Quantum-beat phenomena described by quantum electrodynamics and neoclassical theory*

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We propose and analyze the use of quantum-beat phenomena to test neoclassical radiation theory (NCT) and quantum electrodynamics (QED). For a beam-foil type of experiment with atoms having one upper level and two closely spaced lower levels, all coherently excited, NCT predicts the presence of quantum beats in the emitted radiation; beats are not expected in QED. QED predicts beats when many atoms are present, in agreement with recent photon-echo experiments. An experiment to test NCT and QED is suggested.

I. INTRODUCTION

At present, quantum electrodynamics (QED) represents an apex of modern theoretical physics, for it gives, with the help of certain renormalization prescriptions, remarkable agreement with experiments to many significant figures. Of course it suffers from the presence of infinities. In this regard the foundations of QED, for example, the quantization of the electromagnetic field, and the necessity of this quantization have been subjects of recent discussion.[1] For instance, the self-consistent combination of Maxwell and Schrödinger equations has been used successfully to treat many effects which are often thought to require QED.[2-7] The procedure involves treating the expectation value of the dipole moment operator as the electric dipole of an ensemble of radiating atoms. Neoclassical theory[1] (NCT) carries this procedure further by associating the expectation value of the dipole moment operator with the actual dipole of an atom, thus allowing one to consider each atom individually. So far experiments in areas where differences in the predictions of NCT and QED exist support QED.[8] In this paper we show that quantum-beat phenomena provide an example in which the predictions of QED and NCT are qualitatively different.

One difference[1] between the predictions of QED and NCT concerns the shape of the spontaneous emission pulse from an atom. Since NCT considers the expectation value of the dipole moment operator as an actual dipole which radiates according to classical electrodynamics, it predicts different pulse shapes for different degrees of initial excitation of the atom. According to NCT, an atom purely in an excited state will not decay. An atom that is excited almost entirely to the excited state will decay with the emission of a chirped hyperbolic-secant pulse that has peak intensity at the time when this atom has equal probability of being in the excited and the ground states. Furthermore, when the excitation leaves an atom predominately in the ground state, then NCT predicts the emission of an exponentially decaying pulse. Contrary to NCT, QED predicts the emission of an exponentially decaying pulse for all degrees of initial excitation of the atom. Consequently, to see the difference in the predictions of QED and NCT, one requires atoms that are predominately in the excited state; as proponents of NCT point out,[9] this condition is experimentally difficult to fulfill. In the present work this difficulty is circumvented by using quantum beats to test QED and NCT.[10]

Quantum beats have been observed in beam-foil experiments where each atom is impulsively excited by passage through a carbon foil to a superposition of two closely spaced upper levels and a lower level [Fig. 1(a), henceforth referred to as atom of type I].[11] A typical experimental geometry and apparatus is shown in Fig. 2. From this figure, one sees that the instant when an excited atom radiates light is determined by the point in space at which radiation occurred. Thus beam images recorded on the photographic film in the spectrograph show a decline in the blackening from one end to the other because of the finite lives of the excited levels. Time-dependent oscillations (quantum beats) are transformed into spatial variations in the intensities of the beam images. The sharply defined origin in space (time) required for the observation of quantum beats is provided by the exit surface of the foil. A point whose importance will become clear later is that at normal beam current; the photographic film rarely receives light from more than one atom at a time. For the above experiment the presence of quantum beats is predicted by both QED and NCT.

However, if for the above experiment the atom is excited to a superposition of one upper and two closely spaced lower states [Fig. 1(b), henceforth referred to as atom of type II], then QED does not predict quantum beats while NCT does. No serious search has been made for lower-state beats; however, an experiment designed to look for beats from type-II atoms provides a straightforward test of QED and NCT.

It should be emphasized that the recent photon-echo experiments seem to indicate the presence of beats from atomic ensembles of type II.[12,13] However, one must be careful in the analysis of these experiments, for they are different from the beam-foil experiments in that here one looks at emissions from many atoms simultaneously. In this situation we show that there is essentially no difference between the QED and NCT predictions.

In Sec. II we present the QED and NCT treatments of quantum beats from a single type-I atom. We show that both QED and NCT predict the presence of beats in this case. We show in Sec. III that QED and NCT differ in their predictions of quantum beats from a single type-II atom. In Sec. IV we present results of calculations which indicate that QED distinguishes between single-atom beats and many-atom beats, a detail that is not present in NCT. Finally, in Sec. V we propose an experiment, using type-II atoms, that would serve as a test for NCT and QED.

II. QUANTUM BEATS FROM A SINGLE TYPE-I ATOM

Quantum beats have been observed when an atom is excited to a coherent superposition of closely spaced upper levels [Fig. 1(a)].[11,14] NCT describes the modulation of the radiated intensity as a result of two dipoles created between $|a\rangle$ and $|c\rangle$, and between $|b\rangle$ and $|c\rangle$. That is, an atom excited at time t_0 has at time t the state vector

$$|\psi(t,t_0)\rangle = A(t)e^{-i\omega_\alpha(t-t_0)}|a\rangle + B(t)e^{-i\omega_\beta(t-t_0)}|b\rangle + C(t)|c\rangle .$$

Thus

$$\langle\psi(t,t_0)|r|\psi(t,t_0)\rangle = A^*(t)C(t)e^{i\omega_\alpha(t-t_0)}\langle a|r|c\rangle + B^*(t)C(t)e^{i\omega_\beta(t-t_0)}\langle b|r|c\rangle + \text{c.c.}$$

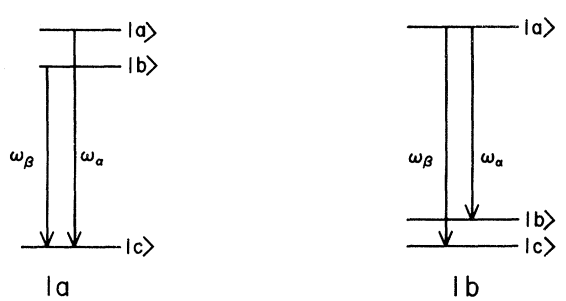

FIG. 1. Energy levels for (a) an atom (type I) with two upper and one lower state; (b) an atom (type II) with two lower and one upper state.

