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Multiatom effects in resonance fluorescence

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The intensity spectrum of the scattered radiation in two-atom resonance fluorescence is investigated and compared with the one-atom spectrum. For intense fields the spectra are found to be proportional to one another. A conjecture is made for the N-atom spectrum.

I. INTRODUCTION

The intensity spectrum of the radiation scattered by a single two-level atom, in the presence of a strong incident field tuned precisely to the atomic transition frequency ω_0, exhibits a three-peak structure[1-15]

$$I_1(\omega)=\frac{\omega_0}{8\pi}\left(\frac{4\gamma^2}{(\omega-\omega_0)^2+\gamma^2}+\frac{3\gamma^2}{(\omega-\omega_0-2k)^2+(\frac{3}{2}\gamma)^2}+\frac{3\gamma^2}{(\omega-\omega_0+2k)^2+(\frac{3}{2}\gamma)^2}\right), \tag{1.1}$$

where $2\gamma=4|\vec{d}|^2\omega_0^3/3c^3$ is the Einstein A coefficient, with $\vec{d}$ being the transition dipole matrix element (we take $\hbar=1$). $k=\frac{1}{2}\vec{d}\cdot\vec{E}_0$, where $\vec{E}_0\cos\omega_0 t$ is the applied field in a semiclassical treatment. In a fully quantum-mechanical treatment, on the other hand, $k=[(n+1)|g|^2]^{1/2}$, where n is the number of photons initially in the pump mode and the coupling of the atom to the field is taken to be $g\sigma^\dagger b$ + H.c.; here σ is the usual atomic lowering operator and b is the annihilation operator for the resonant field mode. Equation (1.1) follows in both cases. The total frequency-integrated intensity is readily found to be

$$I_1=\int_{-\infty}^{+\infty}d\omega I_1(\omega)=\gamma\omega_0. \tag{1.2}$$

For two or more atoms the spectrum is more difficult to obtain, even in the approximation of closely spaced, but not directly interacting atoms. Recently Agarwal, Brown, Narducci, and Vetri (ABNV)[16] have obtained some numerical results for the two- and three-atom problem.

In Sec. II of the present paper the two-atom problem is analyzed. Some results are obtained for the total (frequency-integrated) intensity I_2 for arbitrary field strengths and the complete $I_2(\omega)$ is explicitly obtained in a closed form in the limit of intense fields. In Sec. III we discuss our results and present a conjecture for the N-atom intensity spectrum $I_N(\omega)$.

II. CALCULATION OF THE FLUORESCENCE SPECTRUM

For two identical two-level atoms very close to each other, in the dipole and rotating-wave approximation, the master equation for the atomic density operator can be written as[5]

$$\frac{\partial\rho}{\partial t}=-\tfrac{1}{2}i\omega_0[S^z,\rho]+\tfrac{1}{2}ik[S^\dagger e^{-i\omega_L t}+Se^{i\omega_L t},\rho]-\gamma(S^\dagger S\rho+\rho S^\dagger S-2S\rho S^\dagger), \tag{2.1}$$

where $S=\sigma_1+\sigma_2$ is the collective atomic dipole operator; $S^z=[S^\dagger,S]$, and ω_L is the applied field frequency.[17] The intensity spectrum $I_2(\omega)$ is found by using the quantum regression theorem[18]

$$I_2(\omega)=\frac{\gamma\omega}{\pi}\int_{-\infty}^{+\infty}d\tau\, e^{-i\omega\tau}\langle S^\dagger(t+\tau)S(t)\rangle_{ss}, \tag{2.2}$$

where the subscript ss denotes the steady-state value of the atomic correlation function.

For a complete set of atomic operators we have used the 16 operators arising in forming the products $(1, \sigma_1, \sigma_1^\dagger, \sigma_1^\dagger\sigma_1) \otimes (1, \sigma_2, \sigma_2^\dagger, \sigma_2^\dagger\sigma_2)$. In taking expectation values we have permitted the atomic system to be in an arbitrary initial state, rather than restricting consideration from the outset to just the triplet set of states symmetric under the interchange $1 \leftrightarrow 2$:

$$|1\rangle = |e_1\rangle\,|e_2\rangle\,,$$

$$|2\rangle = (1/\sqrt{2})(|e_1\rangle\,|g_2\rangle + |g_1\rangle\,|e_2\rangle), \tag{2.3a}$$

$$|3\rangle = |g_1\rangle\,|g_2\rangle\,.$$

The fact that the singlet state

$$|4\rangle = (1/\sqrt{2})(|e_1\rangle\,|g_2\rangle - |g_1\rangle\,|e_2\rangle) \tag{2.3b}$$

does not couple, in the dipole approximation, to the triplet states then shows up as a decoupling of the full set of 16 equations (for as many unknowns) into groups of ten equations and six equations for the symmetric and antisymmetric combinations, respectively. Of the ten symmetric operators, however, one (the identity operator) is trivial, so there is actually a set of nine simultaneous equations to be solved.

