Quantum beats in correlation functions of spontaneously emitted radiation and the transition rates of a multilevel atom

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Spontaneous emission from a multilevel atom is studied from the viewpoint of quantum beats. The master equation describing the atomic dynamics is shown to have some parameters that oscillate at the beat frequencies, and because of this a Pauli type of master equation for the diagonal elements of the density matrix is not obtained. As a result, the transition rates in the atomic system show the characteristic beats. The relation of the rate of change of the energy of the atomic system to the total power radiated in the far zone is also established. The normally and antinormally ordered correlation functions of the spontaneously emitted radiation are calculated under the approximation that the level widths are much smaller than the beat frequencies. The contribution arising from the interference of the free field and the source terms to antinormally ordered correlations is carefully evaluated. The nature of beats present in normally and antinormally ordered correlations is discussed, and their detection from the viewpoint of causing transitions in another atom is considered. The change in the nature of beats due to atomic collective effects is studied. Finally, it is shown that because of quantum effects, intensity correlations do not show any beats.

I. INTRODUCTION

Since the successful observation of the quantum beats[1,2] (and also the related experiments on beam foil spectroscopy[3-5]) and the realization that these provide a new method of laser spectroscopy, several theoretical papers[6-10] have examined the quantum beats from different points of view. The existence or nonexistence of a particular type of beat has been considered to be a test of the different theories of spontaneous emission.[11,12] Herman *et al.*[6] examined the beats which may be present in the Poynting vector of the radiation emitted by a multilevel atom. They also calculated the transition rates in a four-level atom with certain restrictions on dipole moments and found no beats. Senitzky[7] analyzed the mean amplitude and the intensity of the radiation emitted from a multilevel atom. Using his second quantized boson formalism, he also discussed the transition to neoclassical theory. Chow[8] *et al.* discussed the type of beats present in the intensity of the radiation emitted by a system of three-level atoms under the assumption that the radiation field contains only one photon.

The purpose of the present study is to develop a self-consistent theory, in which the radiation reaction effects are properly taken into account. Our description does not rely on one photon approximation, and collective effects are properly incorporated in the theory. We examine the nature of beats present in different types of transition rates and correlation functions. An important aspect of the present work is the examination of the antinormally ordered correlations of the emitted radiation. The antinormally ordered correlations are found to exhibit the so-called lower-state beats in addition to the upper-state beats, which are present in the normally ordered correlations. In contrast to the above, we show that the normally ordered intensity correlations do not show any beats. Such normally and antinormally ordered correlations, as is well known in quantum optics, are relevant to different types of detection processes.[13,14]

The outline of the present paper is as follows: In Sec. II we derive the master equation describing the dynamics of a multilevel atom interacting with the zero-point fluctuations. Using the master equation we calculate the transition probability, per unit time, that the atom makes a downward transition. Such transition rates are shown to have upper-state beats. We also comment on the relation between the Poynting vector of the emitted radiation and the rate at which the atom loses its energy. Finally, an approximate solution of the master equation is presented. In Secs. III and IV, we calculate the normally and the antinormally ordered correlations of the field emitted by the atom. We discuss the nature of beats present in such correlations. In Sec. V, the influence of the collective atomic effects on beating phenomena is discussed.

II. MASTER EQUATION FOR SPONTANEOUS EMISSION FROM A MULTILEVEL ATOM AND BEATS IN THE TRANSITION RATES

In this section we discuss the master equation which describes the dynamics of a multilevel atom interacting with zero-point fluctuations. The master equation for the reduced density operator in the interaction picture is given by [Eqs. (2.12) and (2.13) of Ref. 15; the atom is assumed to be located at the origin]

$$\frac{\partial \rho}{\partial t} = -\int_0^\infty d\tau\,(\mathcal{E}^{(s)}_{\alpha\beta}(0,0,\tau)[P_\alpha(t),[P_\beta(t-\tau),\rho(t)]] + \chi''_{\alpha\beta EE}(0,0,\tau)[P_\alpha(t),\{P_\beta(t-\tau),\rho(t)\}]), \tag{2.1}$$

$$P_\alpha(t) = \sum_{ij} A_{ij}\, d^{ij}_\alpha e^{i\omega_{ij}t}, \quad A_{ij} = |i\rangle\langle j|;$$

$$\mathcal{E}^{(s)}_{\alpha\beta}(\vec{r},\vec{r}',\tau) = \tfrac{1}{2}\langle\{E_{0\alpha}(\vec{r},\tau),E_{0\beta}(\vec{r}',0)\}\rangle, \tag{2.2}$$

$$\chi''_{\alpha\beta EE}(\vec{r},\vec{r}',\tau) = \tfrac{1}{2}\langle\{E_{0\alpha}(\vec{r},\tau),E_{0\beta}(\vec{r}',0)\}\rangle,$$

where the E_0's are the free-field operators. We rewrite (2.1) so as to exhibit its t dependence explicitly:

$$\frac{\partial \rho}{\partial t} = -\int_0^\infty d\tau \sum d^{ij}_\alpha d^{kl}_\beta \exp[i(\omega_{ij}+\omega_{kl})t - i\omega_{kl}\tau] \times\{\mathcal{E}^{(s)}_{\alpha\beta}(0,0,\tau)[A_{ij},[A_{kl},\rho(t)]] + \chi''_{\alpha\beta EE}(0,0,\tau)[A_{ij},\{A_{kl},\rho(t)\}]\}. \tag{2.3}$$

In an earlier work[11] the rotating-wave approximation (RWA) was used, i.e., all the rapidly oscillating terms from (2.3) were ignored, and it was found that the off-diagonal elements decay according to

$$\dot\rho_{ij} = -\Gamma_{ij}\rho_{ij}, \quad \Gamma_{ij} = \tfrac{1}{2}(\Gamma_i+\Gamma_j), \quad \Gamma_i = 2\sum_k \gamma_{ki}, \tag{2.4}$$

and the diagonal elements satisfy a Pauli type of master equation,

$$\dot\rho_{ii} = -\sum 2\gamma_{ki}\rho_{ii} + \sum 2\gamma_{ik}\rho_{kk}, \tag{2.5}$$

where $2\gamma_{ij}$ is the transition probability per unit time that an atom make a transition from the level $|j\rangle$ to $|i\rangle$ by emitting a photon. Since we are considering beats, the RWA has to be made very carefully. By keeping only the frequency differences in (2.3), we find that the reduced density operator satisfies the master equation (the density operator ρ is in the interaction picture)

$$\begin{aligned}\frac{\partial \rho}{\partial t} = &-\sum_{\substack{i<j\\k>l}} (\vec{d}^{ij}\cdot\vec{d}^{kl}) \exp[i(\omega_{ij}+\omega_{kl})t]\left(\frac{2}{3}\frac{\omega^3_{lk}}{c^3}\right)[A_{ij},A_{kl}\rho]\\ &+\frac{i}{\pi}\mathrm{P}\sum_{\substack{i>j\\k<l}} \vec{d}^{ij}\cdot\vec{d}^{kl} e^{i(\omega_{ij}+\omega_{kl})t}\int_0^\infty d\omega'\left(\frac{2}{3}\frac{\omega'^3}{c^3}\right)(\omega'+\omega_{kl})^{-1}[A_{ij},A_{kl}\rho]\\ &+\frac{i}{\pi}\mathrm{P}\sum_{\substack{i<j\\k>l}} (\vec{d}^{ij}\cdot\vec{d}^{kl}) e^{i(\omega_{ij}+\omega_{kl})t}\int_0^\infty d\omega'\,\frac{2}{3}\frac{\omega'^3}{c^3}\,(\omega'-\omega_{lk})^{-1}[A_{ij},A_{kl}\rho] + \mathrm{H.c.}\end{aligned} \tag{2.6}$$

