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Resonance fluorescence of two adatoms near a metal surface

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The resonance fluorescence spectrum of two atoms near a perfectly conducting metal is investigated. A quantum-mechanical version of the image method, taking into account the dipole-dipole interaction, is adopted to study the excitation spectrum. Within the framework of this approach, each single atom and its corresponding image are kinematically correlated, while dynamically they are actually independent. The effects of the surface are directly exhibited in the decay rates, the frequency shift, and the spectrum. The influence of the surface is found to be large for weak fields but becomes very small for high intensities of the pump field.

I. INTRODUCTION

There has been a series of studies of the interaction of atoms or molecules absorbed at solid surfaces with radiation fields during the past two decades.[1-8] Especially with the technical advances in the fatty-acid monolayer assembly technique,[9,10] the distance between the adatom and surface can be easily changed. By means of measurements of the fluorescent lifetime of oriented dye molecules held at well-controlled distances from a metal mirror,[11-13] it is found that for large distances from the metal surface the fluorescent lifetime oscillates as a function of distance, while for small distances the lifetime approaches zero, monotonically.

So far, a number of investigations have been made into the resonance fluorescence behavior of an adatom near a metal surface. Kuhn utilized Sommerfeld's classical electromagnetic treatment for radio waves propagating along the earth's surface to consider the surface effect, and obtained good agreement with experimental results.[14-16] Within the framework of his theory, the emitting molecule acts as an oscillating dipole near a solid surface. On the other hand, Morawitz used a quantum-mechanical approach to investigate the emission by a two-level atom at a distance comparable to the radiation wavelength from a metal surface.[17] Assuming the metal to be a perfect conductor, and replacing the metal mirror by an image behind the mirror, the same results were obtained as those from the classical approach. Later, making use of the master equation, Agarwal and George calculated the resonance fluorescence spectrum of an adatom near a metal solid surface.[2,4-7]

Dicke first pointed out that changes in lifetime should be expected when two adatoms, one of which is excited, are separated by a distance $R < \lambda$, where λ is the wavelength of the transition from the ground state to the excited state.[18] The collective atomic effects on resonance fluorescence have recently been discussed by Agarwal.[19] By use of the master equation, the scattered spectrum of light from the collective system containing two or three atoms has been calculated and compared with the one-atom system. The difference is found to be significant for weak fields but less pronounced at high intensities of the driving field. Soon afterwards, Mavroyannis used the Green's-function method to study the problem of a two-atom system in a vacuum.

However, the surface effects have still not been considered for the systems of two and more atoms. In this paper, we shall consider two two-level atoms absorbed on the surface of a perfect metal. In our calculation of the scattering spectrum, we shall assume that the overlap of the wave function is very small and may be neglected, and that the interaction between the atoms is of the dipole-dipole type. Effects associated with the translational motion of the atoms, such as recoil, are ignored. Meanwhile, we assume the metal to have perfect conductivity, so that the role played by the metal surface with respect to the radiation is nothing more than a reflecting mirror. The radiation emitted from one atom close to the surface can reach another one by either direct transmission or through reflection from the surface, as though it were emitted by the image of the first atom. (See Fig. 1.) In fact we cannot differentiate between the two processes, since these fields carry no information about which process has occurred. Therefore the atom and the corresponding image are considered on the same footing.[16,17] The interaction between atoms and images can also be replaced by the dipole-dipole interaction.

This paper is organized as follows. In Sec. II we introduce the atomic and radiation-field operators. This is followed by the construction of atom-image correlated states and the effective Hamiltonian for the system; this Hamiltonian is then used to derive the equation of motion for the Green's functions describing the symmetric and antisymmetric modes in Sec. III. Using a decoupling approximation to truncate the hierarchy of

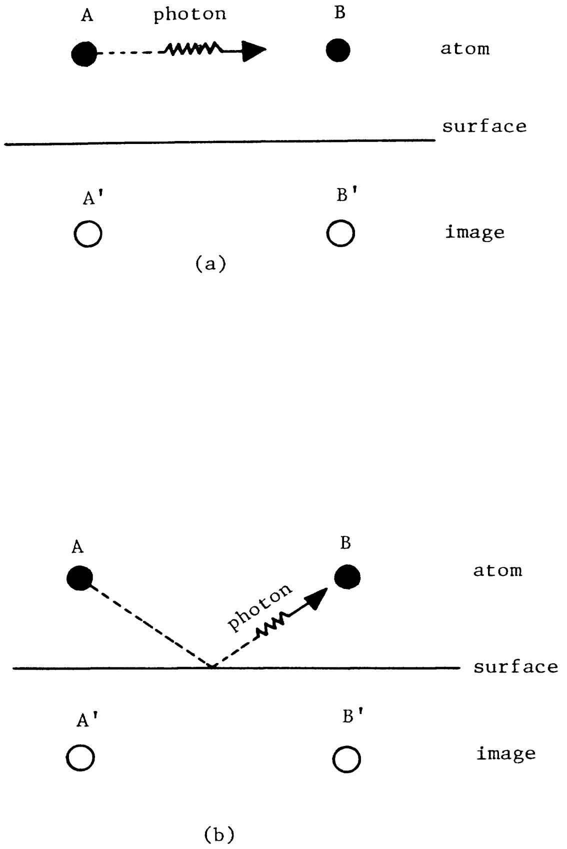

FIG. 1. Plots of two-level atoms located near a perfectly conducting metal surface. Both electronic dipoles are assumed to be perpendicular to the surface plane. (a) Direct photon exchange. (b) Photon exchange via reflection from the surface.

the Green's functions appearing in the equations of motion, expressions are derived for the Green's functions of the symmetric and antisymmetric modes. In Sec. IV the excitation spectra of the symmetric and antisymmetric modes is considered. In Sec. V, we give some concluding remarks.

