Spontaneous emission of two interacting atoms near an interface

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The spontaneous emission rate of two interacting excited atoms near a dielectric interface is studied using the photon closed-orbit theory and the dipole image method. The total emission rate of one atom during the emission process is calculated as a function of the distance between the atom and the interface. The results suggest that the spontaneous emission rate depends not only on the atomic-interface distances, but also on the orientation of the two atomic dipoles and the initial distance between the two atoms. The oscillation in the spontaneous emission rate is caused by the interference between the outgoing electromagnetic wave emitted from one atom and other waves arriving at this atom after traveling along various classical orbits. Each peak in the Fourier transformed spontaneous emission rate corresponds with one action of photon classical orbit.

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The spontaneous emission properties of atoms in environments have been studied in detail both theoretically and experimentally. Using quantum electrodynamics method, Urbach *et al.* calculated the spontaneous emission rate of an atom near a nonabsorbing dielectric film[1,2]. Later, Wang *et al.* studied the spontaneous emission properties of an assembly of atoms in symmetric and asymmetric slabs[3,4]. Since the oscillation in the spontaneous emission rate of an atom is quite similar to the oscillation in the atomic absorption spectra[5], Du *et al.* extended the concepts of the closed-orbit theory from electron to photon and provided a general framework to understand the oscillations in the spontaneous emission rate for atoms in the environment. They calculated the spontaneous emission rate of an atom near an dielectric interface and inside a dielectric slab[6−8]. It has been demonstrated that the oscillations in the spontaneous emission rate are associated with the photon closed orbits going away from and returning to the emitting atom and are interpreted as interferences between outgoing emitted electromagnetic wave and returning electromagnetic wave traveling along different closed orbits. Recently, the spontaneous emission of two excited atoms in environments has attracted much attention. It is expected that the electromagnetic interaction between two excited atoms will be substantial when they are located within the effective mode diameter while the interaction will be weak when they are far apart. Aiello *et al.* have studied this problem in terms of the spontaneous decay of one of the two dipoles and found an expression for the mutual interaction including correct retardation times to represent the multiple reflections between the cavity mirrors[9]. Later, Takada *et al.* analyzed the spontaneous emission by two atoms in a planner microcavity under the dipole approximation and the rotation wave approximation[10]. In these earlier studies, the two atoms are all considered to belocated on the central line of the cavity so that the two atomic dipoles are parallel. As to other orientation of the dipoles, the researchers did not give a discussion. In this letter, by using the photon closed-orbit theory and the dipole image method, we give a vivid physical description and an exact formula about the spontaneous emission of two excited atoms near a dielectric interface for different orientations of the dipoles, and especially discuss the influence of the photon classical orbits on the spontaneous emission rate.

The schematic of the system is shown in Fig. 1. Two semi-infinite nonabsorbing dielectric materials with real refractive indices n_1 and n_2, respectively, form a single plane interface. It is assumed that the medium is laid on the xy plane and the normal of the medium is chosen as the z direction. The two excited atoms 1 and 2 are located near the interface. They are two-level atoms of the same transition frequency ω_0 and the same dipole moment magnitude d_0, but with different dipole moment directions. The distances from these two atoms to the interface are l_1 and l_2, respectively. The horizontal distance between the two atoms is denoted as r_0. $1'$ and $2'$ are the mirror images of atoms 1 and 2.