This master equation exhibits different types of beats. It is clear from (2.6) that the diagonal elements do not satisfy a Pauli type of master equation, i.e., the equation for the diagonal elements contains off-diagonal elements of ρ. We thus see that the small energy separations between the levels leads to certain types of coherence effects in the equation of motion. To see this more explicitly, let us imagine a situation in which a set of upper levels (designated by index μ) decays to the ground level (denoted by g). (This is in fact the situation in the experimental work of Haroche *et al.*[1]) In this case (2.6) reduces to

$$\begin{aligned}\frac{\partial \rho}{\partial t} = &-\sum_{\mu\mu'} \vec{d}^{\mu g}\cdot\vec{d}^{g\mu'} e^{i\omega_{\mu\mu'}t}\left(\frac{2}{3}\frac{\omega^3_{\mu' g}}{c^3}\right)[A_{\mu g},A_{g\mu'}\rho]\\ &+\frac{i}{\pi}\mathrm{P}\sum_{\mu\mu'} \vec{d}^{\mu g}\cdot\vec{d}^{g\mu'} e^{i\omega_{\mu\mu'}t}\int_0^\infty d\omega'\left(\frac{2}{3}\frac{\omega'^3}{c^3}\right)(\omega'-\omega_{\mu' g})^{-1}[A_{\mu g},A_{g\mu'}\rho]\\ &+\frac{i}{\pi}\mathrm{P}\sum_{\mu\mu'} \vec{d}^{g\mu}\cdot\vec{d}^{\mu' g} e^{-i\omega_{\mu\mu'}t}\int_0^\infty d\omega'\left(\frac{2}{3}\frac{\omega'^3}{c^3}\right)(\omega'+\omega_{\mu' g})^{-1}[A_{g\mu},A_{\mu' g}\rho] + \mathrm{H.c.}\end{aligned} \tag{2.7}$$

Using (2.6) we can calculate the transition probability per unit time that the atom decays to the ground state, if initially it was in an arbitrary state $\rho(0)$. This rate is easily obtained from (2.7) by taking the matrix element $\langle g|\dot\rho|g\rangle$ and by setting $\rho(t)\approx\rho(0)$ on the right-hand side of (2.7). The transition rate is found to be

$$\begin{aligned}R_\downarrow \equiv \frac{\partial}{\partial t}\rho_{gg} = &\sum_{\mu\mu'} (\vec{d}^{\mu g}\cdot\vec{d}^{g\mu'}) e^{i\omega_{\mu\mu'}t}\rho_{\mu'\mu}(0)\\ &\times\left(\frac{2}{3c^3}(\omega^3_{\mu' g}+\omega^3_{\mu g}) - \frac{i}{\pi}\int_0^\infty d\omega'\left(\frac{2}{3}\frac{\omega'^3}{c^3}\right)\right.\\ &\left.\times[(\omega'-\omega_{\mu' g})^{-1} - (\omega'-\omega_{\mu g})^{-1}]\right),\end{aligned} \tag{2.8}$$

which clearly shows beats at the frequencies $\omega_{\mu\mu'}$ provided the initial state of the atom is such that $\rho_{\mu'\mu}\neq 0$. In the earlier work such beating terms in transition rates were not found because a time averaging was done. However, in the present case the time averaging has to be done rather carefully since $\omega_{\mu\mu'}\approx 100$ MHz or so, i.e., the times involved are in the nanosecond regime. It should also be noted that the beat at $\omega_{\mu\mu'}$ in the transition rate will also be absent if $\vec{d}^{\mu g}\cdot\vec{d}^{g\mu'}=0$ (which, for example, happens in the situation considered by Herman *et al.*[6]).

The description of our radiating system is complete once we know how to calculate the properties of the radiation field. It can be easily shown that the positive frequency parts of the electric field and the magnetic field operator in the radiation zone are related to the atomic operators by[11,15]

$$E_m^{(+)}(\vec{r},t)\approx E_{0m}^{(+)}(\vec{r},t)+\sum_{j<i}A_{ij}^{(I)}\left(t-\frac{r}{c}\right)b_{e,m}^{ij}(\vec{r},t), \tag{2.9}$$

$$H_m^{(+)}(\vec{r},t)\approx H_{0m}^{(+)}(\vec{r},t)+\sum_{j<i}A_{ij}^{(I)}\left(t-\frac{r}{c}\right)b_{h,m}^{ij}(\vec{r},t), \tag{2.10}$$

where the operators A_{ij} are in the interaction picture and the coefficients are given by

$$b_{e,m}^{ij}(\vec{r},t)=\sum_{\alpha}d_{\alpha}^{ij}\tilde{\chi}_{m\alpha EE}(\vec{r},0,\omega_{ji})\,e^{it\omega_{ij}}, \tag{2.11}$$

$$b_{h,m}^{ij}(\vec{r},t)=\sum_{\alpha}d_{\alpha}^{ij}\tilde{\chi}_{m\alpha HE}(\vec{r},0,\omega_{ji})\,e^{it\omega_{ij}}, \tag{2.12}$$

$$\tilde{\chi}_{m\alpha EE}(\vec{r},0,\omega)=(\omega^2/c^2)(\delta_{m\alpha}-\hat{r}_m\hat{r}_\alpha)\,e^{i\omega r/c}/r, \tag{2.13}$$

$$\tilde{\chi}_{m\alpha HE}(\vec{r},0,\omega)=(\omega^2/c^2)(\hat{r}\times\vec{I})_{m\alpha}\,e^{i\omega r/c}/r. \tag{2.14}$$

The Poynting vector in the far zone will be given by

$$S_\alpha=\frac{c}{4\pi}\langle(:\vec{E}\times\vec{H}:)_\alpha\rangle=\frac{c}{4\pi}\sum\epsilon_{\alpha\beta\gamma}\langle:E_\beta H_\gamma:\rangle=\frac{c}{4\pi}\sum\epsilon_{\alpha\beta\gamma}\langle E_\beta^{(-)}H_\gamma^{(+)}+H_\gamma^{(-)}E_\beta^{(+)}\rangle$$

or

$$\vec{S}=\frac{c}{4\pi}\langle\vec{E}^{(-)}\times\vec{H}^{(+)}-\vec{H}^{(-)}\times\vec{E}^{(+)}\rangle,$$

and hence the power radiated per unit solid angle in the far zone will be

$$\frac{dP}{d\Omega}=r^2\hat{r}\cdot\vec{S}=\frac{c}{4\pi}\sum_{\substack{j<i\\ l<k}}(\vec{d}^{ij}\cdot\vec{d}^{kl*}-(\hat{r}\cdot\vec{d}^{ij})(\hat{r}\cdot\vec{d}^{kl*}))\times\frac{\omega_{ji}^2\omega_{lk}^2}{c^4}\exp[i(\omega_{ij}-\omega_{kl})t]\times\langle A_{lk}^{(I)}(t)A_{ij}^{(I)}(t)\rangle+\text{c.c.},$$

which on using the operator algebra reduces to

$$\frac{dP}{d\Omega}=-\frac{1}{4\pi}\sum_{\substack{j<i\\ l<i}}\frac{\omega_{ji}^2\omega_{li}^2}{c^3}\vec{d}^{il*}\cdot\{\hat{r}\times(\hat{r}\times\vec{d}^{il})\}\,e^{i\omega_{lj}t}\times\langle A_{lj}^{(I)}(t)\rangle+\text{c.c.} \tag{2.15}$$

The expression (2.15), in the special case considered earlier, reduces to

$$\frac{dP}{d\Omega}=-\frac{1}{4\pi}\sum_{\mu\mu'}\frac{\omega_{\mu g}^2\,\omega_{\mu' g}^2}{c^3}\vec{d}^{\mu' g}\cdot\{\hat{r}\times(\hat{r}\times\vec{d}^{g\mu})\}\,e^{i\omega_{\mu'\mu}t}\times\langle A_{\mu'\mu}^{(I)}(t)\rangle+\text{c.c.} \tag{2.16}$$

Hence the total power radiated in the far zone will be

$$P=\int\frac{dP}{d\Omega}d\Omega=\sum_{\mu\mu'}(\vec{d}^{g\mu}\cdot\vec{d}^{\mu' g})\,\frac{2}{3}\,\frac{\omega_{\mu g}^2\,\omega_{\mu' g}^2}{c^3}\times e^{i\omega_{\mu'\mu}t}\langle A_{\mu'\mu}^{(I)}(t)\rangle+\text{c.c.} \tag{2.17}$$

The power radiated in the far zone shows beats. We will now compare the expression for P with that for the rate of change of the energy of the atomic system:

$$W=\sum_j E_j\langle A_{jj}\rangle=\sum_\mu\omega_{\mu g}\langle A_{\mu\mu}\rangle+E_g. \tag{2.18}$$

Using the master equation (2.6) one immediately finds that

$$-\dot{W}=\sum_{\mu\mu'}(\vec{d}^{\mu g}\cdot\vec{d}^{g\mu'})\,\omega_{\mu g}\rho_{\mu'\mu}e^{i\omega_{\mu\mu'}t}\times\left[\frac{2}{3}\frac{\omega_{\mu' g}^3}{c^3}-\frac{i}{\pi}\int_0^\infty d\omega'\left(\frac{2}{3}\frac{\omega'^3}{c^3}\right)\times(\omega'-\omega_{\mu' g})^{-1}\right]+\text{c.c.}, \tag{2.19}$$

which bears close resemblance to (2.17).