II. THEORY

We shall restrict ourselves to the case where each atom near the metal surface has just two energy levels. Let $|v\rangle_\mu$ denote the excited level for the atom labeled by the indes μ, and $|0\rangle_\mu$ the ground state, where normalization and orthogonality require that $_\mu\langle v|v\rangle_\mu=1$, $_\mu\langle 0|0\rangle_\mu=1$, and $_\mu\langle v|0\rangle_\mu=0$. The translation frequency is denoted by $E_{v0}=E_v-E_0$. The translation between the ground and excited states can be facilitated by introducing excitation and deexcitation operators $b_\mu^\dagger$ and b_μ, respectively, with the properties that $b_\mu^\dagger|0\rangle_\mu=|v\rangle_\mu$, $b_\mu|v\rangle_\mu=0$, $b_\mu|0\rangle_\mu=0$, and $b_\mu|v\rangle_\mu=|0\rangle_\mu$. As a result, it can be readily recognized that $b_\mu^\dagger$ and b_μ obey the anticommutation relation $\{b_\mu,b_\mu^\dagger\}=1$. The nonoverlapping property of the two atoms also required that the operators representing different atoms commute, $[b_\mu^\dagger,b_v^\dagger]=[b_\mu,b_v]=0$ and $[b_\mu,b_v^\dagger]=0$ for $\mu\neq v$. Thus according to the model of atom-image correlation in the presence of a perfect metal surface, we may describe the emission by a single atom in the vicinity of a metal surface as a two-atom problem with complete uncertainty as to which atom is excited. In this paper we consider only the transition dipole perpendicular to the metal surface. In this case the dipole of the atom and its image have the same phase.

As in Refs. 20 and 21, we can write down an effective Hamiltonian for the system of two identical atoms with a distance R and surface-atom separation d. The atoms are then excited by the external laser field with frequency $\omega_0=E_{v0}$, and coupled to the remaining modes of the electromagnetic field. Units in which $\hbar=1$ are used throughout. We have

$$H=H_s+H_c+H_{rc}+H_{rs}\ , \tag{1}$$

$$\begin{aligned} H_s=\omega_0(b_A^\dagger b_A+b_{AI}^\dagger b_{AI}+b_b^\dagger b_B+b_{BI}^\dagger b_{BI}+\beta_0^\dagger\beta_0) \\ +\tfrac{1}{2}i\omega_p f^{1/2}[(b_A+b_{AI}+b_B+b_{BI})\beta_0^\dagger-\text{H.c.}]\ , \end{aligned} \tag{2}$$

$$\begin{aligned} H_c=V_{AB}(b_A^\dagger b_B+b_{AI}^\dagger b_{BI}+b_B^\dagger b_A+b_{BI}^\dagger b_{AI}) \\ +V_{ABI}(b_A^\dagger b_{BI}+b_{AI}^\dagger b_B+b_B^\dagger b_{AI}+b_{BI}^\dagger b_A) \\ +V(b_A^\dagger b_{AI}+b_{AI}^\dagger b_A+b_B^\dagger b_{BI}+b_{BI}^\dagger b_B)\ , \end{aligned} \tag{3}$$

$$H_{rc}=\sum_{\mathbf{k},\lambda} ck\beta_{\mathbf{k}\lambda}^\dagger\beta_{\mathbf{k}\lambda}\ , \tag{4}$$

$$\begin{aligned} H_{rs}=\tfrac{1}{2}i\omega_p\sum_{\mathbf{k},\lambda}(fE_{v0}/ck)^{1/2}[(e^{-i\mathbf{k}\cdot\mathbf{R}_A}b_A+e^{-i\mathbf{k}\cdot\mathbf{R}_B}b_B \\ +e^{-i\mathbf{k}\cdot\mathbf{R}_{AI}}b_{AI} \\ +e^{-i\mathbf{k}\cdot\mathbf{R}_{BI}}b_{BI})\beta_{\mathbf{k}\lambda}^\dagger-\text{H.c.}]\ , \end{aligned} \tag{5}$$

$$V_{AB}=\mathbf{P}_A\cdot\mathbf{P}_B/R^3-3(\mathbf{P}_A\cdot\mathbf{R})(\mathbf{P}_B\cdot\mathbf{R})/R^5\ , \tag{6}$$

$$V_{ABI}=\mathbf{P}_A\cdot\mathbf{P}_B/R_{ABI}^3-3(\mathbf{P}_A\cdot\mathbf{R}_{ABI})(\mathbf{P}_B\cdot\mathbf{R}_{BAI})/R_{ABI}^5\ , \tag{7}$$

$$V=-P_A^2/(2d)^3=-P_B^2/(2d)^3\ . \tag{8}$$

Here P_A and P_B denote the dipole moments associated with transitions from the ground states to excited states of the atoms A and B, respectively; R is the distance between atom A and atom B; R_{ABI} (R_{BAI}) refers to the distance between atom A (B) and image B (A); f is the oscillator strength for the electronic transition $0\rightarrow v$; ω_p is the plasma frequency; $\beta_0^\dagger$ and β_0 are boson creation and annihilation operators for the pump mode $\omega_0=E_{v0}$; $\beta_{\mathbf{k}\lambda}^\dagger$ and $\beta_{\mathbf{k}\lambda}$ are the corresponding operators describing the electromagnetic field with wave vector $\mathbf{k}$, frequency ck, and transverse polarization $\lambda(1,2)$.