The physical picture for the spontaneous emission process of this system can be described by using the photon closed-orbit theory. For the case of two identical atoms,

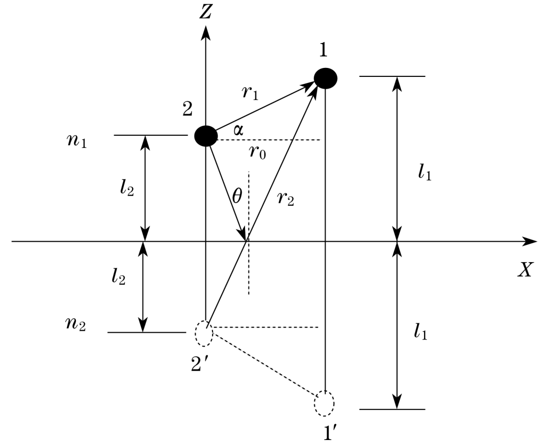

Fig. 1. Two atoms near a dielectric interface.

the electromagnetic wave emitted by the interacting atom is also very important. Some of the electromagnetic wave will propagate directly from the second atom to the first one and others will be reflected by the dividing interface and return to the first atom. The interference effect between these electromagnetic waves emitted from the second atom and the outgoing waves emitted from the first atom also produces the oscillation in the spontaneous emission rate.

Since the spontaneous emission for an excited atom can be modeled as a dipole interaction with the electrical field at the location of the atom, we use the radiation damping of a dipole antenna to simulate the spontaneous emission of the atom. Suppose the transition dipole moments of the two atoms are $\vec{d}_1 = \vec{d}_{10}\mathrm{e}^{-\mathrm{i}\omega_0 t}$, $\vec{d}_2 = \vec{d}_{20}\mathrm{e}^{-\mathrm{i}\omega_0 t}$, where ω_0 is the frequency of the oscillation and $d_{10} = d_{20}$. The radiation damping rate for one dipole antenna can be written as[11]

$$W = \frac{\omega_0}{2U}\mathrm{Im}(\vec{d}^* \cdot \vec{E}), \tag{1}$$

where U is the energy of the antenna, $\vec{d}^*$ is the complex conjugate of the dipole moment $\vec{d}$ of the atom, and $\vec{E}$ is the electric field at the position of the dipole antenna. For a single atom near an interface, $\vec{E}$ can be decomposed into a direct part $\vec{E}_0$ and a returning part $\vec{E}_{\mathrm{ret}}$. While for two interacting atoms, besides the above two parts, the electric field $\vec{E}_{21}$ caused by the interacting atom is also very important. In the following, we take the atom 1 as an example and derive its spontaneous emission rate in front of an interface.

Using $\vec{r}$ to denote the vector of a point relative to the dipole position of the atom 1, the direct electric field is

$$\begin{aligned}\vec{E}_1^{\mathrm{dir}} = \frac{d_1 k^3}{4\pi\varepsilon}\Bigg\{&(\hat{r}\times\hat{d}_1)\times\hat{r}\left(\frac{1}{kr}\right)\\ &+[\hat{d}_1 - 3\hat{r}(\hat{r}\cdot\hat{d}_1)]\\ &\times\left(\frac{\mathrm{i}}{(kr)^2} - \frac{1}{(kr)^3}\right)\Bigg\}\mathrm{e}^{\mathrm{i}(kr-\omega_0 t)},\end{aligned} \tag{2}$$

where ε is the dielectric index and k is the wave number. If the dipole antenna of the atom 1 is at a distance l_1 from the interface and is parallel to the interface, $\vec{E}_1^{\mathrm{ret}}$ can be seen as the electric field emitted from its mirror dipole $1'$ and can be written as

$$\begin{aligned}\vec{E}_1^{\mathrm{ret}} = -\frac{\vec{d}_1 k^3}{4\pi\varepsilon}R_0\Bigg(&\frac{1}{2kl_1} + \frac{\mathrm{i}}{(2kl_1)^2}\\ &-\frac{1}{(2kl_1)^3}\Bigg)\mathrm{e}^{\mathrm{i}(2kl_1-\omega_0 t)},\end{aligned} \tag{3}$$

where R_0 is the reflecting coefficient of the interface for the case of normal incidence.

