

Two-atom resonance fluorescence*

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The excitation spectrum arising from the interaction between two identical atoms (molecules), one of which is excited in the presence of a resonant strong electromagnetic field (pump field), is investigated. The excitation spectrum is found to consist of those describing the symmetric and antisymmetric modes, respectively. The form of both spectra depends on the relation between the distance R separating the atoms and the wavelength λ of the transition from the ground state to the excited state. For $R < \lambda$, and when certain conditions prevail, the spectral function for the symmetric modes consists of three Lorentzian lines describing the central peak and the two sidebands whose radiative widths are equal to γ_0 , $3\gamma_0/2$, and $3\gamma_0/2$, which are twice the corresponding ones arising from an isolated single atom interacting with the pump field. For the antisymmetric modes the central peak has a δ -function distribution indicating the stability of the mode in question, while, when certain conditions are satisfied, the two sidebands are described by Lorentzian lines each having a radiative width of the order of $\gamma_0/2$, which is equal to the natural linewidth for a photon spontaneously emitted from an isolated atom. For both the symmetric and antisymmetric modes, the dipole-dipole interaction between the atoms brings about small energy shifts. For $R > \lambda$, apart from the small energy shifts caused by the dipole-dipole interaction, the spectral functions for symmetric and antisymmetric modes are similar to that for the single atom interacting with the pump field.

I. INTRODUCTION

Dicke¹ first pointed out that changes in the lifetimes should be expected when two atoms, one of which is excited, are separated by a distance $R < \lambda$, where λ is the wavelength of the transition from the ground state to the excited state. He considered¹ the initial state of the system where one atom is excited while the other is in its ground state. This initial state may be taken as a superposition of states which are symmetric and antisymmetric with respect to interchange of the atoms; these states are formally analogous to the singlet and triplet states of two spins. The antisymmetric state is stable while the symmetric state has a lifetime one-half that of the isolated atom. These states have been called by Dicke¹ super-radiant states. A detailed calculation for the energy of interaction between two similar atoms (molecules) has been given by Stephen² and also by Hutchinson and Hamerka³ and McLachlan.⁴

The spectrum of the light scattered by an isolated atom driven at resonance by a strong electromagnetic field consists of three peaks: a central peak at the excitation frequency and two symmetrically placed sidebands. This so-called dynamic, or ac, Stark effect has been theoretically predicted by Mollow⁵ and observed experimentally by Schuda *et al.*⁶ The three peaks are described by Lorentzian lines⁵⁻¹² whose radiative widths are $\frac{1}{2}\gamma_0$, $\frac{3}{4}\gamma_0$, and $\frac{3}{4}\gamma_0$, respectively, where $\frac{1}{2}\gamma_0$ is the natural linewidth for a photon spontaneously emitted from an isolated atom. The ratio of the central peak height to the heights of the sidebands is 3:1.⁵⁻¹²

There have been many theoretical treatments of the subject in question and we refer to a recent one by Kimble and Mandel¹³ where details as well as references can be found.

Collective atomic effects in resonance fluorescence have recently been discussed by Agarwal *et al.*¹⁴ Using the master equation, the scattered light spectrum from two- and three-atom collective systems has been calculated and compared with the one-atom spectrum. The differences are found¹⁴ to be significant for weak fields but become less pronounced at high intensities of the driving field. The effects arising from the dipole-dipole interaction between the atoms have not been considered.¹⁴ The purpose of the present study is to investigate the excitation spectrum arising from the interaction between two identical atoms, one of which is excited in the presence of a strong resonant electromagnetic field.

The problem is formulated in Sec. II where a model Hamiltonian has been considered describing the physical processes where two identical two-level atoms interact between themselves through the dipole-dipole interaction as well as with two fields, a strong resonant (pump) and a weak photon field, respectively. This Hamiltonian is then used to derive the equations of motion for the Green's functions describing the symmetric and antisymmetric modes, respectively. Using a decoupling approximation to truncate the hierarchy of the Green's functions that appear in the equations of motion, expressions are derived for the Green's functions of the symmetric and antisymmetric modes. The derived formulas for the Green's

functions are reduced to those known in the literature in the limits in which either there is no resonant pump field or only a single isolated atom interacts with the pump field.

The excitation spectrum of the symmetric and antisymmetric modes is considered in Sec. III. The corresponding spectral functions have been calculated and discussed for the limiting cases when the atoms are close together $R < \lambda$ (wave-zone region) or when they are far apart, $R > \lambda$. A detailed discussion of the spectra is given in Sec. IV.

II. FORMULATION OF THE PROBLEM

We consider a simple model consisting of two identical atoms (molecules) A and B a distance R apart. The atoms are assumed to have two non-degenerate electronic states, the ground and the excited state, denoted by 0 and ν , respectively. The spectroscopic transition $0 \rightarrow \nu$ is electric-dipole allowed and the transition frequency is denoted by $E_{\nu 0} = E_\nu - E_0$; units in which $\hbar = 1$ are used throughout. The distance between the atoms is taken such that the overlap of the wave functions is very small and may be neglected and the interaction between the atoms is of the dipole-dipole type. Effects associated with the translational motion of the atoms such as recoil, etc., are neglected. The atoms are resonantly pumped by a strong-field mode at a frequency $\omega_0 = E_{\nu 0}$, and simultaneously coupled to the remaining modes of the electromagnetic field, these being initially unpopulated. The Hamiltonian for such a system may be taken to be

$$\mathcal{H} = \mathcal{H}_s + \mathcal{H}_c + \mathcal{H}_r + \mathcal{H}_{sr}, \quad (1)$$

where

$$\begin{aligned} \mathcal{H}_s = & \omega_0 (b_A^\dagger b_A + b_B^\dagger b_B + \beta_0^\dagger \beta_0) \\ & + \frac{1}{2} i \omega_p \sqrt{f} [(b_A + b_B) \beta_0^\dagger - (b_A^\dagger + b_B^\dagger) \beta_0], \end{aligned} \quad (2)$$