The complete solution of the master equation (2.6) is rather complicated. This is in contrast to the solution of (2.4) and (2.5). In what follows we will present an approximate solution of (2.6), valid in case when the beat frequencies are such that $\omega_{\mu\mu'}\gg\Gamma$, where Γ^{-1} is the characteristic decay time. This condition is certainly satisfied in the experiments of Haroche *et al.*[1] ($\omega_{\mu\mu'}\approx 50$ MHz and larger, $\Gamma\sim 10^9/135$). Using Bogoliubov's method of time averaging,[16] one can show that the solution of (2.7) can be written

$$\rho(t)=\rho^{(0)}(t)+\rho^{(1)}(t)+\cdots, \tag{2.20}$$

where $\rho^{(0)}(t)$ is the solution of the master equation in the absence of beat terms [the principle-value in (2.21) are the frequency-shift terms],

$$\dot{\rho}^{(0)}=-\sum_{\mu}|\vec{d}^{\mu g}|^2\left(\frac{2}{3}\frac{\omega_{\mu g}^3}{c^3}-\frac{i}{\pi}\int_0^\infty d\omega'\,\frac{2}{3}\frac{\omega'^3}{c^3}(\omega'-\omega_{\mu g})^{-1}\right)[A_{\mu g},A_{g\mu}\rho^{(0)}]$$
$$+\frac{i}{\pi}\sum\int_0^\infty d\omega'\left(\frac{2}{3}\frac{\omega'^3}{c^3}\right)(\omega'+\omega_{\mu g})^{-1}|\vec{d}^{\mu g}|^2[A_{g\mu},A_{\mu g}\rho^{(0)}]+\text{H.c.}\,, \tag{2.21}$$

and $\rho^{(1)}(t)$ contains the information about beats and is given in terms of $\rho^{(0)}$ by

$$\rho^{(1)}(t)=-\sum_{\mu\neq\mu'}\vec{d}^{\mu g}\cdot\vec{d}^{g\mu'}\,\frac{e^{i\omega_{\mu\mu'}t}}{\omega_{\mu\mu'}}\left[\frac{2}{3}\frac{\omega_{\mu' g}^3}{c^3}-\frac{i}{\pi}\int_0^\infty d\omega'\left(\frac{2}{3}\frac{\omega'^3}{c^3}\right)(\omega'-\omega_{\mu' g})^{-1}\right][A_{\mu g},A_{g\mu'}\rho^{(0)}]$$
$$+\frac{i}{\pi}\sum_{\mu\neq\mu'}\vec{d}^{\mu g}\cdot\vec{d}^{g\mu'}\,\frac{e^{i\omega_{\mu\mu'}t}}{\omega_{\mu\mu'}}\int_0^\infty d\omega'\left(\frac{2}{3}\frac{\omega'^3}{c^3}\right)(\omega'+\omega_{\mu' g})^{-1}[A_{g\mu},A_{\mu' g}\rho^{(0)}]+\text{H.c.} \tag{2.22}$$

It is clear from (2.22) that the perturbation parameter[17] is $\Gamma/\omega_{\mu\mu'}$. The result (2.8) for the downward transition rate is recovered from (2.20). It it clear from (2.20) that if we calculate the mean value of the atomic operator $\langle\vec{P}(t)\rangle$, then $\rho^{(1)}(t)$ terms lead simply to a renormalization of the coefficients of various beat terms. Such a renormalization can be ignored since $\Gamma/\omega_{\mu\mu'}$ is small. Hence in a sense we can still describe the atomic dynamics by Eqs. (2.4) and (2.5). However, care has to be used in the calculation of the time derivatives of such atomic expectation values. In time derivatives, $\rho^{(1)}(t)$ terms can make a contribution which is of the same order of magnitude as the one from $\rho^{(0)}$—this happens, for instance, in the calculation of transition rates and the rate of change of the energy of the atomic system.

Finally, it should be noted that there is also a renormalization of the beat frequencies—this is easily seen from the structure of the equation of motion for the off-diagonal elements $\rho_{\mu g}$, which follows from (2.7):

$$\dot{\rho}_{\mu g}=-i\omega_{\mu g}\rho_{\mu g}-\sum_{\mu'}\beta_{\mu\mu'}\rho_{\mu' g}\,. \tag{2.23}$$

However, such renormalization effects can again be neglected if $\Gamma/\omega_{\mu\mu'}\ll 1$.

III. NORMALLY ORDERED CORRELATION FUNCTIONS OF THE SPONTANEOUSLY EMITTED RADIATION AND THE BEATS

In this section we examine the normally ordered correlation functions of the electric field operator (2.9) in the light of the beating phenomena. Since the radiation field at time $t=0$ is in vacuum state, it follows immediately from (2.9) that

$$G_{mn}^{(N)}(\vec{r}_1,t_1,\vec{r}_2,t_2)\equiv\langle E_m^{(-)}(\vec{r}_1,t_1)E_n^{(+)}(\vec{r}_2,t_2)\rangle$$
$$=\sum_{\substack{j<i\\ l<k}}b_{e,m}^{ij\,*}(\vec{r}_1,t_1)\,b_{e,n}^{kl}(\vec{r}_2,t_2)\times\langle A_{ji}^{(I)}(t_1-r_1/c)A_{kl}^{(I)}(t_2-r_2/c)\rangle\,. \tag{3.1}$$

In view of (2.4) and the quantum-regression theorem,[18] we have

$$\langle A_{ji}^{(I)}(t_1)A_{kl}^{(I)}(t_2)\rangle=\exp[-\Gamma_{ji}(t_1-t_2)]\times\langle A_{ji}^{(I)}(t_2)A_{kl}^{(I)}(t_2)\rangle$$
$$=\exp[-\Gamma_{ji}(t_1-t_2)]\langle A_{jl}^{(I)}(t_2)\rangle\delta_{ik}\,,\quad(t_1\geq t_2)\,, \tag{3.2}$$

where we have used the closure relation

$$A_{ij}A_{kl}=\delta_{jk}A_{il}\,. \tag{3.3}$$

On using (3.2) in (3.1), we obtain for the correlation function

$$G_{mn}^{(N)}(\vec{r}_1,t_1+r_1/c,\vec{r}_2,t_2+r_2/c)$$
$$=\sum_{\substack{j<i\\ l<i}}b_{e,m}^{ij\,*}(\vec{r}_1,t_1+r_1/c)\,b_{e,n}^{il}(\vec{r}_2,t_2+r_2/c)\times\exp[-\Gamma_{ji}(t_1-t_2)]\langle A_{jl}^{(I)}(t_2)\rangle\,. \tag{3.4}$$

In the special case one has from (3.4) for the intensity of the radiation emitted

$$I(\vec{r},t+r/c)=\sum_{\substack{j<i\\ l<i}}\left(\frac{\omega_{ji}^2\omega_{li}^2}{c^4r^2}\right)\langle A_{jl}^{(I)}(t)\rangle\times e^{i\omega_{jl}t}[\vec{d}^{ji}\cdot\vec{d}^{il}-(\hat{r}\cdot\vec{d}^{ji})(\hat{r}\cdot\vec{d}^{il})]\,. \tag{3.5}$$

It should be noted that (3.5), in general, has two types of contributions: (i) from the diagonal terms $\langle A_{jj}(t)\rangle$, and (ii) from the off-diagonal terms

$\langle A_{jl}(t)\rangle$, $(l\neq j)$. This latter contribution oscillates at the frequency ω_{jl} $(j<l)$ and thus shows beats. It is clear from the structure of (3.5) that such beating terms are there provided we have a situation in which the transitions starting from two upper levels $|j\rangle$, $|l\rangle$ end at the common lower level $|i\rangle$ (upper state beats). Note further that in order that the amplitude associated with the beating term to be nonzero, the initial state of the atom must be such that $\langle A_{jl}(0)\rangle\neq 0$, i.e., initially there should be some coherence between the jth and lth level. In particular, if we consider a three-level atom, then the beats will be there at the frequency ω_{12} provided the transitions are allowed between the level $|1\rangle$ and the ground level $|3\rangle$ and between level $|2\rangle$ and the ground level $|3\rangle$ and provided the initial state of the atom does involve the linear superposition of the states $|1\rangle$ and $|2\rangle$ so that $\langle A_{12}(0)\rangle\neq 0$.