The first five terms in Eq. (2) describe the free fields of atom A, atom B, their images, and the pump field, while the remaining ones represent the coupling between them. Equation (3) describes the dipole-dipole interaction between the atoms and images; Eqs. (4) and (5) represent the free electromagnetic fields. If the terms describing the fields of images are discarded in Eqs. (1)–(5), then the remaining terms are identical to those used in Ref. 20 to study the system of two-atom without surface, and therefore our results will be valid in this limit.

III. DERIVATION OF GREEN'S FUNCTION

The Fourier transform of the retarded double-time Green's function is defined as following:

$$G_{AB}^{(\pm)}(\omega)=\langle\langle (b_A+b_{AI}\pm(b_B+b_{BI});b_A^\dagger+b_{AI}^\dagger\rangle\rangle \tag{9}$$

The equation of motion for $G_{AB}^{(\pm)}(\omega)$ is given by

$$\omega G_{AB}^{(\pm)}(\omega)=\langle[(b_A+b_{AI})\pm(b_B+b_{BI},b_A^\dagger+b_{AI}^\dagger]\rangle + \langle\langle[(b_A+b_{AI})\pm(b_B+b_{BI}),H];b_A^\dagger+b_{AI}^\dagger\rangle\rangle \; . \tag{10}$$

We also define the following sets of Green's functions:

$$\begin{aligned}\Gamma_1^{(\pm)}(\omega)&=\langle\langle[(b_A^\dagger b_A+b_{AI}^\dagger b_{AI})\\&\qquad\pm(b_B^\dagger b_B+b_{BI}^\dagger b_{BI})]\beta_0;b_A^\dagger+b_{AI}^\dagger\rangle\rangle\;,\\ \Gamma_2^{(\pm)}(\omega)&=\langle\langle[(b_A^\dagger b_B+b_{AI}^\dagger b_{BI})\\&\qquad\pm(b_B^\dagger b_A+b_{BI}^\dagger b_{AI})]\beta_0;b_A^\dagger+b_{AI}^\dagger\rangle\rangle\;,\\ \Gamma_3^{(\pm)}(\omega)&=\langle\langle[(b_A+b_{AI})\\&\qquad\pm(b_B+b_{BI})]\beta_0^\dagger\beta_0;b_A^\dagger+b_{AI}^\dagger\rangle\rangle\;,\\ \Gamma_4^{(\pm)}(\omega)&=\langle\langle[(b_A^\dagger b_{BI}+b_{AI}^\dagger b_B)\\&\qquad\pm(b_B^\dagger b_{AI}+b_{BI}^\dagger b_A)]\beta_0;b_A^\dagger+b_{AI}^\dagger\rangle\rangle\;,\\ \Gamma_5^{(\pm)}(\omega)&=\langle\langle[(b_A^\dagger b_{AI}+b_{AI}^\dagger b_A)\\&\qquad\pm(b_B^\dagger b_{BI}+b_{BI}^\dagger b_B)]\beta_0;b_A^\dagger+b_{AI}^\dagger\rangle\rangle\;,\end{aligned} \tag{11}$$

where + and − refer to the symmetric and antisymmetric modes, respectively. Using Eq. (10) and the Hamiltonian (1)–(5), we obtain the equation of motion:

$$[\omega-\omega_\pm-\tfrac{1}{2}\gamma_\pm-\omega_p^2 f(1\pm1)/4(\omega-\omega_0)]G_{AB}^{(\pm)}(\omega) =2(1-2n_\nu)+i\omega_p f^{1/2}\Gamma_1^{(\pm)}(\omega)\;, \tag{12}$$

where

$$\omega_\pm=\omega_0+V\pm(V_{AB}+V_{ABI})\;, \tag{13}$$

$$\gamma_\pm=\gamma+\gamma_2\pm(\gamma_{AB}+\gamma_3)\;, \tag{14}$$

$$\gamma_{AB}=\tfrac{1}{2}\omega_p^2\sum_{\mathbf{k},\lambda}\left[\frac{fE\nu_0}{ck(\omega-ck)}\right]\exp(i\mathbf{k}\cdot\mathbf{R}_{AB})\;. \tag{15}$$

The expression for $\gamma=\gamma_{AA}=\gamma_{BB}$ can be obtained from γ_{AB} in Eq. (15), if we replace the exponential factor $\exp[i(\mathbf{k}\cdot\mathbf{R}_{AB})]$ by unity, and

$$\gamma_2=\tfrac{1}{2}\omega_p^2\sum_{\mathbf{k},\lambda}\left[\frac{fE_{\nu0}}{ck(\omega-ck)}\right]\exp(i\mathbf{k}\cdot2\mathbf{d})\;, \tag{16}$$

$$\gamma_3=\tfrac{1}{2}\omega_p^2\sum_{\mathbf{k},\lambda}\left[\frac{fE_{\nu0}}{ck(\omega-ck)}\right]\exp(i\mathbf{k}\cdot\mathbf{R}_{ABI})\;, \tag{17}$$

$$\nu_\nu=\langle b_A^\dagger b_A\rangle=\langle b_B^\dagger b_B\rangle=\langle b_{AI}^\dagger b_{AI}\rangle=\langle b_{BI}^\dagger b_{BI}\rangle\;. \tag{18}$$