$$\mathcal{H}_c = V_{AB} (b_A^\dagger b_B + b_B^\dagger b_A), \quad (3)$$

$$\mathcal{H}_r = \sum_{\vec{k}, \lambda} c k \beta_{\vec{k}, \lambda}^\dagger \beta_{\vec{k}, \lambda}, \quad (4)$$

$$\begin{aligned} \mathcal{H}_{sr} = & \frac{1}{2} i \omega_p \sum_{\vec{k}, \lambda} \left(f_{\omega}(\vec{k}, \lambda) \frac{E_{\nu 0}}{c k} \right)^{1/2} \\ & \times [(e^{-i\vec{k} \cdot \vec{R}_A} b_A + e^{i\vec{k} \cdot \vec{R}_B} b_B) \beta_{\vec{k}, \lambda}^\dagger \\ & - (e^{i\vec{k} \cdot \vec{R}_A} b_A^\dagger + e^{-i\vec{k} \cdot \vec{R}_B} b_B^\dagger) \beta_{-\vec{k}, \lambda}], \end{aligned} \quad (5)$$

$$V_{AB} = V_{AB}(R) = \frac{\vec{P}_A \cdot \vec{P}_B}{R^3} - \frac{3(\vec{P}_A \cdot \vec{R})(\vec{P}_B \cdot \vec{R})}{R^5} \quad (6)$$

with $R = R_{AB}$. \vec{P}_A and \vec{P}_B designate the transition dipole moments to the excited states of the atoms

(molecules) A and B , respectively, $f = f_{0\nu}$ is the oscillator strength for the electronic transition $0 \rightarrow \nu$ and ω_p is the plasma frequency. β_0^\dagger and β_0 are the boson creation and annihilation operators for the pump mode $\omega_0 = E_{\nu 0}$, while $\beta_{\vec{k}, \lambda}^\dagger$ and $\beta_{\vec{k}, \lambda}$ are the corresponding ones describing the electromagnetic field with wave vector \vec{k} , frequency ck , and transverse photon polarization $\lambda (= 1, 2)$. The compound operators b_A^\dagger and b_A are defined as $b_A^\dagger = \alpha_{A\nu}^\dagger \alpha_{A0}$ and $b_A = \alpha_{A0}^\dagger \alpha_{A\nu}$, where $\alpha_{A\nu}^\dagger$ and α_{A0} are the creation and annihilation operators describing the electron states ν and 0 , respectively, and satisfy Fermi statistics; effects resulting from the electron spin are neglected. The operators b_A^\dagger and b_A describe the creation and annihilation of excitations of the atomic field, and for the two-level system under consideration, satisfy Pauli statistics; the operators b_A^\dagger and b_A commute with those of b_B^\dagger and b_B .

The first three terms in Eq. (2) describe the free fields of the atom A , the atom B , and that of the strong pump field, while the remaining ones represent the coupling between them. Eq. (3) describes the Coulomb (dipole-dipole) interaction^{2,4} between the atoms A and B . For the sake of simplicity, a term in Eq. (3) of the form $V_{AB}(b_A^\dagger b_B^\dagger + b_B b_A)$, which describes the simultaneous creation or annihilation of two excitations, has been neglected. Eqs. (4) and (5) represent the free electromagnetic field and its coupling with the atomic fields A and B , respectively. This photon field, which may be considered to be the weak field in comparison with the strong pump field, can be either an external field or the field produced by the radiating atoms themselves. If the terms describing the atomic field B are discarded in Eqs. (1)–(6), then the remaining terms are identical to those used in the literature⁸ to study the dynamic Stark effect for a single two-level atomic system. We are interested in the physical processes that occur when a strong pump field acts at resonance $\omega_0 = E_{\nu 0}$ on the atomic systems A and B , and therefore our results will be valid in this extreme limit.

We shall make use of the retarded double-time Green's functions defined by^{15,16}

$$\langle\langle X(t); Y(t') \rangle\rangle = -i \theta(t - t') \langle [X(t), Y(t')]_{-\eta} \rangle, \quad (7a)$$

where

$$\langle U \rangle = \text{tr} U e^{-\beta \mathcal{H}} / \text{tr} e^{-\beta \mathcal{H}}, \quad \beta = (k_B T)^{-1}, \quad (7b)$$

k_B is Boltzman's constant, T is the absolute temperature, and \mathcal{H} is the total Hamiltonian of the system. $\theta(t)$ is the usual step function and the operators are in the Heisenberg representation and η is taken to be either $+1$ or -1 depending upon considerations of convenience. The equation of

motion of the Green's function $\langle\langle X(t); Y(t') \rangle\rangle_{(\omega)}$ is given by

$$\omega \langle\langle X(t); Y(t') \rangle\rangle_{(\omega)} = (1/2\pi) \langle[X(t), Y(t)]_{-\eta}\rangle + \langle\langle [X(t), \mathcal{H}]; Y(t') \rangle\rangle_{(\omega)}. \quad (8)$$

The subscript (ω) as well as the time arguments of the operators will be suppressed for convenience.

If we define the Fourier transform of the Green's function as $G_{AB}^{(\pm)}(\omega) = \langle\langle b_A \pm b_B; b_A^\dagger \rangle\rangle$ then using Eq. (8) and the Hamiltonian (1) we derive the equation of motion

$$\left(\omega - \omega_{\pm} - \frac{1}{2}\gamma_{\pm} - \frac{\omega_p^2 f(1 \pm 1)}{4(\omega - \omega_0)}\right) G_{AB}^{(\pm)}(\omega) = \frac{(1 - 2n_p)}{2\pi} + i\omega_p \sqrt{f} \langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_0; b_A^\dagger \rangle\rangle, \quad (9)$$

where

$$\omega_{\pm} = \omega_0 \pm V_{AB}, \quad (10)$$

$$\gamma_{\pm} = \gamma \pm \gamma_{AB}, \quad (11)$$

$$\gamma_{AB} \equiv \gamma_{AB}(\omega) = \frac{1}{2}\omega_p^2 \sum_{\vec{k}, \lambda} \frac{f_{0\nu}(\vec{k}, \lambda)}{(\omega - ck)} \frac{\omega_0}{ck} \times \exp[i(\vec{k} \cdot \vec{R}_{AB})], \quad (12)$$

