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Cooperative atomic effects in two-photon spontaneous emission and resonance fluorescence

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In this paper, we study cooperative two-photon transitions in a system of two (nonoverlapping) atoms for two processes: two-photon spontaneous emission and two-photon resonance fluorescence. Expressions are obtained for the energy shifts and decay rates due to the interaction of the atoms with and via the field. The altered decay rates signal the existence of superradiance and subradiance in the two-atom system. The energy shifts of the single-excitation states are found to be proportional to R^{-6} for small atomic separations R, and to R^{-2} for large separations.

I. INTRODUCTION

The subject of cooperative radiative transitions by a system of two or more identical atoms (or molecules) has received considerable attention over the years, beginning with an initial study by Dicke [1]. Many authors since have dealt with coherent single-quantum electric dipole excitations of systems of two [2,3] or more [3,4] atoms. A few exact analytical studies exist of the collective spontaneous emission by fully inverted systems [5], and a very large literature exists involving approximate (and usually numerical) treatments of "Dicke superradiance" [6]. The effects of the atomic interaction include in general the splitting of each atomic energy level into a number of sublevels, the shift of each sublevel from the single-atom energy, and a change in the lifetime of each sublevel from the single-atom radiative lifetime.

With the exception of a single paper dealing with cooperative electric quadrupole transitions in atoms [7], all the work performed to date has dealt with single-photon, electric dipole transitions. It is our purpose in this paper to address the question of cooperative two-photon (2-γ) spontaneous emission and resonance fluorescence in multiatom systems. We study the simplest example of such a system: two identical atoms, separated by a distance R (large compared to the atomic diameter so that overlap can be ignored), interacting with a common radiation field, each atom capable of making a 2-γ transition between two of its levels (of the same parity) via nonresonant intermediate levels (of the opposite parity). Two cases are studied: (i) the atomic system initially in a single-excitation level, interacting with the vacuum field (2-γ spontaneous emission); and (ii) the atomic system initially in its ground level, interacting with the vacuum field and with a weak probe field (2-γ resonance fluorescence).

In studying the evolution in time of the atoms-plus-field system, we do not make the rotating-wave approximation [8], and we include two types of "essential states": states that are resonant in energy with the initial state, which both alter the decay rates and shift the energies of the atomic single-excitation levels; and nonresonant essential states, which contribute only to the energy shifts through the exchange of virtual photons. We find that the effects of the atomic interaction are analogous to its effects for (two-atom) single-photon transitions [2,3].

(i) The energy level corresponding to a single atomic excitation is split into two sublevels, corresponding to the symmetric ($|+\rangle$) and antisymmetric ($|-\rangle$) states of the two-atom system.

(ii) The decay rates of the sublevels are functions of k_0R, where $\hbar k_0 c$ is the energy difference between the single-atom levels $|b\rangle$ and $|a\rangle$ (see Fig. 1). These decay rates first appear as the convolution of two functions, representing the retarded dipole-dipole interaction between the two pairs of oscillating dipoles that characterize each atomic (2-γ) transition. Integration yields a general expression which consists of oscillating functions of k_0R, multiplied by inverse powers of k_0R ranging from $(k_0R)^{-7}$ to $(k_0R)^{-2}$. For small k_0R, the $|+\rangle$ state is superradiant, with a decay rate double that of the single atom γ_0; the $|-\rangle$ state is subradiant, with a decay rate of order $(k_0R)^2\gamma_0$ [9]. For large k_0R, the effects of the atomic interaction become negligible, and the sublevels merge into a single level with frequency and decay rate identical to those of a single atom.

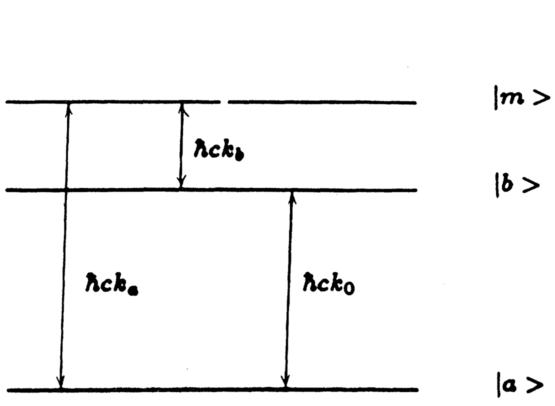

FIG. 1. Three-level atom, with a 2-γ transition between levels $|a\rangle$ and $|b\rangle$ via the intermediate state $|m\rangle$.

(iii) The energies of the sublevels are shifted from the single-atom energy. The shifts are also oscillating functions of k_0R, with leading terms proportional to $(k_0R)^{-6}$ for small k_0R and to $(k_0R)^{-2}$ for large k_0R.

(iv) In 2-γ resonance fluorescence, the total probability of absorption from the ground state is the sum of the probabilities of absorption to the levels $|+\rangle$ ($|-\rangle$), which are shifted in energy and broadened (narrowed) by the vacuum field as they are in spontaneous emission. The energy of the ground state is shifted by the laser-induced shift as well as by the second-order Casimir-London dispersion energy [10].

In Sec. II, we present the theory of two-atom, 2-γ spontaneous emission. In Secs. III–V we evaluate the decay rates and shifts of the two-atom energy levels, and the distribution of energy in the system as a function of time. In Sec. VI, we present the theory of resonance fluorescence by the two-atom system irradiated by a weak probe beam. In Sec. VII, we discuss the results of this paper. Finally, in the Appendix, we give details regarding the evaluation of some of the integrals in the paper.

II. SPONTANEOUS EMISSION

The calculations in this paper are based on the Heitler-Ma (HM) treatment of 2-γ transitions [11], developed as part of a program to extend radiation theory to 2-γ processes.

We consider two identical atoms, centered at $\mathbf{R}_s$ $(s=1,2)$ with $|\mathbf{R}_1-\mathbf{R}_2|=R$. Atom s can make a 2-γ transition between its levels $|b_s\rangle$ and $|a_s\rangle$ via a nonresonant intermediate level $|m_s\rangle$ (see Fig. 1) [12]. We denote by $|C\rangle$ (with energy E_C) states of the two-atom system,

$$|C\rangle=\begin{cases}|B\rangle=|b_1\rangle|b_2\rangle,\quad E_B=2E_b\\ |M\rangle=|m_1\rangle|m_2\rangle,\quad E_M=2E_m\\ |A\rangle=|a_1\rangle|a_2\rangle,\quad E_A=2E_a\\ |\pm\rangle=\dfrac{1}{\sqrt{2}}(|a_1\rangle|b_2\rangle\pm|b_1\rangle|a_2\rangle),\quad E_\pm=E_a+E_b,\end{cases}\tag{1}$$

and we include as well the nonresonant state $|M\rangle$ (with $E_m>E_b>E_a$), which contributes to the energy shifts. The states $|C\rangle$ satisfy the eigenvalue equation

$$H_A|C\rangle=(H_{A_1}+H_{A_2})|C\rangle=E_C|C\rangle\ ,\tag{2}$$

where H_A is the atomic Hamiltonian, which is the sum of those of the two atoms. The Hamiltonian of the field is denoted by H_F, with the eigenvalue equation

$$H_F|\cdots n_k\cdots\rangle=\left[\sum_k \hbar kc(n_k+\tfrac{1}{2})\right]|\cdots n_k\cdots\rangle\ .\tag{3}$$

Here $|\cdots n_k\cdots\rangle$ is the state in which n_k photons are present in field mode k, and so on. The Hamiltonian of the combined atom-field system is

$$H=H_0+V=H_A+H_F+V^1+V^2\ ,\tag{4}$$

where V^s is the interaction of atom s with the field (in the electric dipole approximation),

$$V^s=-\boldsymbol{\mu}(s)\cdot\mathbf{E}(\mathbf{R}_s)\ ,\tag{5}$$

$$\mathbf{E}(\mathbf{R})=i\sum_{k,\varepsilon}\left[\frac{2\pi\hbar ck}{L^3}\right]^{1/2}[\boldsymbol{\varepsilon}(\mathbf{k})a_k e^{i\mathbf{k}\cdot\mathbf{R}}-\boldsymbol{\varepsilon}^*(\mathbf{k})a_k^\dagger e^{-i\mathbf{k}\cdot\mathbf{R}}]\ .\tag{6}$$

The eigenstates of H_0 are the product states $|C\rangle|\cdots n_k\cdots\rangle$. In particular, we will be involved with the states $|C0\rangle\equiv|C\rangle|\cdots 0_k\cdots\rangle$, where $|\cdots 0_k\cdots\rangle$ is the vacuum state of the field, and the states $|Ckl\rangle$ [with energy $E_{Ckl}=E_C+\hbar c(k+l)$], in which the atoms are in the state $|C\rangle$ and one photon is present in each of modes k and l. We shall omit $|0\rangle$ from $|C0\rangle$ when no confusion can result.