In deriving Eq. (12), we have discarded the Green's function

$$\langle\langle[(b_A^\dagger b_A+b_{AI}^\dagger b_{AI})\pm(b_B^\dagger b_B+b_{BI}^\dagger b_{BI})]\beta_{\mathbf{k}\lambda};b_A^\dagger+b_{AI}^\dagger\rangle\rangle$$

as being unimportant compared to $\Gamma_1^{(\pm)}(\omega)$. This is in agreement with our original assumption that the pump field is the strongest. We go on with deriving the equation of motion for $\Gamma_1^{(\pm)}(\omega)$:

$$\begin{aligned}(\omega-\omega_0-\gamma)\Gamma_1^{(\pm)}(\omega)&=-\tfrac{1}{2}i\omega_p f^{1/2}\Gamma_3^{(\pm)}(\omega)\\&\quad+[\tfrac{1}{2}\gamma_{AB}(1\pm1)+V_{AB}(1\pm1)]\Gamma_2^{(\pm)}(\omega)\\&\quad+[\tfrac{1}{2}\gamma_3(1\pm1)+V_{ABI}(1\mp1)]\Gamma_4^{(\pm)}(\omega)+\gamma_2\Gamma_5^{(\pm)}(\omega)\;,\end{aligned} \tag{19}$$

where a term of the form

$$-\tfrac{1}{2}i\omega_p f^{1/2}\langle\langle[(b_A\pm b_B)-(b_{AI}\pm b_{BI})]\beta_0\beta_0-(b_A^\dagger b_A b_B\pm b_B^\dagger b_B b_A)-(b_{AI}^\dagger b_{AI} b_{BI}\pm b_{BI}^\dagger b_{BI} b_{AI});b_A^\dagger+b_{AI}^\dagger\rangle\rangle \tag{20}$$

has been omitted on the right-hand side of Eq. (19). The first term in Eq. (20) describes the physical process where two photons of the pump field act simultaneously on the atomic system, while the remaining ones represent the similar process arising from the action of the dipole atomic field A and B on the number density operators $b_A^\dagger b_A$ and $b_B^\dagger b_B$, respectively. Both processes are less important than $\Gamma_3^{(\pm)}(\omega)$ caused by the photon density of the pump field acting on the atomic system A and B.

Through the same procedure, we derive the following equations for the Green's functions:

$$\begin{aligned}(\omega-\omega_0-\gamma)\Gamma_2^{(\pm)}(\omega)&=\mp\tfrac{1}{2}\omega_p f^{1/2}\Gamma_3^{(\pm)}(\omega)\\&\quad+[\tfrac{1}{2}\gamma_{AB}(1\pm1)+V_{AB}(1\mp1)]\Gamma_1^{(\pm)}(\omega)+\gamma_2\Gamma_r^{(\pm)}(\omega)\\&\quad+[\tfrac{1}{2}\gamma_3(1\pm1)+V_{ABI}(1\mp1)]\Gamma_5^{(\pm)}(\omega)\;,\end{aligned} \tag{21}$$

$$\begin{aligned}(\omega-\omega\pm-\tfrac{1}{2}\gamma\pm)\Gamma_3^{(\pm)}(\omega)&=2(1-2n\nu)+2i\Omega n_0^{1/2}\Gamma_2^{(\pm)}(\omega)\\&\quad+[\Omega^2(1\pm1)/(\omega-\omega_0)]G_{AB}^{(\pm)}(\omega)\;,\end{aligned} \tag{22}$$

where

$$\Omega^2=\omega_p^2 f n_0 , \tag{23}$$

and

$$n_0=\langle \beta_0^\dagger \beta_0 \rangle \tag{24}$$

are the energy shift (Rabi frequency) and the average number density of the photon pump field, respectively. In deriving Eq. (22), use has been made of the following decoupling approximation:

$$\langle\langle [(b_A^\dagger b_A+b_{AI}^\dagger b_{AI})\pm(b_B^\dagger b_B+b_{BI}^\dagger b_{BI})]\beta_0^\dagger\beta_0\beta_0;b_A^\dagger+b_{AI}^\dagger\rangle\rangle \approx 2n_0\langle\langle [(b_A^\dagger b_A+b_{AI}^\dagger b_{AI})\pm(b_B^\dagger b_B+b_{BI}^\dagger b_{BI})]\beta_0;b_A^\dagger+b_{AI}^\dagger\rangle\rangle , \tag{25}$$

$$\langle\langle \beta_0^\dagger\beta_0\beta_0;b_A^\dagger+b_{AI}^\dagger\rangle\rangle \approx 2n_0\langle\langle \beta_0;b_A^\dagger+b_{AI}^\dagger\rangle\rangle=[i\omega_p f^{1/2}/(\omega-\omega_0)]n_0 G_{AB}^{(+)}(\omega) . \tag{26}$$

This decoupling approximation indicates that all dynamic effects arising from photon-photon interactions for the pump field are completely discarded, and a photon is influenced by only the average field of all others (static effect) which is described by the average number of photons $n_0=\langle\beta_0^\dagger\beta_0\rangle$. This approximation is asymptotically valid for large values and high photon densities of the pump field. Its physical meaning is similar to the random-phase approximation (RPA).