with $n_p = \langle b_A^\dagger b_A \rangle = \langle b_B^\dagger b_B \rangle$. The expression for $\gamma = \gamma_{AA}(\omega) = \gamma_{BB}(\omega)$ can be obtained from γ_{AB} Eq. (12), if we replace the exponential $\exp i(\vec{k} \cdot \vec{R}_{AB})$ by unity. In deriving Eq. (9), we have discarded the Green's function $\langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_{\vec{k}\lambda}; b_A^\dagger \rangle\rangle$ as being unimportant compared to $\langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_0; b_A^\dagger \rangle\rangle$. This is in accordance with our original assumption that the pump field is the strongest of the two. Using the Hamiltonian (1) and Eq. (8), we derive the equation of motion for the Green's function $\langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_0; b_A^\dagger \rangle\rangle$ as

$$\begin{aligned} (\omega - \omega_0 - \gamma) \langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_0; b_A^\dagger \rangle\rangle \\ = -\frac{1}{2}i\omega_p \sqrt{f} \langle\langle (b_A \pm b_B) \beta_0^{\dagger} \beta_0; b_A^\dagger \rangle\rangle \\ + \left[\frac{1}{2}\gamma_{AB}(1 \pm 1) + V_{AB}(1 \mp 1)\right] \langle\langle (b_A^\dagger b_B \pm b_B^\dagger b_A) \beta_0; b_A^\dagger \rangle\rangle, \end{aligned} \quad (13)$$

where a term of the form

$$-\frac{1}{2}i\omega_p \sqrt{f} \langle\langle (b_A \pm b_B) \beta_0 \beta_0 - (b_A^\dagger b_A b_B \pm b_B^\dagger b_B b_A); b_A^\dagger \rangle\rangle \quad (14)$$

has been omitted on the right-hand side of Eq. (13). The first term in Eq. (14) describes the physical process where two photons of the pump field act simultaneously on the atomic systems A and B , while the last one represents the analogous process arising from the action of the dipole atomic

field A and B on the number density operators $b_B^\dagger b_B$ and $b_A^\dagger b_A$, respectively. Both processes are much less important than that caused by the photon density of the pump field acting on the atomic systems A and B and described by the Green's function $\langle\langle (b_A \pm b_B) \beta_0^{\dagger} \beta_0; b_A^\dagger \rangle\rangle$.

In the same spirit we derive the following expressions for the Green's functions:

$$\begin{aligned} (\omega - \omega_0 - \gamma) \langle\langle (b_A^\dagger b_B \pm b_B^\dagger b_A) \beta_0; b_A^\dagger \rangle\rangle \\ = \mp \frac{1}{2}i\omega_p \sqrt{f} \langle\langle (b_A \pm b_B) \beta_0^{\dagger} \beta_0; b_A^\dagger \rangle\rangle \\ + [\gamma_{AB} \frac{1}{2}(1 \pm 1) + V_{AB}(1 \mp 1)] \\ \times \langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_0; b_A^\dagger \rangle\rangle, \end{aligned} \quad (15)$$

$$\begin{aligned} (\omega - \omega_{\pm} - \frac{1}{2}\gamma_{\pm}) \langle\langle (b_A \pm b_B) \beta_0^{\dagger} \beta_0; b_A^\dagger \rangle\rangle \\ = \frac{(1 - 2n_p)}{2\pi} n_0 + 2(i\Omega \sqrt{n_0}) \langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_0; b_A^\dagger \rangle\rangle \\ + \frac{\Omega^2(1 \pm 1)}{2(\omega - \omega_0)} G_{AB}^{(\pm)}(\omega), \end{aligned} \quad (16)$$

where

$$\Omega = \omega_p (f n_0)^{1/2}, \quad (17a)$$

and

$$n_0 = \langle \beta_0^{\dagger} \beta_0 \rangle \quad (17b)$$

are the energy shift and the average number density of the photon pump field, respectively. In deriving Eq. (16) use has been made of the following decoupling approximations:

$$\begin{aligned} \langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_0^{\dagger} \beta_0; b_A^\dagger \rangle\rangle \\ \approx 2n_0 \langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_0; b_A^\dagger \rangle\rangle, \end{aligned} \quad (18a)$$

$$\begin{aligned} \langle\langle \beta_0^{\dagger} \beta_0 \beta_0; b_A^\dagger \rangle\rangle \approx 2n_0 \langle\langle \beta_0; b_A^\dagger \rangle\rangle \\ = [i\omega_p \sqrt{f}] / (\omega - \omega_0) n_0 G_{AB}^{(+)}(\omega). \end{aligned} \quad (18b)$$

This decoupling approximation indicates that all dynamic effects arising from photon-photon interactions for the pump field are completely discarded and the photon in question sees only the average field of all others (static effect) which is described by the average number of photons $n_0 = \langle \beta_0^{\dagger} \beta_0 \rangle$. This approximation is asymptotically valid for large values of ω and high photon densities of the pump field.

