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Spectrum of resonance fluorescence from a two-level atom interacting with 100% amplitude-modulated intense radiation

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We give analytical results for the fluorescence spectrum of a two-level atom interacting with fully amplitude-modulated field at resonance. The results are valid for unequal longitudinal and transverse relaxation rates and follow the experimentally observed pattern. The intensity-dependent changes in the peak widths and peak heights are presented. Novel transient effects are predicted.

Interaction of a two-level atom (TLA) with a 100% (fully) amplitude-modulated (FAM) field is of current interest and importance.[1,2] The importance of this interaction lies in the suspected role it plays in the dynamics of certain optically pumped dye lasers.[3] Far from explaining the complex dynamics of these dye lasers, the purpose of this Rapid Communication is to discuss the fundamentally important spectral response of a TLA interacting with a FAM field. We give the first analytical results for the observed[2] time-averaged portion of the time-dependent spectrum from a TLA interacting with a FAM field. Our results are worked out for a resonant situation, with $T_1 \neq T_2$ and $\Omega > T_1^{-1}, T_2^{-1}$, where T_1^{-1} and T_2^{-1} are the longitudinal and transverse relaxation rates and Ω is the modulation frequency.

The spectrum shows several peaks at the multiples of the modulation frequency Ω. Strikingly, there are no Rabi sidebands.[4] The intensity-dependent changes in the peak heights of this multipeaked spectrum are shown to follow a pattern similar to the one observed recently by Zhu, Lezama, Wu, and Mossberg.[2] The intensity-dependent changes in the widths and heights of the peaks at odd and even multiples of Ω are demonstrated. Novel transient effects are predicted and modifications in the spectrum with respect to the known cases are discussed.

Consider a TLA with eigenstates $|1\rangle$ and $|2\rangle$ having energy eigenvalues E_1 and E_2 ($\hbar\omega_0 = E_1 - E_2$), respectively, and interacting with a FAM field $E(t) = \varepsilon\cos(\Omega t + \phi_\Omega)\cos(\omega_L t + \phi_L)$, where $\Omega(\omega_L)$, $\phi_\Omega(\phi_L)$ are, respectively, the frequency of modulation (carrier wave) and arbitrary initial phase of the modulation (carrier) vibration of the radiation. The mean values of the atomic variables, viz. $\langle S^\pm\rangle, \langle S^z\rangle$, in the frame rotating at the carrier frequency ω_L, and in the rotating wave approximation obey the equations

$$\langle \dot{S}^\pm\rangle = \mp i\Delta\langle S^\pm\rangle + i a e^{\mp i\phi_L}\cos(\Omega t + \phi_\Omega)\langle S^z\rangle - T_2^{-1}\langle S^\pm\rangle , \qquad (1)$$

$$\langle \dot{S}^z\rangle = [i\tfrac{1}{2} a e^{i\phi_L}\cos(\Omega t + \phi_\Omega)\langle S^+\rangle + \text{c.c.}] - T_1^{-1}(\langle S^z\rangle - \langle S^z_{\text{eq}}\rangle) , \qquad (2)$$

$$\langle S^+\rangle = e^{-i\omega_L t}\text{Tr}(\rho S^+), \quad a = 2|d_{12}\varepsilon e^{i\phi_L}| . \qquad (3)$$

S^+, S^-, and S^z are the atomic raising, lowering, and inversion operators of TLA, Δ is the detuning, $\omega_L - \omega_0$, d_{12} is the dipole matrix element between the states $|1\rangle$ and $|2\rangle$, ρ is the density matrix of the TLA, $\langle S^z_{\text{eq}}\rangle$ is the value of atomic inversion to which the system relaxes in the absence of any excitation, $\chi(t)$ $[= a\cos(\Omega t + \phi_\Omega)]$ is called the instantaneous Rabi frequency, a is a constant which we prefer to use as the Rabi frequency of modulation, and ε is the constant amplitude of the FAM field. It is assumed that the two phases ϕ_Ω and ϕ_L can be controlled experimentally. The control on ϕ_L is used to demonstrate novel transient effects.