Treating the expectation value of the dipole moment operator as an actual dipole which radiates according to classical electrodynamics, we have the radiated intensity as being proportional to

$$|A^*(t)C(t)\langle a|r|c\rangle|^2 + |B^*(t)C(t)\langle b|r|c\rangle|^2 + 2\,\mathrm{Re}[A^*(t)B(t)|C(t)|^2\langle a|r|c\rangle \times\langle c|r|b\rangle e^{i(\omega_\alpha-\omega_\beta)(t-t_0)}] .$$

However, each measurement for the radiation involves an atom which is excited at a time different from the others. Hence, the beat intensity is actually proportional to

$$2\,\mathrm{Re}\left(A^*(t)B(t)|C(t)|^2\langle a|r|c\rangle\langle c|r|b\rangle \times\frac{\sin[(\omega_\alpha-\omega_\beta)\tau/2]}{(\omega_\alpha-\omega_\beta)\tau/2}\, e^{i(\omega_\alpha-\omega_\beta)t}\right),$$

where τ is the range of excitation times. We note that for beats to be seen the necessary coherence in the wave function is achieved by limiting the range of excitation times to $\tau < 1/(\omega_\alpha - \omega_\beta)$. In foil excitation[11] and in optical excitation[14]

$$\frac{\sin[(\omega_\alpha-\omega_\beta)\tau/2]}{(\omega_\alpha-\omega_\beta)\tau/2}\simeq 1 ,$$

where in foil excitation τ is the foil thickness per average velocity of the atoms.

That QED also predicts quantum beats may readily be seen. Initially, the atom is in a coherent superposition of the upper states shown in Fig. 1(a), and the electromagnetic field is in the vacuum state. Then,

$$|\psi(t_0)\rangle = A_0|a0\rangle + B_0|b0\rangle + C_0|c0\rangle ,$$

where $|\psi(t)\rangle$ is the total wave function, $|j\,0\rangle$ means atomic state $|j\rangle$ and the vacuum electromagnetic field, and A_0, B_0, and C_0 are constants. Since dipole transitions are allowed between $|a\rangle$ and $|c\rangle$ and between $|b\rangle$ and $|c\rangle$, $|\psi(t_0)\rangle$ evolves into

$$|\psi(t)\rangle = A(t)|a0\rangle + B(t)|b0\rangle + C(t)|c0\rangle + A_1(t)|c1_\alpha\rangle + B_1(t)|c1_\beta\rangle .$$

Writing the electric field operator as

$$E(\vec{x},t) = \sum_{\vec{k}} \mathcal{E}_k (a_{\vec{k}} e^{i\vec{k}\cdot\vec{x}-i\omega_k t} + a_{\vec{k}}^\dagger e^{-\vec{k}\cdot\vec{x}+i\omega_k t}),$$

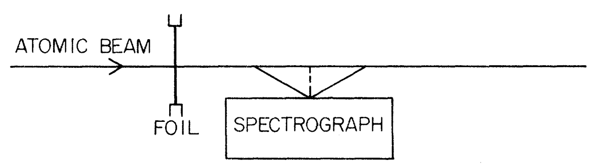

FIG. 2. Experimental geometry and apparatus for a beam-foil type of quantum beat experiment.

where $\mathcal{E}_k$ is the electric field per photon, we see that the presence of beats is indicated by the matrix element

$$\langle\, \psi(t)|\, a_\alpha^\dagger a_\beta|\, \psi(t)\rangle = A_1^*(t)B_1(t)\,,$$

which is nonzero.

III. QUANTUM BEATS FROM A SINGLE TYPE-II ATOM

Application of NCT to treating an analogous problem with a type-II atom again leads to the prediction of quantum beats. In this case it is the dipoles created between $|\,a\rangle$ and $|\,b\rangle$ and $|\,a\rangle$ and $|\,c\rangle$ that interfere to produce a beat. That is, an atom excited at time t_0 has at time t the state vector

$$|\,\psi(t,t_0)\rangle = A(t)e^{-i\omega_\beta(t-t_0)}|\,a\rangle + B(t)e^{-i(\omega_\beta-\omega_\alpha)(t-t_0)}|\,b\rangle + C(t)|\,c\rangle\,.$$

Thus

$$\langle\,\psi(t,t_0)|\,r|\,\psi(t,t_0)\rangle = A^*(t)B(t)e^{i\omega_\alpha(t-t_0)}\langle a|\,r|\,b\rangle + A^*(t)C(t)e^{i\omega_\beta(t-t_0)}\langle\,a|\,r|\,c\rangle + \text{c.c.}$$

According to NCT, we have the radiated intensity as being proportional to

$$|\,A^*(t)B(t)\langle\,a|\,r|\,b\rangle|^2 + |\,A^*(t)C(t)\langle\,a|\,r|\,c\rangle|^2 + 2\,\mathrm{Re}[|\,A(t)|^2B(t)C^*(t)\langle\,a|\,r|\,b\rangle\langle\,a|\,r|\,a\rangle \times e^{i(\omega_\alpha-\omega_\beta)(t-t_0)}]\,.$$