In a recent work, Khoo and Eberly[9,10] have analyzed the beating phenomena from the point of veiw of the detection of the radiation emitted from the multilevel atom. They have carried out a perturbation theory in Heisenberg picture[19] and calculated various transition rates. In the present paper we adopt a different approach—which is similar to the one used by the author in the treatment of Lippmann fringes.[20] In this approach, we regard the field as having a prescribed statistics and then calculate the transition rates using the generalization of Fermi's "golden rule." This generalization concerns transitions in an electromagnetic field of arbitrary coherence. In the present case the coherence of the field is given by Eqs. (2.6) and (2.9).

Let us first consider a detector which works by the absorption of photons. It is well known[13] that such a detector measures the normally ordered correlation functions of the field. To be more specific let us consider the transitions in the detector atom which is located at $\vec{R}$. Assume that the detector is in the far zone of the radiation emitted by the multilevel atom (to be referred to as atom A) located at $\vec{r}=0$. Assume that the detector atom (to be referred to as atom B) is in the ground state $|\chi_g\rangle$ at the time R/c, i.e., when the radiation from the source atom reaches the detector. For the dipole allowed transitions in atom B, the interaction with the field can be written

$$H_1=-\vec{P}^{(B)}\cdot\vec{E}(\vec{R},t)\,. \tag{3.6}$$

Then it can be shown [cf. Ref. 21, Eq. (3.10)] that the probability per unit time that the detector atom makes a transition from the initial state $|\chi_g\rangle$ at time t_0 to some final state $|\chi_f\rangle$ at t_0+T is ($|\chi_g\rangle$ and $|\chi_f\rangle$ are the stationary states of the atom B, with energies $\mathcal{E}_i$ and $\mathcal{E}_f$, respectively)

$$\begin{aligned}R_{fg}(t_0+T,t_0)&=\sum_{mn}\langle\chi_g|P_m^{(B)}|\chi_f\rangle\langle\chi_f|P_n^{(B)}|\chi_g\rangle\\&\quad\times\int_0^T d\tau\,\langle E_m(\vec{R},T+t_0)E_n(\vec{R},T+t_0-\tau)\rangle\\&\quad\times e^{i\Omega_{gf}\tau}+\text{c.c.}\,,\\&\qquad\Omega_{gf}=\mathcal{E}_g-\mathcal{E}_f\,.\end{aligned} \tag{3.7}$$

For upward transitions (detector atom in the ground state) (3.7) simplifies to

$$\begin{aligned}R_{fg}(t_0+T,t_0)&=\sum_{mn}\langle\chi_g|P_m^{(B)}|\chi_f\rangle\langle\chi_f|P_n^{(B)}|\chi_g\rangle\\&\quad\times\int_0^T d\tau\,e^{i\Omega_{gf}\tau}G_{mn}^{(N)}(\vec{R},T+t_0,\vec{R},T+t_0-\tau)+\text{c.c.}\end{aligned} \tag{3.8}$$

In what follows we show explicitly the contribution to R coming from the beat terms in (3.4). On substituting (3.4) into (3.8) and on taking the long time limit and letting $t_0=R/c$, we get for the transition rate

$$\begin{aligned}R_{fg}^{(b)}\left(\frac{R}{c}+T,\frac{R}{c}\right)&=\sum_{\substack{j<i\\l<i}}\frac{(\omega_{li}\omega_{ji})^2}{c^4R^2}\{\vec{P}_{gf}^{(B)}\cdot[\hat{R}\times(\hat{R}\times\vec{d}^{ji})]\}\{\vec{P}_{fg}^{(B)}\cdot[\hat{R}\times(\hat{R}\times\vec{d}^{il})]\}\\&\quad\times\exp[iT(\omega_{jl}+i\Gamma_{jl})]\langle A_{jl}(0)\rangle(i\omega_{il}-i\Omega_{gf}+\tfrac{1}{2}(\Gamma_i-\Gamma_l))^{-1}+\text{c.c.}\end{aligned} \tag{3.9}$$

In deriving (3.9) we have ignored the rapidly oscillating terms. We thus see that the transition rates in the detector atom do show the modulation at the beat frequency.[9,10] It is also interesting to note that the width of each of the level $|j\rangle$, $|l\rangle$ of atom A gives the decay of the transition rates. One should note that the effects of the radiation reaction are included throughout in this treatment. One should also note that in the earlier works[20] one had computed (3.8) for the case when the source atom was a two level atom. The normally ordered intensity correlations are also easily computed using (2.9) and the quantum regression theorem. For example for the experimental situation of Haroche *et al.* one has

$$\langle :I(t)I(t+\tau):\rangle$$

$$=\sum_{\mu_1,\mu_2,\mu_3,\mu_4}(\vec{b}^{g\mu_1 *}\cdot\vec{b}^{g\mu_4})(\vec{b}^{*g\mu_2}\cdot\vec{b}^{g\mu_3})$$

$$\times\langle A_{\mu_1 g}(t)A_{\mu_2 g}(t+\tau)A_{g\mu_3}(t+\tau)A_{g\mu_4}(t)\rangle$$

$$=0\,, \qquad (3.10)$$

which is simply a reflection of the fact that in the problem there is only one photon. Note that $\langle :I(t)I(t+\tau):\rangle$ is proportional to the joint probability of detecting one photon at time t and another one at time $(t+\tau)$. For cascade type of transitions ($|1\rangle\to|2\rangle\to|3\rangle$) the normally ordered intensity correlation function is nonzero, e.g.,

$$\langle :I(t)I(t+\tau):\rangle$$
$$=|\hat{R}\times(\hat{R}\times\vec{d}^{12})|^2|\hat{R}\times(\hat{R}\times\vec{d}^{23})|^2R^{-4}\langle A_{11}(0)\rangle$$
$$\times\exp(-2\gamma_{32}\tau-2\gamma_{21}t)\,, \qquad (3.11)$$

which is as expected.

IV. QUANTUM BEATS AND THE ANTINORMALLY ORDERED CORRELATION FUNCTIONS OF THE SPONTANEOUSLY EMITTED RADIATION

The antinormally ordered correlation functions of the spontaneously emitted radiation are also expected to have beats. We will now examine the type of beats such correlation functions have. It is also well known[14] that a quantum counter measures the antinormally ordered correlation functions because such a counter works by the emission of photons. The probability per unit time that the atom B makes a downward transition is obtained from (3.7) and is given by ($\Omega_{if}>0$)

$$R_{fi}(t_0+T,t_0)=\sum_{mn}\langle\chi_i|P_m^{(B)}|\chi_f\rangle\langle\chi_f|P_n^{(B)}|\chi_i\rangle\int_0^T d\tau\, e^{i\Omega_{if}\tau}\langle E_m^{(+)}(\vec{R},T+t_0)E_n^{(-)}(\vec{R},T+t_0-\tau)\rangle+\text{c.c.} \qquad (4.1)$$