The total intensity I_2 is readily obtained:

$$I_2 = \int_{-\infty}^{+\infty} d\omega\, I_2(\omega)$$

$$= 2\gamma\omega_0 \langle S^\dagger(t) S(t)\rangle_{ss}$$

$$= 2\gamma\omega_0(1-B)\frac{1+R}{1+R+\frac{3}{4}R^2}R\,, \tag{2.4}$$

where $R = k^2/\gamma^2$ and B is a constant depending on the initial atomic state. $B=0$ for an initial triplet state and $B=1$ for an initial singlet state, giving $I_2 = 0$ for the singlet state, as expected. For the initially triplet state, our result (2.4) for the total intensity is essentially contained in ABNV.[16]

The solution of the set of nine equations is substantially more involved. Agarwal, Brown, Narducci, and Vetri[16] have coped with this situation[19] by finding numerical solutions for several values of the field strength. By explicitly expanding the determinantal condition for the eigenvalues λ of the 9×9 coefficient matrix, and in the limit of intense applied field strengths ($k \gg \gamma$), we get

$$\lambda_1 = 0,\ \lambda_2 = -\gamma,\ \lambda_3 = -3\gamma,\ \lambda_4 = 2ik - \tfrac{3}{2}\gamma,$$

$$\lambda_5 = -2ik - \tfrac{3}{2}\gamma,\ \lambda_6 = 2ik - \tfrac{7}{2}\gamma,\ \lambda_7 = -2ik - \tfrac{7}{2}\gamma, \tag{2.5}$$

$$\lambda_8 = 4ik - 5\gamma,\ \lambda_9 = -4ik - 5\gamma.$$

Just as in the one-atom problem, $\mathrm{Im}\lambda$ gives the locations and $-\mathrm{Re}\lambda$ determines the widths of the possible peaks in the spectrum $I_2(\omega)$.

From Eq. (2.5) the most that could be said is that $I_2(\omega)$ *might* have peaks at (i) the central frequency $\omega = \omega_L$, with up to two incoherent components; at (ii) an up-shifted sideband at frequency $\omega = \omega_L + 2k$, with possibly two components of different widths ($\frac{3}{2}\gamma$ and $\frac{7}{2}\gamma$) within the peak; at (iii) an up-shifted sideband at frequency $\omega = \omega_L + 4k$ with width 5γ; and the corresponding down-shifted sidebands. Without the detailed solution of the equations for the symmetric combination of atomic operators, however, it is impossible to predict by this approach the *weights* of the various components and hence to write down a closed-form expression for $I_2(\omega)$.

On the other hand, since the region of greatest interest is that of intense field strengths, the elegant dressed-atom formalism of Cohen-Tannoudji and Reynaud[15] (CTR) can be applied directly to the present problem. In our treatment we will exclude the antisymmetric (singlet) states from the outset since they play no role in determining the steady-state spectrum, assuming the atoms are not initially prepared in the singlet state.

Thus, if we assume that $\omega_L = \omega_0$, for simplicity, and $n \gg 1$, then the CTR formalism shows that the matrix element $\langle 3, n-1\,|D\,|1, n\rangle = 0$, meaning that the sideband at $\omega_0 + 4k$ is absent,[20] since the only transition possible at this frequency is $|1,n\rangle \rightarrow |3,n-1\rangle$. The symmetric partner at $\omega_0 - 4k$ is, of course, also absent from the spectrum.

Next consider the degenerate sideband at $\omega_0 + 2k$ contributed to by the two transitions $|1,n\rangle \rightarrow |2,n-1\rangle$ and $|2,n\rangle \rightarrow |3,n-1\rangle$. On diagonalizing the appropriate 2×2 matrix we find that the sideband $\omega_0 + 2k$ corresponds to two peaks, one of width $\frac{3}{2}\gamma$, the other of width $\frac{7}{2}\gamma$, in agreement with our roots λ_4 and λ_6 [Eq. (2.5)]. The net contribution of this lateral component to the steady-state spectrum is computed to be

$$\frac{\omega_0}{\pi}\,\frac{\gamma^2}{(\omega-\omega_0-2k)^2+(\frac{3}{2}\gamma)^2}\,. \tag{2.6}$$

The other peak of width $\frac{7}{2}\gamma$ is not present.