Since there is a range of excitation times, the beat intensity is actually proportional to

$$2\,\mathrm{Re}\left(|\,A(t)|^2B(t)C^*(t)\langle\,a|\,r|\,b\rangle\langle\,c|\,r|\,a\rangle \times\frac{\sin[(\omega_\alpha-\omega_\beta)\tau/2]}{(\omega_\alpha-\omega_\beta)\tau/2}e^{i(\omega_\alpha-\omega_\beta)t}\right)\,,$$

where again, in foil excitation and in optical excitation,

$$\tau \ll 1/(\omega_\alpha-\omega_\beta)$$

such that

$$\frac{\sin[(\omega_\alpha-\omega_\beta)\tau/2]}{(\omega_\alpha-\omega_\beta)\tau/2}\simeq 1\,.$$

However, the result derived by using QED is that quantum beats are not to be expected. To see this, we start with the initial state

$$|\,\psi(t_0)\rangle = A_0|\,a0\rangle + B_0|\,b0\rangle + C_0|\,c0\rangle\,,$$

which at a later time becomes

$$|\,\psi(t)\rangle = A(t)|\,a0\rangle + B(t)|\,b0\rangle + C(t)|\,c0\rangle + A_1(t)|\,b1_\alpha\rangle + A_2(t)|\,c1_\beta\rangle\,.$$

Consequently, the matrix element

$$\langle\,\psi(t)|\,a_\alpha^\dagger a_\beta|\,\psi(t)\rangle = \langle\,1_\alpha|\,a_\alpha^\dagger a_\beta|\,1_\beta\rangle\langle\,b|\,c\rangle A_1^*(t)A_2(t)$$

is zero since $|\,c\rangle$ is orthogonal to $|\,b\rangle$. This absence of coherent beats was noted by Breit.[15]

The following argument based on the "quantum theory of measurement" provides some physical insight concerning the "missing" beats. A type-I atom when coherently excited will decay via the emission of a photon with frequency ω_α or ω_β. Since both transitions lead to the same atomic state, one cannot determine the emitted photon's frequency. Analogous to the Young's double-slit problem, this uncertainty in the photon's frequency leads to an interference of photons with frequencies ω_α and ω_β, giving rise to quantum beats. A coherently excited type-II atom will also decay via the emission of a photon with frequency ω_α or ω_β. However, after the emission is long past, an observation of the atom would now tell us which decay channel (α or β) was taken. Consequently, we expect no beats in this case.

However, recent experiments involving photon echo showed sinusoidal modulations in the intensity of echos involving atoms with nearly degenerate ground states.[12,13] To see that the results of these experiments do not conflict with QED, we have to look at beats resulting from many atoms.

IV. MANY-ATOM BEATS

If instead of one atom there are two atoms separated by a distance small compared to an optical wavelength, then

$$|\,\psi(0)\rangle = A_0|\,aa0\rangle + B_0|\,ab0\rangle + C_0|\,ac0\rangle + D_0|\,ba0\rangle + E_0|\,bb0\rangle + F_0|\,bc0\rangle + G_0|\,ca0\rangle + H_0|\,cb0\rangle + I_0|\,cc0\rangle\,.$$

The interaction gives rise to states with nonzero photon occupation number, in particular, the states $|\,bc1_\alpha\rangle$ and $|\,bc1_\beta\rangle$, with probability amplitudes $C(t)$ and $D(t)$, respectively. This time the matrix element

$$\langle\,\psi(t)|\,a_\alpha^\dagger a_\beta|\,\psi(t)\rangle = C^*(t)D(t)$$

is nonzero, and QED predicts beats. Thus no contradiction exists between QED and the photon-echo observations.

In line with the physical argument given at the end of Sec. III, we see that if two of these atoms are present, one possible final state has both atoms in different lower levels. For this atomic state it is impossible to determine the photon frequency since one does not know which atom emitted the photon. Hence quantum beats will be present. Extension to the many-atom case leads to a prediction in agreement with the photon-echo observations.

Using QED, we have calculated, in the appendices, the beat intensity from an arbitrary number of type-II atoms. We present the results in the remainder of this section. First, we treat the atoms as being effectively stationary, for example, as in a crystal. Then, we will treat them as being nonstationary, for example, as in an atomic beam.

We begin by looking at two identical atoms, located (e.g., in a crystal) at polar coordinates (r_1, ϕ_1, θ_1) and (r_2, ϕ_2, θ_2). These atoms are excited by a laser pulse that is incident in the y direction. This pulse is polarized in the x direction, and it has a duration that is much less than the beat period. Assuming that all levels are equally populated by the excitation, the radiated intensity from these two atoms at the detector is derived in Appendix A. The result is

$$\begin{aligned}\langle \Psi(t)|\, E(0,t)^2|\,\Psi(t)\rangle = (e^2/4\pi^2\epsilon_0^2 r^2)\tfrac{1}{9}\{&2e^{-4\gamma_{ab}t}(X_{ab}^2k_\alpha^4 + X_{ac}^2k_\beta^4) + e^{-2\gamma_{ab}t}[4(X_{ab}^2k_\alpha^4 + X_{ac}^2k_\beta^4) + 2X_{ab}^2k_\alpha^4\cos k_\alpha(r_1 - r_2 + y_1 - y_2)\\ &+ 2X_{ac}^2k_\beta^4\cos k_\beta(r_1 - r_2 + y_1 - y_2)]\\ &+ 2e^{-2\gamma_{ab}t}X_{ab}X_{ac}k_\alpha^2k_\beta^2[\cos(\omega_\beta(t - r_2/c) - \omega_\alpha(t - r_1/c) + k_\alpha(y_1 - y_2))\\ &+ \cos(\omega_\beta(t - r_1/c) - \omega_\alpha(t - r_2/c) + k_\alpha(y_2 - y_1))]\}\ ,\end{aligned}$$

which describes an exponential curve modulated by a sinusoidal function with periodicity $2\pi/(\omega_\beta - \omega_\alpha)$. Here,

$$\gamma_{ab} = (e^2/6\pi\epsilon_0\hbar)(X_{ab}^2k_\alpha^2 + X_{ac}^2k_\beta^2)\ ,$$

r is the average distance of atoms from detector;

$$y_i = r_i\sin\phi_i\sin\theta_i\ ;\quad X_{ab} = \langle a|\,X|\,b\rangle\ ,\quad X_{ac} = \langle a|\,X|\,c\rangle\ ;\quad k_\alpha = \omega_\alpha/c,\quad k_\beta = \omega_\beta/c\ .$$