To have the Green's functions closed, we can derive the finite hierachy equations as follows:

$$(\omega-\omega_0-\gamma)\Gamma_4^{(\pm)}(\omega)=\mp\tfrac{1}{2}i\omega_p f^{1/2}\Gamma_3^{(\pm)}(\omega)+[\tfrac{1}{2}\gamma_3(1\pm1)+V_{ABI}(1\mp1)]\Gamma_1^{(\pm)}(\omega) +\gamma_2\Gamma_2^{(\pm)}(\omega)+[\tfrac{1}{2}\gamma_{AB}(1\pm1)+V_{AB}(1\mp1)]\Gamma_5^{(\pm)}(\omega) , \tag{27}$$

$$(\omega-\omega_0-\gamma)\Gamma_5^{(\pm)}(\omega)=-\tfrac{1}{2}i\omega_p f^{1/2}\Gamma_3^{(\pm)}(\omega)+[\tfrac{1}{2}\gamma_3(1\pm1)+V_{ABI}(1\mp1)]\Gamma_2^{(\pm)}(\omega) +\gamma_2\Gamma_1^{(\pm)}(\omega)+[\tfrac{1}{2}\gamma_{AB}(1\pm1)+V_{AB}(1\mp1)]\Gamma_4^{(\pm)}(\omega) . \tag{28}$$

Solving Eqs. (19), (21), (22), (27), and (28), we obtain

$$[(\omega-\omega_0-\gamma_+)(\omega-\omega_+-\tfrac{1}{2}\gamma_+)-\Omega^2]\Gamma_1^{(+)}(\omega)=-\tfrac{1}{2}i\omega_p f^{1/2}\left[2(1-2n_\nu)n_0+\frac{2\Omega^2}{\omega-\omega_0}G_{AB}^{(+)}(\omega)\right] , \tag{29}$$

$$[(\omega-\omega_0-\tfrac{1}{2}\gamma_-)(\omega-\omega_0-\gamma-\gamma_2+2V_{AB}+2V_{ABI})-\Omega^2]\Gamma_1^{(-)}(\omega)=-i\omega_p f^{1/2}n_0(1-2n_\nu) . \tag{30}$$

Substituting Eq. (29) into Eq. (12) and retaining only those energy shifts induced by the pump field which are of the order of Ω, we have

$$G_{AB}^{(+)}(\omega)=\frac{2(1-2n_\nu)}{\omega-\omega_+-\frac{1}{2}\gamma_+}\left[1+\frac{\frac{1}{2}\Omega^2}{(\omega-\omega_+-\frac{1}{2}\gamma_+)(\omega-\omega_0-\gamma_+)-\Omega^2}\right] , \tag{31}$$

which describes the spectrum of the symmetric modes, while substituting Eq. (30) into Eq. (12) yields

$$G_{AB}^{(-)}(\omega)=\frac{2(1-2n_\nu)}{\omega-\omega_--\frac{1}{2}\gamma_-}\left[1+\frac{\frac{1}{2}\Omega^2}{(\omega-\omega_--\frac{1}{2}\gamma_-)(\omega-\omega_0-\gamma-\gamma_2+2V_{AB}+2V_{ABI})-\Omega^2}\right] , \tag{32}$$

which describes the spectrum of the antisymmetric modes. The Green's functions (31) and (32), first of all, will be used to compare with the results obtained by former authors. In the absence of the pump field and the surface, i.e., in the limit in which $\Omega=0(n_0=0)$, Eqs. (31) and (32) reduce to

$$G_{AB}^{(\pm)}(\omega)=\frac{2(1-2n_\nu)}{\omega-\omega_\pm-\frac{1}{2}\gamma_\pm} , \tag{33}$$

where

$$\omega_\pm=\omega_0\pm V_{AB} , \tag{34}$$

$$\gamma_\pm=\gamma\pm\gamma_{AB} . \tag{35}$$

Equation (33) describes the excitation spectrum of the symmetric (+) and antisymmetric (−) energy modes for the physical process arising from the interaction of two similar atoms, one of which is excited. When the appropriate limits are taken for $R<\lambda$ and $R>\lambda$, where $\lambda=c/E_{\nu 0}$, the results obtained from Eq. (33) for the spectrum of the symmetric and antisymmetric modes are identical to those in Refs. 22–24.

In the absence of the atom B and surface, i.e., $\gamma_{AB}=\gamma_2=\gamma_3=0$ and $V=V_{AB}=V_{ABI}=0$, then Eqs. (31) and (32) become identical,

$$G_{AB}^{(+)}(\omega)=G_{AB}^{(-)}(\omega)=G_{AA}(\omega) ,$$

$$G_{AA}(\omega)=\frac{2(1-2n_\nu)}{\omega-\omega_0-\frac{1}{2}\gamma}\times\left[1+\frac{\frac{1}{2}\Omega^2}{(\omega-\omega_0-\gamma)(\omega-\omega_0-\frac{1}{2}\gamma)-\Omega^2}\right] , \tag{36}$$

where[20]

$$\begin{aligned}&\gamma(\omega)\rightarrow -i\,\mathrm{Im}\gamma(\omega)=-i\gamma_0\ ,\\&\gamma_0=\tfrac{4}{3}(\omega_0/c)^3|\mathbf{P}_A|^2\ ,\end{aligned}\tag{37}$$

which is the spontaneous-emission probability. Then taking the imaginary part of $G_{AA}(\omega)$, we have

