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LETTER TO THE EDITOR

A comment on the quantum treatment of spontaneous emission from a strongly driven two-level atom

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Abstract. A quantum-mechanical master equation approach to the spontaneous emission from a two-level atom in the presence of a strong pump field is reported. Access is available to both a one-photon treatment and a treatment incorporating photon cascades. A peak height ratio of 3:1 for the complete many photon approach is reduced to 2:1 in a one-photon approximation. Broadening of the sidebands is also lost through this approximation.

A recent experiment (Schuda *et al* 1974) measuring the spectrum of light scattered from an atomic beam illuminated by intense laser radiation has heightened interest in the theoretical treatment of the resonant Stark effect in the optical regime. Although this problem has been discussed extensively by earlier authors (Heitler 1954, Apanasevich 1964, Bergmann 1967, Newstein 1968, Morozov 1969, Mollow 1969, 1970, Chang and Stehle 1971, Stroud 1971, Oliver *et al* 1971, Gush and Gush 1972) disagreement has been considerable and even extends to such gross features as the general shape of the spectrum. In this particular respect Schuda *et al* report evidence for a three-peaked spectrum consisting of a central peak at the excitation frequency and symmetrically placed sidebands. This is in agreement with the predictions of a number of authors (Apanasevich 1964, Newstein 1968, Mollow 1969, 1970, Stroud 1971, Oliver *et al* 1971). Amongst these authors there is still however a lack of concord concerning the finer details of the spectrum such as linewidths and peak heights on which no conclusive experimental results have as yet been reported. In particular we are concerned with the contrasting results of Mollow (1969) and Oliver *et al* (1971) on the one hand and Stroud (1971) on the other. In this comparison we find conflicting views, both with regard to the relative heights of the spectral peaks and the respective peak linewidths. We wish to communicate here the results of recent theoretical studies which enable us to move towards the resolution of these differences.

It is recognized by Stroud himself (Stroud 1972) that the method he has adopted must admit to a fundamental shortcoming: 'In spite of the wide range of problems which can be treated efficiently by these techniques the method has a deficiency whose seriousness is yet to be determined. While we have allowed any number of stimulated absorptions and emissions we have described only a single fluorescence.' Stroud describes a one-photon process while the physical circumstances allow for photon cascades. By pursuing a formalism which does not of necessity resort to a one-photon approximation, but which nonetheless does allow for a one-photon treatment as a

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special case, we gain a perspective from which the seriousness of Stroud's approximation can be viewed.

The system we describe consists of a single fixed two-level atom, resonantly pumped by a strong field mode at a frequency ω_0, and simultaneously coupled to the remaining modes of the radiation field, these being initially unpopulated. If we view the many modes of the radiation field as a reservoir R into which the coupled atom–field system S spontaneously emits, the Hamiltonian for the composite system $S \oplus R$ may be written

$$H = H_S + H_R + H_{SR} \tag{1}$$

where

$$\begin{aligned} H_S &= \hbar\omega_0 a^\dagger a + \tfrac{1}{2}\hbar\omega_0\sigma_z + \hbar(\kappa a\sigma_+ + \kappa^* a^\dagger\sigma_-) \\ H_R &= \sum_{\mathbf{k}\lambda} \hbar\omega_k r^\dagger_{\mathbf{k}\lambda} r_{\mathbf{k}\lambda} \\ H_{SR} &= \sum_{\mathbf{k}\lambda} \hbar(\kappa_{\mathbf{k}\lambda} r_{\mathbf{k}\lambda}\sigma_+ + \kappa^*_{\mathbf{k}\lambda} r^\dagger_{\mathbf{k}\lambda}\sigma_-). \end{aligned} \tag{2}$$

Here $a^\dagger$ and a are boson creation and annihilation operators for the pump mode, $r^\dagger_{\mathbf{k}\lambda}$ and $r_{\mathbf{k}\lambda}$ are boson creation and annihilation operators for the radiation mode with wavevector $\mathbf{k}$ (frequency ω_k) and polarization λ, σ_z, σ_+ and σ_- are the familiar atomic operators, and κ and $\kappa_{\mathbf{k}\lambda}$ are coupling constants.

The formal apparatus necessary for our calculation is then as follows. Specifying the field operator for the spontaneous emission by

$$\boldsymbol{E}_{\mathrm{sp}}(\boldsymbol{r}, t) = \boldsymbol{E}^{(-)}_{\mathrm{sp}}(\boldsymbol{r}, t) + \boldsymbol{E}^{(+)}_{\mathrm{sp}}(\boldsymbol{r}, t) \tag{3}$$

where

$$\boldsymbol{E}^{(-)}_{\mathrm{sp}}(\boldsymbol{r}, t) = \boldsymbol{E}^{(+)}_{\mathrm{sp}}(\boldsymbol{r}, t)^\dagger \tag{4}$$

the spectrum for the scattered field is given by

$$I(\omega, \boldsymbol{r}, T) = I(\boldsymbol{r}, T)\frac{1}{2\pi}\frac{\int_{r/c}^{T} \mathrm{d}t_2 \int_{r/c}^{T} \mathrm{d}t_1 \exp[\mathrm{i}\omega(t_2 - t_1)] G^{(1)}_{\mathrm{sp}}(\boldsymbol{r}, t_1; \boldsymbol{r}, t_2)}{\int_{r/c}^{T} \mathrm{d}t I(\boldsymbol{r}, t)}. \tag{5}$$