Equations (1) and (2) have been solved exactly for $\Delta = 0$ and $T_1 = T_2$.[5] The improved steady-state solutions for $T_1 \neq T_2$ and $\Delta = 0$ have been obtained by Ruyten[6] using an iterative procedure on the parameter $\gamma_- = (T_1^{-1} - T_2^{-1})/2$. Using the same procedure and dropping terms of the order of and higher powers in $\gamma_\pm/\Omega$, for $\Omega \gg \gamma_\pm$, with $\gamma_+ = (T_1^{-1} + T_2^{-1})/2$, we have obtained solutions which are valid in the transient domain as well. These have been used to calculate the spectrum of the emitted fluorescence, by first determining the two-time correlation function $\langle S^+(\tau + t)S^-(t)\rangle$ using the quantum-mechanical regression theorem in a well-known way,[7] and then using this correlation function in the Eberly-Wodkiewicz definition for the physical spectrum.[8] We get for the time-averaged spectrum, for $\phi_\Omega = \phi_L = 0$, the following expression,

$$\bar{S}(\infty,\omega,\Gamma) = \frac{C_{\text{inc}}}{(\frac{1}{2}\Gamma + \frac{1}{2}\gamma)^2 + D^2} + \frac{C^R_{\text{inc}}}{(\frac{1}{2}\Gamma + \gamma_I)^2 + D^2} + \sum_{n=1}^{\infty}\left[\left(\frac{S^{(n)}_{\text{coh}}}{(\frac{1}{2}\Gamma)^2 + (D - n\Omega)^2} + \frac{S^{(n)}_{\text{inc}}}{(\frac{1}{2}\Gamma + \gamma_I)^2 + (D - n\Omega)^2}\right) + (n \rightarrow -n)\right] , \qquad (4)$$

$$D = \omega - \omega_L, \quad \gamma_I = \gamma_+ + \gamma_- J_0(2z), \quad z = a/\Omega, \quad \gamma = T^{-1} . \qquad (5)$$

Γ and ω are the width and the central frequency of the passband of the Fabry-Pérot cavity used to collect the data in the measurement of the physical spectrum, D is the detuning of the Fabry-Pérot with respect to the laser frequency ω_L at $\Delta=0$. $J_n(z)$ is the Bessel function of order n. The weight functions $C_{\rm inc}$ and $C_{\rm inc}^R$ of the central peak and $S_{\rm coh}^{(n)}$ and $S_{\rm inc}^{(n)}$ of the nth side peak are functions of Γ, γ, γ_I, and $J_n(z)$. These weight functions are responsible for the varying shapes of the peaks. Their explicit forms are discussed later.

Equation (4) differs from the Mollow triplet[4] of resonance fluorescence from a TLA interacting with a constant amplitude resonant intense radiation. Equation (4) does not predict the three peaks of the Mollow triplet, but several peaks at the multiples of the modulation frequency Ω, i.e., $D=\pm n\Omega$. The peak heights and the peak widths are intensity dependent while the peak positions, for a constant modulation frequency Ω, are fixed. This is in contrast to the intensity-dependent positions of the Rabi sidebands of the Mollow triplet,[4] which have their widths and heights independent of the intensity—at high intensities.