The electric field $\vec{E}_{21}$caused by the atom 2 is also composed of two parts, one is the direct part and the other is the reflecting one caused by the interface. Using $\vec{r}_1$ to denote the vector from the atom 2 to the atom 1 and supposing that the dipole moments of the two atoms are parallel, the direct electric field from the atom 2 to the atom 1 can be written as

$$\begin{aligned}\vec{E}_{21}^{\mathrm{dir}} = \frac{d_2 k^3}{4\pi\varepsilon}\Bigg\{&\hat{d}_2\left[\frac{1}{kr_1} + \frac{\mathrm{i}}{(kr_1)^2} - \frac{1}{(kr_1)^3}\right]\\ &-\cos\alpha\hat{r}_1\left[\frac{1}{kr_1} + \frac{3\mathrm{i}}{(kr_1)^2} - \frac{3}{(kr_1)^3}\right]\Bigg\}\cdot\\ &\mathrm{e}^{\mathrm{i}(kr_1-\omega_0 t)},\end{aligned} \tag{4}$$

where α is the angle between $\vec{r}_1$ and the electric dipole moment $\vec{d}_1$. The reflecting electric field $\vec{E}_{21}^{\mathrm{ret}}$ can be calculated by using the dipole image method. It can be seen as the electric field emitted from its mirror image $2'$. Using $\vec{r}_2$ to denote the vector from the mirror image $2'$ to the atom 1, $\vec{E}_{21}^{\mathrm{ret}}$ can be written as

$$\begin{aligned}\vec{E}_{21}^{\mathrm{ret}} = -\frac{d_2 k^3}{4\pi\varepsilon}R\Bigg\{&\hat{d}_2\left[\frac{1}{kr_2} + \frac{\mathrm{i}}{(kr_2)^2} - \frac{1}{(kr_2)^3}\right]\\ &-\sin\theta\hat{r}_2\left[\frac{1}{kr_2} + \frac{3\mathrm{i}}{(kr_2)^2} - \frac{3}{(kr_2)^3}\right]\Bigg\}\cdot\\ &\mathrm{e}^{\mathrm{i}(kr_2-\omega_0 t)},\end{aligned} \tag{5}$$

where θ is the incident angle of the electric field emitted from atom 2; R is the reflecting coefficient for the case of oblique incidence, which depends on the refractive indices n_1 and n_2, and the incident angle θ[12].

Therefore, the whole electric field acting on the position of the atom 1 can be described as the sum of the above four parts:

$$\vec{E} = \vec{E}_1^{\mathrm{dir}} + \vec{E}_1^{\mathrm{ret}} + \vec{E}_{21}^{\mathrm{dir}} + \vec{E}_{21}^{\mathrm{ret}}. \tag{6}$$

By substituting Eq. (6) into Eq. (1), the damping rate for the first dipole near an interface can be written as

$$W^{//} = W_0 + W_1^{\mathrm{ret}} + W_{21}^{\mathrm{dir}} + W_{21}^{\mathrm{ret}}, \tag{7}$$

where $W^{//}$ denotes the spontaneous emission when the dipole moments of the two atoms are parallel. $W_0 = nW_{\mathrm{vac}}$, where W_{vac} is the spontaneous emission rate of an atom in the vacuum[7], n is the refractive index of the medium, which equals n_1 or n_2; W_1^{ret} is the spontaneous emission rate caused by the returning electric field E_1^{ret} of the atom 1:

$$\begin{aligned}W_1^{\mathrm{ret}} = -\frac{3}{2}W_0 R_0\Bigg[&\frac{\sin(2nk_0 l_1)}{2nk_0 l_1}\\ &+\frac{\cos(2nk_0 l_1)}{(2nk_0 l_1)^2} - \frac{\sin(2nk_0 l_1)}{(2nk_0 l_1)^3}\Bigg];\end{aligned} \tag{8}$$

