \Omega_2 Y_1^{(l-1)} \\ & + \Omega_1 Y_2^{(l-1)} + \Omega_2 Y_2^{(l+1)}], \end{aligned} \quad (6.1c)$$

where we have assumed the long-time limit of the raising operator $X_2(t)$ given by Eq. (2.1). This system of equations is ideally suited for our goal of computing the time-averaged fluorescence, because the zero frequency (that is, time-averaged) component of $Y_1(t, \tau)$ has been placed solely in the $l=0$ component by the constraints chosen for this partitioning scheme.

We solve this system with Fourier transform methods, using the convention that a function $f(\tau)$ is related to its one-sided Fourier transform $\hat{f}(\omega)$ by

$$\hat{f}(\omega) = \int_0^\infty f(\tau) e^{i(\omega - \omega_s)\tau} d\tau. \quad (6.2)$$

Here, the frequency ω is offset by the field frequency ω_s because our starting equations describe dynamics in this rotating frame. Applying this transformation to Eqs. (6.1), we find that the components of \hat{Y}_3 obey a three-term recurrence relation,

$$A_l \hat{Y}_3^{(l)}(t, \omega) + B_l \hat{Y}_3^{(l-2)}(t, \omega) + D_l \hat{Y}_3^{(l+2)}(t, \omega) = G_l, \quad (6.3)$$

where

$$\begin{aligned} A_l = & -i(\omega - \omega_s) + \Gamma + il\delta + \frac{1}{2} \Omega_1^2 \left[\frac{1}{P_{l+1}(\omega)} + \frac{1}{Q_{l-1}(\omega)} \right] \\ & + \frac{1}{2} \Omega_2^2 \left[\frac{1}{P_{l-1}(\omega)} + \frac{1}{Q_{l+1}(\omega)} \right], \end{aligned} \quad (6.4a)$$

$$B_l = \frac{1}{2} \Omega_1 \Omega_2 \left[\frac{1}{P_{l-1}(\omega)} + \frac{1}{Q_{l-1}(\omega)} \right], \quad (6.4b)$$

$$D_l = \frac{1}{2} \Omega_1 \Omega_2 \left[\frac{1}{P_{l+1}(\omega)} + \frac{1}{Q_{l+1}(\omega)} \right], \quad (6.4c)$$

and

$$\begin{aligned} G_l = & Y_3^{(l)}(t, \tau=0) - \frac{\Gamma}{2} X_2^{(l)} \left\{ \pi \delta[\omega - \omega_s - l\delta] \right. \\ & \left. - \frac{1}{i(\omega - \omega_s - l\delta)} \right\} - \frac{1}{2} \left[\frac{\Omega_2}{P_{l-1}(\omega)} Y_1^{(l-1)}(t, \tau=0) \right. \\ & \left. + \frac{\Omega_1}{P_{l+1}(\omega)} Y_1^{(l+1)}(t, \tau=0) \right], \end{aligned} \quad (6.4d)$$

where we have defined

$$P_l(\omega) = -i(\omega - \omega_s) + il\delta + \frac{1}{2} \Gamma + i\Delta \quad (6.5a)$$

and

$$Q_l(\omega) = -i(\omega - \omega_s) + il\delta + \frac{1}{2} \Gamma - i\Delta. \quad (6.5b)$$

The initial conditions are found to be

$$Y_1^{(l)}(t, \tau=0) = \frac{1}{2} \delta_{l,0} + X_3^{(l)}, \quad (6.6a)$$

$$Y_2^{(l)}(t, \tau=0) = 0, \quad \text{and} \quad (6.6b)$$

$$Y_3^{(l)}(t, \tau=0) = -\frac{1}{2} X_2^{(l)}. \quad (6.6c)$$

Note that the recurrence relation described by Eqs. (6.3)–(6.5) and the initial conditions in Eqs. (6.6) are, in fact, independent of the time t . This fortuitous result tells us that the time-averaged resonance fluorescence is simply proportional to the quantity $\text{Re } \hat{Y}_1^{(0)}(t, \omega)$, without the need to average this function over time explicitly.

The coefficients $\hat{Y}_3^{(l)}(t, \omega)$ can be found numerically from this recurrence relation using continued-fraction or matrix methods. The components of \hat{Y}_1 and \hat{Y}_2 are then given by

$$\begin{aligned} \hat{Y}_1^{(l)}(t, \omega) = & \frac{1}{P_l(\omega)} [Y_1^{(l)}(\tau=0) \\ & + \Omega_1 \hat{Y}_3^{(l-1)}(t, \omega) + \Omega_2 \hat{Y}_3^{(l+1)}(t, \omega)] \end{aligned} \quad (6.7a)$$

and

$$\hat{Y}_2^{(l)}(t, \omega) = \frac{1}{Q_l(\omega)} [\Omega_1 \hat{Y}_3^{(l+1)}(t, \omega) + \Omega_2 \hat{Y}_3^{(l-1)}(t, \omega)], \quad (6.7b)$$

so the time-averaged (dc) resonance-fluorescence spectrum $\overline{S}(t, \omega) \propto \text{Re } \hat{Y}_1^{(0)}(t, \omega)$ can be determined with Eq. (6.7a), thereby completing our solution.

Note that in previous work [1,2], systems of equations similar to Eqs. (6.1) were solved using Laplace transforms (using the integration kernel $e^{-s\tau}$, with $\text{Re } s > 0$), then the resulting expressions were evaluated for purely imaginary values of the transform variable s . On one hand, such a procedure misses the Dirac δ function term that we report in our expression for G_l , above. On the other hand, this procedure is consistent with calculating the physical spectrum $S(t, \omega, \Gamma)$, as defined by Eberly and Wodkiewicz [5], in the limit of narrow detector spectral width Γ . We prefer to apply the transform defined in Eq. (6.2) and convolve the predicted resonance-fluorescence spectrum with the detector response separately.

In the end, our prescription for computing the resonance fluorescence, Eqs. (6.3)–(6.7), is quite similar to that in Ref. [2], differing only in the form of the coefficient G_l . Specifically, our expression (6.4d) involves a quantity proportional to $\Gamma X_2^{(l)}/[2i(\omega - \omega_s - l\delta)]$, whereas in the cited reference it appears (in the present notation) as proportional to $\Gamma X_2^{(0)}\delta_{l,0}/[2i(\omega - \omega_s)]$. Since the Floquet expansion for $X_2(t)$ is dominated by the $l=0$ term for the examples given in Refs. [1] and [2], this justifies our initial observation that the discrepancies are typically small.

VII. RECONCILIATION AND CONCLUSION

Reference [1] computes only the so-called inelastic component of the resonance-fluorescence spectrum. The two-time correlation Bloch vector pertinent to the inelastic spectrum [see Ref. [1], Eq. (2)] satisfies a partial differential equation that lacks the decay term \mathbf{v} . In such situations, partitioning the expansion (4.1) either by the “ τ frequencies” or the “ t frequencies” leads to the same final prescription for determining the fluorescence spectrum. So the results of Ref. [1] are unaffected by the subtleties in solving the optical Bloch equations described here.

Finally, we have confirmed that our solution of the full resonance-fluorescence spectrum, Eqs. (6.3)–(6.7), agrees with the prescription from Ref. [1], Eqs. (5)–(9), in all cases given as examples in Refs. [1] and [2], to within the precision of the computer calculations. Thus we have reconciled the discrepancies in the two predictions of the resonance-fluorescence spectrum.

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