Note that the spectral response of an atom is the direct evidence of the energy-level separations existing in the atom. Thus the modified energy-level separations, for a TLA interacting with an FAM field, occur at the integral multiples of the modulation frequency Ω. Such energy-level separations indicate the existence of the Floquet energy eigenvalues of the periodic Hamiltonian with period $2\pi/\Omega$, in the interaction of the FAM field with a TLA.[9]

The recently reported experimental observations[2] for the spectrum of resonance fluorescence from TLA interacting with the FAM field are thus a direct experimental demonstration of the Floquet energy eigenvalues in this system. The observed spectrum available to us shows that peaks alternate in their heights and as well in their widths in that the peaks at odd multiples of the modulation frequency are higher and narrower. Both features are explained by the weight functions of the side peaks, given by

$$S_{\rm coh}^{(n)}=J_n^2(z)J_0^2(z)\gamma^2\Gamma[1-(-1)^n](8\gamma_I^2)^{-1}\,, \qquad (6)$$

$$S_{\rm inc}^{(n)}=J_n^2(z)(\Gamma+2\gamma_I)\{2\gamma_I^2-J_0^2(z)\gamma^2 \times[1-(-1)^n]\}(8\gamma_I^2)^{-1}\,, \qquad (7)$$

which show that (i) the peak heights are proportional to $J_n^2(z)$, (ii) the heights of two consecutive odd-even peaks satisfy (with m as an integer) the relation

$$J_{2m+2}^2(z)\Gamma\gamma_I P_{2m+1} = J_{2m+1}^2(z)[\Gamma\gamma_I+2\gamma^2J_0^2(z)]P_{2m+2}\,, \qquad (8)$$

P_n being the height of the nth peak, and (iii) the full width at half maximum (FWHM) for all even peaks are $(\Gamma+2\gamma_I)$, for all odd peaks the FWHM are given by the solution of a quadratic equation (because each odd peak is made up of two Lorentzians, one with a coherent width $\Gamma/2$, and the second with an incoherent intensity-dependent width $\Gamma/2+\gamma_I$). In Fig. 1 the FWHM of the odd and even peaks are plotted for different values of z. Clearly all odd peaks are narrower or equal in width to the even peaks for the values of z shown in Fig. 1. The results of Eq. (4) are plotted in Fig. 2, at five different values of z. The curve at $z=4.6$ corresponds closely to the available parameters of the reported experiments. The agreement between the theory and the experiment is fair. Anomalous features in the spectrum are predicted at different values of z. For example, the curves at $z=3.7$, 5.0, and 6.2 do not show, respectively, the peaks at $n=1$, 2, and 3 because the Bessel functions $J_1(z)$, $J_2(z)$, and $J_3(z)$ are zero for these peaks. Similarly, the curve at $z=2.4$ [having $J_0(z)=0$] shows all peaks (even and odd) to have the same width, and pure $J_n^2(z)$ peak-height variation. These variations in the peak heights and peak widths are the result of an equilibrium which is reached in a time-dependent steady state of a TLA due to the excitation by the FAM field and the dissipation caused by the phenomenon of spontaneous emission in the system.

Before comparing the spectrum of Eq. (4) with the spectrum of other known cases, we digress here to discuss a novel transient effect in this system. We have shown that just as in the interaction of the constant field case,[10]

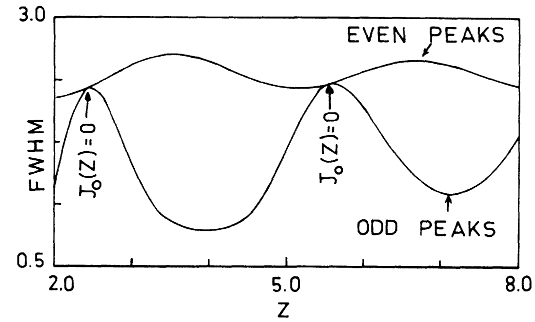

FIG. 1. The intensity-dependent variation in the widths of odd and even peaks are shown for $\Gamma=0.5\gamma$ and $\Omega=5\gamma$.