Here the intensity $I(\boldsymbol{r}, t)$ is given by

$$I(\boldsymbol{r}, t) = G^{(1)}_{\mathrm{sp}}(\boldsymbol{r}, t; \boldsymbol{r}, t) \tag{6}$$

and the first-order correlation function $G^{(1)}_{\mathrm{sp}}(\boldsymbol{r}, t_1; \boldsymbol{r}, t_2)$ is defined by

$$G^{(1)}_{\mathrm{sp}}(\boldsymbol{r}, t_1; \boldsymbol{r}, t_2) = \langle \boldsymbol{E}^{(-)}_{\mathrm{sp}}(\boldsymbol{r}, t_1) \boldsymbol{E}^{(+)}_{\mathrm{sp}}(\boldsymbol{r}, t_2) \rangle. \tag{7}$$

For simplicity we may neglect the normalization factors in equation (5) and simply write

$$I(\omega, \boldsymbol{r}, T) \propto \frac{1}{2\pi}\int_{r/c}^{T} \mathrm{d}t_2 \int_{r/c}^{T} \mathrm{d}t_1 \exp[\mathrm{i}\omega(t_2 - t_1)] G^{(1)}_{\mathrm{sp}}(\boldsymbol{r}, t_1; \boldsymbol{r}, t_2). \tag{8}$$

It can be shown that for times not too short (Mollow 1969)

$$\boldsymbol{E}^{(+)}_{\mathrm{sp}}(\boldsymbol{r}, t) = \boldsymbol{\mathscr{E}}^{(+)}_{\mathrm{sp}}(\boldsymbol{r}, t) - \frac{\omega_0^2}{4\pi\mathscr{E}_0 c^2 r}\left[\left(\boldsymbol{\mu} \times \frac{\boldsymbol{r}}{r}\right) \times \frac{\boldsymbol{r}}{r}\right]\sigma_-\left(t - \frac{r}{c}\right) \tag{9}$$

where $\boldsymbol{\mathscr{E}}^{(+)}_{\mathrm{sp}}(\boldsymbol{r}, t)$ is simply the freely propagating part of the field and $\boldsymbol{\mu}$ is the atomic dipole matrix element. Hence, the scattered spectrum admits expression in terms of two-time averages of atomic operators alone.

Corresponding to the Hamiltonian of equations (1) and (2) a Markoffian master equation is available (see for example Haake 1973) for the reduced density operator ρ of S. This may be written formally as

$$\frac{\partial \rho}{\partial t} = \mathscr{L}\rho. \tag{10}$$

The two-time average required for the calculation of the autocorrelation function then finds formal expression in terms of the generalized Liouville operator $\mathscr{L}$:

$$\langle \sigma_+(t_1 - r/c)\sigma_-(t_2 - r/c)\rangle = Tr_s\{\exp[\mathscr{L}(t_2 - t_1)][\rho(t_1 - r/c)\sigma_+]\sigma_-\} \qquad t_2 \geqslant t_1 \tag{11}$$

with the relation

$$\langle \sigma_+(t_1 - r/c)\sigma_-(t_2 - r/c)\rangle = \langle \sigma_+(t_2 - r/c)\sigma_-(t_1 - r/c)\rangle^* \tag{12}$$

providing definition for $t_2 \leqslant t_1$.

Utilization of the scheme outlined above is possible once the explicit form of the master equation (8) is established. The eigenstates of H_S defined by

$$\begin{aligned} H_S|E_n^2\rangle &= E_n^2|E_n^2\rangle \\ H_S|E_n^1\rangle &= E_n^1|E_n^1\rangle \end{aligned} \tag{13}$$

are well known. If $|E_2\rangle$ and $|E_1\rangle$ are respectively the excited and ground states of our atom, then

$$\begin{aligned} |E_n^2\rangle &= \frac{1}{\sqrt{2}}(|n, E_2\rangle + |n+1, E_1\rangle) \\ |E_n^1\rangle &= \frac{1}{\sqrt{2}}(|n, E_2\rangle - |n+1, E_1\rangle), \end{aligned} \tag{14}$$

the Fock states $|n\rangle$ being energy eigenstates for the free pump field. The corresponding eigenvalues read

$$\begin{aligned} E_n^2 &= \hbar(\omega_0 + 2n^{1/2}\kappa) \\ E_n^1 &= \hbar(\omega_0 - 2n^{1/2}\kappa). \end{aligned} \tag{15}$$