$$\begin{aligned}-2\mathrm{Im}G_{AA}(\omega)=4(1-2n_\nu)\Biggl[&\frac{\tfrac{1}{2}\gamma_0\left[1-\dfrac{\tfrac{1}{2}\Omega^2}{(\Omega^2+\gamma_0^2/16)}\right]}{(\omega-\omega_0)^2+(\tfrac{1}{2}\gamma_0)^2}\\&+\frac{\tfrac{1}{4}\Omega^2}{\Omega^2+\gamma_0^2/16}\left[\frac{\tfrac{3}{4}\gamma_0-(\omega-\omega_0-\Omega)\gamma_0/4\Omega}{(\omega-\omega_0-\Omega)^2+(\tfrac{3}{4}\gamma_0)^2}+\frac{\tfrac{3}{4}\gamma_0+(\omega-\omega_0+\Omega)\gamma_0/4\Omega}{(\omega-\omega_0+\Omega)^2+(\tfrac{3}{4}\gamma_0)^2}\right]\Biggr]\ ,\end{aligned}\tag{38}$$

which describes the spectral function for the interacting system in question. In the limiting case when $\Omega^2 \gg \gamma_0^2/16$, Eq. (38) reduces to

$$-2\mathrm{Im}G_{AA}(\omega)=(1-2n\nu)\left[\frac{2(\tfrac{1}{2}\gamma_0)}{(\omega-\omega_0)^2+(\tfrac{1}{2}\gamma_0)^2}+\frac{\tfrac{3}{4}\gamma_0}{(\omega-\omega_0-\Omega)^2+(\tfrac{3}{4}\gamma_0)^2}+\frac{\tfrac{3}{4}\gamma_0}{(\omega-\omega_0+\Gamma)^2+(\tfrac{3}{4}\gamma_0)^2}\right]\ ,\tag{39}$$

which is identical to that known in Refs. 1 and 25–30, except for the differences between the factors which have no influence on the conclusion.

In the absence of the surface, i.e., $\gamma_2=\gamma_3=0$ and $V=V_{ABI}=0$, Eqs. (31) and (32) become

$$G_{AB}^{(+)}(\omega)=\frac{2(1-2n_\nu)}{\omega-\omega_+-\tfrac{1}{2}\gamma_+}\left[1+\frac{\tfrac{1}{2}\Omega^2}{(\omega-\omega_0-\gamma_+)(\omega-\omega_+-\tfrac{1}{2}\gamma_+)-\Omega^2}\right]\ ,\tag{40}$$

$$G_{AB}^{(-)}(\omega)=\frac{2(1-2n_\nu)}{\omega-\omega_--\tfrac{1}{2}\gamma_-}\left[1+\frac{\tfrac{1}{2}\Omega^2}{(\omega-\omega_0-\gamma+2V_{AB})(\omega-\omega_--\tfrac{1}{2}\gamma_-)-\Omega^2}\right]\ ,\tag{41}$$

where

$$\omega_\pm=\omega_0\pm V_{AB}\ ,\tag{42}$$

$$\gamma_\pm=\gamma\pm\gamma_{AB}\ .\tag{43}$$

Equations (40) and (41) are not different from Ref. 20 which discusses in more detail the problem of the two-atom system without a surface. Thus it is unnecessary to go into details.

In the absence of atom B, i.e., $\gamma_{AB}=\gamma_3=0$ and $V_{AB}=V_{ABI}=0$,

$$G_{AB}^{(\pm)}(\omega)=\frac{2(1-2n_\nu)}{\omega-\omega_0-V-\tfrac{1}{2}(\gamma+\gamma_2)}\left[1+\frac{\tfrac{1}{2}\Omega^2}{(\omega-\omega_0-\gamma-\gamma_2)(\omega-\omega_0-V-\tfrac{1}{2}\gamma-\tfrac{1}{2}\gamma_2)-\Omega^2}\right]\ ,\tag{44}$$

comparing Eq. (44) with Eq. (36), we find that the effects of the surface are obviously on the frequency shift and decay rate. Especially in the case in which the atom is close to the surface, the frequency shift and decay rate become large, so that the spectral widths become wider. This is identical to Ref. 8.

IV. EXCITATION SPECTRUM

For the sake of convenience, we shall consider the excitation spectrum for the symmetric and antisymmetric modes separately.

A. Excitation spectrum of the symmetric modes

The expression (31) for $G_{AB}^{(+)}(\omega)$ may be rewritten as

$$G_{AB}^{(+)}(\omega)\approx 2(1-2n_\nu)\left[\frac{1-\Omega^2/2/(\Omega_+-\gamma_+/2)(\Omega_-+\gamma_+/4)}{\omega-\omega_+-\tfrac{1}{2}\gamma_+}+\frac{\Omega/4/(\Omega_-+\gamma_+/4)}{\omega-\omega_0-\Omega_+-\tfrac{3}{4}\gamma_+}+\frac{\Omega/4/(\Omega_+-\gamma_+/2)}{\omega-\omega_0+\Omega_--\tfrac{3}{4}\gamma_+}\right]\tag{45}$$

where

$$\Omega_{\pm}=\Omega\pm\tfrac{1}{2}(V_{AB}+V_{ABI})+\tfrac{1}{2}V\ . \tag{46}$$

According to Refs. 18, 22, 24, and 31, we have the following conclusions.