Solving Eqs. (13), (15) and (16), we obtain

$$\begin{aligned} [(\omega - \omega_0 - \gamma_+)(\omega - \omega_{\pm} - \frac{1}{2}\gamma_{\pm}) - \Omega^2] \langle\langle (b_A^\dagger b_A \pm b_B^\dagger b_B) \beta_0; b_A^\dagger \rangle\rangle \\ = -\frac{1}{2}i\omega_p \sqrt{f} \left(\frac{(1 - 2n_p)}{2\pi} n_0 + \frac{\Omega^2}{(\omega - \omega_0)} G_{AB}^{(+)}(\omega) \right), \end{aligned} \quad (19)$$

$$[(\omega - \omega_0 - \gamma + 2V_{AB})(\omega - \omega_- - \frac{1}{2}\gamma_-) - \Omega^2] \\ \times \langle\langle (b_A^\dagger b_A - b_B^\dagger b_B)\beta_0; b_A^\dagger \rangle\rangle = -\frac{1}{2}i\omega_p \sqrt{f} \frac{(1-2n_\nu)}{2\pi} n_0 \quad (20)$$

Substituting Eq. (19) into Eq. (9) and retaining only those energy shifts induced by the pump field which are of the order of Ω , i.e., considering the limit in which $n_0 \gg 1$, we have

$$G_{AB}^{(+)}(\omega) = \frac{(1-2n_\nu)}{2\pi(\omega - \omega_+ - \frac{1}{2}\gamma_+)} \\ \times \left(1 + \frac{\frac{1}{2}\Omega^2}{(\omega - \omega_0 - \gamma_+)(\omega - \omega_+ - \frac{1}{2}\gamma_+) - \Omega^2} \right), \quad (21)$$

which describes the spectrum of the symmetric modes, while substitution of Eq. (20) into Eq. (9) yields

$$G_{AB}^{(-)}(\omega) = \frac{(1-2n_\nu)}{2\pi(\omega - \omega_- - \frac{1}{2}\gamma_-)} \left(1 + \frac{\frac{1}{2}\Omega^2}{(\omega - \omega_0 - \gamma + 2V_{AB})(\omega - \omega_- - \frac{1}{2}\gamma_-) - \Omega^2} \right), \quad (22)$$

the spectrum of the antisymmetric modes. The Green's functions (21) and (22) will be used in Sec. III to discuss the excitation spectrum of the system under investigation.

III. EXCITATION SPECTRUM

In the absence of the pump field, i.e., in the limit in which $\Omega=0$ ($n_0=0$), Eqs. (21) and (22) are reduced to

$$G_{AB}^{(\pm)}(\omega) = \frac{(1-2n_\nu)}{(2\pi)(\omega - \omega_\pm - \frac{1}{2}\gamma_\pm)}, \quad (23)$$

which describes the excitation spectrum of the symmetric (+) and antisymmetric (-) energy modes for the physical process arising from the interaction of two similar atoms one of which is excited. When the appropriate limits are taken for $R < \lambda$ and $R > \lambda$, where $\lambda = (c/E_{\nu 0})$, the results obtained from Eq. (23) for the spectrum of the symmetric and antisymmetric modes are identical to those known in the literature.²⁻⁴

In the absence of the atom B , i.e., when $\gamma_{AB}=0$ and $V_{AB}=0$, then Eqs. (21) and (22) become identical, $G_{AB}^{(+)}(\omega) = G_{AB}^{(-)}(\omega) = G_{AA}(\omega)$, which is given by

$$G_{AA}(\omega) = \frac{(1-2n_\nu)}{2\pi(\omega - \omega_0 - \frac{1}{2}\gamma)} \\ \times \left(1 + \frac{\frac{1}{2}\Omega^2}{(\omega - \omega_0 - \gamma)(\omega - \omega_0 - \frac{1}{2}\gamma) - \Omega^2} \right), \quad (24)$$

which may be rewritten as

$$G_{AA}(\omega) = \frac{1-2n_\nu}{2\pi} \left[\frac{1 - \Omega^2/2(\Omega^2 - \frac{1}{16}\gamma^2)}{\omega - \omega_0 - \frac{1}{2}\gamma} + \frac{\Omega}{4(\Omega^2 - \frac{1}{16}\gamma^2)} \right. \\ \left. \times \left(\frac{\Omega - \frac{1}{4}\gamma}{\omega - \omega_0 - \Omega - \frac{3}{4}\gamma} + \frac{\Omega + \frac{1}{4}\gamma}{\omega - \omega_0 + \Omega - \frac{3}{4}\gamma} \right) \right]. \quad (25)$$

In calculating $\gamma \equiv \gamma(\omega)$ from Eq. (12) with $\exp(\vec{k} \cdot \vec{R}_{AB}) = 1$, we discard the real part and consider only the imaginary part so that

$$\gamma(\omega) - i\text{Im}\gamma(\omega) = -i\gamma_0, \quad (26)$$

where

$$\gamma_0 = \frac{4}{3}(\omega_0/c)^3 |\vec{P}_A|^2, \quad (27)$$

which is the spontaneous-emission probability. Substituting Eq. (26) into (25) and then taking the imaginary part of $G_{AA}(\omega)$ we find

$$-2\text{Im}G_{AA}(\omega) = \frac{1-2n_\nu}{\pi} \left[\frac{\frac{1}{2}\gamma_0 [1 - \Omega^2/2(\Omega^2 + \frac{1}{16}\gamma_0^2)]}{(\omega - \omega_0)^2 + (\frac{1}{2}\gamma_0)^2} + \frac{\Omega^2}{4(\Omega^2 + \frac{1}{16}\gamma_0^2)} \right. \\ \left. \times \left(\frac{\frac{3}{4}\gamma_0 - (\omega - \omega_0 - \Omega)\gamma_0/4\Omega}{(\omega - \omega_0 - \Omega)^2 + (\frac{3}{4}\gamma_0)^2} + \frac{\frac{3}{4}\gamma_0 + (\omega - \omega_0 + \Omega)\gamma_0/4\Omega}{(\omega - \omega_0 + \Omega)^2 + (\frac{3}{4}\gamma_0)^2} \right) \right]. \quad (28)$$

Equation (28) describes the spectral function for the interacting system in question. In the limiting case when $\Omega^2 \gg \frac{1}{16}\gamma_0^2$ then Eq. (28) is reduced to

$$-2\text{Im}G_{AA}(\omega) \approx \frac{1-2n_\nu}{4\pi} \left(\frac{2(\frac{1}{2}\gamma_0)}{(\omega-\omega_0)^2+(\frac{1}{2}\gamma_0)^2} + \frac{(\frac{3}{4}\gamma_0)}{(\omega-\omega_0-\Omega)^2+(\frac{3}{4}\gamma_0)^2} + \frac{(\frac{3}{4}\gamma_0)}{(\omega-\omega_0+\Omega)^2+(\frac{3}{4}\gamma_0)^2} \right), \quad (29)$$

which is identical to that known in the literature.^{5,8-13} For the sake of convenience, we will consider the excitation spectrum for the symmetric and antisymmetric modes separately.

