Analytical solution for the spectrum of resonance fluorescence of a cooperative system of two atoms and the existence of additional sidebands

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The analytical solution for the spectrum of resonance fluorescence from a collective system of two atoms is presented. It is shown that the spectrum is a decreasing function of frequency in the neighborhood $\omega - \omega_L = \pm 2\Omega$ (Ω is the Rabi frequency) for moderately large Ω and exhibits resonant structure only for extremely large $\Omega \approx 100\gamma$. For moderately large Ω the resonant contributions at $\omega - \omega_L = \pm 2\Omega$ are destroyed by the nonresonant contributions; hence the additional structures at $\pm 2\Omega$ can be resolved only by filtering the nonresonant contributions.

Recently the structure of the spectrum of resonance fluorescence of a cooperative system of atoms has been studied in detail[1-6] and several different authors have reached different conclusions. The master equation which describes resonance fluorescence from a collective atomic system has not been solved analytically,[1] however very careful numerical studies[1,2] for two, three and, five atoms show that the incoherent part of the spectrum in the limit of large Rabi frequencies Ω, consists of the peaks at $\omega = \omega_L$, $\omega_L \pm \Omega$. Numerical computations also show that in the limit of large Ω, the spectrum for a cooperative situation is identical to the single-atom case[7] except for a scaling factor. This scaling factor has also been discussed by Amin and Cordes.[3] Mavroyannis[4] has done a Green's-function calculation and has reached a similar conclusion for the two-atom case. A recent paper by Senitzky[5,6] also examines this equation, however Senitzky has found that the spectrum of a cooperative system would consist not only of the usual peaks but also of additional sidebands at the harmonics of the Rabi frequency. Senitzky has arrived at this conclusion by using a certain decoupling approximation. One might ask, are these additional sidebands merely an artifact of the decoupling approximation[8] or a reality, and if they really exist, how the earlier numerical studies missed them. It can, of course, be argued that the numerical studies might not have been accurate enough to see these weak additional sidebands. In view of these differences, we have reexamined the cooperative system of two atoms and we have obtained the analytical solution for the master equation, valid for all values of the laser field strengths. For moderately large laser field strength ($\Omega \approx 50\gamma$), the analytical solution shows no additional sidebands at $\omega_L \pm 2\Omega$ (Senitzky's argument when used for the two-atom case only shows two additional peaks). The approximate form of the analytical result is also examined. The approximate form shows the resonant structure at $\omega_L \pm 2\Omega$. However, there are also nonresonant contributions which kill these additional resonant structures so that the resultant approximate form shows no additional peaks at $\omega_L \pm 2\Omega$. For extremely intense fields ($\Omega \gtrsim 100\gamma$) the analytical results show well-defined additional sidebands in the resonance fluorescence spectrum. Our analytical results shed new light on the range of Ω values for which additional sidebands are present and also indicate why in the early numerical work such bands were missed.

We follow the development of Paper I very closely and use the same notation as in Paper I. The master equation describing the atomic dynamics for a cooperative system under the influence of

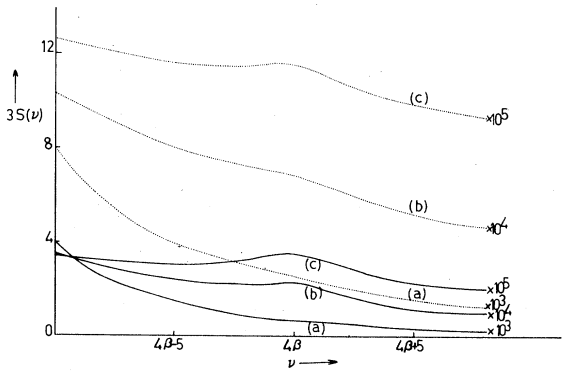

FIG. 1. Spectrum $3S(\nu)$ as a function of ν for $\beta = 10$, 20, 50 in the neighborhood of $\nu = 4\beta$: the solid and dotted curves correspond respectively to exact results [Eqs. (9) and (12)] and approximate result [Eq. (13)].