The beat signal for n atoms is (from Appendix B)

$$\frac{e^2X_{ab}X_{ac}k_\alpha^2k_\beta^2}{2\pi^2\epsilon_0^2r^2}\frac{1}{3^n}\left(\sum_{i=0}^{n-2} e^{-2\gamma_{ab}t(1+i)}\frac{1}{i!}\frac{(n-2)!}{(n-2-i)!}2^{(n-2-i)}\right)\sum_{\substack{l=1\\ l\neq j}}^{n}\sum_{j=1}^{n}\cos[\omega_\beta(t - r_l/c) - \omega_\alpha(t - r_j/c) + \omega_\alpha(y_j - y_l)]\ .$$

If the positions of the atoms are not fixed (e.g., in an atomic beam), then it is necessary to average the intensity over all positions occupied by each atom before it decays. If each atom is confined in a cube of volume δ^3, then

$$\begin{aligned}\langle \psi(t)|\,E(0,t)^2|\,\psi(t)\rangle_{\text{beats}} = &\frac{e^2X_{ab}X_{ac}k_\alpha^2k_\beta^2}{2\pi^2\epsilon_0^2r^2}\frac{1}{3^n}\left(\sum_{i=0}^{n-2} e^{-2\gamma_{ab}t(1+i)}\frac{1}{i!}\frac{(n-2)!}{(n-2-i)!}2^{n-2-i}\right)\\ &\times\frac{\sin^4(k_\alpha\delta/2)}{(k_\alpha\delta/2)^4}\sum_{\substack{l=1\\ l\neq j}}^{n}\sum_{j=1}^{n}\cos[\omega_\beta(t - R_l/c) - \omega_\alpha(t - R_j/c)]\ ,\end{aligned}$$

where R_i is the distance between the detector and the center of the cube for the ith atom. This means that in order to see an appreciable beat signal at each wavelength λ each atom must be localized in a volume of λ^3.

If the n atoms are in a flask, then δ^3, the volume of the flask, must be less than λ^3 for beats to be seen. It would be difficult to see quantum beats with such an experimental setup. However, with an atomic beam, it is possible to select a velocity distribution such that each atom maintains its position relative to its neighbors within a distance λ during the time it is in an excited state. With such a velocity distribution QED predicts quantum beats for a density of at least 1 atom/λ^3. NCT predicts beats for any density greater than 1 atom/$(k_\beta - k_\alpha)^{-3}$. This is because, according to NCT, the electric field from n atoms is

$$\sum_{l=1}^{n}\left(k_\alpha^2p_\alpha\frac{e^{ik_\alpha r_l}}{r_l} + k_\beta^2p_\beta\frac{e^{ik_\beta r_l}}{r_l}\right)\ ,$$

where p_α and p_β are the dipole moments. Thus the beat signal is

$$2k_\alpha^2k_\beta^2p_\alpha p_\beta^*\left(\sum_{l\neq j}\frac{e^{ik_\alpha r_l - ik_\beta r_j}}{r_lr_j} + \sum_l\frac{e^{i(k_\alpha - k_\beta)r_l}}{r_l^2}\right)\ .$$

If the positions of the atoms are not fixed so that each atom can be anywhere inside a volume δ^3, the beat signal becomes

$$2k_\alpha^2k_\beta^2\frac{p_\alpha p_\beta^*}{R^2}\left(\sum_{l\neq j}e^{ik_\alpha R_l - ik_\beta R_j}\frac{\sin^2(k_\alpha\delta/2)}{(k_\alpha\delta/2)^2} + \sum_l e^{i(k_\alpha - k_\beta)R_l}\frac{\sin[(k_\alpha - k_\beta)\delta/2]}{(k_\alpha - k_\beta)\delta/2}\right)\ ,$$

which is small only when $1/(k_\beta - k_\alpha) < \delta$. [$R_i$ is the average distance between the detector and the ith atom and $R = (1/n)\sum_i R_i$.] Hence, in nonlocalized atoms, we have another difference between the predictions of QED and NCT. We note that this difference arises because in QED quantum beats from type-II atoms occur only as a cooperative effect, while in NCT they occur also as a single-atom phenomena. Next we propose an experiment that may serve as a test for NCT and QED.

