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ADDITIONAL SIDEBANDS IN TIME DEPENDENT SPECTRA FOR A SPACIALLY DISTRIBUTED COLLECTION OF TWO-LEVEL ATOMS DRIVEN BY A RESONANT COHERENT FIELD

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We derive the time-dependent spectra of a laser-driven, interacting, collection of spacially distributed two-level atoms. We assume a system with dimensions large relative to a wavelength which is initially in the atomic and electromagnetic field ground state and at the initial time begins to interact with the coherent, externally applied radiation field. The explicit spacial distribution of the two-level atoms affects the time-dependent spectra of the transmitted radiation and the many-body interaction leads to the appearance of additional sidebands.

The fluorescence spectrum of a system of a large number of two-level atoms driven by a strong resonant field has been treated for atoms that are contained in a region that is small relative to a wavelength. For high laser intensities, resonances displaced from the line center by twice the Rabi frequency were predicted [1–3] in addition to the usual Stark sidebands [4,5]. The intensity of the additional sidebands in resonance fluorescence experiments is expected to be very weak and, so far, they have not been observed.

In this communication, we treat the appearance of additional sidebands in a time-dependent spectrum [6–9] for a nonstationary state of the two-level system. We follow here a previous model [10] which is based upon a quantum electrodynamical representation of a spacial distribution of a large number of two-level atoms interacting with the quantized electromagnetic field. In the previous work [10] we studied the steady-state conditions for this system. We picture that different individual atoms follow Rabi oscillations with the same frequency but different phases so that by ensemble-averaging we get many atom steady states. In the present work we assume a sample of two-level atoms with dimensions very large relative to a wavelength which is initially in the ground state and at time $t = 0$ begins to interact with the coherent externally applied resonant radiation field. We study the spectrum of the radiation transmitted through the sample of two-level atoms and show that the explicit spacial distribution of the atoms affects the shape of the radiation spectrum and can lead to the appearance of additional sidebands [11]. The additional sidebands should follow from modulation of the collective dipole oscillations at frequency $2|\omega_R|$ in addition to modulation at frequency $|\omega_R|$ which leads to the fundamental bands. We simplify the present treatment for time-dependent spectra by assuming that the filter time [6–9] is short relative to the relaxation and dephasing times, but long relative to the period time of Rabi oscillations. Under these conditions we get a quasistationary spectrum which is damped slowly in time. In the present approach we describe the time-dependent collective dipole oscillations by using a semiclassical model and treat the nonlinear terms by a perturbative approach in which the reaction field is taken as small relative to the externally applied field. Following the above considerations, the present analysis is good as long as the magnitude of the collective oscil-

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lations is large relative to other sources of fluctuations including those related to the Langevin terms that represent quantum fluctuations.

The hamiltonian which describes the system is given in the rotating wave and in the electric dipole approximation by

$$H = H_0 + H' ,$$

$$H_0 = \tfrac{1}{2}\hbar\omega \sum_i^N \sigma_{ii}^{(i)} + \hbar \sum_k \omega_k a_k^\dagger a_k ,$$

$$H' = -\frac{i\hbar}{2} \sum_k \sum_i^N g_k^{(i)} a_k \sigma_{+-}^{(i)} \exp(i\boldsymbol{k}\cdot\boldsymbol{r}_i)$$

$$-\frac{i\hbar}{2} \sum_i^N \omega_R \sigma_{+-}^{(i)} \exp[-(i\omega_0 t - \boldsymbol{k}\cdot\boldsymbol{r}_i)] + \text{h.c.} , \quad (1)$$

where h.c. denotes hermitian conjugate and

$$g_k^{(i)} = (2\pi\omega_k/\hbar V)^{1/2} P \hat{\boldsymbol{p}}_i \cdot \hat{\boldsymbol{x}} .$$

H_0 includes the free atoms and free field hamiltonians. We use here the usual operators related to the SU(2) algebra for a single atom: $\sigma_{ii}^{(i)}$ represents the inversion of population operator and $\sigma_{+-}^{(i)}$ are the raising and lowering operators for atom i with coordinates r_i. ω_R is the Rabi rate associated with the applied coherent field amplitude. ω_0 and k are the carrier frequency and wavevector of the applied field, respectively. V is the quantization volume for the field. ω_k is the frequency of the mode k, and $\hat{\boldsymbol{x}}$ is a unit vector in the direction of polarization of the applied radiation field that is being taken to be in a coherent state and propagating along the z-axis. P is the dipole matrix element and $\hat{\boldsymbol{p}}_i$ is the unit vector for the dipole of atom i.