1. Excitation spectrum of the symmetric modes

The expression (21) for $G_{AB}^{(*)}(\omega)$ may be written

$$G_{AB}^{(*)}(\omega) \approx \frac{1-2n_\nu}{2\pi} \left(\frac{1-\Omega^2/2(\Omega_+ - \frac{1}{4}\gamma_+)(\Omega_- + \frac{1}{4}\gamma_+)}{\omega-\omega_+ - \frac{1}{2}\gamma_+} + \frac{\Omega/4(\Omega_- + \frac{1}{4}\gamma_+)}{\omega-\omega_0-\Omega_+ - \frac{3}{4}\gamma_+} + \frac{\Omega/4(\Omega_+ - \frac{1}{4}\gamma_+)}{\omega-\omega_0+\Omega_- - \frac{3}{4}\gamma_+} \right), \quad (30)$$

where

$$\Omega_\pm = \Omega \pm \frac{1}{2}V_{AB}. \quad (31)$$

We first consider the case in which the atoms are close together.

(a) $R < \lambda$ ($\lambda = c/\omega_0 = c/E_{\nu 0}$). The expression for $\gamma_\pm = \gamma_\pm(\omega)$ can be calculated from Eqs. (11) and (12); the results of such a calculation are known in the literature.²⁻⁴ In general, for the real part of $\gamma_{AB}(\omega)$ we have²

$$\frac{1}{2}\text{Re}\gamma_{AB}(\omega) = -V_{AB} + \omega_{AB}(R), \quad (32)$$

where the expression for $\omega_{AB}(R)$ decays exponentially with R/λ . For $R < \lambda$, an expansion in powers of R/λ yields,² $\omega_{AB}(R) \approx V_{AB}$ so that, in this limit, $\text{Re}\gamma_{AB}(\omega) \approx 0$. For the imaginary part of $\gamma_{AB}(\omega)$ for $R < \lambda$ is given by^{2-4,17}

$$\text{Im}\gamma_{AB}(\omega) = \gamma_0[1 + O(R^2/\lambda^2)] \approx \gamma_0. \quad (33)$$

Hence,

$$\text{Im}\gamma_+(\omega) \approx 2\gamma_0. \quad (34)$$

Substituting Eqs. (32)–(34) into Eq. (30) and then taking the imaginary part of $G_{AB}^{(*)}(\omega)$, we have

$$-2\text{Im}G_{AB}^{(*)}(\omega) = \frac{1-2n_\nu}{\pi} \left(\frac{\gamma_0(1 - \frac{1}{2}D) + (\omega - \omega_+) \gamma_0 V_{AB} D / (4\Omega^2 - V_{AB}^2 + \gamma_0^2)}{(\omega - \omega_+)^2 + \gamma_0^2} \right. \\ \left. + \frac{1}{4}D_- \frac{(\frac{3}{2}\gamma_0) - (\omega - \omega_0 - \Omega_-) \gamma_0 / (2\Omega - V_{AB})}{(\omega - \omega_0 - \Omega_-)^2 + (\frac{3}{2}\gamma_0)^2} + \frac{1}{4}D_+ \frac{\frac{3}{2}\gamma_0 + (\omega - \omega_0 + \Omega_-) \gamma_0 / (2\Omega + V_{AB})}{(\omega - \omega_0 + \Omega_-)^2 + (\frac{3}{2}\gamma_0)^2} \right), \quad (35)$$

where

$$D = \frac{4\Omega^2(4\Omega^2 - V_{AB}^2 + \gamma_0^2)}{(4\Omega^2 - V_{AB}^2 + \gamma_0^2)^2 + 4\gamma_0^2 V_{AB}^2}, \quad (36)$$

$$D_\pm = \frac{2\Omega(2\Omega \pm V_{AB})}{(2\Omega \pm V_{AB})^2 + \gamma_0^2}.$$

Equation (35) for $-2\text{Im}G_{AB}^{(*)}(\omega)$ describes the spectral function of the system under consideration as a function of ω and for any values of the parameters Ω , V_{AB} , and γ_0 .

In the limit when $\Omega_\pm^2 \gg \frac{1}{4}\gamma_0^2$ and $\Omega \gg V_{AB}$, Eq. (35) is reduced to

$$-2\text{Im}G_{AB}^{(*)}(\omega) \approx \frac{1-2n_\nu}{4\pi} \left(\frac{2\gamma_0}{(\omega - \omega_+)^2 + \gamma_0^2} \right. \\ \left. + \frac{\frac{3}{2}\gamma_0}{[\omega - (\omega_0 + \Omega_+)]^2 + (\frac{3}{2}\gamma_0)^2} \right. \\ \left. + \frac{\frac{3}{2}\gamma_0}{[\omega - (\omega_0 - \Omega_-)]^2 + (\frac{3}{2}\gamma_0)^2} \right), \quad (37)$$

for $\Omega_\pm^2 \gg \frac{1}{4}\gamma_0^2$, $\Omega \gg V_{AB}$, and $R < \lambda$.