V. PROPOSED EXPERIMENT

We propose this illustrative experiment in order to emphasize the features that must be present in an experiment using quantum beats to test QED and NCT. In Fig. 3 we sketch a possible experimental setup wherein laser radiation is used to put atoms in a coherent superposition of states.[16] The atoms will interact with the laser field $E(t)$ and acquire off-diagonal contributions to the density matrix $\rho_{ac}(t)$ and $\rho_{ab}(t)$. If we have a thermal distribution initially, then these two off-diagonal density-matrix elements arise from the first-order process involving the interaction of the atom with the two applied fields. That is,

$$\rho_{ac}(t) = \frac{p_\beta}{\hbar}\int_{t_0}^{t} dt_1 E(t_1) e^{i\omega_\beta t_1}[\rho_{aa}(t_0) - \rho_{cc}(t_0)]$$

and

$$\rho_{ab}(t) = \frac{p_\alpha}{\hbar}\int_{t_0}^{t} dt_1 E(t_1) e^{i\omega_\alpha t_1}[\rho_{aa}(t_0) - \rho_{bb}(t_0)] ,$$

where p_α and p_β are the dipole matrix elements, and $E(t)$ is the applied electric field. It is to be noted that atoms excited at different times would see slightly different electromagnetic fields due to the finite laser linewidth. Consequently, one has to limit each measurement to times less than the coherence time of the light, which may be of order of 10^{-4} sec. However, with an atomic lifetime of 10^{-8} sec we may repeat the (single-atom) experiment many times during the coherence time. An experiment of this type should provide a rather direct test of NCT vs QED. Furthermore, one can extend this experiment to look for beats at higher atomic densities. The presence of beats at these higher densities will confirm that the excitation mechanism coherently excites the atoms.

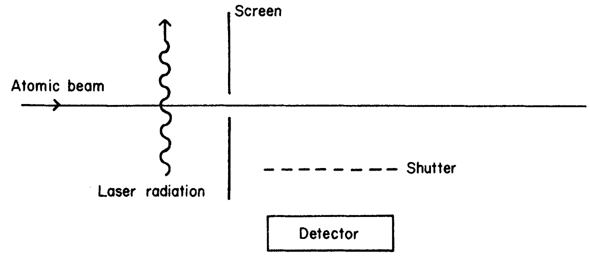

FIG. 3. Experimental geometry and apparatus for proposed experiment. A screen is used to shield the detector from scattered laser radiation. The shutter limits the measurement to less than the laser coherence time.

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APPENDIX A: CALCULATION FOR BEAT INTENSITY FROM TWO TYPE-II ATOMS

The atoms are located by the spherical coordinates (r_1, ϕ_1, θ_1) and (r_2, ϕ_2, θ_2). Each atom has the energy levels shown in Fig. 1(b), and the upper state can decay to either lower states via electric dipole transition. In the interaction picture the Hamiltonian is

$$V(t) = \sum_{\vec{k}\lambda}\sum_{j=1}^{2}\frac{\mathcal{E}_k}{\sqrt{2}}\Big(e\vec{X}_{ab}\cdot\hat{\epsilon}_{\vec{k}\lambda}|a\rangle_j\langle b|\,a_{\vec{k}\lambda}e^{i(\omega_\alpha-\omega_k)t+i\vec{k}\cdot\vec{r}_j}$$
$$+ e\vec{X}_{ac}\cdot\hat{\epsilon}_{\vec{k}\lambda}|a\rangle_j\langle c|\,a_{\vec{k}\lambda}e^{i(\omega_\beta-\omega_k)t+i\vec{k}\cdot\vec{r}_j}\Big) + \text{H.c.},$$

where $\mathcal{E}_k = \hbar\omega_k/L^3\epsilon_0$.

The atoms are excited by a laser pulse with its electric field polarized in the x direction and a pulse duration that is much less than $1/(\omega_\beta - \omega_\alpha)$. With the pulse incident in the y-direction, the state of the atom-field system after the pulse has reached both atoms is

$$|\psi(0)\rangle \simeq \big(A_0 e^{-ik_\beta(2y_0-y_1-y_2)}|aa\rangle + B_0 e^{-ik_\beta(y_0-y_1)-i(k_\beta-k_\alpha)(y_0-y_2)}|ab\rangle + C_0 e^{-ik_\beta(y_0-y_1)}|ac\rangle$$
$$+ D_0 e^{-(k_\beta-k_\alpha)(y_0-y_1)}e^{-ik_\beta(y_0-y_2)}|ba\rangle + F_0 e^{-ik_\beta(y_0-y_2)}|ca\rangle + G_0 e^{-i(k_\beta-k_\alpha)(2y_0-y_1-y_2)}|bb\rangle + H_0|cc\rangle$$
$$+ I_0 e^{-i(k_\beta-k_\alpha)(y_0-y_1)}|bc\rangle + J_0 e^{-i(k_\beta-k_\alpha)(y_0-y_2)}|cb\rangle\big)|\{0_{k\lambda}\}\rangle ,$$

where y_0 is the position of the wave front of the pulse along the y axis at time $t=0$. Since the volume looked at by the detector is small, the probability that the first atom that is excited has decayed before the second atom is excited is negligible.

To first order the state vector at a later time is

$$|\psi(t)\rangle = (A|aa\rangle + B|ab\rangle + C|ac\rangle + D|ba\rangle + F|ca\rangle)|\{0_{k\lambda}\}\rangle + \sum_{\vec{k}\lambda}(G_{\vec{k}\lambda}|ab\rangle + H_{\vec{k}\lambda}|ba\rangle + I_{\vec{k}\lambda}|bb\rangle + J_{\vec{k}\lambda}|cb\rangle$$
$$+ K_{\vec{k}\lambda}|cc\rangle + L_{\vec{k}\lambda}|bc\rangle + M_{\vec{k}\lambda}|ac\rangle + N_{\vec{k}\lambda}|ca\rangle)|1_{k\lambda}\{0_{k\lambda}\}\rangle\ ,$$

where the coefficients on the right-hand side satisfy differential equations obtained from

$$i\hbar\frac{\partial|\psi(t)\rangle}{\partial t} = V(t)|\psi(t)\rangle\ .$$

The equations for A to F can be solved by the method used in Weisskopf-Wigner decay:

$$A(t) = A_0 e^{-2\gamma_{ab}t + ik_\beta(y_1 - y_2)}\ ,$$

$$[B(t)/B_0]e^{ik_\beta(y_0-y_1)+i(k_\beta-k_\alpha)(y_0-y_2)}$$
$$= [C(t)/C_0]e^{ik_\beta(y_0-y_1)}$$
$$= [D(t)/D_0]e^{i(k_\beta-k_\alpha)(y_0-y_1)+ik_\beta(y_0-y_2)}$$
$$= [F(t)/F_0]e^{ik_\beta(y_0-y_2)} = e^{-\gamma_{ab}t}\ ,$$

where

$$\gamma_{ab} = (e^2/6\pi\epsilon_0\hbar)(X_{ab}^2 k_\alpha^2 + X_{ac}^2 k_\beta^2)\ .$$