In the previous work [10] we developed the Heisenberg equations of motion in the bad cavity limit for this system. The rapid time and spacial dependence of the dipole operators has been eliminated by the transformation:

$$\sigma_{+0}^{(i)}(t) = \sigma_{+-}^{(i)}(t) \exp[-i(\omega_0 t - \boldsymbol{k}_0\cdot\boldsymbol{r}_i)] . \quad (2)$$

By using the coupled hierarchy of the Heisenberg equations of motions, and by adiabatically eliminating the variables associated with the field modes one gets terms in the equations of "spontaneous", "cooperative", "induced", and "Langevin force" origin. While the "spontaneous", "induced" and "Langevin force" terms depend only on single atom response, the "cooperative" terms include factors of operators belonging to pairs of different atoms with arguments expressive of retardation. The basic equations are obtained by taking the expectation values and factorizing the multiplication of the dipole operators. Factorization is a justifiable approximation in a many-body theory as the correlation between each pair of atoms tends to be overwhelmed by the total interaction of each atom with the many-atom system. The basic equations for a long sample of two-level atoms with dimensions larger than a wavelength are obtained [10] by describing the expectation values of the atomic operators as continuous variables and defining $\langle \sigma_{+0}(\boldsymbol{r}, t) \rangle$ and $\langle \sigma_{ii}(\boldsymbol{r}, t) \rangle$, respectively, as the complex dipole and the inversion of population per unit volume at point $\boldsymbol{r}$. Following the previous work [10] we get the following equations

$$d\langle \sigma_{ii}(\boldsymbol{r}, t) \rangle / dt = -\beta_1 [\langle \sigma_{ii}(\boldsymbol{r}, t) \rangle + n]$$

$$- i[2\theta^*(\boldsymbol{r}, t) + \omega_R] \langle \sigma_{+0}(\boldsymbol{r}, t) \rangle + \text{c.c.} , \quad (3)$$

$$d\langle \sigma_{+0}(\boldsymbol{r}, t) \rangle / dt = (i\Delta - \beta_2) \langle \sigma_{+0}(\boldsymbol{r}, t) \rangle$$

$$+ [\theta(\boldsymbol{r}, t) + \omega_R^*/2] \langle \sigma_{ii}(\boldsymbol{r}, t) \rangle , \quad (4)$$

where $\theta(\boldsymbol{r}, t)$ is a continuous variable corresponding to the reaction field and is given by [10]

$$\theta(\boldsymbol{r}, t) = (3\pi\beta/k^3)$$

$$\times \int_0^\infty dz' \langle \sigma_{+0}(z', t - (z - z')/c) \rangle , \quad (5)$$

where n is the number of two-level atoms per unit volume and $\Delta = \omega - \omega_0$ is the deviation of the applied field frequency from resonance. Self-consistently from the quantum model $\beta_1 = \beta$, $\beta_2 = \beta/2$ where β is the spontaneous decay constant. We may introduce additional homogeneous broadening by considering β_1 and β_2 as empirical constants. Eqs. (3) – (5) lead to a set of equations which are of the form of the usual Maxwell Bloch equations if we perform the space derivative in eq. (5) and neglect the renormalization of the resonance frequency caused by coherent dipole-dipole interaction [10].

We calculate the time dependence of the total electromagnetic field transmitted at the end of a short sample for which the conditions

$|\theta(r, t)| \ll |\omega_R|, \quad \beta_{1,2} \ll |\omega_R|$,

are fulfilled. Under these conditions the reaction field is given approximately by

$$\theta(r, t) \simeq \alpha \langle \sigma_{-0}(z, t) \rangle , \tag{6}$$

where

$$\alpha = (3\pi\beta/k^2) z . \tag{7}$$

In order to explain further the use of eq. (6), we substitute this equation in the ordinary Maxwell equation:

$$c^{-1} \partial\theta(z, t)/\partial t + \partial\theta(z, t)/\partial z = (3\pi\beta/k^2) \langle \sigma_{-0}(z, t) \rangle \tag{8}$$

and find that eq. (6) is valid if

$$(\partial \langle \sigma_{-0}(z, t) \rangle / \partial z)_t \simeq 0 , \tag{9a}$$

or more explicitly if consistent with our first order perturbation theory:

$$|z[\partial \log \langle \sigma_{-0} \rangle / \partial z]_t| \ll 1 . \tag{9b}$$

where $t' = t - z/c$ is the retarded time. Since α is a small parameter, the derivative of $\langle \sigma_{-0} \rangle$ relative to z will contribute only to second order corrections which can be neglected. The restriction of the discussion to a strong field in which our approximations are valid simplifies the present treatment for the appearance of the additional sidebands.