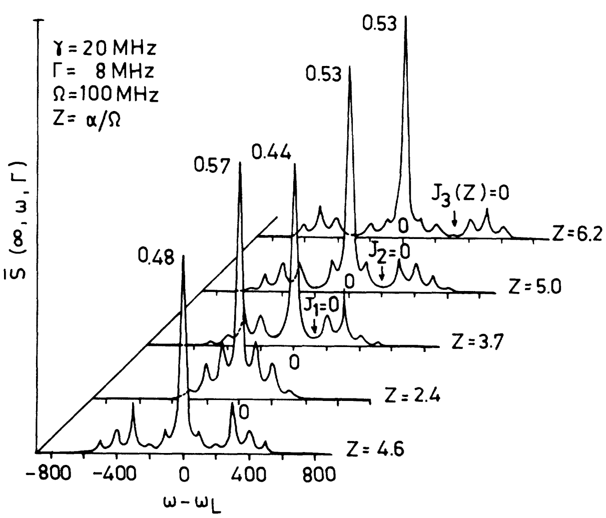

FIG. 2. The spectra for five values of z (marked along the right-hand side) are shown. $z=4.6$ curve corresponds closely to the observed spectrum. The curves with $z=2.4$, 3.7, 5.0, and 6.2 correspond to the zero in $J_0(z)$, $J_1(z)$, $J_2(z)$, and $J_3(z)$.

it may be possible to place a TLA interacting with a phase-controlled resonant FAM field into one of its dressed states. This may be achieved by suddenly changing the phase ϕ_L of the FAM field by $-\pi/2(+\pi/2)$ at time t_1, determined by the condition

$$\int_{t_0}^{t_1} \alpha\cos(\Omega t'+\phi_\Omega)\partial t' = \pi/2, \ \alpha > \Omega \,. \qquad (9)$$

It has been assumed that the atom is in its ground state at time t_0. The fact that the atom has been placed in one of its dressed states implies that the Bloch vector and the pseudomagnetic field vector of this system, hereafter called **V** and **H**, respectively, have become parallel (antiparallel). However, note that as time proceeds the vector $\mathbf{H}=(\chi(t)\cos(\phi_L\mp\pi/2), \chi(t)\sin(\phi_L\mp\pi/2), 0)$, for $\phi_\Omega=0$ and $T/4<t<3T/4$, becomes antiparallel (parallel) to vector **V**, which must remain stationary because $\dot{\mathbf{V}}=0$ in the dressed state. Thus, though the Bloch vector does not change, the atom finds itself in a stationary state that has become antiparallel (parallel) to the new position of the vector **H**. This remarkable situation can be tested only by examining the transient spectrum of the system, as the total intensity of the emitted light remains constant in spite of full modulation. Using our formulas, one predicts a transient spectrum shown in Fig. 3 for a system placed initially in the antiparallel state. We have shown the spectrum after one full cycle, at the end of which the system has returned to the original $(t=t_1)$ antiparallel dressed state (with $\phi_\Omega=0$). The peaks on the $D<0$ [Fig. 3(a)] side of the spectrum arise due to the spontaneous emission transitions originating from the antiparallel state. The increase in the peak height for $D<0$ [Fig. 3(b)] implies the buildup of population in the antiparallel eigenstate. After the time $T+T/4$ $(T=2\pi/\Omega)$, one notes that it is the $D>0$ peaks that start to build up [Fig. 3(c)]. They even become higher than the $D<0$ peak for time $5T/4<t<7T/4$. The behavior in the time period depicted in Figs. 3(d)–3(f) also shows similar peak-height changes from the right-hand to the left-hand side. Thus this shows that the system shifts between the two dressed states due to the FAM field. Experimental demonstration of these novel transient effects shall allow us to draw two conclusions, viz., (i) that a system with periodic Hamiltonian may be placed in its dressed state or Floquet state, and (ii) that the time dependence of the external field (in addition to the spontaneous emission process) transfers portions of the population from one dressed state to another. Note that for the constant field[11] the transfer of population from one dressed state to another only occurs by spontaneous emission.