(i) In the limit $R<\lambda(\lambda=c/\omega_0 c/E_{\nu 0})$,

$$\tfrac{1}{2}\mathrm{Re}\gamma_{AB}=-V_{AB}+\omega_{AB}(R);\quad \mathrm{Im}\gamma_{AB}\approx\gamma_0 \tag{47}$$

where the expression for $\omega_{AB}(R)$ decays exponentially with R/λ. For $R<\lambda$, an expansion in powers of R/λ yields $\omega_{AB}(R)\approx V_{AB}$, so that, in this limit

$$\mathrm{Re}\gamma_{AB}\approx 0;\quad \mathrm{Im}\gamma_{AB}\approx\gamma_0\ . \tag{48}$$

(ii) In the limit $R>\lambda$, $\omega_{AB}(R)$ and $\mathrm{Im}\gamma_{AB}$ are negligibly small in comparison with V_{AB} and γ_0. Neglecting $\omega_{AB}(R)$ and $\mathrm{Im}\gamma_{AB}$, we obtain from Eq. (47)

$$\tfrac{1}{2}\mathrm{Re}\gamma_{AB}=-V_{AB};\quad \mathrm{Im}\gamma_{AB}=0\ . \tag{49}$$

Since the expression for γ_2 and γ_3 are similar to that for γ_{AB}, we can draw the same conclusion for γ_2 and γ_3 as for γ_{AB}. Using the above approximate expression, when $R,2d<\lambda$, Eq. (45) becomes

$$-2\mathrm{Im}G_{AB}^{(+)}(\omega)=4(1-2n_\nu)\left[\frac{2\gamma_0(1-\frac{1}{2}A)+2(\omega-\omega_+)\gamma_0 AV_+/(4\Omega^2-V_+^2+\gamma_0^2)}{(\omega-\omega_+)^2+(2\gamma_0)^2}\right.$$
$$+\tfrac{1}{4}A_-\frac{3\gamma_0-2(\omega-\omega_0-\Omega_+)\gamma_0/(2\Omega-V_+)}{(\omega-\omega_0-\Omega_+)^2+(3\gamma_0)^2}$$
$$\left.+\tfrac{1}{4}A_+\frac{3\gamma_0+2(\omega-\omega_0+\Omega_-)\gamma_0/(2\Omega+V_+)}{(\omega-\omega_0+\Omega_-)^2+(3\gamma_0)^2}\right]\ , \tag{50}$$

where

$$A=4\Omega^2(4\Omega^2-V_+^2+4\gamma_0^2)/[(4\Omega^2-V_+^2+4\gamma_0^2)+16\gamma_0^2V_+^2]\ , \tag{51}$$

$$A_{\pm}=2\Omega(2\Omega\pm V_+)/[(2\Omega\pm V_+)^2+4\gamma_0^2]\ , \tag{52}$$

$$V_+=V_{AB}+V_{ABI}+V\ . \tag{53}$$

Equation (50) describes the spectrum function of the symmetric mode. In comparison with Eq. (39), one can find the peculiar feature that the three peaks show asymmetry, and the height of the right sideband is not equal to that of the left. Doubtless, it is due to the effects of the atom B and the metal surface.

In the limit $\Omega_{\pm}^2\gg\gamma_0^2$ and $\Omega\gg V_+$, Eq. (50) reduces to

$$-2\,\mathrm{Im}G_{AB}^{(+)}(\omega)=2(1-2n_\nu)\{4\gamma_0/[(\omega-\omega_+)^2+4\gamma_0^2]+3\gamma_0/[(\omega-\omega_0-\Omega_+)^2+(3\gamma_0)^2]$$
$$+3\gamma_0/[(\omega-\omega_0+\Omega_-)^2+(3\gamma_0)^2]\}\ , \tag{54}$$

which describes three Lorentzian lines peaked at the frequencies $\omega=\omega_+$, $\omega=\omega_0+\Omega_+$, and $\omega=\omega_0-\Omega_-$, having spectral widths of ν_0, $3\gamma_0$, and $3\gamma_0$, respectively. The ratio of the central-peak height to those of the sidebands is 3:1 and the linewidth ratio is 1:1.5, which is in agreement with the conclusion derived by Agarwal and Brown.[19] The decay rates and frequency shifts of the spectrum become larger because of cooperative effects due to the presence of atom and the metal surface. We also conclude that the decay rates and energy shifts arising from the surface are greater than that from the additional atom B.

Equation (45) has been calculated numerically for different cases, and the results are shown in Figs. 2(a)–2(d). Figures 2(a) and 2(b) describe the spectrum for the cases in which the external field is not strong, where we can see that the decay rates, frequency shift, and symmetry are different from those for the cases without a surface.[1,20,25–30] Comparing Figs. 2(a) with 2(b), we find that the effect of the surface on the spectrum is stronger than that of the additional atom B.

On the other hand, Figs. 2(c) and 2(d) are the spectrum for the cases in which the driving field is strong. Comparing those with Figs. 2(a) and 2(b), the effects of the surface and the atom B have been restrained enormously, and the asymmetry of each peak almost disappears. When two atoms are located far from the surface as shown in Fig. 2(d), the influence of the second atom diminishes. Comparing Figs. 2(d) with 2(b), the height of the right sideband is practically the same as that of the left, and the symmetry is restored. It is evident that the strong field can remove the effects of the additional atom more effectively.