We take for the initial state $|I\rangle$ of our system the single-excitation state

$$|I\rangle=c_+|+0\rangle+c_-|-0\rangle,\quad |c_+|^2+|c_-|^2=1\ .\tag{7}$$

(For example, if one atom is initially in state $|b\rangle$ and the other in state $|a\rangle$, $c_\pm=1/\sqrt{2}$.) In HM, we study $G(E)|I\rangle$, the Fourier transform of the wave function $|\psi(t)\rangle$ of the atom-field system,

$$|\psi(t)\rangle=-(2\pi i)^{-1}\int_{-\infty}^{\infty}dE\ G(E)|I\rangle\exp(-iEt/\hbar)\ .\tag{8}$$

The basic (exact) equation of the HM method for 2-γ processes is [11]

$$[E-H_0-K(E)]G(E)|I\rangle=[1+V(E-H_0)^{-1}]|I\rangle\ ,\tag{9}$$

where the effective operator K is given by

$$K(E)\equiv\sum_{s,s'=1}^{2}V^s(E-H_A-H_F)^{-1}V^{s'}\ .\tag{10}$$

Our objective is to find the dependence on the interatomic separation R of the decay rates and energy shifts of the system, to the lowest nonvanishing order in V. The diagonal elements of $K(E)$ (e.g., $K_{\pm,\pm}$) are found to give second-order R-independent (single-atom) shifts and are absorbed into the corresponding energies $(E_\pm)$. Multiplying Eq. (9) by $\langle C|$ and by $\langle Ckl|$ in turn, and denoting $\langle C|G|I\rangle$ by G_C and so on, we obtain the equations

$$(E-E_C)G_C-\sum_{D\neq C}K_{C,D}G_D-\sum_{D,k,l}K_{C,Dkl}G_{Dkl}=\langle C|I\rangle\ ,\tag{11}$$

$$(E-E_{Ckl})G_{Ckl}-\sum_{D\neq C}K_{Ckl,Dkl}G_{Dkl}-\sum_{D}K_{Ckl,D}G_D=0\ .\tag{12}$$

In Eq. (12) we have neglected the coupling between G_{Ckl} and $G_{Dklk'l'}$, which gives rise to higher-order corrections.

If the initial state $|I\rangle$ is $|\pm 0\rangle(c_\pm=1,c_\mp=0)$, we can proceed exactly as in HM [11]. For the more general state $|I\rangle$ of Eq. (7), however, a modified approach is required. We make the substitution

$$G_{Ckl}(E)=U_{Ckl}(E)\zeta(E-E_{Ckl})\tag{13}$$

in Eqs. (11) and (12) (i.e., G_{Ckl} cannot be taken proportional to G_I in this case), where $\zeta(x)=\mathcal{P}(1/x)-i\pi\delta(x)$. We thus obtain a system of linear, inhomogeneous equations for G_C and U_{Ckl}:

$$(E-E_C)G_C-\sum_{D\neq C}K_{C,D}G_D-\sum_{Dkl}K_{C,Dkl}\zeta(E-E_{Dkl})U_{Dkl}=\langle C|I\rangle \ , \tag{14}$$

$$U_{Ckl}-\sum_{D\neq C}K_{Ckl,Dkl}\zeta(E-E_{Dkl})U_{Dkl}-\sum_{D}K_{Ckl,D}G_D=0 \ , \tag{15}$$

which we solve using the continued-fraction (CF) rules of Swain [13], obtaining the results

$$G_\pm(E)=\frac{c_\pm}{E-E_\pm+(i/2)\hbar\Gamma_\pm(E)} \ , \tag{16}$$

$$U_{Akl}(E)=K_{Akl,+}(E)G_+(E)+K_{Akl,-}(E)G_-(E) \ , \tag{17}$$

$$\Gamma_\pm(E)=\frac{2i}{\hbar}\left[\sum_{C\neq\pm}\frac{|K_{\pm,C}(E)|^2}{E-E_C}+\sum_{Ckl}|K_{\pm,Ckl}(E)|^2\zeta(E-E_{Ckl})\right] . \tag{18}$$

Furthermore, using Eq. (7), we write G_I in the form

$$G_I(E)=c_+G_+(E)+c_-G_-(E) \ . \tag{19}$$

The decay rates $\gamma_\pm$ and energy shifts $\Delta_\pm$ of levels $|\pm\rangle$ are obtained, respectively, from the real and imaginary parts of $\Gamma_\pm(E)$, evaluated at $E=E_\pm$ (the pole approximation [14]):

$$\gamma_\pm\equiv\mathrm{Re}[\Gamma_\pm(E_\pm)]=\frac{2\pi}{\hbar}\sum_{k,l}|K_{\pm,Akl}(E_\pm)|^2\delta(E_\pm-E_{Akl}) \ , \tag{20}$$

$$\Delta_\pm\equiv\frac{\hbar}{2}\,\mathrm{Im}[\Gamma_\pm(E_\pm)]=\frac{|K_{\pm,M}(E_\pm)|^2}{E_\pm-E_M}+\sum_{C,k,l}\frac{|K_{\pm,Ckl}(E_\pm)|^2}{(E_\pm-E_{Ckl})} \ , \tag{21}$$

in which the factors $1/(E_\pm-E_{Ckl})$ are understood as principal values. In Eq. (21), we have dropped the quantities $|K_{\pm,C}|^2/(E_\pm-E_C)$ (for $C=\mp,B,A$), which contribute R-independent (fourth-order) shifts.

The time evolution of the system can be obtained from the Fourier transform (8) and the solutions for the G functions. The amplitudes $\langle\pm|\psi(t)\rangle$ and $\langle I|\psi(t)\rangle$ are found to be

$$\langle\pm|\psi(t)\rangle=c_\pm\exp\left[-\frac{\gamma_\pm}{2}t-\frac{it}{\hbar}(E_\pm+\Delta_\pm)\right] , \tag{22}$$

$$\langle I|\psi(t)\rangle=c_+\langle+|\psi(t)\rangle+c_-\langle-|\psi(t)\rangle \ , \tag{23}$$

and the probability amplitude for the state $|Akl\rangle$ for times $t\gg1/\gamma_+,1/\gamma_-$ is

$$\langle Akl|\psi(\infty)\rangle=\frac{c_+K_{Akl,+}(E_{Akl})}{E_{Akl}-E_+-\Delta_++(i/2)\hbar\gamma_+}+\frac{c_-K_{Akl,-}(E_{Akl})}{E_{Akl}-E_--\Delta_-+(i/2)\hbar\gamma_-} \ . \tag{24}$$

Equations (20), (21), and (24) describe the effects of the atomic interaction on the decay rates, energy shifts, and photon distribution for two-atom 2-γ spontaneous emission processes.

To evaluate these quantities, we need the matrix elements of the effective operator $K(E)$. We define the single-atom operators $K^s(E)$ ($s=1,2$) by

$$K^s(E)\equiv V^s\frac{1}{E-H_{A_s}-H_F}V^s \ , \tag{25}$$

and the two-atom operators $K^{ss'}(E)(s\neq s')$ by

$$K^{ss'}(E)\equiv V^s\frac{1}{E-H_A-H_F}V^{s'} \ . \tag{26}$$

Using Eqs. (1) and (10), we find

$$K_{\pm,Akl}(E_\pm)=\frac{1}{\sqrt{2}}[K^2_{b,akl}(E_b)\pm K^1_{b,akl}(E_b)] \ , \tag{27}$$

$$K_{\pm,Bkl}(E_\pm)=\frac{1}{\sqrt{2}}[K^1_{a,bkl}(E_a)\pm K^2_{a,bkl}(E_a)] \ , \tag{28}$$

$$\begin{aligned}K_{+,\pm kl}(E_+)&=K_{-,\mp kl}(E_-)\\&=\tfrac{1}{2}[K^1_{a,akl}(E_a)+K^2_{b,bkl}(E_b)\\&\qquad\pm K^1_{b,bkl}(E_b)\pm K^2_{a,akl}(E_a)] \ ,\end{aligned} \tag{29}$$

$$K_{\pm,Mn'}(E_\pm)=\frac{1}{\sqrt{2}}\sum_{s\neq s'}[K^{ss'}_{a_1b_2,Mn'}(E_\pm)\pm K^{ss'}_{b_1a_2,Mn'}(E_\pm)] \ , \tag{30}$$

where $|n'\rangle$ is either the vacuum state $|0\rangle$ or the two photon state $|kl\rangle$. The single-atom matrix elements $K^s_{r,tkl}(E)$ are evaluated in terms of the matrix elements of V in Eq. (5):

$$K^s_{r,tkl}(E)=-\left(\frac{2\pi\hbar c}{L^3}\right)\sum_{i,j}\varepsilon_j(\mathbf{k})\varepsilon_i(\mathbf{l})\sqrt{kl}\,\alpha^{ji}_{rt}(E,k,l;s)\times e^{i(\mathbf{k}+\mathbf{l})\cdot\mathbf{R}_s} \ , \tag{31}$$

$$\begin{aligned}\alpha^{ji}_{rt}(E,k,l;s)&\equiv\left(\frac{\mu^j_{rm}(s)\mu^i_{mt}(s)}{E-E_m-\hbar ck}+\frac{\mu^i_{rm}(s)\mu^j_{mt}(s)}{E-E_m-\hbar cl}\right)\\&=\alpha^{ij}_{rt}(E,l,k;s) \ ,\end{aligned} \tag{32}$$

where $\{r,t\}=\{a,b\}$, i and j are the $\{x,y,z\}$ directions, and $\mu_{rm}(s)=\langle r_s|\mu^s|m_s\rangle$. The matrix elements of the two-atom operator $K^{ss'}$ are evaluated similarly.