Finally, we treat the triply degenerate central component at ω_0. Diagonalizing the relevant 3×3 matrix gives the three roots: $\omega = \omega_0$, $\omega = \omega_0 - i\gamma$, $\omega = \omega_0 - 3i\gamma$. Thus the remaining roots of Eq. (2.5)

are accounted for. The contribution of these central transitions to the spectrum $I_2(\omega)$ is, however, simply

$$\frac{4\omega_0}{3\pi}\frac{\gamma^2}{(\omega-\omega_0)^2+\gamma^2}. \tag{2.7}$$

Combining the various contributions, we finally have

$$I_2(\omega)=\frac{\omega_0}{3\pi}\left(\frac{4\gamma^2}{(\omega-\omega_0)^2+\gamma^2}+\frac{3\gamma^2}{(\omega-\omega_0-2k)^2+(\tfrac{3}{2}\gamma)^2}+\frac{3\gamma^2}{(\omega-\omega_0+2k)^2+(\tfrac{3}{2}\gamma)^2}\right). \tag{2.8}$$

Comparing with the single-atom spectrum (1.1), we see that

$$I_2(\omega)=\tfrac{8}{3}I_1(\omega). \tag{2.9}$$

The total integrated intensity for the two-atom case is thus

$$I_2(\omega)=\tfrac{8}{3}\gamma\,\omega_0 \quad \text{(strong-field limit)} \tag{2.10}$$

in agreement with our general expression (2.4).

III. DISCUSSION OF RESULTS

The proportionality found in Eq. (2.9) between the two-atom and single-atom spectra is highly suggestive; one is naturally inclined to speculate that the N-atom spectrum may be simply a multiple of $I_1(\omega)$. Further evidence in support of this may be found in the numerical calculations of ABNV[16] who find that, for strong fields, both $I_2(\omega)$ and $I_3(\omega)$ tend to the same shape as $I_1(\omega)$. Whether or not the *shape* of $I_N(\omega)$ is the same as that of $I_1(\omega)$, for strong fields the total intensity I_N can be calculated as follows. The fraction of the time spent in each of the $N+1$ (bare) symmetric atomic states will be $1/(N+1)$. In each of these states the rate of radiation is given by[21]

$$2\gamma\omega_0 N_e(N_g+1), \tag{3.1}$$

where $N_e(N_g)$ is the number of excited- (ground-) state atoms in the state. Thus the total radiation rate should be given by[22]

$$I_N=\frac{2\gamma\omega_0}{N+1}\sum_{N_g=0}^{N}N_e(N_g+1)$$

$$=\frac{2\gamma\omega_0}{N+1}\frac{N(N+1)(N+2)}{6}$$

$$=\tfrac{1}{3}N(N+2)\gamma\omega_0. \tag{3.2}$$

This agrees with our earlier results for $N=1,2$. If the speculation concerning the shape of $I_N(\omega)$ is taken seriously and combined with the above expression for I_N, we could write

$$I_N(\omega)=\tfrac{1}{3}N(N+2)I_1(\omega) \tag{3.3}$$

for the steady-state, intense field spectrum for the N-atom case. Verification (or otherwise) of Eq. (3.3) must await either further numerical calculations or, perhaps, a dressed N-atom calculation.

Very recently another calculation[23] which attempts to obtain a closed-form solution for the two-atom spectrum $I_2(\omega)$ has come to our attention. The result is a three-peak spectrum but with widths differing from the one-atom peak widths; the calculation is thus in disagreement with the results of both ABNV[16] and ourselves.

Other recent calculations[24-26] have attempted to calculate the N-atom fluorescence spectrum by a different method. For very strong applied fields these calculations result in a three-peaked spectrum of the same shape as $I_1(\omega)$, but with a linear dependence on the number of atoms N, in contrast to our conjectured $I_N(\omega)$. Presumably this discrepancy is owing to the factorization and linearization approximations made in these model calculations. It would be very interesting to see if the appropriate multiatom intensity spectrum measurements could be made.

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