W_{21}^{dir} is the spontaneous emission rate caused by the direct electric field E_{21}^{dir} of the atom 2:

$$\begin{aligned}W_{21}^{\mathrm{dir}} = \frac{3}{2}W_0\Bigg\{&\Bigg[\frac{\sin(nk_0 r_1)}{nk_0 r_1} + \frac{\cos(nk_0 r_1)}{(nk_0 r_1)^2}\\ &-\frac{\sin(nk_0 r_1)}{(nk_0 r_1)^3}\Bigg] - \cos^2\alpha\Bigg[\frac{\sin(nk_0 r_1)}{nk_0 r_1}\\ &+\frac{3\cos(nk_0 r_1)}{(nk_0 r_1)^2} - \frac{3\sin(nk_0 r_1)}{(nk_0 r_1)^3}\Bigg]\Bigg\};\end{aligned} \tag{9}$$

$W_{21}^{\rm ret}$ is the spontaneous emission rate induced by the returning electric field $E_{21}^{\rm ret}$ of the atom 2:

$$W_{21}^{\rm ret} = -\frac{3}{2}W_0R\Bigg\{\Bigg[\frac{\sin(nk_0r_2)}{nk_0r_2}+\frac{\cos(nk_0r_2)}{(nk_0r_2)^2} -\frac{\sin(nk_0r_2)}{(nk_0r_2)^3}\Bigg] - \sin^2\theta[\frac{\sin(nk_0r_2)}{nk_0r_2} +\frac{3\cos(nk_0r_2)}{(nk_0r_2)^2}-\frac{3\sin(nk_0r_2)}{(nk_0r_2)^3}\Bigg]\Bigg\}. \quad (10)$$

In Eqs. (8)–(10), k_0 is the wave number of the emitted light in vacuum.

If the electric dipole moment of the atom 2 is antiparallel to the atom 1, by using the same method, the spontaneous emission rate can be described as

$$W_{\rm anti}^{//} = W_0 + W_1^{\rm ret} - W_{21}^{\rm dir} - W_{21}^{\rm ret}. \quad (11)$$

For the case that the electric dipole moment of the atom 2 is perpendicular to that of the atom 1, the electric field $\vec{E}_{21}$ caused by the atom 2 can be written as

$$\vec{E}_{21}^{\rm dir\perp} = \frac{d_2k^3}{4\pi\varepsilon}\Bigg\{\hat{d}_2\Bigg[\frac{1}{kr_1}+\frac{\rm i}{(kr_1)^2}-\frac{1}{(kr_1)^3}\Bigg] -\sin\alpha\hat{r}_1\Bigg[\frac{1}{kr_1}+\frac{3\rm i}{(kr_1)^2}-\frac{3}{(kr_1)^3}\Bigg]\Bigg\}\cdot {\rm e}^{{\rm i}(kr_1-\omega_0t)}. \quad (12)$$

The reflecting electric field $\vec{E}_{21}^{\rm ret}$ can be described as

$$\vec{E}_{21}^{\rm ret\perp} = \frac{d_2k^3}{4\pi\varepsilon}R\Bigg\{\hat{d}_2\Bigg[\frac{1}{kr_2}+\frac{\rm i}{(kr_2)^2}-\frac{1}{(kr_2)^3}\Bigg] -\cos\theta\hat{r}_2\Bigg[\frac{1}{kr_2}+\frac{3\rm i}{(kr_2)^2}-\frac{3}{(kr_2)^3}\Bigg]\Bigg\}\cdot {\rm e}^{{\rm i}(kr_2-\omega_0t)}. \quad (13)$$