III. DECAY RATES

We square $K_{\pm,Akl}(E_\pm)$ of Eq. (27) and divide Eq. (20) into two terms:

$$\gamma_{\pm}=\gamma_0+\gamma_{12}\ , \tag{33}$$

$$\gamma_0=\frac{\pi}{\hbar}\sum_{s,k,l}|K^{s}_{b,akl}(E_b)|^2\delta(E_b-E_{akl})\ , \tag{34}$$

$$\gamma_{12}=\frac{2\pi}{\hbar}\mathrm{Re}\sum_{k,l}K^{1}_{b,akl}(E_b)K^{2}_{akl,b}(E_b)\delta(E_b-E_{akl})\ . \tag{35}$$

The summation $\sum_k$ over the field modes is then replaced in the usual way:

$$\sum_k \Longrightarrow \left[\frac{L}{2\pi}\right]^3\int dk\, k^2 d\Omega_k \sum_{\varepsilon(\mathbf{k})}\ , \tag{36}$$

and a factor $(\frac{1}{2})$ is included to prevent double counting over the frequencies of the photons [15]. The expression (34) for γ_0 is independent of R; after performing the summations over the polarizations and integrations over the solid angles and l, we obtain the expression

$$\gamma_0=\frac{4c}{9\pi}\sum_{i,j}\int_0^{k_0}dk\, k^3(k_0-k)^3|\alpha^{ji}_{ba}(E_b,k,k_0-k)|^2\ , \tag{37}$$

where we have set $k_0=(E_b-E_a)/\hbar c$, and omitted s in α^{ji}_{ba} because the two atoms are identical. The quantity γ_0 is recognized as the 2-γ decay rate of the excited state for an isolated atom (c.f. Ref. [11] and references therein). The quantity γ_{12}, on the other hand, is R dependent, and gives the effect of the cooperative atomic interaction on the damping rate of the system:

$$\begin{aligned}\gamma_{12}(R)=\frac{c}{\pi}\mathrm{Re}\sum_{i,j,p,q}\int_0^{k_0}dk\, k^3(k_0-k)^3&F_{jq}(kR)\\ &\times F_{ip}((k_0-k)R)\\ &\times\alpha^{ji}_{ba}(E_b,k,k_0-k;2)\\ &\times\alpha^{pq}_{ab}(E_b,k,k_0-k;1)\ .\end{aligned} \tag{38}$$

Here the function $F_{jq}(kR)$ is given by

$$\begin{aligned}F_{jq}(kR)&\equiv\frac{1}{4\pi}\int d\Omega_k(\delta_{jq}-\hat{k}_j\hat{k}_q)\exp(\pm i\mathbf{k}\cdot\mathbf{R})\\ &=(\delta_{jq}-\hat{R}_j\hat{R}_q)\frac{\sin kR}{kR}+(\delta_{jq}-3\hat{R}_j\hat{R}_q)\left[\frac{\cos kR}{k^2R^2}-\frac{\sin kR}{k^3R^3}\right]\\ &=\delta_{jq}[\tfrac{2}{3}j_0(kR)-\tfrac{1}{3}j_2(kR)]+\hat{R}_j\hat{R}_q j_2(kR)\ ,\end{aligned} \tag{39}$$

where $j_n(x)$ is the spherical Bessel function (of the first kind) of order n, and $\hat{R}_j$ $(\hat{k}_j)$ is the jth component of $\mathbf{R}/R$ $(\mathbf{k}/k)$. The expression $\sum_{j,q}k^3\mu^j(2)F_{jq}(kR)\mu^q(1)$ represents the retarded interaction of a transition dipole at $\mathbf{R}_1$, oscillating with frequency kc, with a transition dipole at $\mathbf{R}_2$ [3]. Thus it is seen that γ_{12} is the convolution of two such dipole-dipole interaction expressions, one each at frequencies kc and $(k_0-k)c$. We recall that the decay rates of the two-atom system for single-photon transitions are expressed as $\gamma_0\pm\gamma'_{12}$, where γ'_{12} is proportional to $F_{jq}(k_0R)$ [2,3]. Thus γ_{12} is the analog for 2-γ transitions of γ'_{12}.

Equations (37) and (38) are valid for all separations of the atoms; the general expressions involve exponential integrals. To obtain a closed approximate result, we replace the denominators in the α functions by their values at $k=k_0/2$ [16]. We are then able to immediately evaluate Eq. (37) for γ_0:

$$\gamma_0=\frac{4ck_0^7}{315\pi}\sum_{i,j}\left|\alpha^{ji}_{ba}\left[E_b,\frac{k_0}{2},\frac{k_0}{2}\right]\right|^2\ . \tag{40}$$

To evaluate γ_{12}, we introduce the A integrals:

$$\begin{aligned}A(n,n')&=A(n',n)\\ &=\frac{2}{\pi}\int_0^{x_0}dx\, x^3(x_0-x)^3 j_n(x)j_{n'}(x_0-x)\\ &=\int_0^{x_0}dx\, x^{5/2}(x_0-x)^{5/2}J_{n+1/2}(x)\\ &\qquad\times J_{n'+1/2}(x_0-x)\ ,\end{aligned} \tag{41}$$

where $J_n(x)$ is a (cylindrical) Bessel function, and $x_0=k_0R$. In terms of these integrals, we rewrite Eq. (38) in the form

$$\begin{aligned}\gamma_{12}(R)=\frac{2c}{9R^7}\mathrm{Re}\sum_{i,j,p,q}\alpha^{ji}_{ba}&\left[E_b,\frac{k_0}{2},\frac{k_0}{2};2\right]\alpha^{pq}_{ab}\left[E_b,\frac{k_0}{2},\frac{k_0}{2};1\right]\\ &\times\{4\delta_{jq}\delta_{ip}A(0,0)+(\delta_{jq}-3\hat{R}_j\hat{R}_q)(\delta_{ip}-3\hat{R}_i\hat{R}_p)A(2,2)\\ &\quad-2[\delta_{jq}(\delta_{ip}-3\hat{R}_i\hat{R}_p)+\delta_{ip}(\delta_{jq}-3\hat{R}_j\hat{R}_q)]A(2,0)\}\ .\end{aligned} \tag{42}$$

The integrals $A(n,n')$ are evaluated using the recurrence relation

$$J_{1/2}(x)=\frac{3}{x}J_{3/2}(x)-J_{5/2}(x) \tag{43}$$

and the tabulated integral (for $\mu,\nu>-\frac{1}{2}$) [17]

$$\int_0^{x_0}dx\, x^{\mu}(x_0-x)^{\nu}J_{\mu}(x)J_{\nu}(x_0-x)=\frac{\Gamma(\mu+\frac{1}{2})\Gamma(\nu+\frac{1}{2})}{\sqrt{2\pi}\Gamma(\mu+\nu+1)}x_0^{\mu+\nu+1/2}J_{\mu+\nu+1/2}(x_0)\ , \tag{44}$$

where $\Gamma(\mu)$ is the gamma function. We thus obtain

$$\gamma_{12}(R)=\frac{2\sqrt{2}c}{3\sqrt{\pi}R^7}\mathrm{Re}\sum_{i,j,p,q}\alpha_{ba}^{ji}\left[E_b,\frac{k_0}{2},\frac{k_0}{2};2\right]\alpha_{ab}^{pq}\left[E_b,\frac{k_0}{2},\frac{k_0}{2};1\right]$$
$$\times\left[\delta_{jq}\delta_{ip}(k_0R)^{7/2}J_{7/2}(k_0R)+(\delta_{jq}-\hat{R}_j\hat{R}_q)(\delta_{ip}-\hat{R}_i\hat{R}_p)\frac{(k_0R)^{11/2}J_{11/2}(k_0R)}{20}\right.$$
$$\left.-[\delta_{jq}(\delta_{ip}-\hat{R}_i\hat{R}_p)+\delta_{ip}(\delta_{jq}-\hat{R}_j\hat{R}_q)]\frac{(k_0R)^{9/2}J_{9/2}(k_0R)}{4}\right] . \tag{45}$$