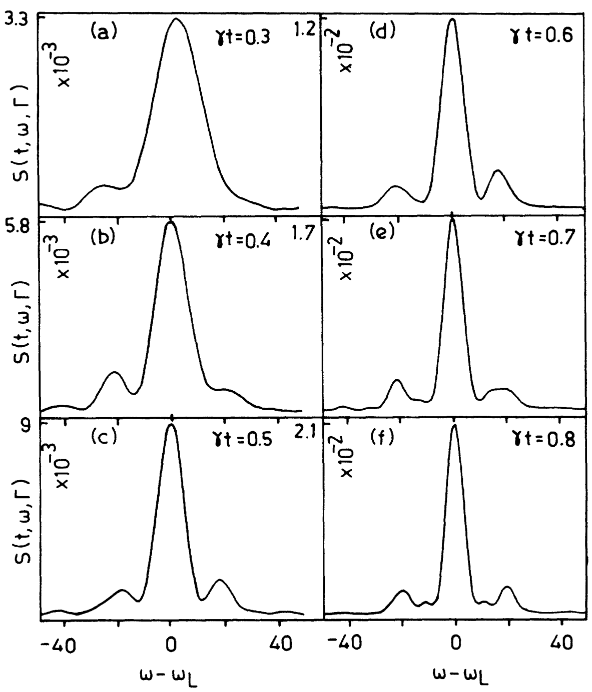

FIG. 3. The transient spectrum for the system prepared initially in the antiparallel dressed state are shown for different values of γt and $\Gamma=0.1\gamma$, $\Omega=20\gamma$, $\alpha=20\gamma$, and $\gamma T=\pi/10$. Note the difference in the relative peak height of the left-hand and right-hand sides of the central peak at $\gamma t=0.4$ and $\gamma t=0.6$. The system stayed for a sufficiently long time in the antiparallel and parallel dressed states, respectively, until these times.

Returning to Eq. (4), note that the central peak is made up of two Lorentzians. There is no Lorentzian with a coherent width of $\Gamma/2$, being an even term $(n=0)$. Each of the two Lorentzians has a different kind of incoherent width. The FWHM are $(\Gamma+\gamma)$ and $(\Gamma+2\gamma_I)$ with their respective weight functions given by

$$C_{\rm inc}=(\Gamma+\gamma)[\gamma_I-\gamma J_0^2(z)](4\gamma_I)^{-1}\,, \qquad (10)$$

$$C_{\rm inc}^R=(\Gamma+2\gamma_I)J_0^2(z)(\gamma_I-\gamma)(4\gamma_I)^{-1}\,. \qquad (11)$$

To understand this situation, note that Eq. (4) differs in many ways from the resonance fluorescence spectrum of a TLA interacting with a partially amplitude-modulated field[12,13] with instantaneous Rabi frequency $\chi'(t)[=\alpha_0+\alpha\cos(\Omega t+\phi_\Omega)]$. Equation (4) contains effects of γ_- not included in earlier calculations. Notice that in Eq. (4) the coherent and the incoherent terms are placed at $(D\pm n\Omega)=0$. This is quite unlike the partially modulated case in which the coherent side peaks are symmetrically placed at $(D\pm n\Omega)=0$ and the two sets of incoherent peaks are placed at $D\pm\alpha_0\pm n\Omega=0$; each incoherent set is symmetrically placed about the corresponding Rabi sideband at the average Rabi frequency α_0, of $\chi'(t)$. We have shown[14] that in the limit $\alpha_0\to 0$, $\alpha\neq 0$, Eq. (4) can be obtained as a limit of the spectrum from a TLA interacting with the partially modulated case. The Rabi sidebands at $\alpha_0\to 0$ merge into the central peak at $D=0$ giving rise to the second kind of incoherent peak, other than the one with width $(\frac{1}{2}\Gamma+\frac{1}{2}\gamma)$, at $D=0$. A merger of several coherent peaks also takes place with their corresponding peaks coming from two sets of incoherent peaks (of $\pm\alpha_0$) thus leading to Eq. (4) in the above-mentioned limits. In this very limit a portion of the coherent peak associated with the steady-state values (proportional to $|\alpha_0|^2$) has also vanished.