the laser field is given by

$$\frac{\partial\rho}{\partial t} = -ig[S^+ + S^-, \rho] + 2\gamma(S^-\rho S^+ - \tfrac{1}{2}\rho S^+ S^- - \tfrac{1}{2}S^+ S^-\rho), \quad (1)$$

where ρ is in a frame rotating with the laser frequency ω_L which is taken to be on resonance with the atomic frequency. 2γ is the Einstein-A coefficient and $2g$ is the Rabi frequency Ω; $S^{\pm}$ are the collective operators. It has been shown in Paper I (Appendix A) that in the representation $|j,m\rangle$, $j=1$, $m=\pm 1,0$, Eq. (1) can be written in the matrix form as

$$\frac{d\chi}{d\tau} = A\chi,$$

$$A = \begin{bmatrix} -2 & 0 & g' \\ 2 & -1 & -g' \\ g' & -g' & -1 \end{bmatrix}, \quad g' = i\beta\sqrt{2} \quad (2)$$

$$\beta = g/2\gamma,$$

$$\tau = 2\gamma t,$$

$$\chi_1 = \rho_{1,0} + \rho_{0,1}, \quad \chi_2 = \rho_{0,-1} + \rho_{-1,0}, \quad \chi_3 = \rho_{1,-1} - \rho_{-1,1},$$

$$\frac{d\Phi}{d\tau} = B\Phi + I,$$

$$I_j = -2g'\delta_{j2}, \quad j = 1,\ldots,5 \quad (3)$$

$$B = \begin{bmatrix} -2 & 0 & g' & 2g' & -2g' \\ 2 & -1 & -g' & 2g' & 4g' \\ g' & -g' & -1 & 0 & 0 \\ g' & 0 & 0 & -2 & 0 \\ -g' & g' & 0 & 2 & -2 \end{bmatrix}, \quad (4)$$

$$\Phi_1 = \rho_{1,0} - \rho_{0,1}, \quad \Phi_2 = \rho_{0,-1} - \rho_{-1,0},$$

$$\Phi_3 = \rho_{1,-1} + \rho_{-1,1},$$

$$\Phi_4 = \rho_{1,1}, \quad \Phi_5 = \rho_{0,0}. \quad (5)$$

Using Eqs. (2)–(5), one can write the time dependence of the dipole moment as

$$\langle\hat{S}^+(z)\rangle = \int_0^\infty e^{-z\tau}\langle S^+(\tau)\rangle d\tau$$

$$= \frac{1}{\sqrt{2}}(\hat{\chi}_1 + \hat{\chi}_2 - \hat{\Phi}_1 - \hat{\Phi}_2)$$

$$= \frac{1}{\sqrt{2}}\sum_{j=1}^{3}[(z-A)^{-1}_{1j}\chi_j(0) + (z-A)^{-1}_{2j}\chi_j(0)] - \frac{1}{\sqrt{2}}\sum_{j=1}^{5}[(z-B)^{-1}_{1j}\Phi_j(0) + (z-B)^{-1}_{2j}\Phi_j(0) - (z-B)^{-1}_{1j}z^{-1}I_j - (z-B)^{-1}_{2j}z^{-1}I_j]. \quad (6)$$

Using the quantum-regression theorem and the steady-state solutions of Eqs. (2), and (3), viz.,

$$\chi_1 = \chi_2 = \chi_3 = 0,$$

$$\Phi_1 = 2g'^3/D, \ \Phi_2 = -2g'(1-g'^2)/D, \ \Phi_3 = 2g'^2/D, \ \Phi_4 = g'^4/D, \ \Phi_5 = -g'^2(1-g'^2)/D, \quad (7)$$

$$D = 3g'^4 - 2g'^2 + 1,$$

we obtain the form of the steady-state correlation function

$$\hat{\Gamma}^{(1)}(z) = \int_0^\infty d\tau\, e^{-z\tau}\lim_{t\to\infty}\langle S^+(t+\tau)S^-(t)\rangle$$

$$= \frac{g'^4}{D}\left[[(z-B)^{-1}_{12} + (z-B)^{-1}_{22}]\left(1 - \frac{2}{g'^2} + \frac{4}{z} - \frac{2}{zg'^2}\right) + (z-B)^{-1}_{11} + (z-B)^{-1}_{21} + \frac{1}{g'}[(z-B)^{-1}_{13} + (z-B)^{-1}_{23} - (z-B)^{-1}_{15} - (z-B)^{-1}_{25} + (z-A)^{-1}_{13} + (z-A)^{-1}_{23}] + (z-A)^{-1}_{11} + (z-A)^{-1}_{12} + (z-A)^{-1}_{21} + (z-A)^{-1}_{22}\right]. \quad (8)$$