Thus our first object is the evaluation of the antinormally ordered correlation function

$$G_{mn}^{(A)}(\vec{r}_1,t_1;\vec{r}_2,t_2)=\langle E_m^{(+)}(\vec{r}_1,t_1)E_n^{(-)}(\vec{r}_2,t_2)\rangle\,, \qquad (4.2)$$

with $\vec{E}^{(\pm)}$ given by (2.9). On substituting (2.9) into (4.2), we find that

$$G_{mn}^{(A)}(\vec{r}_1,t_1,\vec{r}_2,t_2)=\sum_{i=1}^{4}C_{mn}^{(i)}(\vec{r}_1,t_1,\vec{r}_2,t_2);\; R_{fi}(t_0+T,t_0)=\sum_{i=1}^{4}R_{fi}^{(i)}(t_0+T,t_0)\,, \qquad (4.3)$$

where the different types of contributions are given by

$$C_{mn}^{(1)}(\vec{r}_1,t_1,\vec{r}_2,t_2)=\langle E_{0m}^{(+)}(\vec{r}_1,t_1)E_{0n}^{(-)}(\vec{r}_2,t_2)\rangle\,, \qquad (4.4)$$

$$C_{mn}^{(2)}(\vec{r}_1,t_1,\vec{r}_2,t_2)=\left\langle E_{0m}^{(+)}(\vec{r}_1,t_1)\sum_{j<i}A_{ji}^{(I)}\left(t_2-\frac{r_2}{c}\right)\right\rangle b_{e,n}^{ij\,*}(\vec{r}_2,t_2)\,, \qquad (4.5)$$

$$C_{mn}^{(3)}(\vec{r}_1,t_1,\vec{r}_2,t_2)=\left\langle\sum_{j<i}A_{ij}^{(I)}\left(t_1-\frac{r_1}{c}\right)E_{0n}^{(-)}(\vec{r}_2,t_2)\right\rangle b_{e,m}^{ij}(\vec{r}_1,t_1)=[C_{nm}^{(2)}(\vec{r}_2,t_2,\vec{r}_1,t_1)]^*\,, \qquad (4.6)$$

$$C_{mn}^{(4)}(\vec{r}_1,t_1,\vec{r}_2,t_2)=\sum_{\substack{j<i\\ l<k}}b_{e,m}^{ij}(\vec{r}_1,t_1)b_{e,n}^{kl\,*}(\vec{r}_2,t_2)\left\langle A_{ij}^{(I)}\left(t_1-\frac{r_1}{c}\right)A_{lk}^{(I)}\left(t_2-\frac{r_2}{c}\right)\right\rangle. \qquad (4.7)$$

It is clear that $C^{(1)}$ is the usual free-field contribution and leads to the usual spontaneous decay, i.e., in the absence of the atom A. We now examine $C^{(4)}$. The analysis similar to that used in connection with (3.1) shows that

$$C_{mn}^{(4)}\left(\vec{r}_1,t_1+\frac{r_1}{c},\vec{r}_2,t_2+\frac{r_2}{c}\right)=\sum_{\substack{i>j\\ k>j}}b_{e,m}^{ij}\left(\vec{r}_1,t_1+\frac{r_1}{c}\right)b_{e,n}^{kj\,*}\left(\vec{r}_2,t_2+\frac{r_2}{c}\right)\exp[-\Gamma_{ij}(t_1-t_2)]\langle A_{ik}^{(I)}(t_2)\rangle\,, \qquad (4.8)$$

and in particular for the same point $\vec{r}$, one has

$$C_{mn}^{(4)}\left(\vec{r},t+\frac{r}{c},\vec{r},t+\frac{r}{c}-\tau\right)=C_{mn,0}^{(4)}\left(\vec{r},t+\frac{r}{c},t+\frac{r}{c}-\tau\right)+C_{mn,b}^{(4)}\left(\vec{r},t+\frac{r}{c},\vec{r},t+\frac{r}{c}-\tau\right)\,, \qquad (4.9)$$

where $C_{mn}^{(4)},0[C_{mn,b}^{(4)}]$ denote the slowly varying [rapidly varying] terms in t. More explicitly one has

$$C^{(4)}_{mn,\mathrm{b}}\left(\vec{r}, t+\frac{r}{c}, \vec{r}, t+\frac{r}{c}-\tau\right) = \sum_{\substack{i>j\\k>j\\i\neq k}} \frac{(\omega_{ji}\omega_{jk})^2}{c^4 r^2}[\hat{r}\times(\hat{r}\times\vec{d}^{\,ij})]_m[\hat{r}\times(\hat{r}\times\vec{d}^{\,jk})]_n\langle A_{ik}(0)\rangle \exp[-it(\omega_{ki}-i\Gamma_{ki})]$$
$$\times\exp[-i\tau\omega_{jk}-\tfrac{1}{2}\tau(\Gamma_j-\Gamma_k)]. \tag{4.10}$$

Thus the contribution $C^{(4)}$ to the antinormally ordered correlation functions shows beats at ω_{ki} provided our multilevel atom is such that the transitions starting from a common upper level end at two lower levels and provided the initial state of the multilevel atom is such that there is an off diagonal matrix element between the two lower levels. Thus for a three level atom $C^{(4)}$ will show beats at ω_{23} provided the allowed transitions are $|1\rangle - |3\rangle$, $|1\rangle - |2\rangle$ and provided the initial state is such that $\rho_{23}\neq 0$. Note that the retardation character of (2.9) implies that $C^{(4)}$ is nonvanishing provided $t_1 > r_1/c$, $t_2 > r_2/c$. Such type of beats are not found in normally ordered correlations (cf. Ref. 22).

Let us now examine the terms $C^{(2)}$, $C^{(3)}$. These arise from the interference of the zero-point fluctuations and the source fields. The computation of the correlation function of the free field and atomic operators is rather involved. We relegate the details of this calculation to the Appendix. Using the Eq. (A8) we find that

$$C^{(2)}_{mn}\left(\vec{r}_1, t_1, \vec{r}_2, t_2+\frac{r_2}{c}\right) = -\sum_{j<i}\sum_{\alpha\beta q} b^{ij*}_{e,n}\left(\vec{r}_2, t_2+\frac{r_2}{c}\right) d^{\alpha\beta}_q$$
$$\times\exp(-i\omega_{ji}t_1)\Big[\delta_{\beta j}\tilde{\chi}_{mqEE}(\vec{r}_1, 0, \omega_{j\alpha})\Big\langle A_{\alpha i}\left(t_1-\frac{r_1}{c}\right)\Big\rangle e^{i\omega_{\alpha i}r_1/c}$$
$$-\delta_{i\alpha}\tilde{\chi}_{mqEE}(\vec{r}_1, 0, \omega_{\beta i})\Big\langle A_{j\beta}\left(t_1-\frac{r_1}{c}\right)\Big\rangle e^{i\omega_{j\beta}r_1/c}\Big] \tag{4.11}$$

$$= -\sum_{j<i}\sum_{\alpha>j}[\hat{r}_1\times(\hat{r}_1\times\vec{d}^{\,\alpha j})]_m[\hat{r}_2\times(\hat{r}_2\times\vec{d}^{\,ji})]_n\frac{\omega^2_{ji}\omega^2_{j\alpha}}{c^4 r_1 r_2}\Big\langle A^{(I)}_{\alpha i}\left(t_1-\frac{r_1}{c}\right)\Big\rangle$$
$$\times\exp\left(i\omega_{\alpha i}t_1 - i\omega_{ji}(t_1-t_2)+i\omega_{j\alpha}\frac{r_1}{c}\right)$$
$$+\sum_{j<i}\sum_{\beta<i}[\hat{r}_1\times(\hat{r}_1\times\vec{d}^{\,i\beta})]_m[\hat{r}_2\times(\hat{r}_2\times\vec{d}^{\,ji})]_n\frac{\omega^2_{ji}\omega^2_{\beta i}}{c^4 r_1 r_2}\Big\langle A^{(I)}_{j\beta}\left(t_1-\frac{r_1}{c}\right)\Big\rangle$$
$$\times\exp\left(i\omega_{j\beta}t_1 + i\omega_{\beta i}\frac{r_1}{c} - i\omega_{ji}(t_1-t_2)\right). \tag{4.12}$$

Equation (4.12) is seen to have two different kinds of beats with respect to the time t_1 [keeping (t_1-t_2) constant]. The first term is characteristic of the lower-state beats, whereas the second term shows beats at the upper-state frequencies.