The spectral function (37) describes three Lorentzian lines peaked at frequencies $\omega = \omega_+$, $\omega = \omega_0 + \Omega_+$, and $\omega = \omega_0 - \Omega_-$ having spectral widths of the

order of γ_0 , $\frac{3}{2}\gamma_0$, and $\frac{3}{2}\gamma_0$, respectively. Equation (33) implies that the damping constants are larger by a factor of 2 than the corresponding ones of Eq. (29). Hence, there is a cooperative effect due to the presence of the atom B . There is also an additional small energy shift arising from the dipole-dipole interaction between the atoms. The ratio of the central-peak height to the heights of the sidebands is 3:1, the linewidth ratio is 1:1.5 and remains the same in both cases; this is in agreement with the conclusion derived by Agarwal *et al.*¹⁴

The spectral function (35) implies that the profile of the three lines becomes asymmetric for frequencies occurring far from the center of the lines. The asymmetry of the central peak for $\omega \neq \omega_+$ depends on the value of the dipole-dipole interaction V_{AB} as well as of that of the ratio $(4\Omega^2 \pm V_{AB}^2)/\gamma_0^2$. The asymmetry of the sidebands occurs for frequencies $\omega \neq \omega_0 + \Omega_+$ and $\omega \neq \omega_0 - \Omega_-$ and depends on the values of $(2\Omega - V_{AB})/\gamma_0$ and $(2\Omega + V_{AB})/\gamma_0$, respectively. It is pointed out that one of the three terms of Eq. (35) may be negative or zero for values of ω for which the numerator of the corresponding term becomes negative or vanishes. For example, considering that V_{AB} is an attractive interaction so that $V_{AB} = -|V_{AB}|$, then the first term of Eq. (35) for the central peak becomes negative

or zero for values of ω satisfying the condition

$$\omega - \omega_+ \geq \frac{(4\Omega^2 - V_{AB}^2 + \gamma_0^2)}{|V_{AB}|} \left(\frac{1}{D} - \frac{1}{2} \right). \quad (38)$$

Similarly, the second and third terms in Eq. (35) which describe the two sidebands become negative or zero for values of ω satisfying the expressions

$$4\Omega - V_{AB} \leq \omega - \omega_0 \leq -4\Omega - V_{AB}. \quad (39)$$

Negative values of the spectral function indicate amplification rather than attenuation of the signal field and corresponds to the physical process of stimulated emission.^{5,18} Thus, if ω satisfies one of the inequalities (38)–(39) then the phenomenon of stimulated emission occurs. Antiresonance is obtained when ω satisfies one of the equalities (38)–(39), for which value the corresponding term in Eq. (35) vanishes.¹⁹

(b) $R > \lambda$. In this limit, $\omega_{AB}(R)$ and $\text{Im}\gamma_{AB}(\omega)$ are negligibly small in comparison with that of V_{AB} and γ_0 and are given by Eqs. (33) and (34) of Ref. (2), respectively. Neglecting $\omega_{AB}(R)$ and $\text{Im}\gamma_{AB}(\omega)$, we obtain from Eqs. (32)–(34), for $R > \lambda$,

$$\frac{1}{2}\text{Re}\gamma_{AB}(\omega) \approx -V_{AB}, \quad \text{Im}\gamma_+(\omega) \approx \gamma_0. \quad (40)$$

Substituting Eq. (40) into Eq. (30) and then taking the imaginary part of $G_{AB}^{(*)}(\omega)$, we have

$$\begin{aligned} -2\text{Im}G_{AB}^{(*)}(\omega) = \frac{1-2n_v}{2\pi} & \left(\frac{\frac{1}{2}\gamma_0(1-\frac{1}{2}D') + (\omega-\omega_0)\gamma_0 V_{AB} D'/4(\Omega^2 - V_{AB}^2 + \frac{1}{16}\gamma_0^2)}{(\omega-\omega_0)^2 + (\frac{1}{2}\gamma_0)^2} \right. \\ & \left. + \frac{1}{4}D'_- \frac{\frac{3}{4}\gamma_0 - (\omega-\omega_- - \Omega)\gamma_0/4(\Omega - V_{AB})}{(\omega-\omega_- - \Omega)^2 + (\frac{3}{4}\gamma_0)^2} + \frac{1}{4}D'_+ \frac{\frac{3}{4}\gamma_0 + (\omega-\omega_- + \Omega)\gamma_0/4(\Omega + V_{AB})}{(\omega-\omega_- + \Omega)^2 + (\frac{3}{4}\gamma_0)^2} \right), \quad (41) \end{aligned}$$

where

$$D' = \frac{\Omega^2(\Omega^2 - V_{AB}^2 + \frac{1}{16}\gamma_0^2)}{(\Omega^2 - V_{AB}^2 + \frac{1}{16}\gamma_0^2)^2 + \frac{1}{4}\gamma_0^2 V_{AB}^2}, \quad (42)$$

$$D'_\pm = \frac{\Omega(\Omega \pm V_{AB})}{(\Omega \pm V_{AB})^2 + \frac{1}{16}\gamma_0^2}.$$

Eq. (41) represents the spectral function for our system for distances $R > \lambda$ as a function of ω and for general values of the parameters Ω , V_{AB} , and γ_0 . In the limit when $(\Omega \pm V_{AB})^2 \gg \frac{1}{16}\gamma_0^2$ and $\Omega \gg V_{AB}$, we derive from Eq. (42) the expression

$$\begin{aligned} -2\text{Im}G_{AB}^{(*)}(\omega) \approx \frac{1-2n_v}{4\pi} & \left(\frac{2(\frac{1}{2}\gamma_0)}{(\omega-\omega_0)^2 + (\frac{1}{2}\gamma_0)^2} \right. \\ & \left. + \frac{\frac{3}{4}\gamma_0}{(\omega-\omega_- - \Omega)^2 + (\frac{3}{4}\gamma_0)^2} \right. \\ & \left. + \frac{\frac{3}{4}\gamma_0}{(\omega-\omega_- + \Omega)^2 + (\frac{3}{4}\gamma_0)^2} \right), \quad (43) \end{aligned}$$

for

$$(\Omega \pm V_{AB})^2 \gg \frac{1}{16}\gamma_0^2, \quad \Omega \gg V_{AB}, \quad R > \lambda.$$

In this limit, the damping of the modes in Eq. (43) is identical to that of Eq. (29). The central peak is not affected at all, while there is a small frequen-

cy shift of the sidebands due to the dipole-dipole interaction.