Therefore, this explains the absence of the Mollow triplet in Eq. (4). It also makes it clear why the spectrum of a TLA interacting with pure time-dependent pulses of pseudomagnetic field of a general class represented by $\mathbf{H}=(\alpha\cos\phi_L f(t),\ \alpha\sin\phi_L f(t),\ 0)$ (the FAM field case is of this class) does not have the Rabi sidebands of the Mollow triplet.[15] The discussions of the transient spectrum in this Rapid Communication suggest that for pulses defined by $f(t)$, the time-dependent transfer of population from one dressed state to another becomes important. The dressed states in such $(\alpha_0=0)$ situations are defined by $\mathbf{H}=(\alpha\cos\phi_L,\alpha\sin\phi_L,0)$. Note that it is possible even in such cases to identify the coherent and incoherent effects separately. The Rabi-frequency effects in the pulsed case are seen in the height of the peaks, just as in the FAM field case studied in this Rapid Communication where the effects of the Rabi frequency of modulation α are seen in the peak-height function $J_n^2(z)$.

In conclusion, the spectrum of resonance fluorescence from a TLA interacting with the FAM field exhibits several peaks, demonstrating the presence of Floquet energy eigenvalues in this system. The control of the FAM field in the transfer of the system from one dressed state to another is shown to lead to novel transient spectra. Typical variation in the peak height and peak widths observed recently have been explained. Several important comparisons are made and anomalous absence of the Mollow triplet in spectra from time-dependent pulses is commented upon. Use of these results with the dye-laser problem shall be reported elsewhere.

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[1]S. Chakmakjian, K. Koch, and C. R. Stroud, Jr., J. Opt. Soc. Am. B **5**, 2015 (1988).

[2]Y. Zhu, A. Lezama, Q. Wu, and T. W. Mossberg, in Proceedings of the Sixth Rochester Conference on Coherence and Quantum Optics, June, 1989 (unpublished).

[3]L. W. Hillman, J. Krasinski, R. W. Boyd, and C. R. Stroud, Jr., Phys. Rev. Lett. **52**, 1605 (1984).

[4]B. R. Mollow, Phys. Rev. **188**, 1969 (1969).

[5]C. J. Favre and E. Geneux, Phys. Lett. **8**, 190 (1964); see also G. S. Agarwal and N. Nayak, J. Phys. B **19**, 3385 (1986).

[6]W. M. Ruyten, J. Opt. Soc. Am. B **6**, 1796 (1989).

[7]M. Lax, Phys. Rev. **172**, 350 (1968).

[8]J. H. Eberly and K. Wodkiewicz, J. Opt. Soc. Am. **67**, 1252 (1977).

[9]See, for example, the review article by N. L. Manakov, V. D. Ovsiannikov, and L. P. Rapoport, Phys. Rep. **141**, 319 (1986).

[10]Y. S. Bai, A. G. Yodh, and T. W. Mossberg, Phys. Rev. Lett. **55**, 1277 (1985); Lu Ning, P. R. Berman, A. G. Yodh, Y. S. Bai, and T. W. Mossberg, Phys. Rev. A **33**, 3956 (1986).

[11]J. E. Golub and T. W. Mossberg, Phys. Rev. Lett. **59**, 2149 (1987).

[12]B. Blind, P. R. Fontana, and P. Thomann, J. Phys. B **13**, 2717 (1980).

[13]Surya P. Tewari and M. Krishna Kumari, in Proceedings of the Sixth Rochester Conference on Coherence and Quantum Optics (Ref. 2).

[14]M. Krishna Kumari and Surya P. Tewari (unpublished).

[15]P. A. Rodgers and S. Swain, J. Mod. Opt. **34**, 643 (1987); K. Rzazewski and M. Florjanczyk, J. Phys. B **17**, L509 (1984).