In general, γ_{12} involves terms oscillating at frequency k_0R, multiplied by inverse powers of k_0R ranging from $(k_0R)^{-7}$ to $(k_0R)^{-2}$. In the two limiting cases of $k_0R \ll 1$ and $k_0R \gg 1$, we find

$$\gamma_{12}\Longrightarrow\begin{cases}\gamma_0[1-O(k_0R)^2] & (k_0R \ll 1)\\ -D(k_0R)^{-2}\cos(k_0R) & (k_0R \gg 1)\ ,\end{cases} \tag{46}$$

where the coefficient D is given by

$$D=\frac{ck_0^2}{15\pi}\mathrm{Re}\sum_{i,j,p,q}\alpha_{ba}^{ji}\left[E_b,\frac{k_0}{2},\frac{k_0}{2};2\right]\alpha_{ab}^{pq}\left[E_b,\frac{k_0}{2},\frac{k_0}{2};1\right]$$
$$\times(\delta_{jq}-\hat{R}_j\hat{R}_q)(\delta_{ip}-\hat{R}_i\hat{R}_p)\ . \tag{47}$$

Thus the decay rates for the $|\pm\rangle$ states become

$$\gamma_+\Longrightarrow\begin{cases}2\gamma_0 & (k_0R \ll 1)\\ \gamma_0 & (k_0R \gg 1),\end{cases} \tag{48}$$

$$\gamma_-\Longrightarrow\begin{cases}O(k_0R)^2\gamma_0 & (k_0R \ll 1)\\ \gamma_0 & (k_0R \gg 1)\ .\end{cases} \tag{49}$$

We see that the symmetric single-excitation state of the two-atom system is superradiant and the antisymmetric state subradiant for small atomic separations, exactly as is the case for one-photon processes. For large atomic separations, the effects of the atomic interaction on the decay rates vanish as $(k_0R)^{-2}$, in analogy with the $(k_0R)^{-1}$ behavior of the corresponding quantity for one-photon emission.

IV. LEVEL SHIFTS

In this section, we calculate the contribution of the atomic interaction to the level shifts. All single-atom shifts (which involve terms independent of the separation R) are absorbed into the atomic energies E_a and E_b. We divide the shifts in Eq. (21) into $\Delta_\pm^{\mathrm{r}}$ and $\Delta_\pm^{\mathrm{nr}}$:

$$\Delta_\pm=\Delta_\pm^{\mathrm{r}}+\Delta_\pm^{\mathrm{nr}}\ , \tag{50}$$

$$\Delta_\pm^{\mathrm{r}}\equiv\sum_{k,l}\frac{|K_{\pm,Akl}(E_\pm)|^2}{(E_\pm-E_{Akl})}\ , \tag{51}$$

$$\Delta_\pm^{\mathrm{nr}}\equiv\frac{|K_{\pm,M}(E_\pm)|^2}{E_\pm-E_M}+\sum_{C\neq A}\sum_{k,l}\frac{|K_{\pm,Ckl}(E_\pm)|^2}{(E_\pm-E_{Ckl})}\ . \tag{52}$$

The shifts $\Delta_\pm^{\mathrm{r}}$ arise from 2-γ transitions from $|\pm\rangle$ to the states $|Akl\rangle$, which are resonant for $k+l\simeq k_0$, while $\Delta_\pm^{\mathrm{nr}}$ arise from transitions to other nonresonant states. Substituting into the matrix elements of $K(E)$, denoting $k_{b(a)}$ by

$$k_{b(a)}\equiv E_{m,b(a)}/\hbar c\equiv(E_m-E_{b(a)})/\hbar c\ , \tag{53}$$

and making use of the symmetry properties of $F_{jq}(kR)F_{ip}(lR)$ with respect to exchanges of the indices and of k and l, we calculate the R-dependent terms in $\Delta_\pm^r$ and $\Delta_\pm^{\mathrm{nr}}$ to lowest (fourth) order in V:

$$\Delta_\pm^r=\mp\frac{2}{\pi^2\hbar c}\mathrm{Re}\sum_{i,j,p,q}\int_0^\infty\int_0^\infty dk\,dl\,k^3l^3F_{jq}(kR)F_{ip}(lR)\left[\frac{\mu_{bm}^j(2)\mu_{ma}^i(2)}{(k+k_b)(k+l-k_0)}\left[\frac{\mu_{am}^p(1)\mu_{mb}^q(1)}{k+k_b}+\frac{\mu_{am}^q(1)\mu_{mb}^p(1)}{l+k_b}\right]\right]\ , \tag{54}$$

$$\Delta_\pm^{\mathrm{nr}}=-\frac{1}{2\pi^2\hbar c(k_a+k_b)}\left|\sum_{j,q}\int_0^\infty dk\,k^3F_{jq}(k,R)\left[\left[\frac{\mu_{am}^j(1)\mu_{bm}^q(2)}{k+k_a}+\frac{\mu_{bm}^j(2)\mu_{am}^q(1)}{k+k_b}\right]\pm(1\Longleftrightarrow 2)\right]\right|^2$$
$$-\frac{2}{\pi^2\hbar c}\mathrm{Re}\sum_{i,j,p,q}\int_0^\infty\int_0^\infty dk\,dl\,k^3l^3F_{jq}(kR)F_{ip}(lR)$$
$$\times\left\{\frac{2\mu_{bm}^j(2)\mu_{mb}^i(2)}{(k+k_b)(k+l)}\left[\frac{\mu_{am}^p(1)\mu_{ma}^q(1)}{k+k_a}+\frac{\mu_{am}^q(1)\mu_{ma}^p(1)}{l+k_a}\right]\right.$$
$$\pm\frac{\mu_{am}^j(2)\mu_{mb}^i(2)}{(k+k_a)(k+l+k_0)}\left[\frac{\mu_{bm}^p(1)\mu_{ma}^q(1)}{k+k_a}+\frac{\mu_{bm}^q(1)\mu_{ma}^p(1)}{l+k_a}\right]$$
$$+\frac{\mu_{am}^j(2)\mu_{bm}^p(1)}{(k+k_a)(l+k_b)}\left[\mu_{ma}^i(2)\mu_{mb}^q(1)\left[\frac{1}{k+k_b}+\frac{1}{l+k_a}\right]\right.$$
$$\left.\left.\pm\mu_{mb}^i(2)\mu_{ma}^q(1)\left[\frac{1}{k+k_a}+\frac{1}{l+k_b}\right]\right]\right\}\ . \tag{55}$$

The integrals in $\Delta_{\mp}^{\mathrm{nr}}$ are similar to those in the calculation of the Casimir-Polder (CP) dispersion interaction energy between two atoms in the ground state [10,15]; the only real difference is that one of our atoms is in an excited state $|b\rangle$. The integral in $\Delta_{\pm}^{\mathrm{r}}$ is new in that the integrand contains the factor $\mathcal{P}/(k+l-k_0)$, which has poles at $k+l-k_0\pm i\sigma=0$. Because all integrals extend from 0 to ∞, we cannot in general approximate the k's and l's in their denominators by $\frac{1}{2}k_0$, as we did for the decay rates. However, the integrals can all be expressed in terms of three types of integrals:

$$I_1(k_1,k_2,k_3)\\ \equiv\int_0^\infty\int_0^\infty dk\,dl\,k^3l^3F_{jq}(kR)F_{ip}(lR)\\ \times\frac{1}{(k+k_1)(k+k_2)(k+l+k_3)}\,, \tag{56}$$

$$I_2(k_1,k_2,k_3)\\ \equiv\int_0^\infty\int_0^\infty dk\,dl\,k^3l^3F_{jq}(kR)F_{ip}(lR)\\ \times\frac{1}{(k+k_1)(l+k_2)(k+l+k_3)}\,, \tag{57}$$

and $I_3(k_1)$ or the square of I_3, where

$$I_3(k_1)\equiv\int_0^\infty dk\,k^3F_{jq}(kR)\frac{1}{k+k_1}\,, \tag{58}$$

with $\{k_1,k_2\}=\{k_a,k_b\}$ and $k_3=\{0,\pm k_0\}$. We have, therefore, to evaluate the integrals in Eqs. (56)–(58). For positive values of k_3, these integrals are evaluated routinely by contour integration in the complex k and l planes; for negative and zero values of k_3, additional care must be taken for poles of the integrands. We have succeeded in finding analytical expressions for these integrals for the limiting cases of small and large k_0R. Details of the evaluation are given in the Appendix. Here we present the main results and give expressions for the leading terms in $\Delta_\pm$ for small and for large atomic separations.