The signal at the detector is proportional to

$$\langle\psi(t)|\vec{E}^2(0t)|\psi(t)\rangle = 2\left|\sum_{\vec{k}\lambda}\mathcal{E}_k\hat{\epsilon}_{\vec{k}\lambda}e^{-i\omega_k t}J_{\vec{k}\lambda}\right|^2 \quad (1)$$

plus similar terms with $H_{\vec{k}\lambda}$, $I_{\vec{k}\lambda}$, $G_{\vec{k}\lambda}$, $L_{\vec{k}\lambda}$, $M_{\vec{k}\lambda}$, and $N_{\vec{k}\lambda}$ in place of $J_{\vec{k}\lambda}$.

Looking at the first term on the right-hand side, we have, from the Schrödinger equation,

$$\sum_{\vec{k}\lambda}\mathcal{E}_k\hat{\epsilon}_{\vec{k}\lambda}e^{-i\omega_k t}J_{\vec{k}\lambda} = \sum_{\vec{k}\lambda}\mathcal{E}_k^2\hat{\epsilon}_{\vec{k}\lambda}e^{-i\omega_k t}\frac{-i}{\hbar}\int_0^t dt_1 e^{-\gamma_{ab}t_1}\Big\{F_0 e\vec{X}_{ab}\cdot\hat{\epsilon}_{\vec{k}\lambda}\frac{1}{\sqrt{2}}\exp[-ik_\beta(y_0-y_2) - i\vec{k}\cdot\vec{r}_2 - i(\omega_\alpha-\omega_k)t_1]$$
$$+ B_0 e\vec{X}_{ac}\cdot\hat{\epsilon}_{\vec{k}\lambda}\frac{1}{\sqrt{2}}\exp[-ik_\beta(y_0-y_1) - i(k_\beta-k_\alpha)(y_0-y_2)$$
$$- i\vec{k}\cdot\vec{r}_1 - i(\omega_\beta-\omega_k)t_1]\Big\}\ . \quad (2)$$

Integrations involving the first term of Eq. (2) can easily be done in a frame where $\vec{r}_2$ is along the z axis. In this frame the atomic polarization vector is no longer in the x direction, but is along

$$R\begin{pmatrix}1\\0\\0\end{pmatrix} \equiv \begin{pmatrix}P_x\\P_y\\P_z\end{pmatrix} = \begin{pmatrix}\cos\theta_2\cos\phi_2\\ \sin\phi_2\\ \sin\theta_2\cos\phi_2\end{pmatrix}.$$

Furthermore, the summation on $\vec{k}$ is replaced by an integral,

$$\sum_{\vec{k}} \to \left(\frac{L}{2\pi c}\right)^3\int_0^\infty d\omega\,\omega^2\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\phi\ .$$

Choosing the field polarization vectors to be

$$\hat{\epsilon}_{\vec{k}1} = (\sin\phi,\ -\cos\phi,\ 0)$$
$$\hat{\epsilon}_{\vec{k}2} = (\cos\theta\cos\phi,\ \cos\theta\sin\phi,\ -\sin\theta)\ ,$$

where θ and ϕ define the direction of the wave vector $\vec{k}$, the polarization sum and the angular integrations are straightforward. Dropping terms which go as $1/(kr_2)^2$, we are left with the following t_1 and ω integrals:

$$-\frac{F_0 eX_{ab}}{(2\pi c)^3\epsilon_0\sqrt{2}}2\pi\int_0^t dt_1 e^{-\gamma_{ab}t_1 - ik_\beta(y_0-y_2)}\int_0^\infty d\omega\,\omega^3 e^{-i\omega t - i(\omega_\alpha-\omega)t_1}\frac{1}{kr_2}(e^{ikr_2} - e^{-ikr_2})\begin{pmatrix}P_x\\P_y\\0\end{pmatrix}.$$

Replacing the slowly varying quantities in ω by their average, we have

$$-\frac{F_0 eX_{ab}}{(2\pi)^2c^3\epsilon_0\sqrt{2}}\int_0^t dt_1 e^{-\gamma_{ab}t_1 - ik_\beta(y_0-y_2)}k_\alpha^2 e^{-i\omega_\alpha t}\int_{-\omega_\alpha}^\infty d\Omega\, e^{i\Omega(t_1-t)}\sin\frac{(\Omega+\omega_\alpha)r_2}{c}\begin{pmatrix}P_x\\P_y\\0\end{pmatrix}.$$

Since $\omega_\alpha \gg 1$, the Ω integral may be approximated by two Dirac δ functions with arguments $(t_1 - t + r_2/c)$ and $(t_1 - t - r_2/c)$. Integrating over t_1, one gets

$$-\frac{F_0 e X_{ab}}{2\pi\epsilon_0\sqrt{2}}\frac{k_\alpha^2}{r_2}\exp[-\gamma_{ab}t - i\omega_\alpha(t-r_2/c) - ik_\beta(y_0-y_2)]\begin{pmatrix}P_x\\P_y\\0\end{pmatrix}.$$