By assuming exact resonance ($\Delta = 0$) and substituting eq. (6) into eqs. (3) and (4), we get

$$\langle \dot{\sigma}_z \rangle = -\beta_1 [\langle \sigma_z \rangle + n] - \omega_R \langle \sigma_{-0} \rangle - \omega_R^* \langle \sigma_{+0} \rangle - 4\alpha \langle \sigma_{-0} \rangle \langle \sigma_{+0} \rangle , \tag{10}$$

$$\langle \dot{\sigma}_{-0} \rangle = -\beta_2 \langle \sigma_{-0} \rangle + \tfrac{1}{2} \omega_R^* \langle \sigma_z \rangle + \alpha \langle \sigma_{-0} \rangle \langle \sigma_z \rangle , \tag{11}$$

where the dot represents the time derivative $\partial/\partial t'$. Here we have used a short notation in which it is understood that all expectation values are functions of retarded time t' and at position z. The explicit dependence of the expectation values on z enters only via the parameter α.

Assuming $\beta_{1,2} \ll |\omega_R|$, $\alpha n \ll |\omega_R|$, we solve eqs. (10) and (11) by iterative substitutions in which we neglect all terms which are of second order or higher in α and $\beta_{1,2}$. We get

$$\langle \ddot{\sigma}_z \rangle + |\omega_R|^2 \langle \sigma_z \rangle - 3\alpha \langle \sigma_z \rangle \langle \dot{\sigma}_z \rangle + (\beta_1 + \beta_2) \langle \dot{\sigma}_z \rangle = 0 , \tag{12}$$

with the first-order solution

$$\langle \sigma_z(t) \rangle = \sigma_{z0} \cos(|\omega_R| t) \exp\{-[(\beta_1 + \beta_2/2)]t'\} + \tfrac{1}{2}(\alpha/|\omega_R|)\sigma_{z0}^2 \sin(2|\omega_R| t) \exp[-(\beta_1 + \beta_2)t'] . \tag{13}$$

We have assumed here that $\langle \sigma_z(\infty) \rangle = 0$, and $\langle \dot{\sigma}_z(\infty) \rangle = 0$ for a strong field at exact resonance.

Eqs. (12) and (13) derived here are similar to the equations derived by Senitzky [1]. The assumption of conservation of total angular momentum made by him is not valid in our treatment due to the damping mechanism. The parameter α derived here is for a sample with dimensions large relative to a wavelength. It depends on z via eq. (7) and is essentially different from a corresponding parameter α derived in ref. [1] for a sample with dimensions smaller than a wavelength. The broadening mechanism which was neglected there [1] is introduced here via the parameter $\beta_1 + \beta_2$.

The solution for $\langle \sigma_{-0}(t) \rangle$ of first order is given by

$$\langle \sigma_{-0} \rangle = \tfrac{1}{2} \omega_R^* \sigma_{z0} \exp\{-[(\beta_1 + \beta_2)/2]t'\} \times \left(\frac{\sin(|\omega_R| t')}{|\omega_R|} + \frac{\beta_1 - \beta_2}{2} \frac{1 - \cos(|\omega_R| t')}{|\omega_R|^2} \right) + \alpha\sigma_{z0}^2 (\omega_R^*/2) \exp[-\beta_1 + \beta_2)t'] \times \left(\frac{1 - \cos(2|\omega_R| t')}{2|\omega_R|^2} \right) . \tag{14}$$

According to eqs. (13) and (14) the oscillation is modulated at frequency $2|\omega_R|$ in addition to the modulation at frequency $|\omega_R|$.

Assuming that the CW laser is activated at $t' = 0$ and that the two-level atoms are initially in the lower state, the time-dependent spectrum is given by [7]

$$S=4\gamma\,\mathrm{Re}\int_0^{t'}\mathrm{d}\tau\,\exp[(\gamma-\mathrm{i}\Delta\omega)\tau]$$

$$\times\int_0^{t'}\mathrm{d}t_2\,\exp[-2\gamma(t-t_2)]$$

$$\times\langle\langle E^*(t+\tau)\,E(t_2)\rangle\rangle\,, \tag{15}$$

where, for our case

$$E(t)=[\omega_R+2\theta^*(z,t)]\hbar/P\,, \tag{16}$$

and $1/\gamma$ is the filter time.