The three lines corresponding to the three terms in Eq. (41) become asymmetric for frequencies $\omega \neq \omega_0$ and $\omega \neq \omega_{\pm} \pm \Omega$, respectively. One of the three terms in Eq. (41) becomes negative (zero) if ω satisfies one of the following inequalities (equalities)

$$\omega - \omega_0 \geq \left(\frac{2}{D'} - 1 \right) \frac{(\Omega^2 - V_{AB}^2 + \frac{1}{16}\gamma_0^2)}{|V_{AB}|}, \quad (44)$$

$$4(\Omega - V_{AB}) \leq \omega - \omega_0 \leq -4(\Omega + V_{AB}). \quad (45)$$

Eqs. (44) and (45) are analogous to Eq. (38) and (39), respectively.

2. Excitation spectrum of the antisymmetric modes

We may rewrite Eq. (22) for $G_{AB}^{(-)}(\omega)$ as

$$G_{AB}^{(-)}(\omega) = \frac{1-2n_\nu}{2\pi} \left(\frac{1-\Omega^2/2(\Omega_- + \frac{1}{2}\gamma_- - \frac{1}{4}\gamma_-)(\Omega_+ - \frac{1}{2}\gamma_+ + \frac{1}{4}\gamma_-)}{(\omega - \omega_- - \frac{1}{2}\gamma_-)} + \frac{\Omega/4(\Omega_- + \frac{1}{2}\gamma_- - \frac{1}{4}\gamma_-)}{\omega - \omega_- - \Omega_- - \frac{1}{2}\gamma_- - \frac{1}{4}\gamma_-} + \frac{\Omega/4(\Omega_+ - \frac{1}{2}\gamma_+ + \frac{1}{4}\gamma_-)}{\omega - \omega_+ + \Omega_+ - \frac{1}{2}\gamma_+ - \frac{1}{4}\gamma_-} \right). \quad (46)$$

(a) $R < \lambda$. In this limit, the imaginary part of $\gamma_-(\omega)$ is given by $2 \operatorname{Im} \gamma_-(\omega) \approx \gamma_0(R^2/\lambda^2)$, which is small in comparison to γ_0 and may be neglected. Using the fact that $\operatorname{Re} \gamma_{AB}(\omega) \approx 0$ and $\operatorname{Im} \gamma(\omega) = \gamma_0$, Eq. (46) becomes

$$G_{AB}^{(-)}(\omega) = \frac{1-2n_\nu}{2\pi} \left(\frac{1-\Omega^2/2(\Omega_- - \frac{1}{2}i\gamma_0)(\Omega_+ + \frac{1}{2}i\gamma_0)}{(\omega - \omega_-)} + \frac{\Omega(\Omega_- + \frac{1}{2}i\gamma_0)/4(\Omega_-^2 + \frac{1}{4}\gamma_0^2)}{(\omega - \omega_- - \Omega_- + \frac{1}{2}i\gamma_0)} + \frac{\Omega(\Omega_+ - \frac{1}{2}i\gamma_0)/4(\Omega_+^2 + \frac{1}{4}\gamma_0^2)}{(\omega - \omega_+ + \Omega_+ + \frac{1}{2}i\gamma_0)} \right). \quad (47)$$

Taking the imaginary part of Eq. (47), we have

$$-2 \operatorname{Im} G_{AB}^{(-)}(\omega) = \frac{1-2n_\nu}{\pi} \left[\left(1 - \frac{1}{2}\Omega^2 \frac{(\Omega_+ \Omega_- + \frac{1}{4}\gamma_0^2)}{(\Omega_+ \Omega_- + \frac{1}{4}\gamma_0^2)^2 + \frac{1}{4}\gamma_0^2 V_{AB}^2} \right) \pi \delta(\omega - \omega_-) + \frac{\Omega \Omega_-}{4(\Omega_-^2 + \frac{1}{4}\gamma_0^2)} \frac{\frac{1}{2}\gamma_0}{(\omega - \omega_- - \Omega_-)^2 + (\frac{1}{2}\gamma_0)^2} - \frac{(\omega - \omega_- - \Omega_-)\gamma_0/2\Omega_-}{(\omega - \omega_- - \Omega_-)^2 + (\frac{1}{2}\gamma_0)^2} + \frac{\Omega \Omega_+}{4(\Omega_+^2 + \frac{1}{4}\gamma_0^2)} \frac{\frac{1}{2}\gamma_0 + (\omega - \omega_+ + \Omega_+)\gamma_0/2\Omega_+}{(\omega - \omega_+ + \Omega_+)^2 + (\frac{1}{2}\gamma_0)^2} \right]. \quad (48)$$

In limit when $\Omega_{\pm}^2 \gg \frac{1}{4}\gamma_0^2$ and $\Omega \gg V_{AB}$, Eq. (48) becomes

$$-2 \operatorname{Im} G_{AB}^{(-)}(\omega) = \frac{1-2n_\nu}{4\pi} \left(2\pi \delta(\omega - \omega_-) + \frac{\frac{1}{2}\gamma_0}{(\omega - \omega_- - \Omega_-)^2 + (\frac{1}{2}\gamma_0)^2} + \frac{\frac{1}{2}\gamma_0}{(\omega - \omega_+ + \Omega_+)^2 + (\frac{1}{2}\gamma_0)^2} \right), \quad (49)$$

for $\Omega_{\pm}^2 \gg \frac{1}{4}\gamma_0^2$, $\Omega \gg V_{AB}$.

The spectral function (49) indicates that the central peak has a δ -function distribution and peaked at $\omega = \omega_-$. This is in agreement with the results obtained in the absence of the pump field.¹⁻⁴ The sidebands are described by Lorentzian lines peaked at the frequencies $\omega = \omega_+ + \Omega_+$ and $\omega = \omega_- - \Omega_-$ with each having a spectral width equal to $\frac{1}{2}\gamma_0$. Thus, the energy mode of the central peak is stable and does not radiate, while the modes $\omega = \omega_+ + \Omega_+ = \omega_0 - \frac{3}{2}V_{AB} + \Omega$ and $\omega = \omega_- - \Omega_- = \omega_0 - \frac{3}{2}V_{AB} - \Omega$, corresponding to the sidebands, are damped and have a lifetime equal to that for a single isolated atom, respectively. The lack of radiative cooperative contribution of the atoms *A* and *B* to the sidebands

can be seen from Eqs. (13) and (15) wherein the expressions for the Green's functions $\langle\langle (b_A^\dagger b_A - b_B^\dagger b_B) \beta_0; b_A^\dagger \rangle\rangle$ and $\langle\langle (b_A^\dagger b_B - b_B^\dagger b_A) \beta_0; b_A^\dagger \rangle\rangle$, the cooperative radiative effects (of the type γ_{AB}), are cancelled and only that of $\gamma(\omega) = \gamma_{AA}(\omega) = \gamma_{BB}(\omega)$ survives.