A straightforward but long algebraic manipulation reduces Eq. (8) to

$$\hat{\Gamma}^{(1)}(z) = \frac{g'^4}{D}\left(\frac{N(z)}{D(z)} + \frac{(z+1)(2z+5)+1+8\beta^2}{(z+2)(z+1)^2 + 2\beta^2(2z+1)}\right), \quad (9)$$

$$N(z) = (z+3)(z+1)(z+2)^2 + 4\beta^2(4z^2+9z+6) - z(2z+3)(z+2) - 16\beta^2(2z+3) + [(1+4/z) + 1/\beta^2(1+1/z)][(z+2)^3(z+1) + 4\beta^2(4z^2+11z+8)], \quad (10)$$

$$D(z) = 4\beta^2[z(z+1)^2 + 8\beta^2(2z+3)] + (z+1)^2(z+2)^3 + 2\beta^2(z+2)(8z^2+17z+8). \quad (11)$$

The spectrum of resonance fluorescence is defined by

$$S(\nu) = \mathrm{Re}\hat{\Gamma}^{(1)}(i(\omega-\omega_L)/2\gamma), \quad \nu = \omega - \omega_L/2\gamma. \quad (12)$$

A plot of the spectrum, which follows from Eq. (9), in the neighborhood of $\nu = 4\beta$ for $\beta = 10, 20, 50$ is shown in Fig. 1. We find that (i) for moderately large Ω the spectrum consists of the usual three peaks at $\omega = \omega_L \pm \Omega, \omega_L$. The extra peaks expected at $\omega = \omega_L \pm 2\Omega$ are absent. (ii) For extremely large $\Omega \gtrsim 100\gamma$, the spectrum shows well-defined sidebands. We now present the approximate form which follows from Eq. (9) and which is valid for large β. A careful perturbation analysis of Eq. (9), in the inverse powers of β, leads to the following expression for the incoherent part of the spectrum:

$$3S(\nu) = \frac{\frac{3}{4}(1 - 1/4\beta^2)}{(\nu - 2\beta)^2 + (\frac{3}{4})^2} + \frac{\frac{3}{4}(1 - 1/4\beta^2)}{(\nu + 2\beta)^2 + (\frac{3}{4})^2} + \frac{(1 - 1/16\beta^2)}{\nu^2 + \frac{1}{4}} + \frac{1}{16\beta^2}\left(\frac{\frac{5}{2}}{(\nu - 4\beta)^2 + (\frac{5}{2})^2} + \beta \to -\beta\right)$$
$$- \frac{5}{8\beta^2}\left(\frac{(\nu - 2\beta)}{(\nu - 2\beta)^2 + (\frac{3}{4})^2} + \beta \to -\beta\right) + \frac{1}{16\beta^2}\left(\frac{\frac{7}{4}}{(\nu - 2\beta)^2 + (\frac{7}{4})^2} + \beta \to -\beta\right). \tag{13}$$

The first three terms in Eq. (13) represent the usual three peaks, the next two terms of (13) represent the additional sidebands as discussed by Senitzky, and the remaining terms of Eq. (13) are the nonresonant contributions which hitherto have not been obtained. Expression (13) as such shows the additional resonant structures at $\nu = \pm 4\beta$. However, the nonresonant contributions in Eq. (13) take over the resonant ones in the region $\nu \sim \pm 4\beta$, as a result the complete expression (13) shows *no additional peaks* at $\nu = \pm 4\beta$, for $\beta \sim 10$. The resonant structure at $\nu = \pm 4\beta$ can thus be studied only if one can suppress all the nonresonant contributions. Expression (13) is plotted in Fig. 1 for $\beta = 10, 20, 50$ in the neighborhood of 4β. For very large values of β ($\Omega \approx 200\gamma$) the additional sidebands do appear.

We have thus shown the following: (i) For moderately large Ω, the analytical expression for the spectrum of the resonance fluorescence from a cooperative system of two atoms shows no additional peaks besides the usual three peaks. The approximate expression shows only an *apparent* resonant structure at $\omega - \omega_L = \pm 2\Omega$, which is wiped out by the nonresonant contributions. (ii) For extremely large Ω, the additional sidebands are clearly resolvable. We expect this to happen in general for an arbitrary number of atoms, and we are developing a new technique, for the solution of the master equation[9] based on the principle of detailed balance and deviations from it, which should enable us to calculate all the resonant and nonresonant contributions.

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