The spontaneous emission rate of the atom 1 in front of an interface can be written as

$$W^{\perp} = W_0 + W_1^{\rm ret} + W_{21}^{\rm dir\perp} + W_{21}^{\rm ret\perp}, \quad (14)$$

in which

$$W_{21}^{\rm dir\perp} = \frac{3}{2}W_0\Bigg\{-\sin\alpha\cos\alpha\Bigg[\frac{\sin(nk_0r_1)}{nk_0r_1} +\frac{3\cos(nk_0r_1)}{(nk_0r_1)^2}-\frac{3\sin(nk_0r_1)}{(nk_0r_1)^3}\Bigg]\Bigg\}, \quad (15)$$

$$W_{21}^{\rm ret\perp} = \frac{3}{2}W_0R\Bigg\{-\sin\theta\cos\theta\Bigg[\frac{\sin(nk_0r_2)}{nk_0r_2} +\frac{3\cos(nk_0r_2)}{(nk_0r_2)^2}-\frac{3\sin(nk_0r_2)}{(nk_0r_2)^3}\Bigg]\Bigg\}. \quad (16)$$

In the above equations, $2nk_0l_1$ is the action of the emitted photon going from the atom 1 to the interface and reflected back to the atom 1. This path forms a closed orbit. nk_0r_1 is the action of the emitted photon going directly from the atom 2 to the atom 1, and nk_0r_2 is the action of the emitted photon going from the atom 2 to the interface and reflecting back to the atom 1. These two paths form two open classical orbits.

In our calculation, we take the wavelength of the emitted photon in vacuum as $\lambda_0 = 510$ nm and $k_0 = 2\pi/\lambda_0$. The first dielectric medium is made up with the poly methyl methacrylate (PAMA) material with the refractive index $n_1 = 1.49$, and the second medium is the vacuum with $n_2 = 1.0$[7]. The two atoms are located in the first medium. Thus $n = n_1$ in the spontaneous emission rate formula. Firstly, we calculate the spontaneous emission rate of the first atom without considering the interaction of the second atom, see the dotted lines in Figs. 2–4. Then we calculate the total emission rate of this atom considering the influence of the second atom. Suppose the electric dipole moment of the first atom is parallel to the interface and the electric dipole moment of the atom 2 is parallel to that of the atom 1. Figure 2 shows the spontaneous emission rate when the horizontal distance between the two atoms is zero ($r_0 = 0$), i.e., they are located on the z axis. We keep the atom 2 fixed and move the atom 1 slowly along the z axis. The distances between the atom 2 and the interface are $2.0\lambda_0$ and $5.0\lambda_0$. From this figure, we find that compared with the case of one single atom near an interface[7], the oscillating amplitude in the spontaneous emission rate becomes increased. When the atomic-interface distance l_2 is small, the spontaneous emission rate of the atom 1 becomes weakened with the increase of the distance l_1. In each plot, when l_1 is close to l_2, the amplitude becomes increased. At $l_1 = l_2$, there appears a resonance structure. This is caused by the constructive interference of the electric field emitted from the two dipoles. Figure 3 shows the spontaneous emission rate when the horizontal distance between the two atoms is $r_0 = 0.25\lambda_0$. The oscillating structure of the spontaneous emission rate is the same as Fig. 2. But the amplitude of the oscillation becomes decreased. Figure 4 shows the spontaneous emission rate of the atom 1 when $r_0 = 4.0\lambda_0$ and $l_2 = 5.0\lambda_0$. The amplitude of the oscillation decreases greatly. The total emission rate oscillates around W_0. From this figure, we find that when $l_1 < l_2$, the influence of the atom 2 is little and the total spontaneous emission rate of atom 1 is

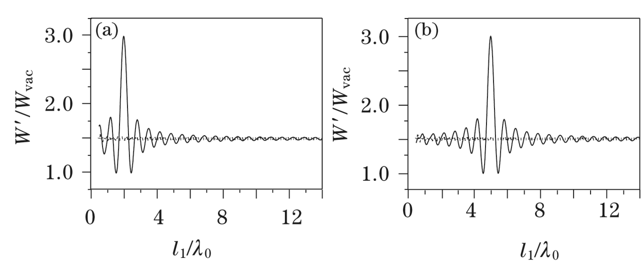

Fig. 2. Spontaneous emission rate of the atom 1 near the interface when the electrical dipole moments of the two atoms are parallel. The horizontal distance of the two atoms is $r_0 = 0$. The distances between the atom 2 and the interface are (a) $l_2 = 2.0\lambda_0$ and (b) $l_2 = 5.0\lambda_0$. The solid lines are the total emission rates of atom 1 including the influence of the atom 2 while the dotted lines are the emission rates without considering the mutual interaction of the two atoms.

nearly the same as the case that there is only one atom. But when $l_1 > l_2$, the influence of the second atom becomes significant. The main contribution to the emission rate comes from the direct part W_{21}^{dir} of the second atom.