The spectral function (48) indicates that, for frequencies $\omega \neq \omega_+ + \Omega_+$ and $\omega \neq \omega_- - \Omega_-$, the spectral lines for the sidebands become asymmetric, the extent of which depends on the values of the ratios $\gamma_0/2\Omega_-$ and $\gamma_0/2\Omega_+$, respectively. When ω satisfies one of the inequalities

$$2(\Omega + V_{AB}) \leq \omega - \omega_0 \leq -2(\Omega + V_{AB}), \quad (50)$$

then one of the last two terms in Eq. (48) becomes negative, indicating that the process of stimulated emission may occur. Antiresonance is obtained when ω satisfies one of the equalities (50).

(b) $R > \lambda$. Neglecting $\omega_{AB}(R)$ and $\text{Im}\gamma_{AB}(\omega)$ and considering that $\text{Re}\gamma_{AB}(\omega) \approx -2V_{AB}$ and $\text{Im}\gamma(\omega) \approx \gamma_0$, we obtain from Eq. (46) the expression for $-2\text{Im}G_{AB}^{(-)}(\omega)$ which is identical to that of Eq. (41) as it should be. Hence, Eqs. (41) and (43) describe the spectral function for case in question since, for $R > \lambda$, $\text{Im}G_{AB}^{(+)}(\omega) = \text{Im}G_{AB}^{(-)}(\omega)$.

IV. DISCUSSION

We have considered the excitation spectrum arising from the interaction between two similar atoms, one of which is excited in the presence of a strong resonant electromagnetic field. It is shown that when $R < \lambda$, the spectral function for the symmetric modes is given by Eq. (35), which consists of three asymmetric lines whose form depends on the relation among the parameters Ω , γ_0 , and V_{AB} . When $\Omega_{\pm}^2 \gg \frac{1}{4}\gamma_0^2$ and $\Omega \gg V_{AB}$, Eq. (35) is reduced to Eq. (37) which is similar to Eq. (29) describing the interaction of an isolated single atom with the pump field with the exception that the damping constants are twice as large and the energies are shifted due to the dipole-dipole interaction between the atoms.

The spectral function for the antisymmetric modes ($R < \lambda$) is given by Eq. (48), where the mode of the central peak is stable while the two sidebands are described by asymmetric Lorentzian lines where the extent of the asymmetry depends on the values of $\gamma_0/2\Omega_-$ and $\gamma_0/2\Omega_+$, respectively; when $\Omega_{\pm} \gg \frac{1}{2}\gamma_0$, both lines for the sidebands become symmetric with a linewidth appropriate to spontaneous emission from a single isolated atom. It is of interest to point out that the central peak corresponds to a stationary state with energy $\omega = \omega_-$, which is in agreement with the literature,¹⁻⁴ while both states corresponding to the sidebands are capable of emitting photons.

For $R > \lambda$, the spectral function for the symmetric and antisymmetric modes is given by Eq. (41) which is reduced to Eq. (43) when $(\Omega \pm V_{AB})^2$

$\gg \frac{1}{16}\gamma_0^2$ and $\Omega \gg V_{AB}$. Eq. (43) is similar to that obtained when an isolated single atom interacts with a strong pump field.

In the expression (35), the central peak is asymmetric at frequencies $\omega \neq \omega_+$ and the asymmetry appears to be proportional to V_{AB} . To investigate this term further, we write the asymmetric part of the numerator of the first term in Eq. (35) as $(\omega - \omega_+)2\Omega^2 S(\Omega^2)$, where the function $S(\Omega^2)$ has the form

$$S(\Omega^2) = \frac{(2\gamma_0 V_{AB})}{(4\Omega^2 - V_{AB}^2 + \gamma_0^2)^2 + (2\gamma_0 V_{AB})^2}. \quad (51)$$

Equation (51) indicates that $S(\Omega^2)$ as a function of Ω^2 is described by a Lorentzian line peaked at $4\Omega^2 = V_{AB}^2 - \gamma_0^2$ and has a width of the order of $2\gamma_0 V_{AB}$. Physically, the function $S(\Omega^2)$ describes the cooperative effect which results from the correlation of the interatomic (dipole-dipole) interactions and the radiation field. Such physical processes arising from correlation effects satisfy the appropriate energy-conservation rules and bring about asymmetric broadening of the spectral lines. The function $S(\Omega^2)$ becomes maximum

$$S_{\max}(\Omega^2) = 1/2\gamma_0 V_{AB}, \quad (52)$$

for

$$4\Omega^2 = V_{AB}^2 - \gamma_0^2. \quad (53)$$

When Eq. (53) is satisfied, then the term describing the asymmetry becomes equal to $(\omega - \omega_+)\Omega^2/\gamma_0 V_{AB}$, an effect which may be observed experimentally for $\omega \neq \omega_+$. Similar discussion holds for the asymmetry that appears in the first term of Eq. (41).

We emphasize that our results are valid when the pump field is strong, that is, when the number of photons n_0 is much greater than one ($n_0 \gg 1$). In this respect, the neglect of the Green's functions like those appearing in Eq. (14) is justifiable. The decoupling approximation given by Eq. (18) is equivalent to the Hartree-Fock self-consistent field approximation²⁰ and is expected to be asymptotically correct for large values of frequency ω and high photon densities of the pump field.

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