At small separations, the integrals I_1 and I_2 are of order R^{-5}, I_3 of order R^{-3}. The terms involving I_1, I_2, and I_3 are therefore negligible compared to the terms involving the square of $I_3(k_1)$ in $\Delta_{\mp}^{\mathrm{nr}}$, which are of order R^{-6}; the latter thus provide the dominant contributions to the shifts at small R. The resulting shifts $\Delta_\pm$,

$$\Delta_\pm\Longrightarrow-\frac{1}{2(E_{m,a}+E_{m,b})R^6}\\ \times\left|\sum_{j,q}[\mu_{am}^j(1)\mu_{bm}^q(2)\pm\mu_{am}^j(2)\mu_{bm}^q(1)]\\ \times(\delta_{jq}-3\hat{R}_j\hat{R}_q)\right|^2\,, \tag{59}$$

are the leading results at short distances, at which retardation of the interaction should be negligible. Indeed, if we replace the retarded interaction of Eq. (5) by the instantaneous (Coulomb) dipole-dipole coupling

$$V_c=\frac{1}{R^3}\sum_{j,q}\mu^j(1)\mu^q(2)(\delta_{jq}-3\hat{R}_j\hat{R}_q)\,, \tag{60}$$

and use second-order perturbation theory [18], we obtain the results

$$\sum_C\frac{|\langle\pm|V_c|C\rangle|^2}{E_\pm-E_C}=\frac{|\langle\pm|V_c|M\rangle|^2}{E_\pm-E_M}\,, \tag{61}$$

which are found to be identical with Eq. (59), as expected. These shifts are of the same order of magnitude as the London–van der Waals dispersion energies [10], which have been extensively investigated experimentally [19].

For large atomic separations, it is found (as with CP) that $\Delta_{\mp}^{\mathrm{nr}}$ yields shifts whose leading terms are proportional to R^{-7}, given by terms involving the integrals $I_1(k_b,k_a,0)$ and $I_2(k_b,k_a,0)$. In our system, however, these are found to be negligible compared to $\Delta_{\pm}^{\mathrm{r}}$, which is of order R^{-2} at large R. The shifts $\Delta_{\pm}^{\mathrm{r}}$, involving $I_1(k_b,k_b,-k_0)$ and $I_2(k_b,k_b,-k_0)$, are absent in the CP theory of the dispersion interaction; they are analogous to the first-order (resonance) dispersion energies arising between an atom in an excited state and an identical atom in the ground state in one-photon transitions (see e.g., Stephen, Ref. 2). The contribution of the shifts $\Delta_{\pm}^{\mathrm{r}}$ at large atomic separations is much larger than that of the $\Delta_{\mp}^{\mathrm{nr}}$ for reasons which can be viewed in the following way: Photons that fail to conserve energy by an amount ΔE can survive only for a time $\hbar/\Delta E$, which is of the order of magnitude $(k_0c)^{-1}$ in the present case. In that time, they can propagate a distance of order k_0^{-1}. Thus, for distances $R\gg k_0^{-1}$, the contribution of these virtual photons is negligible, and the shifts are dominated by $\Delta_{\pm}^{\mathrm{r}}$,

$$\Delta_\pm\Longrightarrow\mp\frac{2\hbar ck_0^7}{15\pi}\frac{\sin(k_0R)}{(k_0R)^2}\mathrm{Re}\sum_{i,j,p,q}\frac{\mu_{bm}^j(2)\mu_{ma}^i(2)[\mu_{am}^p(1)\mu_{mb}^q(1)+\mu_{am}^q(1)\mu_{mb}^p(1)]}{(E_{m,a}+E_{m,b})^2}(\delta_{jq}-\hat{R}_j\hat{R}_q)(\delta_{ip}-\hat{R}_i\hat{R}_p)\,. \tag{62}$$

These shifts vanish as $(k_0R)^{-2}$, and are analogous to those occurring in the corresponding one-photon case, which involve functions oscillating at frequency k_0R and vanishing as $(k_0R)^{-1}$ for large atomic separations

V. ENERGY DISTRIBUTION OF THE SYSTEM

In this section we calculate the probability of excitation as a function of time of the atomic system, and the frequency distribution of the photons in the radiation field.

A. Probability of excitation

When the two-atom system is prepared in the state $|\pm\rangle$, the population of the initial state decays exponentially with a decay rate $\gamma_\pm$. For the general initial state $|I\rangle$, the population does not decay with a single exponential; the probability of finding one of the atoms in the excited state, $P(t)$, is found to be

$$\begin{aligned} P(t) &\equiv |\langle a_1 b_2|\psi(t)\rangle|^2 + |\langle b_1 a_2|\psi(t)\rangle|^2 \\ &= |c_+|^2 e^{-\gamma_+ t} + |c_-|^2 e^{-\gamma_- t} . \end{aligned} \tag{63}$$

In the special but interesting case in which one of the atoms is initially in the excited state and the other in the ground state, the excitation probability becomes

$$P(t) = \exp(-\gamma_0 t)\cosh(\gamma_{12} t) . \tag{64}$$

We see that the lifetime of the excitation in the two-atom system is apparently lengthened. This occurs because the photons emitted from the excited atom can be absorbed by the atom in the ground state, so that the photons do not escape immediately from the system. For small atomic separations, γ_- is very small, and the antisymmetric state $|-\rangle$ is metastable. For times t such that $\gamma_+^{-1} \ll t \ll \gamma_-^{-1}$, (approximately) half of the energy still remains in the atomic system, and half is emitted into the field. For $t \gg \gamma_\pm^{-1}$, $P(t)$ approaches zero, and all the energy is emitted into the field.

B. Photon frequency distribution

Substitution of the matrix elements of K into Eq. (24) yields the probability amplitude for $|Akl\rangle$:

$$\langle Akl|\psi(\infty)\rangle = \frac{1}{\hbar\sqrt{2}} \times \left[\frac{c_+[K^2_{akl,b}(E_{akl}) + K^1_{akl,b}(E_{akl})]}{\omega_k + \omega_l - \Omega_0 - \delta_+ + (i/2)\gamma_+} + \frac{c_-[K^2_{akl,b}(E_{akl}) - K^1_{akl,b}(E_{akl})]}{\omega_k + \omega_l - \Omega_0 - \delta_- + (i/2)\gamma_-} \right] , \tag{65}$$

where $\delta_\pm = \Delta_\pm/\hbar$, $\omega_k = kc$, and $\Omega_0 = k_0 c$. Maxima occur in the distribution for $\omega_k + \omega_l = \Omega_0 + \delta_\pm$, with widths $\gamma_\pm$ and heights $K^2_{akl,b}(E_{akl}) \pm K^1_{akl,b}(E_{akl})$. For $t \gg \gamma_\pm^{-1}$, we see that

$$\sum_{k,l} |\langle Akl|\psi(\infty)\rangle|^2 \Longrightarrow 1 , \tag{66}$$

that is, all the energy is released into the field. For $k_0 R \ll 1$, most of the energy is emitted into the peak having $\omega_k + \omega_l = \Omega_0 + \delta_+$. For $k_0 R \gg 1$, both γ_{12} and $\delta_\pm$ approach zero, and the two peaks merge to give the photon distribution corresponding to that from an isolated atom [11].

The spectrum $P(\omega)$ of the emitted radiation can be obtained by suitably summing and integrating the square of the probability amplitude in Eq. (65), in the same way as in Ref. 11. The results are similar: For the initial state $|\pm\rangle$, the spectrum is proportional to the factor $\omega^3(\Omega_0 + \delta_\pm - \omega)^3$. For general initial states, the spectrum is a superposition of terms proportional to $\omega^3(\Omega_0 + \delta_+ - \omega)^3$ and $\omega^3(\Omega_0 + \delta_- - \omega)^3$.

VI. RESONANCE FLUORESCENCE

In this section, we consider the atomic system in the ground state $|A\rangle$, irradiated by a weak probe beam that is nearly 2-γ resonant. The atoms are excited to $|\pm\rangle$ by the absorption of two photons from the beam, and subsequently emit two photons into modes k and l, returning to $|A\rangle$. At the same time, the probability also exists for ground state atoms to emit virtual photons and become "excited." To calculate the shift of the ground state properly, we must take both these processes into account. (We continue, of course, to include emission of virtual photons from $|\pm\rangle$ as well, so that their shifts too are included properly.)

The Hamiltonian H_F of the vacuum field is given by Eq. (3), including all modes but that of the (laser) probe beam. We denote by H_L, $\hbar c k_L$, and n the Hamiltonian, one-photon energy, and initial occupation number, respectively, of the (single) mode of the probe beam. The intensity of the beam is so low that we need to consider in our calculation only a single (2-γ) absorption-emission event. We separate the interaction Hamiltonian into two parts: $V = \sum_s V^s$, as defined in Eqs. (5) and (6) (with all modes but that of the probe); and the interaction of the atoms with the probe, $W = \sum_s W^s$, defined similarly by

$$W^s = -\boldsymbol{\mu}(s)\cdot\mathbf{E}_L(\mathbf{R}_s) , \tag{67}$$

$$\mathbf{E}_L(\mathbf{R}) = i\left[\frac{2\pi\hbar c k_L}{L^3}\right]^{1/2} (\boldsymbol{\varepsilon}_L a_L e^{i\mathbf{k}_L\cdot\mathbf{R}} - \boldsymbol{\varepsilon}_L^* a_L^\dagger e^{-i\mathbf{k}_L\cdot\mathbf{R}}) . \tag{68}$$

The effective 2-γ interaction of the atoms with the vacuum field is given by Eq. (10) and with the probe beam by

$$K'(E) = \sum_{s,s'=1}^{2} W^s (E - H_A - H_L)^{-1} W^{s'} . \tag{69}$$

The effective interaction operator with the whole field is given by

$$\bar{K}(E) = (V + W)(E - H_0)^{-1}(V + W) , \tag{70}$$

where $H_0 = H_A + H_F + H_L$.