Next, we calculate the spontaneous emission rate when the two atomic dipoles are anti-parallel, as shown in Figs. 5 and 6 for different horizontal distances between the two atoms of $r_0 = 0$ and $r_0 = 0.25\lambda_0$, respectively. From these two figures, we find that when l_1 is around l_2, the oscillation becomes strengthened. But as $l_1 = l_2$, the emission rate W_{21}^{dir} caused by the direct part of the atom 2 is of the order of $-W_0$, which cancels out the background emission rate. Therefore, the whole emission rate is the smallest. This is caused by the destructive interference of the electric field emitted from the two dipoles.

Thirdly, we calculate the spontaneous emission rate when the electric dipole moment of the atom 2 is perpendicular to that of the atom 1. Under this condition, when the horizontal distance between the two atoms is zero ($r_0 = 0$), then $\alpha = 0°$ and $\theta = 0°$. The spontaneous emission rate is the same as the case that there is only one atom. But when $r_0 \neq 0$, the influence of the atom z becomes important. Figure 7 shows the spontaneous emission rate when the horizontal distance $r_0 = 0.25\lambda_0$. The distances between the atom 2 and the interface are $2.0\lambda_0$ and $5.0\lambda_0$. Compared with the parallel case, the oscillation in the spontaneous emission rate becomes decreased. When the two atoms are far from each other, the influence of the atom 2 can be neglected. But as l_1 is close to l_2, due to the direct influence of the atom 2, the oscillation becomes strong. Around $l_1 = l_2$, there is a peak and a valley.

In order to show the correspondence between the oscillation in the spontaneous emission rate with the photon classical orbits, we make a Fourier transform (FT) to the spontaneous emission rate. We use a dimensionless variable γ to measure the system size relative to a standard one. The standard distance l_0 between the atom and the interface is taken as the wavelength of the emitted photon in vacuum, $l_0 = \lambda_0$. For simplicity, we only consider the case that the two distances between the atoms and the interface are equal. The real distances between the atoms and the interface can be written as $l_1 = l_2 = \gamma l_0$. The horizontal distance between the two atoms is $r_0 = \gamma l_0$.

We define the FT spontaneous emission rate as

$$W(S) = \int_{\gamma_1}^{\gamma_2} \frac{W - W_0}{W_0} \exp(-\mathrm{i}\gamma S)\gamma \mathrm{d}\gamma. \tag{17}$$

In our calculations, we take $\gamma_1 = 0.5$, $\gamma_2 = 16$, and $\Delta\gamma = 0.01$.

Figure 8(a) shows the Fourier transformed emission rate of atom 1 when the electric dipole moments of the two atoms are parallel to each other. There are three peaks in this plot and each peak corresponds to one photon classical orbit. In our present problem, for the excited atom 1, there is only one closed orbit of the emitted photon, which goes out from the atom, and then is reflected by the interface, and finally returns to the emitting atom 1. The action for this closed orbit is $S_1 = 2n_1k_0l_1 = 4\pi n_1\gamma$. For the standard system size $\gamma = 1$, $S_1 = 18.72$. Therefore, this closed orbit corresponds to the second peak in the Fourier transformed emission rate. Besides this closed orbit, there are two short classical orbits. The first photon classical orbit is emitted from the atom 2 and propagates directly to the atom 1. The action of this orbit is $S_2 = n_1k_0r_1 = 9.36$, which corresponds to the first peak in the Fourier