Finally, we have to transform back to the original frame, thus obtaining the expression

$$-F_0\exp[-ik_\beta(y_0-y_2)-\gamma_{ab}t - i\omega_\alpha(t-r_2/c)]\frac{eX_{ab}}{2\sqrt{2}\pi\epsilon_0}\frac{k_\alpha^2}{r_2}\begin{pmatrix}\cos^2\theta_2\cos^2\phi_2+\sin^2\phi_2\\-\frac{1}{2}\sin^2\theta_2\sin 2\phi_2\\-\frac{1}{2}\sin 2\theta_2\cos\phi_2\end{pmatrix}.$$

Application of the same procedure in the evaluation of the second term in Eq. (2) gives

$$-B_0\exp[-ik_\beta(y_0-y_1)-i(k_\beta-k_\alpha)(y_0-y_1)]\frac{eX_{ac}}{2\sqrt{2}\pi\epsilon_0}\frac{k_\beta^2}{r_1}\exp[-\gamma_{ab}t - i\omega_\beta(t-r_1/c)]\begin{pmatrix}\cos^2\theta_1\cos^2\phi_1+\sin^2\phi_1\\-\frac{1}{2}\sin^2\theta_1\sin 2\phi_1\\-\frac{1}{2}\sin 2\theta_1\cos\phi_1\end{pmatrix}.$$

If the detector is placed far from the two atoms compared to the separation between the atoms, then the above terms are essentially in the x direction. Hence

$$2\left|\sum_{\vec{k}\lambda}\mathcal{E}_k\hat{\epsilon}_{\vec{k}\lambda}J_{\vec{k}\lambda}\right|^2=\frac{e^2}{4\pi^2\epsilon_0^2 r^2}e^{-2\gamma_{ab}t}\{X_{ab}^2k_\alpha^4F_0^2+X_{ac}^2k_\beta^4B_0^2$$
$$+2X_{ab}X_{ac}k_\alpha^2k_\beta^2F_0B_0\cos[\omega_\beta(t-r_1/c)-\omega_\alpha(t-r_2/c)+k_\alpha(y_2-y_1)]\}.$$

The remaining terms of Eq. (1) may be evaluated similarly. Upon combining these terms, we have, for the radiated intensity,

$$\langle\psi(t)|\,E(0t)^2|\,\psi(t)\rangle=\frac{e^2}{4\pi^2\epsilon_0^2r^2}\{2e^{-4\gamma_{ab}t}A_0^2(X_{ab}^2k_\alpha^4+X_{ac}^2k_\beta^4)+e^{-2\gamma_{ab}t}[(B_0^2+D_0^2)(X_{ab}^2k_\alpha^4+X_{ac}^2k_\beta^4)+(C_0^2+F_0^2)(X_{ac}^2k_\beta^4+X_{ab}^2k_\alpha^4)$$
$$+2X_{ab}^2k_\alpha^4B_0D_0\cos(k_\alpha(r_1-r_2)+k_\alpha(y_1-y_2))$$
$$+2X_{ac}^2k_\beta^4C_0F_0\cos(k_\beta(r_1-r_2)+k_\beta(y_1-y_2))]$$
$$+2e^{-2\gamma_{ab}t}X_{ab}X_{ac}k_\alpha^2k_\beta^2[C_0D_0\cos(\omega_\beta(t-r_2/c)-\omega_\alpha(t-r_1/c)+k_\alpha(y_1-y_2))$$
$$+F_0B_0\cos(\omega_\beta(t-r_1/c)-\omega_\alpha(t-r_2/c)+k_\alpha(y_2-y_1))]\}.$$

APPENDIX B: CALCULATION OF BEAT INTENSITY FROM n ATOMS

We will obtain the n-atom beat intensity by deriving the three-atom result and then generalizing it to the n-atom case. The three-atom problem has the initial state

$$|\,\psi(0)\rangle=(A_0\exp[-ik_\beta(3y_0-y_1-y_2-y_3)]\,|\,aaa\rangle+B_0\exp[-ik_\beta(2y_0-y_1-y_3)-i(k_\beta-k_\alpha)(y_0-y_2)]\,|\,aba\rangle$$
$$+C_0\exp[-ik_\beta(2y_0-y_1-y_2)]\,|\,aac\rangle+D_0\exp[-i(k_\beta-k_\alpha)(y_0-y_1)-ik_\beta(2y_0-y_2-y_3)]\,|\,baa\rangle$$
$$+E_0\exp[-ik_\beta(2y_0-y_1-y_3)]\,|\,aca\rangle+\cdots)|\{0_{k\lambda}\}\rangle.$$

At a later time t, the state vector contains eigenstates with one photon present. Some of these states contribute to the beat signal. One such state is $|\,abc1_{\vec{k}\lambda}\{0_{k\lambda}\}\rangle$, and if it has a probability amplitude $F_{\vec{k}\lambda}$, then its contribution to the beat signal, $\langle\psi(t)|\,E^2(0t)|\,\psi(t)\rangle$, is $2|\sum_{\vec{k}\lambda}F_{\vec{k}\lambda}e^{-\omega_k t}\mathcal{E}_k\hat{\epsilon}_{\vec{k}\lambda}|^2$. From the Schrödinger equation,

$$\sum_{\vec{k}\lambda}\mathcal{E}_k\hat{\epsilon}_{\vec{k}\lambda}F_{\vec{k}\lambda}e^{-i\omega_k t}=\sum_{\vec{k}\lambda}\hat{\epsilon}_{\vec{k}\lambda}\mathcal{E}_k^2e^{-i\omega_k t}\frac{-i}{\hbar}\int_0^t dt_1\{eX_{ab}(1/\sqrt{2})(\hat{P}_2\cdot\hat{\epsilon}_{\vec{k}\lambda})e^{-i\vec{k}_1\cdot\vec{r}_2-i(\omega_\alpha-\omega_k)t_1}C_0e^{-(\gamma_a+\gamma_b)t_1-ik_\beta(2y_0-y_1-y_2)}$$
$$+eX_{ac}(1/\sqrt{2})(\hat{P}_1\cdot\hat{\epsilon}_{\vec{k}\lambda})e^{-i\vec{k}_1\cdot\vec{r}_3-i(\omega_\beta-\omega_k)t_1}$$
$$\times B_0e^{-(\gamma_a+\gamma_b)t_1-ik_\beta(2y_0-y_1-y_3)}e^{-i(k_\beta-k_\alpha)(y_0-y_2)}\}.$$