B. Excitation spectrum of the antisymmetric modes

The analytic expressions for this case will not be given here. Since the structure of the spectrum in this case is similar to that of the symmetric models, except that the widths of the spectrum become more narrow, we shall de-

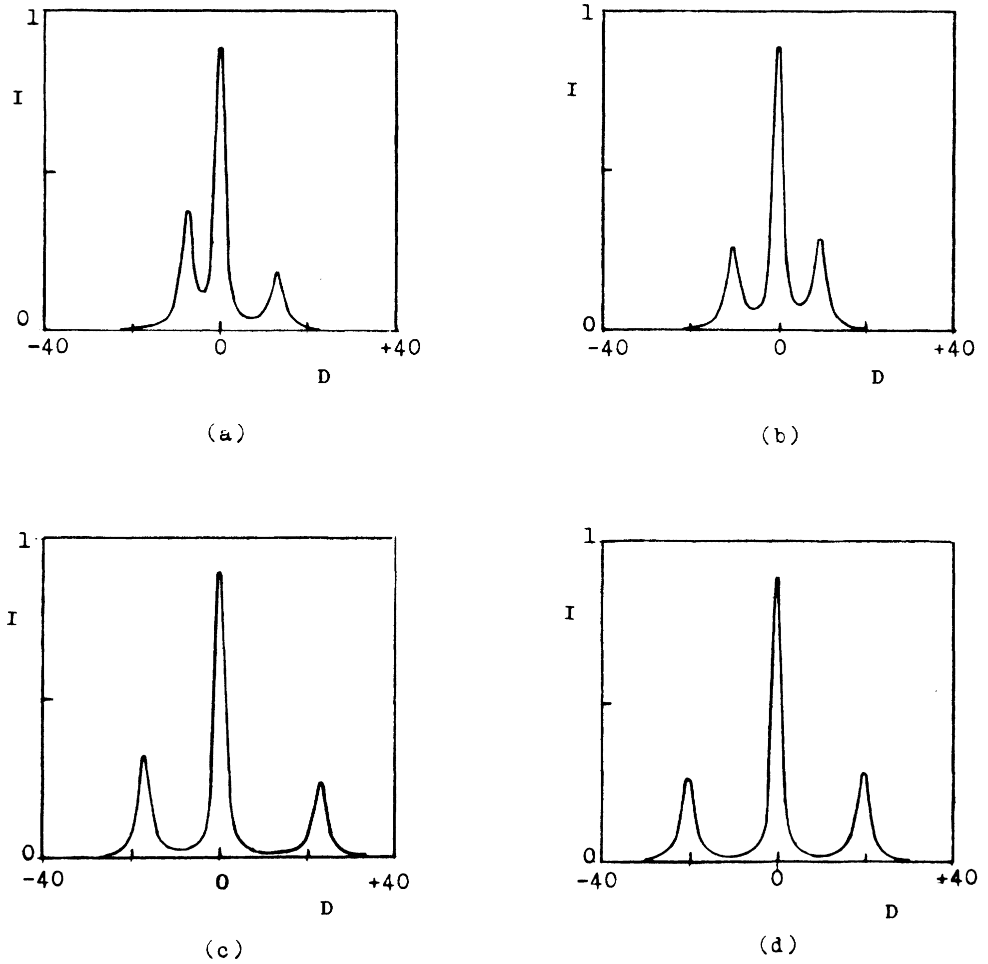

FIG. 2. Resonance-fluorescence spectrum for symmetric models vs $D=(\omega-\omega_{+}-\frac{1}{2}\mathrm{Re}\gamma_{+})/\gamma_0$ for different pump fields. The spectral intensity I is proportional to $-2\,\mathrm{Im}G_{AB}^{(+)}(\omega)$. In the plots, the quantity is dimensionless. (a) $d=0.4\lambda$, $R=2.0\lambda$, $\Omega/\gamma_0=10.0$; (b) $d=\lambda$, $R=0.4\lambda$, $\Omega/\gamma_0=10.0$; (c) $d=0.4\lambda$, $R=2.0\lambda$, $\Omega/\gamma_0=20.0$; (d) $d=\lambda$, $R=0.4\lambda$, $\Omega/\gamma_0=20.0$.

scribe just the numerical results below. When the pump field is not strong and when $R>\lambda$ and $2d<\lambda$, the result is the same as Fig. 2(a). The reason is the following. The two atoms are separated far from each other in both cases, thus the effect of the second atom is so small that it can be neglected, while the influence of the surface plays a leading role. Also, with the atomic dipole moments perpendicular to the surface in both cases, the effects of the surface on the spectrum should be the same in the two cases. Therefore the shapes of the peaks will resemble one another.

The δ peak appears when two atoms are close and far from the surface. It indicates that the central peak does not decay, i.e., the energy mode of the central peak is stable and does not radiate. We also find that the two sidebands have the same height, telling us that the cooperative effects of the atoms A and B and the surface have no contribution to the lifetime of the sidebands. This conclusion can be confirmed from the processes of deriving Green's functions.

When solving Eqs. (19), (21), (22), (27), and (28), we found

$$\Gamma_1^{(-)}(\omega)=-\Gamma_2^{(-)}(\omega)\ , \tag{55}$$

$$\Gamma_4^{(-)}(\omega)=-\Gamma_5^{(-)}(\omega)\ . \tag{56}$$

Those equations imply that the cooperative radiative effects of the atoms A and B and the surface are cancelled, and only $\Gamma_3^{(-)}(\omega)$ and $\gamma=\gamma_{AA}=\gamma_{BB}$, which have no cooperative effects, survive. When the pump field is strong and $R>\lambda$ and $2d<\lambda$, the spectral + shape resembles Fig. 2(c). The reason is the same as that stated above. The influence of a strong field on the spectrum is similar to that in Figs. 2(c) and 2(d).

V. SUMMARY

In this paper we have derived the Green's functions including surface-induced decay rates and frequence shifts. Using the Green's function method, analytic expressions for the resonance-fluorescence spectrum of two atoms near a metal surface are obtained, and the influence of the surface and additional atom *B* on the spectrum are discussed. Depending on the circumstances, the existence of the surface and additional atom *B* can cause the asymmetry of the sidebands to appear or disappear, and cause the widths of the spectrum to become wider. The external field has a strong effect on the spectrum.

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