The basic equation of the HM method for 2-γ resonance fluorescence becomes

$$[E-H_0-\bar{K}(E)]G(E)|I\rangle =[1+(V+W)(E-H_0)^{-1}]|I\rangle , \tag{71}$$

where $|I\rangle=|An\rangle$. The "essential states" in this problem are $\{|C\;n\rangle,\ |C\;n-2\rangle,\ |C\;n\;kl\rangle$, and $|C\;n-2\;kl\rangle\}$, where $|C\;n\rangle$ represent states in which n photons are present in the probe field, and $|C\;n\;kl\rangle$ states in which one additional photon is present in each of modes k and l, etc. The application of Eq. (71) yields

$$(E-E_{C,n})G_{C,n}-\sum_{D}{}' \sum_{n'=n,n-2} \bar{K}_{C,n;D,n'}G_{D,n'} - \sum_{D;k,l} \bar{K}_{C,n;D,n,kl}G_{D,n,kl}=\delta_{C,A} , \tag{72}$$

$$(E-E_{C,n-2})G_{C,n-2}-\sum_{D}{}' \sum_{n'=n,n-2} \bar{K}_{C,n-2;D,n'}G_{D,n'} - \sum_{D;k,l} \bar{K}_{C,n-2;D,n-2,kl}G_{D,n-2,kl}=0 , \tag{73}$$

$$(E-E_{c,n,kl})G_{C,n,kl} -\sum_{D}{}' \sum_{n'=n;n-2,kl;n,kl} \bar{K}_{C,n,kl;D,n'}G_{D,n'}=0 , \tag{74}$$

$$(E-E_{C,n-2,kl})G_{C,n-2,kl} -\sum_{D}{}' \sum_{n'=n-2;n-2,kl;n,kl} \bar{K}_{C,n-2,kl;D,n'}G_{D,n'}=0 , \tag{75}$$

where the primes on the summations indicate that the diagonal elements of $\bar{K}$ (e.g., $\bar{K}_{C,n;C,n}$) have been absorbed into the corresponding energies ($E_{C,n}$). We introduce the U functions by the equations

$$G_{D,n}=U_{D,n}\zeta(E-E_{D,n})G_I \quad (D\neq A) , \tag{76}$$

$$G_{D,n'}=U_{D,n'}\zeta(E-E_{D,n'})G_I \quad (n'=n-2;n-2,kl;n,kl) . \tag{77}$$

The substitution into Eq. (72) yields

$$(G_{An})^{-1} =E-E_{An}-\sum_{D} \sum_{n'=n,n-2} \bar{K}_{An;D,n'}\zeta(E-E_{D,n'})U_{D,n'} - \sum_{D;k,l} \bar{K}_{An;D,n,kl}\zeta(E-E_{D,n,kl})U_{D,n,kl} , \tag{78}$$

and the substitution into Eqs. (72)–(75) yields the equations for the U functions, which we solve by the continued-fraction method. Noting that the matrix elements of the operator $\bar{K}(E)$ [Eq. (70)] can be expressed in terms of the matrix elements of $K(E)$ and $K'(E)$,

$$\bar{K}_{C,n;D,n}(E_{C,n})=K_{C,D}(E_C) , \tag{79}$$

$$\bar{K}_{C,n;D,n-2}(E_{C,n})=K'_{C,n;D,n-2}(E_{C,n}) , \tag{80}$$

$$\bar{K}_{C,n;D,n,kl}(E_{C,n})=K_{C;D,kl}(E_C) , \tag{81}$$

we obtain, from Eq. (78) and the solutions for the U functions, the results

$$G_{An}(E)=\frac{1}{E-E_A-\Delta_A-n\hbar\omega_L+(i\hbar/2)\gamma_A} , \tag{82}$$

$$\gamma_A=\mathrm{Re}\left[\frac{2i}{\hbar}\left[\sum_{C=\pm}\frac{|K'_{An;C,n-2}(E_{An})|^2}{E_A-E_C-\Delta'_C+2\hbar\omega_L+(i\hbar/2)\gamma_C}\right]\right] , \tag{83}$$

$$\Delta_A=\sum_{C}\frac{|K'_{An;C,n-2}(E_{An})|^2}{E_A-E_C+2\hbar\omega_L}+\sum_{C\neq A}\frac{|K_{A,C}(E_A)|^2}{E_A-E_C} +\sum_{Ckl}\frac{|K_{A,Ckl}(E_A)|^2}{E_A-E_C-\hbar c(k+l)} , \tag{84}$$

where $\gamma_\pm$ is given by Eq. (33) and $\Delta'_\pm$ is given by

$$\Delta'_\pm=\Delta_\pm+\sum_{C}\frac{|K'_{\pm n-2;C,n}(E_{\pm n-2})|^2}{E_\pm-E_C-2\hbar\omega_L} . \tag{85}$$

Here $\Delta_\pm$ is given by Eq. (50). We see that γ_A and Δ_A are the width and shift, respectively, of the ground state $|A\rangle$; $\gamma_\pm$ and $\Delta'_\pm$ are the width and shift, respectively, of the state $|\pm\rangle$.

The calculation of the matrix elements of $K'(E)$ gives

$$K'_{An,\pm n-2}(E_{An})=\frac{\hbar\beta_{a,b}(E_a)}{\sqrt{2}}(e^{-2i\mathbf{k}_L\cdot\mathbf{R}_2}\pm e^{-2i\mathbf{k}_L\cdot\mathbf{R}_1}) , \tag{86}$$

where

$$\beta_{a,b}(E_a)\equiv-\frac{2\pi ck_L}{L^3}n(n-1)\frac{(\boldsymbol{\mu}_{am}\cdot\boldsymbol{\varepsilon}_L^*)(\boldsymbol{\mu}_{mb}\cdot\boldsymbol{\varepsilon}_L^*)}{E_a-E_m+\hbar\omega_L} . \tag{87}$$

With these results, Eq. (83) can be rewritten in the form

$$\gamma_A=|\beta_{a,b}(E_a)|^2\left[\frac{\gamma_+[1+\cos(2\mathbf{k}_L\cdot\mathbf{R})]}{(\Omega_0+\delta'_+-2\omega_L)^2+\gamma_+^2/4} +\frac{\gamma_-[1-\cos(2\mathbf{k}_L\cdot\mathbf{R})]}{(\Omega_0+\delta'_--2\omega_L)^2+\gamma_-^2/4}\right] , \tag{88}$$

where $\delta'_\pm=\Delta'_\pm/\hbar$. The width of level A consists of two terms, corresponding to the probabilities of absorption from level $|A\rangle$ to the levels $|\pm\rangle$. For small R, the absorption to level $|+\rangle$ is predominant; for large R, γ_A becomes the absorption probability for two noninteracting atoms.

The shift of level A in Eq. (84) is interpreted as follows. The last two terms are the shifts due to (virtual) 2-γ exchange between the two (ground-state) atoms; as in the calculation of Sec. IV, they produce exactly the London–van der Waals dispersion energy for small R and Casimir-Polder energy for large R [10]. The first term is the laser-induced (fourth-order) shift, which can be written in the form

$$2\hbar\left[|\beta_{a,b}(E_a)|^2\frac{\mathcal{P}}{2\omega_L-\Omega_0} +\frac{|\beta_{a,a,}(E_a)|^2[1+\cos(2\mathbf{k}_L\cdot\mathbf{R})]}{2\omega_L}\right] \tag{89}$$

and is smaller than the other terms for a weak probe field. A similar interpretation applies to the second term of Eq. (85).

Finally, the frequency distribution in modes k and l of the scattered photons is the square of the amplitude $\langle An-2kl|\psi(\infty)\rangle$ [11],

$$\langle An-2kl|\psi(\infty)\rangle = \frac{1}{\hbar(\omega_k+\omega_l)-2\hbar\omega_L-\Delta_A+(i\hbar/2)\gamma_A} \times \sum_{C=\pm} \frac{K_{Akl,C}(E_{Akl})K'_{Cn-2,An}(E_{An})}{\hbar(\omega_k+\omega_l)-\hbar\Omega_0-\Delta'_C+(i\hbar/2)\gamma_C} . \tag{90}$$

We see that, in general, two (resonance) scattering channels exist: $|An\rangle\rightarrow|\pm n-2\rangle\rightarrow|An-2kl\rangle$. These have different probability amplitudes, being proportional to $1\pm e^{2i\mathbf{k}_L\cdot\mathbf{R}}$, in complete analogy with the scattering in two-atom single-photon fluorescence [3]. For small R, the channel $|An\rangle\rightarrow|+n-2\rangle\rightarrow|An-2kl\rangle$ dominates; for large R, both channels contribute, and Eq. (90) effectively describes the scattering by two independent atoms.