The integrals, when evaluated in a manner similar to that used in Appendix A, result in

$$-\hat{i}\frac{e}{2\sqrt{2}\pi\epsilon_0 r}e^{-2\gamma_{ab}t}\left(C_0 e^{-ik_\beta(2y_0-y_1-y_2)}X_{ab}k_\alpha^2 e^{-i\omega_\alpha(t-r_2/c)}+B_0 e^{-ik_\beta(2y_0-y_1-y_3)-i(k_\beta-k_\alpha)(y_0-y_2)}X_{ac}k_\beta^2 e^{-i\omega_\beta(t-r_3/c)}\right).$$

Thus the contribution to the beat signal is

$$\left\{\frac{e^2X_{ab}X_{ac}}{2\pi^2\epsilon_0^2 r^2}k_\alpha^2k_\beta^2\right\}e^{-4\gamma_{ab}t}C_0B_0\cos[\omega_\beta(t-r_3/c)-\omega_\alpha(t-r_2/c)+k_\alpha(y_2-y_3)].$$

All remaining terms may be evaluated similarly, but there is a less tedious method. Note that the factors inside { } will be the same for all terms. What remains is to find the states that will give a nonzero matrix element for the operator $a_\alpha^\dagger a_\beta$. For example, a nonzero matrix element is

$$\langle bca1_\alpha|\,a_\alpha^\dagger a_\beta|\,bca1_\beta\rangle\,.$$

The bra comes from the initial states $|\,aca\{0_{k\lambda}\}\rangle$, while the ket comes from $|\,baa\{0_{k\lambda}\}\rangle$. So the exponential factors associated with this matrix element should be $\exp[-i\vec{k}_\alpha\cdot\vec{r}_1+i\omega_\alpha t+i\vec{k}_\beta\cdot\vec{r}_2-i\omega_\beta t]$. Also, the excitation process imparts the phase factors $\exp[ik_\beta(y_0-y_1)-i(k_\beta-k_\alpha)(y_0-y_1)-ik_\beta(y_0-y_2)]$, and the states with 2 particles in the excited states decay at the rate $2\gamma_{ab}$. Therefore another term in the beat signal is

$$\frac{e^2X_{ab}X_{ac}k_\alpha^2k_\beta^2}{2\pi^2\epsilon_0^2 r^2}e^{-4\gamma_{ab}t}E_0D_0\cos[\omega_\beta(t-r_2/c)-\omega_\alpha(t-r_1/c)+k_\alpha(y_1-y_2)].$$

The final result is obvious. However, we will not need the general expression for the beat intensity, but rather the expression where all states of the three atoms are equally populated by the excitation. For this case the beat intensity reduces to

$$\langle E^2\rangle_{\text{beat}}=\frac{e^2X_{ab}X_{ac}}{2\pi^2\epsilon_0^2 r^2}k_\alpha^2k_\beta^2\,\tfrac{1}{27}|(e^{-4\gamma_{ab}t}+2e^{-2\gamma_{ab}t})\sum_{\substack{i=1\\ i\neq j}}^{3}\sum_{j=1}^{3}\cos[\omega_\beta(t-r_i/c)-\omega_\alpha(t-r_j/c)+k_\alpha(y_j-y_i)].$$

Generalizing the above derivation, we see that the n-atom beat signal is

$$\langle E^2_{\text{beats}}\rangle_n=\frac{e^2X_{ab}X_{ac}k_\alpha^2k_\beta^2}{2\pi^2\epsilon_0^2 r^2}\frac{1}{3^n}\left[\sum_{i=0}^{n-2}e^{-2\gamma_{ab}t(1+i)}\frac{1}{i\,!}\frac{(n-2)!}{(n-2-i)!}2^{(n-2-i)}\right]$$
$$\times\sum_{\substack{l=1\\ l\neq j}}^{n}\sum_{j=1}^{n}\cos[\omega_\beta(t-r_l/c)-\omega_\alpha(t-r_j/c)+k_\alpha(y_j-y_l)],$$

where the 3^{-n} comes from the normalization of the initial state, and the quantity in square brackets containing the decay rates comes from the following argument. Initial states that contribute to the beat are of the form

$$\left|\begin{pmatrix}a\\b\\c\end{pmatrix}\begin{pmatrix}\\ \\ \end{pmatrix}\cdots\begin{pmatrix}a\\b\\c\end{pmatrix}ac\right\rangle.$$

Let $i+1$ be the number of excited particles. The state with $i+1$ particles in $|\,a\rangle$ will decay with the rate $\gamma_{ab}(1+i)$. The number of these states is

$$\frac{(n-2)(n-1)\cdots[n-2-(i-1)]*2^{n-2-i}}{i\,!},$$

since two of the particles are fixed, and we are only allowed to distribute the remaining $i|\,a\rangle$ states among $n-2$ particles. Also, there are $n-2-i$ particles that can be in $|\,b\rangle$ or $|\,c\rangle$.

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