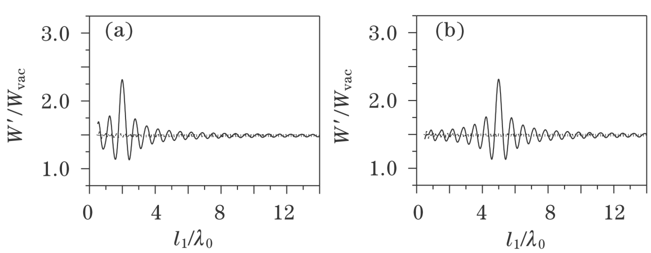

Fig. 3. The same as Fig. 2 except that the horizontal distance between the two atoms r_0 is $0.25\lambda_0$. (a) $l_2 = 2.0\lambda_0$, (b) $l_2 = 5.0\lambda_0$.

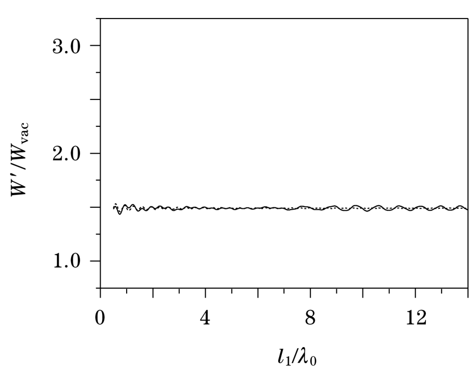

Fig. 4. The same as Fig. 2 except that the horizontal distance between the two atoms r_0 is $4.0\lambda_0$. The distance between the atom 2 and the interface is $l_2 = 5.0\lambda_0$.

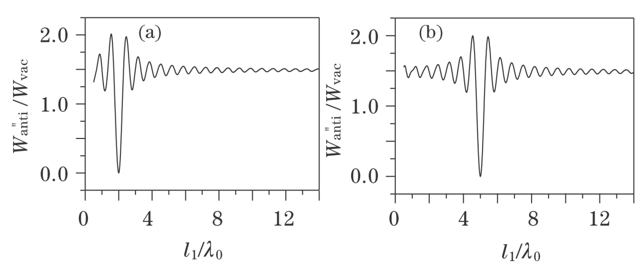

Fig. 5. Spontaneous emission rate of the atom 1 when the electric dipole moment of the atom 2 is anti-parallel to that of the atom 1. The horizontal distance between the two atoms is $r_0 = 0$. The distances between the atom 2 and the interface are (a) $l_2 = 2.0\lambda_0$ and (b) $l_2 = 5.0\lambda_0$.

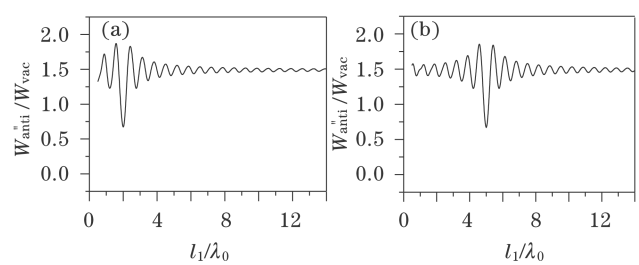

Fig. 6. The same as Fig. 5 except that the horizontal distance between the two atoms r_0 is $0.25\lambda_0$. (a) $l_2 = 2.0\lambda_0$, (b) $l_2 = 5.0\lambda_0$.