VII. DISCUSSION

We have used the Heitler-Ma method to study a system of two atoms undergoing cooperative 2-γ transitions for two cases: (i) the atomic system initially in a single-excitation state, interacting with the vacuum field (2-γ spontaneous emission); and (ii) the atomic system initially in the ground state, interacting with the vacuum field and with a weak probe field (2-γ resonance fluorescence). In both cases, cooperative atomic effects are found analogous to those which are well known for single-photon transitions.

(i) The lifetimes of the $|\pm\rangle$ states are altered by the atomic interaction, having the values $\gamma_\pm=\gamma_0\pm\gamma_{12}$. The cooperative decay rate γ_{12} consists of oscillating functions of k_0R, multiplied by inverse powers of k_0R ranging from $(k_0R)^{-7}$ to $(k_0R)^{-2}$. For small atomic separations, the $|+\rangle$ state is superradiant, with $\gamma_+\rightarrow 2\gamma_0$, while the $|-\rangle$ state is subradiant, with $\gamma_-\rightarrow O(k_0R)^2\gamma_0$. For large separations, γ_{12} vanishes as $(k_0R)^{-2}\gamma_0$, and the sublevels merge into a single level with a decay rate identical with that of an isolated atom.

(ii) The energies of the $|\pm\rangle$ sublevels are shifted from the single-atom energy by an amount $\Delta_\pm$ (or $\Delta'_\pm$ for the case of resonance fluorescence that approaches $\Delta_\pm$ in a weak probe field), which in general also consists of oscillating functions of k_0R multiplied by inverse powers of k_0R. The leading terms in $\Delta_\pm$ are proportional to $(k_0R)^{-6}$ for small separations, and to $(k_0R)^{-2}$ for large separations. The shift of the ground state $|A\rangle$ in 2-γ resonance fluorescence is given by the sum of the laser-induced shift and the Casimir-London energy.

We have studied cooperative atomic effects in a two-atom system undergoing 2-γ transitions, predicting the existence of and obtaining expressions for energy shifts and altered decay rates due to the interaction of the atoms with the radiation field. The two-atom system is, however, merely the simplest example of an N-atom system. The results obtained for the two-atom system can clearly be extended to an N-atom system.

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APPENDIX: EVALUATION OF THE I INTEGRALS

In this appendix we evaluate the integrals I_i $(i=1-3)$ of Eqs. (56)–(58) for small and large atomic separations R.

It is convenient to introduce the functions

$$f_{jq}(x)\equiv(\delta_{jq}-\hat{R}_j\hat{R}_q)(-ix^2)+(\delta_{jq}-3\hat{R}_j\hat{R}_q)(x+i) , \tag{A1}$$

$$A(k,l;k_1,k_2)\equiv\begin{bmatrix} A_1(k;k_1,k_2) \\ A_2(k,l;k_1,k_2 \end{bmatrix} \equiv \begin{bmatrix} \dfrac{1}{(k+k_1)(k+k_2)} \\ \dfrac{1}{(k+k_1)(l+k_2)} \end{bmatrix} . \tag{A2}$$

The integrals I_i are then expressed in the form

$$\begin{bmatrix} I_1(k_1,k_2,k_3) \\ I_2(k_1,k_2,k_3) \end{bmatrix} = \frac{1}{R^6}\int_0^\infty\int_0^\infty dk\,dl \frac{\mathrm{Re}[f_{jq}(kR)e^{ikR}]\mathrm{Re}[f_{ip}(lR)e^{ilR}]}{(k+l+k_3)} \times A(k,l;k_1,k_2) , \tag{A3}$$

$$I_3(k_1)=\frac{1}{R^3}\mathrm{Re}\int_0^\infty dk[f_{jq}(kR)e^{ikR}]\frac{1}{k+k_1} . \tag{A4}$$

We evaluate them by contour integration in the complex k and l planes, choosing a contour in the first quadrant for terms involving e^{ikR}, and in the fourth for those involving e^{-ikR}. For an integrand of the form $B(k)e^{ikR}$, we write (where $\mathcal{R}$ are residues)

$$\int_0^\infty dk\, e^{ikR}B(k)+\int_{i\infty}^0 dk\, e^{ikR}B(k)=2\pi i\sum_{Q_1}\mathcal{R} \tag{A5}$$

or

$$\int_0^\infty dk\, e^{ikR}B(k)=i\int_0^\infty du\, e^{-uR}B(iu)+2\pi i\sum_{Q_1}\mathcal{R} ; \tag{A6}$$

similarly, for terms involving e^{-ikR}, we obtain the result

$$\int_0^\infty dk\, e^{-ikR}B(k)=-i\int_0^\infty du\, e^{-uR}B(-iu)-2\pi i\sum_{Q_4}\mathcal{R} , \tag{A7}$$

where Q_1 (Q_4) is the set of poles of the integrand occurring in the first (fourth) quadrant. Since $\{k_1,k_2\}=\{k_a,k_b\}>0$, the function $A(k,l;k_1,k_2)$ and the integrand of I_3 contain no poles in the first and fourth quadrants; only the factor $1/(k+l+k_3)$ in I_1 and I_2 may contain poles, depending on the value of k_3.

We begin with the case $k_3=k_0$, for which the integrands of I_1 and I_2 contain no poles in the first and fourth quadrants. We introduce the (real) function $B_{jq}(x)$ by the equation

$$B_{jq}(x)\equiv -\mathrm{i}f_{jq}(ix)=(\delta_{jq}-\hat{R}_j\hat{R}_q)x^2+(\delta_{jq}-3\hat{R}_j\hat{R}_q)(x+1)\ , \tag{A8}$$

and change variables k and l into u and u' as in Eqs. (A6) and (A7). For later reference, we define the integral $Y_0(k_1,k_2,k_0)$ as

$$\begin{aligned}Y_0(k_1,k_2,k_0)=\frac{1}{4R^6}\int_0^\infty\int_0^\infty du\,du'e^{-uR-u'R}\Bigg[&B_{jq}(uR)B_{ip}(u'R)\frac{A(iu,iu';k_1,k_2)}{iu+iu'+k_0}\\&+B_{jq}(-uR)B_{ip}(u'R)\frac{A(-iu,iu';k_1,k_2)}{-iu+iu'+k_0}\\&+B_{jq}(uR)B_{ip}(-u'R)\frac{A(iu,-iu';k_1,k_2)}{iu-iu'+k_0}\\&+B_{jq}(-uR)B_{ip}(-u'R)\frac{A(-iu,-iu';k_1,k_2)}{-iu-iu'+k_0}\Bigg]\ ,\end{aligned} \tag{A9}$$

in terms of which we express Eq. (A3) as

$$\begin{bmatrix}I_1(k_1,k_2,k_0)\\I_2(k_1,k_2,k_0)\end{bmatrix}=Y_0(k_1,k_2,k_0)\ . \tag{A10}$$

For $k_3=0$, we first apply Eqs. (A6) and (A7) to the integration over l, and express the right-hand side of Eq. (A3) in the form

$$\begin{aligned}&-\frac{1}{2R^6}\int_0^\infty dk[f_{jq}(kR)e^{ikR}+f_{jq}^*(kR)e^{-ikR}]\\&\qquad\times\int_0^\infty du'e^{-u'R}\left[B_{ip}(u'R)\frac{A(k,iu';k_1,k_2)}{k+iu'}+B_{ip}(-u'R)\frac{A(k,-iu';k_1,k_2)}{k-iu'}\right]\ .\end{aligned} \tag{A11}$$

The factors $(k\pm iu')^{-1}$ represent poles at $k=\mp iu'$, on the imaginary axis. Performing the integration for k as in Eqs. (A6) and (A7), we obtain

$$\begin{aligned}\begin{bmatrix}I_1(k_1,k_2,0\\I_2(k_1,k_2,0)\end{bmatrix}=Y_0(k_1,k_2,0)+\frac{\pi}{R^6}\int_0^\infty du'e^{-2u'R}[&B_{jq}(u'R)B_{ip}(-u'R)A(iu',-iu';k_1,k_2)\\&+B_{jq}(-u'R)B_{ip}(u'R)A(-iu',iu';k_1,k_2)]\ ,\end{aligned} \tag{A12}$$

where $Y_0(k_1,k_2,0)$ is given by Eq. (A9) with $k_0=0$, and the factor $1/(u-u')$ occurring in Y_0 is understood as a principal value.