Expressed within this representation the master equation describing the dynamics of S reads

$$\frac{\partial \rho_{n,\xi;m,\eta}}{\partial t} = -i\frac{(E_n^\xi - E_m^\eta)}{\hbar}\rho_{n,\xi;m,\eta} + (-1)^{\xi+\eta}\tfrac{1}{4}\gamma \sum_{\mu,\nu} \rho_{n+1,\mu;m+1,\nu} - \tfrac{1}{4}\gamma \sum_{\mu} (\rho_{n,\xi;m,\mu} + \rho_{n,\mu;m,\eta}) \tag{16}$$

where

$$\rho_{n,\xi;m,\eta} = \langle E_n^\xi|\rho|E_m^\eta\rangle \qquad \xi = 1, 2; \eta = 1, 2. \tag{17}$$

Using this master equation we have been able to follow the treatment intimated in equations (3)–(12) for a pump field distributed in the Fock states about a mean $\bar{n} \gg 1$ with a variance $\sigma \ll \bar{n}$ without resorting to further approximation than that which is implied by these conditions. We have also been able to follow the same treatment in a one-photon approximation by only recognizing a reduced basis consisting of the four states $|E_N^2\rangle$, $|E_N^1\rangle$, $|E_{N-1}^2\rangle$ and $|E_{N-1}^1\rangle$. Both calculations reveal a spectrum which for

sufficiently intense pump fields splits into three peaks. However, the threshold at which this splitting occurs is predicted by the multiphoton treatment to be

$$8\bar{n}^{1/2}\kappa = \gamma \tag{18}$$

and in the one-photon approximation is given by

$$4N^{1/2}\kappa = \gamma. \tag{19}$$

Agreement is obtained in the first instance with the work of Mollow (1969) and in the second with Stroud (1971). The factor of two which separates equations (15) and (16) arises from the one-photon approximation.

We now come to consider the spectrum in detail. Specializing to the limit in which many optical nutations occur during a radioactive lifetime, the multiphoton calculation predicts for $P(t)$, the probability of observing the atom in its excited state at time t,

$$P(t) = \tfrac{1}{2}[1+\exp(-\tfrac{3}{4}\gamma t)\cos 2\bar{n}^{1/2}\kappa t]. \tag{20}$$

From this expression we see that for long times the atom settles down to a saturation steady state midway between the ground and excited states. The spectrum in this long time limit does not degenerate into a sharp line at ω_0 as suggested by semiclassical theory (Schuda *et al* 1974, Stroud 1972) but maintains the triple-peaked form

$$I(\omega, \boldsymbol{r}, \infty) \propto 2\frac{\frac{1}{2}\gamma}{(\omega-\omega_0)^2+(\frac{1}{2}\gamma)^2}+\frac{\frac{3}{4}\gamma}{[\omega-(\omega_0+2\bar{n}^{1/2}\kappa)]^2+(\frac{3}{4}\gamma)^2} + \frac{\frac{3}{4}\gamma}{[\omega-(\omega_0-2\bar{n}^{1/2}\kappa)]^2+(\frac{3}{4}\gamma)^2}. \tag{21}$$

This spectrum is in complete agreement with that of Mollow (1969) and Oliver *et al* (1971). The ratio of the central peak height to the heights of the sidebands is 3:1. The linewidth of the central peak is $\frac{1}{2}\gamma$, as for ordinary spontaneous emission, and the sidebands are broadened each having a width $\frac{3}{4}\gamma$. Contrasting this result however, in the one-photon approximation we find a spectrum

$$I(\omega, \boldsymbol{r}, \infty) \propto 2\frac{\frac{1}{2}\gamma}{(\omega-\omega_0)^2+(\frac{1}{2}\gamma)^2}+\frac{\frac{1}{2}\gamma}{[\omega-(\omega_0+2N^{1/2}\kappa)]^2+(\frac{1}{2}\gamma)^2} + \frac{\frac{1}{2}\gamma}{[\omega-(\omega_0-2N^{1/2}\kappa)]^2+(\frac{1}{2}\gamma)^2}. \tag{22}$$

Here deviations from equation (21) are apparent which originate in the one-photon approximation. In particular we notice a ratio of 2:1 rather than 3:1 in the peak heights. This of course arises in close association with the loss of sideband broadening which was exhibited by the multiphoton calculation. Hence we see that the effect of the photon cascades is to broaden the sidebands while maintaining their integrated intensity. The ratio of the central peak height to the height of the sidebands is consequently increased. A one-photon approximation is inadequate in that it neglects photon cascades and hence fails to account for this effect.

We should recognize at this point a recent paper by Smithers and Freedhoff (1974) which treats the same problem as ourselves but disagrees with the above result. These authors claim to include photon cascades and yet arrive at a spectrum which is in exact agreement with Stroud's one-photon result. From the information available in their short publication it appears to the present authors that Smithers and Freedhoff

have not managed to include true photon cascades but have simply followed a series of sequential one-photon emissions as treated by Stroud; hence their exact agreement with Stroud's spectrum.

We have been able to indicate here the consequences of omitting to recognize photon cascades in the treatment of the fluorescence problem. In conclusion we should point out however that in making a one-photon approximation we have not managed an identification with Stroud's results in every detail. Our equation (22) agrees with Stroud (1971) so far as relative peak heights and widths are concerned; it does not however support Stroud's prediction of linewidth narrowing. This is a point to which further consideration must yet be given.

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