The expression for the transition rate is now easily obtained by substituting (4.4), (4.7), (4.10), and (4.12) into (4.1). It is clear that $C^{(2)}=0$ for $t_1 > t_2 + r_1/c$ and thus $C^{(2)}$ will not contribute to the downward transition rate. The explicit expression for $C^{(3)}$ becomes

$$C^{(3)}_{mn}\left(\vec{R}, T+\frac{2R}{c}, \vec{R}, T+\frac{2R}{c}-\tau\right) = -\sum_{j<i}\sum_{\alpha>j}[\hat{R}\times(\hat{R}\times\vec{d}^{\,j\alpha})]_n[\hat{R}\times(\hat{R}\times\vec{d}^{\,ij})]_m\frac{\omega^2_{ji}\omega^2_{j\alpha}}{c^4R^2}$$
$$\times\Big\langle A^{(I)}_{i\alpha}\left(T+\frac{R}{c}-\tau\right)\Big\rangle\exp\left[-i\omega_{ji}\tau - i\omega_{\alpha i}\left(T+\frac{R}{c}-\tau\right)\right]$$
$$+\sum_{j<i}\sum_{\beta<i}\frac{\omega^2_{ji}\omega^2_{\beta i}}{c^4R^2}[\hat{R}\times(\hat{R}\times\vec{d}^{\,\beta i})]_n[\hat{R}\times(\hat{R}\times\vec{d}^{\,ij})]_m$$
$$\times\Big\langle A^{(I)}_{\beta j}\left(T+\frac{R}{c}-\tau\right)\Big\rangle\exp\left[-i\omega_{j\beta}\left(T+\frac{R}{c}-\tau\right)-i\omega_{ji}\tau\right], \tag{4.13}$$

and hence the contribution of $C^{(3)}, C^{(4)}$ to the downward transition rate of the atom B becomes (in what follows we are only writing the contribution from the terms which "beat")

$$R_{fi}^{(4)}=\sum_{\substack{i>j\\k>j}}\langle\chi_i|\vec{P}^{(B)}\cdot[\hat{R}\times(\hat{R}\times\vec{d}^{\,ij})]|\chi_f\rangle\langle\chi_f|\vec{P}^{(B)}\cdot[\hat{R}\times(\hat{R}\times\vec{d}^{\,jk})]|\chi_i\rangle$$
$$\times\frac{\omega_{ji}^2\omega_{jk}^2}{R^2c^4}\langle A_{ik}(0)\rangle\exp(-it\omega_{ki}-\Gamma_{ik}t)[i\omega_{jk}-i\Omega_{if}+\tfrac{1}{2}(\Gamma_j-\Gamma_k)]^{-1}+\text{c.c.}, \tag{4.14}$$

$$R_{fi}^{(3)}=\sum_{\substack{i>j\\\alpha>j}}\langle\chi_i|\vec{P}^{(B)}\cdot[\hat{R}\times(\hat{R}\times\vec{d}^{\,ij})]|\chi_f\rangle\langle\chi_f|\vec{P}^{(B)}\cdot[\hat{R}\times(\hat{R}\times\vec{d}^{\,j\alpha})]|\chi_i\rangle$$
$$\times\frac{\omega_{ji}^2\omega_{j\alpha}^2}{c^4R^2}\exp\left[-i\left(T+\frac{R}{C}\right)(\omega_{\alpha i}-i\Gamma_{i\alpha})\right][i\omega_{\alpha j}+i\Omega_{if}+\Gamma_{i\alpha}]^{-1}\langle A_{i\alpha}(0)\rangle$$
$$-\sum_{j<i}\sum_{\beta<i}\langle\chi_i|\vec{p}^{(B)}\cdot[\hat{R}\times(\hat{R}\times\vec{d}^{\,ij})]|\chi_f\rangle\langle\chi_f|\vec{p}^{(B)}\cdot[\hat{R}\times(\hat{R}\times\vec{d}^{\,\beta i})]|\chi_i\rangle$$
$$\times\frac{\omega_{ji}^2\omega_{\beta i}^2}{c^4R^2}\exp\left[-i\left(T+\frac{R}{c}\right)(\omega_{j\beta}-i\Gamma_{j\beta})\right](i\omega_{i\beta}+i\Omega_{if}+\Gamma_{\beta j})^{-1}\langle A_{\beta g}(0)\rangle+\text{c.c.} \tag{4.15}$$

Note $R^{(2)}=0$ and $R^{(1)}$ denotes the usual transition rate, i.e., in the absence of any source atom. As discussed before in connection of the structure of $C^{(3)}$, $C^{(4)}$ it is clear that $R^{(4)}$ has beats at the lower-level frequencies, whereas $R^{(3)}$ has beats at *both lower-level and upper-level frequencies*. The results (4.14) and (4.15) agree with those of Khoo and Eberly[9,10] obtained by using the method of Heisenberg-picture perturbation theory.

The beats found in Secs. III and IV are very much reflections of the properties of the normally and the antinormally ordered correlation functions of the polarization operators of the noninteracting multilevel atom. If we define

$$\vec{\mathcal{P}}^{(+)}_{,\mu}=\sum_{j<i}\mu_{ij}\vec{d}^{\,ij}A_{ij},\quad \vec{\mathcal{P}}^{(-)}_{,\mu}=\sum_{j>i}\mu_{ij}^*\vec{d}^{\,ij}A_{ij}, \tag{4.16}$$

then

$$\langle\mathcal{P}^{(-)}_{\alpha,\mu}(t)\mathcal{P}^{(+)}_{\beta,\nu}(t-\tau)\rangle=\sum_{\substack{j<i\\l<i}}\nu_{il}\mu_{ji}^*d_\alpha^{ji}d_\beta^{il}\langle A_{jl}\rangle\times e^{-i\omega_{il}\tau+i\omega_{jl}t}, \tag{4.17}$$

$$\langle\mathcal{P}^{(+)}_{\alpha,\mu}(t)\mathcal{P}^{(-)}_{\beta,\nu}(t-\tau)\rangle=\sum_{\substack{j<i\\j<k}}\mu_{ij}\nu_{jk}^*d_\alpha^{ij}d_\beta^{jk}\langle A_{ik}\rangle\times e^{-i\omega_{jk}\tau+i\omega_{ik}t}. \tag{4.18}$$

The correlations (4.17) and (4.18) show, respectively, the upper-state and lower-state beats.

V. ATOMIC COLLECTIVE EFFECTS AND QUANTUM BEATS

So far we have considered the quantum beats in the radiation from a single multilevel atom. We saw that the second quantized property $A_{ij}A_{kl}=\delta_{jk}A_{il}$ essentially decides the types of beats which would be present in a given type of correlation. Here we would like to consider a collection of multilevels atoms (located in region whose linear dimensions are smaller than an optical wavelength) and the beats in the radiation emitted from such a system. Formulas like (2.9), (A8), and (3.1) remain valid, with the difference that A_{ij} in these should be replaced by the collective operators $\mathcal{A}_{ij}$ defined by

$$\mathcal{A}_{ij}=\sum_n A_{ij}^{(n)},\ [\mathcal{A}_{ij},\mathcal{A}_{kl}]=\mathcal{A}_{il}\delta_{jk}-\mathcal{A}_{kj}\delta_{il}, \tag{5.1}$$

however, the second quantized property (3.3) holds no longer for the operators $\mathcal{A}_{ij}$. The atomic dynamics is again governed by Eq. (2.6) with A_{ij} replaced by collective atomic operators. However $\rho^{(0)}$ does not satisfy simple equations[23] like (2.4) and (2.5) and the solution for $\rho^{(0)}$ is rather difficult to obtain except in an approximate sense.[24,25] One has from (3.1) the following result for the intensity at the point $\vec{r}$:

$$I\left(\vec{r},t+\frac{r}{c}\right)=\sum_{\substack{j<i\\l<k}}\frac{\omega_{ji}^2\omega_{lk}^2}{c^4r^2}\times[\hat{r}\times(\hat{r}\times\vec{d}^{\,ji})]\cdot[\hat{r}\times(\hat{r}\times\vec{d}^{\,kl})]\times\langle\mathcal{A}_{ji}(t)\mathcal{A}_{kl}(t)\rangle. \tag{5.2}$$

In general, it is not possible to calculate the mean values appearing in (5.2). To see the qualitative features of the beats present in (5.2), we ignore the effect of the radiation reaction in the calculation of the mean values and thus we make the replacement

$$\langle\mathcal{A}_{ji}(t)\mathcal{A}_{kl}(t)\rangle\approx e^{it(\omega_{ji}+\omega_{kl})}\langle\mathcal{A}_{ji}\mathcal{A}_{kl}\rangle_0, \tag{5.3}$$

where $\langle\ \rangle_0$ denotes the initial state of the system. We assume that the atomic system is prepared initially in an atomic coherent state. In what follows we consider specifically the three level systems, the systems with more than three levels are similarly treated.