For $k_3=-k_0$, we first rewrite the factor $\mathcal{P}/(k+l-k_0)$ as

$$\frac{\mathcal{P}}{k+l-k_0}=\frac{1}{2}\left[\frac{1}{k+l-k_0+i\sigma}+\frac{1}{k+l-k_0-i\sigma}\right]\ ,$$

with poles at $l=k_0-k\pm i\sigma$. The substitutions $l=\pm iu'$ in the l integrations then cause poles to occur in the k integration at $k=k_0\mp iu'$. Taking these into account, we obtain

$$\begin{aligned}\begin{bmatrix}I_1(k_1,k_2,-k_0)\\I_2(k_1,k_2,-k_0)\end{bmatrix}=&\,Y_0(k_1,k_2,-k_0)\\&+\frac{\pi}{R^6}\mathrm{Re}\left[e^{ik_0R}\int_0^\infty du'e^{-2u'R}f_{jq}((k_0+iu')R)f_{ip}^*(-iu'R)A(k_0+iu',-iu';k_1,k_2)\right]\\&+\frac{i\pi}{4R^6}\int_0^{k_0}dk[f_{jq}(kR)e^{ikR}+\mathrm{c.c.}][f_{ip}((k_0-k)R)e^{i(k_0-k)R}-\mathrm{c.c.}]A(k,k_0-k;k_1,k_2)\ ,\end{aligned} \tag{A13}$$

where $Y_0(k_1,k_2,-k_0)$ is given by Eq. (A9) with k_0 replaced by $-k_0$.

Finally, it is straightforward to rewrite Eq. (A4) as

$$I_3(k_1)=-\frac{1}{2R^3}\int_0^\infty du\, e^{-uR}\left[B_{jq}(uR)\frac{1}{iu+k_1}+B_{jq}(-uR)\frac{1}{-iu+k_1}\right]. \tag{A14}$$

It is noted that the upper limits on u and u' in the integrals I_1-I_3 are effectively of the same order of magnitude as either $1/R$ (due to the exponential factors) or k_0 [due to factors such as $1/(u^2+k_0^2)$], whichever is smaller. This allows us to simplify the integrations for two limiting cases: small separations and large separations.

1. Small separations: $k_0R \ll 1$

In this case, only very small uR and $u'R$ contribute significantly to the integrals. It is then sufficient to retain only the leading terms of $B_{jq}(\pm uR)B_{ip}(\pm u'R)$ and $e^{-uR-u'R}$, namely $(\delta_{jq}-3\hat{R}_j\hat{R}_q)(\delta_{ip}-3\hat{R}_i\hat{R}_p)$ and 1, respectively. The symmetry of the resulting integrands allows us to extend the integrations to $-\infty$. For instance, from Eqs. (A9) and (A10), we have

$$\begin{bmatrix} I_1(k_1,k_2,k_0) \\ I_2(k_1,k_2,k_0) \end{bmatrix} \Longrightarrow \frac{1}{4R^6}(\delta_{jq}-3\hat{R}_j\hat{R}_q)(\delta_{ip}-3\hat{R}_i\hat{R}_p) \times\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}du\,du'\frac{A(iu,iu';k_1,k_2)}{iu+iu'+k_0}. \tag{A15}$$

For $I_1(k_1,k_2,k_0)$, we first factor A_1

$$A_1(iu;k_1,k_2)=\begin{cases} \dfrac{1}{k_2-k_1}\left[\dfrac{1}{iu+k_1}-\dfrac{1}{iu+k_2}\right] & (k_1\neq k_2) \\[2ex] -\dfrac{\partial}{\partial k_1}\left[\dfrac{1}{iu+k_1}\right] & (k_1=k_2)\,, \end{cases} \tag{A16}$$

and then perform the resulting integrations, obtaining as the leading term of the result

$$I_1(k_1,k_2,k_0)\Longrightarrow O\left[\frac{1}{R^5}\right]. \tag{A17}$$

For $I_2(k_1,k_2,k_0)$, we factor A_2

$$A_2(iu,iu';k_1,k_2) =\left[\frac{1}{iu+k_1}+\frac{1}{iu'+k_2}\right]\frac{1}{i(u+u')+k_1+k_2}, \tag{A18}$$

and complete the integrations, obtaining

$$I_2(k_1,k_2,k_0)\Longrightarrow O\left[\frac{1}{R^5}\right]. \tag{A19}$$

Proceeding in the same way for Eqs. (A12)–(A14), we find that the leading terms of $I_1(k_1,k_2,0)$, $I_2(k_1,k_2,0)$, $I_1(k_1,k_2,-k_0)$, and $I_2(k_1,k_2,-k_0)$ are all $O(1/R^5)$, while the leading term of $I_3(k_1)$ is

$$I_3(k_1)\Longrightarrow -\frac{\pi}{2R^3}(\delta_{jq}-3\hat{R}_j\hat{R}_q). \tag{A20}$$

It is the square of $I_3(k_1)$ that provides the dominant (R^{-6}) contribution to the shifts for small atomic separations.

2. Large separations: $k_0R \gg 1$

The presence of the exponential in the I integrals ensures that only $u,u'<1/R$, or $u,u'\ll k_0$, contribute significantly to the integrals. We then approximate the integrals by neglecting u and u' in the denominators in comparison with k_1, k_2, and k_0.

We consider $k_3=k_0$ first. From Eqs. (A9) and (A10), we find

$$\begin{aligned} I_1(k_1,k_2,k_0),I_2(k_1,k_2,k_0)&\Longrightarrow\frac{1}{4k_1k_2k_0R^8}\int_0^\infty\int_0^\infty dx\,dy\,e^{-x-y}[B_{jq}(x)+B_{jq}(-x)][B_{ip}(y)+B_{ip}(-y)] \\ &=\frac{1}{k_1k_2k_0R^8}(3\delta_{jq}-5\hat{R}_j\hat{R}_q)(3\delta_{ip}-5\hat{R}_i\hat{R}_p)\,. \end{aligned} \tag{A21}$$

Similarly, $I_3(k_1)$ is found to be

$$I_3(k_1)\Longrightarrow -\frac{1}{k_1R^4}(2\delta_{ip}-5\hat{R}_i\hat{R}_p) \tag{A22}$$

and the square of $I_3(k_1)$ is thus proportional to $1/R^8$ for large separations.

In the same way, we calculate $I_1(k_1,k_2,0)$ and $I_2(k_1,k_2,0)$: these are found to approach zero as $1/R^7$ for large R. We note that it is precisely these terms that yield the well-known R^{-7} results of Casimir and Polder, in direct analogy with their calculations.

There is no analog in the CP calculation, however, to the integrals $I_1(k_1,k_2,-k_0)$ and $I_2(k_1,k_2-k_0)$ in Eq. (A13).

These arise from 2-γ transitions from the states $|\pm\rangle$ to the states $|Akl\rangle$, resonant for $k+l\simeq k_0$. The results are summarized as follows: (i) The term $Y_0(k_1,k_2,-k_0)$ is evaluated in the same way as is $Y_0(k_1,k_2,k_0)$, yielding terms vanishing as R^{-8}. (ii) The integrations of the second term produce results whose leading term is proportional to $(k_0R)^{-5}\cos(k_0R)$. (iii) The last term is calculated as is $\gamma_\pm$; the results are valid for all atomic separations:

$$\begin{aligned}\frac{\pi}{4dR^7}\Bigg[&\frac{(k_0R)^5}{15}\sin(k_0R)(\delta_{jq}-\hat{R}_j\hat{R}_q)(\delta_{ip}-\hat{R}_i\hat{R}_p)\\ &+\left[\frac{(k_0R)^4}{6}\cos(k_0R)-\frac{2(k_0R)^3}{3}\sin(k_0R)+\frac{3(k_0R)^2}{2}\cos(k_0R)-\frac{3k_0R}{2}\sin(k_0R)\right]\\ &\times[(\delta_{jq}-\hat{R}_j\hat{R}_q)(\delta_{ip}-3\hat{R}_i\hat{R}_p)+(\delta_{jq}-3\hat{R}_j\hat{R}_q)(\delta_{ip}-\hat{R}_i\hat{R}_p)]\\ &-\left[\frac{(k_0R)^3}{3}\sin(k_0R)+2(k_0R)^2\cos(k_0R)-2k_0R\sin(k_0R)\right](\delta_{jq}-3\hat{R}_j\hat{R}_q)(\delta_{ip}-3\hat{R}_i\hat{R}_p)\Bigg]\end{aligned} \tag{A23}$$

where $d=(k_1+k_0/2)(k_2+k_0/2)$. These terms oscillate at frequency k_0R and are multiplied by powers of k_0R ranging from $(k_0R)^{-6}$ to $(k_0R)^{-2}$. For small separations, the leading terms of Eq. (A23) are proportional to R^{-5}, and, as discussed in Sec. A 1, are negligible compared to the R^{-6} contribution of the square of $I_3(k_1)$. It is the first term of Eq. (A23) that gives, for large separations, the leading results of Eq. (62) for the shifts, with

$$I_1(k_1,k_2,-k_0),I_2(k_1,k_2,-k_0)\Longrightarrow\frac{\pi k_0^5\sin(k_0R)}{15(2k_1+k_0)(2k_2+k_0)R^2}(\delta_{jq}-\hat{R}_j\hat{R}_q)(\delta_{ip}-\hat{R}_i\hat{R}_p)\ . \tag{A24}$$

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