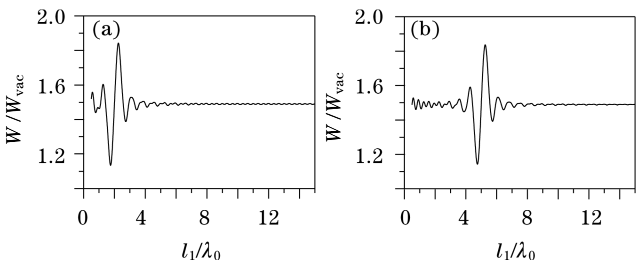

Fig. 7. Spontaneous emission rate of the atom 1 when the electric dipole moment of the atom 2 is perpendicular to that of the atom 1. The horizontal distance between the two atoms is $r_0 = 0.25\lambda_0$. The distances between the atom 2 and the interface are (a) $l_2 = 2.0\lambda_0$ and (b) $l_2 = 5.0\lambda_0$.

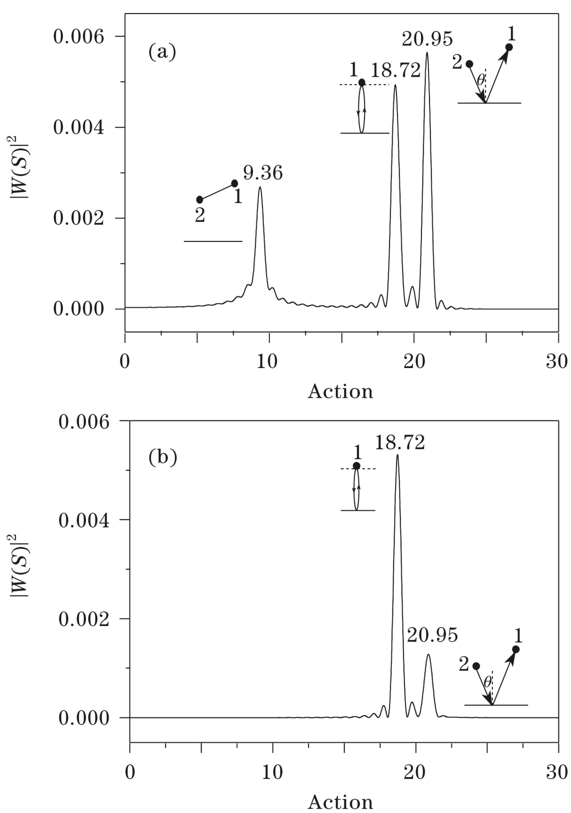

Fig. 8. Fourier transformed spontaneous emission rate of the atom 1. (a) The electric dipole moments of the two atoms are parallel to each other; (b) the electric dipole moments of the two atoms are perpendicular to each other. The corresponding photon classical orbits and the action of the orbits are shown beside each peak.

transformed emission rate. The second photon classical orbit is emitted from the atom 2 and propagates towards the interface; after reflected by the interface, it returns to the atom 1. The action of this orbit is $S_3 = n_1 k_0 r_2 = 20.95$, which corresponds to the third peak in the Fourier transformed emission rate. Figure 8(b) shows the Fourier transformed emission rate when the electric dipole moments of the two atoms are perpendicular to each other. There are two peaks in this plot. One corresponds to the photon closed orbit with the action S=18.72, and the second peak corresponds to the second photon classical orbit as described above, whose action is S=20.95. Under this condition, the direct spontaneous emission rate W_{21}^{dir}caused by the second atom is zero, therefore the influence of the first classical orbit disappears.

In conclusion, we have derived a formula for the spontaneous emission rate of two interacting atoms near a dielectric interface by using the photon closed-orbit theory and the dipole image method. Compared with the case of only one atom, the oscillation in the spontaneous emission rate of two interacting atoms becomes much more complex. The results suggest that the spontaneous emission rate of two interacting atoms not only depends on the distance between the atoms and the interface, but also depends on the orientations of the two electric dipole moments of the atoms. Besides, the horizontal separation of the two atoms also plays an important role. The study suggests that the oscillations in the spontaneous emission rate are caused by the interference between the outgoing emitted electromagnetic wave and other waves traveling along various classical orbits. We hope that our results will be useful in guiding the experimental study of the spontaneous emission rate of two atoms near an interface and in a microcavity structure[13].

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