For the case when the allowed transitions are $|1\rangle \to |2\rangle$, $|1\rangle \to |3\rangle$, one finds from (5.2) and (5.3) that the beat amplitude in the intensity is

$$a=\frac{\omega_{12}^2\omega_{13}^2}{c^4r^2}\{[\hat{r}\times(\hat{r}\times\vec{d}^{21})]\cdot[\hat{r}\times(\hat{r}\times\vec{d}^{13})] \times\langle\alpha_{13}\alpha_{21}\rangle e^{it\omega_{23}}+\text{c.c.}\}\ , \tag{5.4}$$

which on using relations (B3) and (B5) reduces to

$$a=\frac{2\omega_{12}^2\omega_{23}^2}{c^4r^2}|[\hat{r}\times(\hat{r}\times\vec{d}^{21})]\cdot[\hat{r}\times(\hat{r}\times\vec{d}^{13})]| \times\cos(\omega_{23}t+\varphi_0-\theta_2)N(N-1)I_1[I_2(1-I_1-I_2)]^{1/2}\ , \tag{5.5}$$

where φ_0 is the phase of $[\hat{r}\times(\hat{r}\times\vec{d}^{21})]\cdot[\hat{r}\times(\hat{r}\times\vec{d}^{13})]$. Thus the beat amplitude will be nonzero if $N\neq1, z_1\neq0, z_2\neq0$. We thus see that the collective effects make the beat amplitude nonzero. It is also interesting to note that the beat amplitude is proportional to the square of the number of the atoms,[26] which is typical of cooperative effects. This should be compared with the prediction of neoclassical theory, which does in fact predict beats even for a single atom.

Some effects of the radiation reaction can be put in the limit of large N. It has been shown elsewhere[24] that the master equation (2.21) is equivalent to the following Langevin equations (in the atomic coherent state representation)

$$\begin{aligned}\dot{I}_1&=-2\gamma_{21}I_1[1+(N-1)I_2]\\&\quad-2\gamma_{31}I_1[N-(N-1)(I_1+I_2)]\ ,\\ \dot{I}_2&=2\gamma_{21}I_1[1+(N-1)I_2]\\&\quad-2\gamma_{32}I_2[N-(N-1)(I_1+I_2)]\ ,\end{aligned} \tag{5.6}$$

where we have ignored the diffusion terms. This approximation is expected to work in the limit of large N and if the initial state of the atom is not a completely inverted state. The general solution of (5.6) depends on the relation of various γ's to each other. The solutions of Eqs. (5.6) are easily found by quadratures. For the case when $\gamma_{32}=0$, $\gamma_{21}=\gamma_{31}=\Gamma$, we find that

$$\begin{aligned}I_1(t)&=\frac{(N+1)}{2(N-1)}\{1-\tanh(N+1)\Gamma(t-t_0)\}\ ,\\ I_2(t)&=\frac{1}{N-1}\{ce^{2(N+1)\Gamma t_0}-\tfrac{1}{2}(1+ce^{2(N+1)\Gamma t_0})\\&\quad\times[1-\tanh(N+1)\Gamma(t-t_0)]\}\ ,\end{aligned} \tag{5.7}$$

where

$$e^{-2(N+1)\Gamma t_0}=\frac{N+1}{(N-1)I_1'}-1,\quad c=\frac{(N+1)I_2'}{I_1'}+1\ . \tag{5.8}$$

The time dependence of the beat amplitude can now be obtained by substituting (5.7) in (5.5).

For the case when $\gamma_{21}=0$, $\gamma_{31}=\gamma_{32}=\Gamma$, we find that

$$\begin{aligned}I_1(t)&=\frac{1}{4}\left(I_1'-I_2'+\frac{N}{N-1}\right)\\&\quad\times\{1-\tanh\Gamma[N+(N-1)(I_1'-I_2')](t-t_0)\}\ ,\\ I_2(t)&=I_1(t)+I_2'-I_1',\quad I'=I(0)\ .\end{aligned} \tag{5.9}$$

In this case the beat amplitude is given by

$$a=\frac{\omega_{13}^2\alpha_{23}^2}{c^2r^4}|[\hat{r}\times(\hat{r}\times\vec{d}^{31})]\cdot[\hat{r}\times(\hat{r}\times\vec{d}^{23})] \times(\langle\alpha_{23}\alpha_{31}\rangle e^{i\omega_{21}t+i\varphi_0}+\text{c.c.})\ , \tag{5.10}$$

where the relevant expectation value is given by (B4). The time dependence of the beat amplitude can be obtained by substituting (5.9) and (B4) into (5.10). If we ignore terms of order $(1/N)$ and let $I_1(0)=I_2(0)$, then

$$a=\frac{\omega_{13}^2\omega_{23}^2}{4c^2r^4}|[\hat{r}\times(\hat{r}\times\vec{d}^{31})]\cdot[\hat{r}\times(\hat{r}\times\vec{d}^{23})]| \times\text{sech}^2[N\Gamma(t-t_0)] \times\cos[\omega_{21}t+(\theta_1-\theta_2)+\varphi_0]\ . \tag{5.11}$$

We have thus shown that the collective effects can play an important role in the determination of the existence of beats. We have also presented the approximate time dependence of the beat amplitudes. The approximate time dependence of such beat amplitudes is very similar to the one found in neoclassical theory.

APPENDIX A: CALCULATION OF THE CORRELATION FUNCTION $\langle E_{0m}^{(+)}(\vec{R}_1,t_1)A_{ij}(t_2)\rangle$

In this Appendix we will calculate the correlation function $\langle E_{0m}^{(+)}(\vec{R}_1,t_1)A_{ij}(t_2)\rangle$ in the lowest order in the perturbation theory. Our method of proof is similar to that used in our earlier work[15] on the calculation of the radiation reaction field. Any Heisenberg operator can be expressed as

$$A_{ji}(t_2)=\exp(i\mathcal{L}t_2)A_{ji}(0),\quad \mathcal{L}=[H,\ ], \tag{A1}$$

where H is the full Hamiltonian of the system. Now using the identity

$$e^{i\mathcal{L}t}=e^{i\mathcal{L}_0t}+\int_0^t d\tau e^{i\mathcal{L}\tau}i\mathcal{L}_1e^{i\mathcal{L}_0(t-\tau)}, \tag{A2}$$

we can write (A1) as

$$A_{ji}(t_2)=e^{i\omega_{ji}t_2}A_{ji}(0)-i\int_0^{t_2}d\tau\, e^{i\mathcal{L}}\sum_{\alpha\beta n}d_n^{\alpha\beta}e^{i\omega_{ji}(t_2-\tau)}E_n(0,0)\,(A_{\alpha i}\delta_{\beta j}-A_{j\beta}\,\delta_{i\alpha})$$

$$=e^{i\omega_{ji}t_2}A_{ji}(0)-i\sum_{\alpha\beta n}d_n^{\alpha\beta}\int_0^{t_2}d\tau\, e^{i\omega_{ji}(t_2-\tau)}E_n(0,\tau)\,[A_{\alpha i}(\tau)\delta_{\beta j}-A_{j\beta}(\tau)\delta_{i\alpha}]. \quad \text{(A3)}$$

Using (A3) and the fact that the radiation field is initially in vacuum state, the correlation function becomes

$$\Lambda_{mji}(\vec{R}_1,t_1,t_2)=\langle E_{0m}^{(+)}(\vec{R}_1,t_1)A_{ji}(t_2)\rangle$$

$$=-i\sum_{\alpha\beta n}d_n^{\alpha\beta}\int_0^{t_2}d\tau\, e^{i\omega_{ji}(t_2-\tau)}\langle E_{0m}^{(+)}(\vec{R}_1,t_1)[A_{\alpha i}(\tau)\delta_{\beta j}-A_{j\beta}(\tau)\delta_{i\alpha}]E_n(0,\tau)\rangle. \quad \text{(A4)}$$

Since we want to calculate the correlation function in the lowest order in the coupling constant $d_n^{\alpha\beta}$, we can approximate (A4) by

$$\Lambda_{mji}(\vec{R}_1,t_1,t_2)=-i\sum_{\alpha\beta n}d_n^{\alpha\beta}\int_0^{t_2}d\tau\, e^{i\omega_{ji}(t_2-\tau)}\langle E_{0m}^{(+)}(\vec{R}_1,t_1)\,E_{0n}^{(-)}(0,\tau)\rangle[\langle A_{\alpha i}(\tau)\rangle\delta_{\beta j}-\langle A_{j\beta}(\tau)\rangle\delta_{i\alpha}]. \quad \text{(A5)}$$