By assuming the conditions

$$\beta_{1,2}\ll\gamma\ll\omega_R\,, \tag{17}$$

the ensemble average in eq. (15) refers to averaging over the Rabi oscillations where the electromagnetic field at time t' is obtained by using eqs. (6), (14), and (16). We notice that the zeroth order and first-order value for the complex dipole introduce, respectively, first-order and second-order corrections to the zeroth order electromagnetic field which is proportional to ω_R. We can distinguish in the expression for the electromagnetic field between terms that are constants, those that oscillate at frequency $|\omega_R|$ and those that oscillate at frequency $2|\omega_R|$. Due to the ensemble averaging over the Rabi oscillations the terms in the correlation function which couple different oscillation frequencies can be neglected.

In order to simplify the expression for the correlation function we include in each term that oscillates at a given frequency only that part which results from the lowest order in perturbation theory. Therefore we neglect the first- and second-order corrections to the constant term ω_R and we neglect the second-order correction to the terms modulated at frequency $|\omega_R|$.

Following these considerations we obtain

$$\langle\langle E(t_2+\tau)^*E(t_2)\rangle\rangle=(\hbar^2/P^2)|\omega_R|^2$$

$$\times\{1+\tfrac{1}{2}\delta^2\cos(|\omega_R|\tau)\exp[-(\beta_1+\beta_2)(t_2+\tau/2)]$$

$$+\tfrac{1}{8}\delta^4\exp[-(\beta_1+\beta_2)(2t_2+\tau)]\cos(2|\omega_R|\tau)\}\,, \tag{18}$$

where

$$\delta=\alpha\sigma_{10}/|\omega_R|\,. \tag{19}$$

In evaluating the spectrum according to eqs. (15) and (18) we use the approximation $\beta_{1,2}\ll\gamma$ and neglect terms which include exponents with arguments as products of τ and β_1 or β_2 but take into account terms which include exponents having arguments as multiplications of t' and β_1 or β_2 as they will affect the decay of the spectra for long times t'. Also we develop the expression for times $t'>1/\gamma$ so that $\exp(-2\gamma t')\to 0$. By following these approximations [7] we obtain by a straightforward calculation

$$S=\frac{2|\omega_R|^2\hbar^2}{P^2}\left\{\frac{\gamma}{(\omega-\omega_0)^2+\gamma^2}\right.$$

$$+\tfrac{1}{4}\delta^2\exp[-(\beta_1+\beta_2)t']$$

$$\times\left[\frac{\gamma}{(\omega-\omega_0+|\omega_R|)^2+\gamma^2}\right.$$

$$\left.+\frac{\gamma}{(\omega-\omega_0-|\omega_R|)^2+\gamma^2}\right]$$

$$+\tfrac{1}{16}\delta^4\exp[-2(\beta_1+\beta_2)t']$$

$$\times\left[\frac{\gamma}{(\omega-\omega_0+2|\omega_R|)^2+\gamma^2}\right.$$

$$\left.\left.+\frac{\gamma}{(\omega-\omega_0-2|\omega_R|)^2+\gamma^2}\right]\right\}\,. \tag{20}$$

Eq. (20) shows that within our approximations the time-dependent spectra for the transmitted radiation includes the laser line, and the fundamental and the additional sidebands with lorentzian line shapes whose centers are located, respectively, at ω_0, $\omega_0\pm|\omega_R|$ and $\omega_0\pm 2|\omega_R|$. Since according to our conditions γ dominates the correlation function, the width of each band is fixed by the filter time $1/\gamma$ and is different from the Mollow line shape in which the width of each line is related to β. The lorentzian fundamental and additional sidebands are decaying, respectively, with the rates $\beta_1+\beta_2$ and $2(\beta_1+\beta_2)$. By our perturbative theory we find that the ratio of the intensity of the fundamental sideband to the monochromatic laser line, and that of the additional sideband relative to the fundamental is given by $\delta^2/4$ (similar to a relation found in ref. [1]).

In a dressed state approach for a two-level system,

one gets shifted energy levels with the corresponding frequencies ω_0, $\omega_0 \pm |\omega_R|$. The additional sidebands result from transitions in which the dipole moment is doubled. In order that such transitions occur, the dipoles of different atoms must have a non-negligible probability to oscillate in phase with one another. While the probability for this kind of transition is very small for the many-atom steady states, it becomes relatively strong in the case treated here due to the transient cooperation induced by our initial conditions. For long times the system tends to a steady state for which the relative phases of different dipoles become randomized. Therefore we suggest that the observation of additional sidebands can be made in a time-dependent spectrum.

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