On substituting the value of the free-field correlation function [Ref. 21, Eq. (5.15); $\chi''(\tau)=\operatorname{Im}\chi(\tau)$],

$$\langle E_{0m}^{(+)}(\vec{R}_1,t_1)E_{0n}^{(-)}(0,\tau)\rangle=\frac{1}{\pi}\int_0^{\infty}d\omega\,\chi''_{mnEE}(\vec{R}_1,0,\omega)e^{-i\omega(t_1-\tau)} \quad \text{(A6)}$$

in (A5), we obtain the result

$$\Lambda_{mji}(\vec{R}_1,t_1,t_2)=-\frac{i}{\pi}\sum_{\alpha\beta n}d_n^{\alpha\beta}\int_0^{t_2}d\tau\int_0^{\infty}d\omega\,\chi''_{mnEE}(\vec{R}_1,0,\omega)e^{-\omega(t_1-\tau)}e^{i\omega_{ji}(t_2-\tau)}[\langle A_{\alpha i}(\tau)\rangle\delta_{\beta j}-\langle A_{j\beta}(\tau)\rangle\delta_{i\alpha}]. \quad \text{(A7)}$$

The above expression for the correlation function can be simplified in the long-time approximation and in the far zone. The standard procedure (cf. Ref. 15, Sec. VI) then leads to

$$\Lambda_{mji}(\vec{R}_1,t_1,t_2)=-\sum_{\alpha\beta q}d_q^{\alpha\beta}e^{-i\omega_{ji}(t_1-t_2)}\Big[\langle A_{\alpha i}^{(I)}(t_1-R_1/c)\rangle e^{i\omega_{\alpha i}t_1}\delta_{\beta j}\tilde{\chi}_{mqEE}(\vec{R}_1,0,\omega_{j\alpha})-\Big\langle A_{j\beta}^{(I)}\Big(t_1-\frac{R_1}{c}\Big)\Big\rangle e^{i\omega_{j\beta}t_1}$$

$$\times\,\delta_{i\alpha}\tilde{\chi}_{mqEE}(\vec{R}_1,0,\omega_{\beta i})\Big] \quad \text{for } t_1<t_2+\frac{R_1}{c} \quad \text{(A8)}$$

$$=0 \quad \text{for } t_1>t_2+R_1/c.$$

The summation over $j\alpha$ $(i\beta)$ in the first term (second term) is only over those indices such that $j<\alpha$ [$\beta<i$]. It is also obvious from (A7) that Λ is zero unless $t_1>(R_1/c+\text{minimum value of } t_2)$. The appearance of beats in Λ is again to be noted.

APPENDIX B: CALCULATION OF THE EXPECTATION VALUES IN THE ATOMIC COHERENT STATE REPRESENTATION

We have seen in Sec. V that the calculation of the beat amplitude requires the knowledge of the expectation values involving the collective atomic operators. As in the experiments on beats, the atomic system is prepared in an excited state by an external laser beam, and hence it is natural to assume that initially the atomic system is in a coherent state[24, 25] $|z_1,z_2\rangle$ defined by

$$|z_1,z_2\rangle=(1+|z_1|^2+|z_2|^2)^{-N/2}\times\sum\sum\frac{z_1^{n_1}z_2^{n_2-n_1}(N!)^{1/2}}{[(n_1!\,(n_2-n_1)!\,(N-n_2)!)]^{1/2}}|N,n_1,n_2\rangle, \quad \text{(B1)}$$

where the state $|N,n_1,n_2\rangle$ represents a collective state of the system. The matrix elements involving the collective atomic operators can be calculated using the relations[24]

$$
\begin{aligned}
\mathcal{A}_{12}|z_1,z_2\rangle\langle z_1,z_2| &= \left(z_2\frac{\partial}{\partial z_1}+\frac{Nz_1^*z_2}{1+|z_1|^2+|z_2|^2}\right)|z_1,z_2\rangle\langle z_1,z_2|,\\
\mathcal{A}_{21}|z_1,z_2\rangle\langle z_1,z_2| &= \left(z_1\frac{\partial}{\partial z_2}+\frac{Nz_1z_2^*}{1+|z_1|^2+|z_2|^2}\right)|z_1,z_2\rangle\langle z_1,z_2|,\\
\mathcal{A}_{13}|z_1,z_2\rangle\langle z_1,z_2| &= \left(\frac{\partial}{\partial z_1}+\frac{Nz_1^*}{1+|z_1|^2+|z_2|^2}\right)|z_1,z_2\rangle\langle z_1,z_2|,\\
\mathcal{A}_{31}|z_1,z_2\rangle\langle z_1,z_2| &= z_1\left(\frac{N}{1+|z_1|^2+|z_2|^2}-z_1\frac{\partial}{\partial z_1}-z_2\frac{\partial}{\partial z_2}\right)|z_1,z_2\rangle\langle z_1,z_2|,\\
\mathcal{A}_{23}|z_1,z_2\rangle\langle z_1,z_2| &= \left(\frac{\partial}{\partial z_2}+\frac{Nz_2^*}{1+|z_1|^2+|z_2|^2}\right)|z_1,z_2\rangle\langle z_1,z_2|,\\
\mathcal{A}_{32}|z_1,z_2\rangle\langle z_1,z_2| &= z_2\left(\frac{N}{1+|z_1|^2+|z_2|^2}-z_1\frac{\partial}{\partial z_1}-z_2\frac{\partial}{\partial z_2}\right)|z_1,z_2\rangle\langle z_1,z_2|.
\end{aligned}
\tag{B2}
$$

We illustrate the calculation of the following matrix elements, which are used in Sec. V:

$$
\begin{aligned}
\langle\mathcal{A}_{13}\mathcal{A}_{21}\rangle &= \mathrm{Tr}\left(z_1\frac{\partial}{\partial z_2}+\frac{Nz_1z_2^*}{1+|z_1|^2+|z_2|^2}\right)\mathcal{A}_{13}|z_1,z_2\rangle\langle z_1,z_2|\\
&= \left(z_1\frac{\partial}{\partial z_2}+\frac{Nz_1z_2^*}{1+|z_1|^2+|z_2|^2}\right)\left(\frac{\partial}{\partial z_1}+\frac{Nz_1^*}{1+|z_1|^2+|z_2|^2}\right)\mathrm{Tr}\,|z_1,z_2\rangle\langle z_1,z_2| = \frac{N(N-1)|z_1|^2z_2^*}{(1+|z_1|^2+|z_2|^2)^2},
\end{aligned}
\tag{B3}
$$

$$
\langle\mathcal{A}_{23}\mathcal{A}_{31}\rangle = z_1\left(\frac{N}{1+|z_1|^2+|z_2|^2}-z_1\frac{\partial}{\partial z_1}-z_2\frac{\partial}{\partial z_2}\right)\frac{Nz_2^*}{1+|z_1|^2+|z_2|^2} = \frac{Nz_1z_2^*(N+|z_1|^2+|z_2|^2)}{(1+|z_1|^2+|z_2|^2)^2}.
\tag{B4}
$$

It turns out convenient to introduce the intensity and phase variables defined by

$$
z_1=\left(\frac{I_1}{1-I_1-I_2}\right)^{1/2}e^{i\theta_1},\quad z_2=\left(\frac{I_2}{1-I_1-I_2}\right)^{1/2}e^{i\theta_2}.
\tag{B5}
$$

As indicated in the text, the equations of motion acquire rather simple form in terms of such variables.

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[26]Note that if the atomic ensemble is more general than (5.5), i.e., when

$$\rho(0)=\int \Phi(z_1,z_2)|z_1,z_2\rangle\langle z_1,z_2|d^2z_1d^2z_2,$$

then the corresponding beat amplitudes are to be obtained by averaging (5.7) over the distribution of Φ. For the sake of completeness we also record here the condition that ρ represent an equilibrium ensemble, i.e.,

$$\left[\sum E_{jj}A_{jj},\rho\right]=0 \Rightarrow E_1\frac{\partial}{\partial\theta_1}\Phi+E_2\frac{\partial}{\partial\theta_2}\Phi=0,\ \ (E_3=0),$$

and hence in order that Φ represent an equilibrium ensemble, it must be a functional only of $(\theta_1 E_2-E_1\theta_2)$.