Photons in a semibounded dielectric and the surface effect on spontaneous emission in nanostructures

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Quantization of electromagnetic field in isotropic dielectric medium occupying a half space is carried out. For a dielectric having a plane boundary with vacuum, we have constructed a complete orthonormal set of light waves propagating in a whole space. Using this set we calculated the rate of spontaneous emission of photons by electrons in quantum dots and quantum wells. We predict oscillations of photon emission rate as a function of the distance between a quantum nanostructure and the surface.

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I. INTRODUCTION

It is known[1] that the spontaneous emission rate of an atom in a cavity is enhanced compared to that for the atom placed in a free space. For a cavity formed by two mirrors, this effect was measured in Ref. 2. During the past decades much effort had been devoted to the understanding of the influence of confinement of electromagnetic field on emission and absorption of light in semiconductor nanostructure. In semiconductor quantum structures of small lateral dimensions, enhancement of the spontaneous emission was recently observed in Ref. 3. While the role of resonatorlike photon modes that exist in cavities has been analyzed in detail (see, e.g., Refs. 4–6, and references therein), less attention has been focused on important geometrical restrictions, such as the surface of a structure. Spontaneous magnetic dipole transitions in the vicinity of a perfectly reflecting mirror has been studied using the classical electromagnetic Green's function in Ref. 7. Recent experiments[8] revealed strong influence of sample-air interface on radiative dephasing time of excitons in quantum wells. The purpose of the present paper is to study a role of modification of photons, which stems from the presence of a surface, in radiative transitions in semiconductor quantum nanostructures. In order to solve this problem, we first carry out the quantization of electromagnetic field using the standard procedure.[9,10] We construct photon modes in homogeneous isotropic dielectric medium filling up the half space and contacting with a vacuum. The obtained quanta should replace the conventional bulk photons that are found by the help of the periodic boundary conditions. Then, we calculate the rate of photon emission under electron transitions in a quantum well, where quantum dots are placed at a finite distance from the surface.

II. PHOTONS IN A HALF SPACE

We assume that the boundary between the medium ($z>0$) and vacuum ($z<0$) is an infinite plane surface ($z=0$). The medium and the bounding surface have no free electric charges and external currents. In such a source-free system one can set the scalar potential equal to zero. The electric field $\mathbf{E}$ and magnetic field $\mathbf{H}$ are found completely from a vector potential $\mathbf{A}$,

$$\mathbf{E}=-\frac{1}{c}\frac{\partial\mathbf{A}}{\partial t},\quad \mathbf{H}=\mathrm{curl}\ \mathbf{A}. \tag{1}$$

Herein, we use the Coulomb gauge—div $\mathbf{A}=0$—the potential $\mathbf{A}$ is found from the wave equation

$$\nabla^2\mathbf{A}-\frac{\varepsilon}{c^2}\frac{\partial^2\mathbf{A}}{\partial t^2}=0, \tag{2}$$

where ε is the dielectric permittivity of the medium, magnetic permeability is assumed to be equal to unity. Electromagnetic field fills all the space, at the interface $z=0$ the tangential components of $\mathbf{E}$ and $\mathbf{H}$ are continuous.

We seek for the solution in the complex-valued form

$$\mathbf{A}(\mathbf{r},t)=\frac{\mathbf{A}(z)}{\sqrt{S}}e^{i(\mathbf{k}_\tau\mathbf{r}-\omega t)}, \tag{3}$$

where ω is the frequency, and $\mathbf{k}_\tau=(k_x,k_y)$ is the propagation vector. We imply periodic boundary conditions in the x-y plane over the area of normalization S. The continuous eigenvalue ω and quasidiscrete vectors $\mathbf{k}_\tau$ are the same for all modes, the different modes are specified in the terms of their z dependence. The electromagnetic field is represented by a linear superposition of the traveling monochromatic waves. We choose two types of superpositions (see Fig. 1). The first one corresponds to the field created by a wave incident at the surface $z=0$ from the half space $z<0$ (from the medium 1). We will denote the corresponding vector potential as $\mathbf{A}^{(-)}$. The second type of mode, ($\mathbf{A}^{(+)}$), corresponds to the field created by a wave incident at the interface from half space $z>0$ (from the medium 2). An arbitrary vector $\mathbf{A}$ can be written as the sum $\mathbf{A}=\mathbf{A}_s+\mathbf{A}_p$, where $\mathbf{A}_{s(p)}$ is the component perpendicular (parallel) to the plane of incidence, i.e., to the plane defined by a normal to the boundary and wave vector $\mathbf{k}=(\mathbf{k}_\tau,k_z)$. So, the modes are specified by the complex quantum number $\lambda=\{\omega,\mathbf{k}_\tau,j,\nu\}$ where $j=\pm$ stands for above described choice of an incident wave, and $\nu=s,p$ labels polarization of the incident wave.

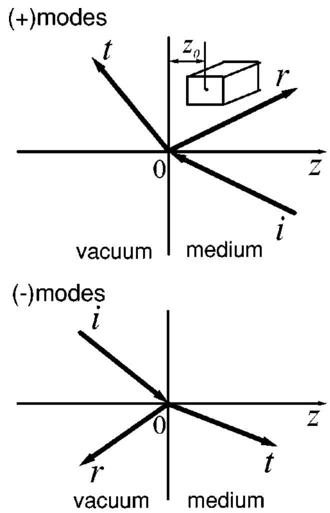

FIG. 1. Schematic diagram of photon modes. The inset shows a quantum dot placed near the boundary with a vacuum.

Using the boundary conditions, the amplitudes of reflected and transmitted waves are expressed through the amplitude $\mathbf{A}_i$ of an incident wave. In the case where the wave incident from vacuum is a s-polarized wave, one gets

$$\mathbf{A}_s^{(-)}(z)=\mathbf{A}_{is}^{(-)}(e^{ik_{1z}z}+r_s e^{-ik_{1z}z}),\quad z\leqslant 0$$
$$=\mathbf{A}_{is}^{(-)}(1+r_s)e^{ik_{2z}z},\quad z\geqslant 0, \tag{4}$$

where

$$k_{1z}=\sqrt{\frac{\omega^2}{c^2}-k_\tau^2},\quad k_{2z}=\sqrt{\frac{\omega^2}{c^2}n^2-k_\tau^2},\quad r_s=\frac{k_{1z}-k_{2z}}{k_{1z}+k_{2z}}, \tag{5}$$

$n=\sqrt{\varepsilon}$ is the index of refraction, and r_s is the amplitude of reflection.

For an incident wave parallel to the plane of incidence, $\mathbf{A}_p^{(-)}$ mode, we get

$$\mathbf{A}_{p\tau}^{(-)}(z)=\mathbf{A}_{ip\tau}^{(-)}(e^{ik_{1z}z}-r_p e^{-ik_{1z}z}),\quad z\leqslant 0$$
$$=\mathbf{A}_{ip\tau}^{(-)}\frac{k_{2z}}{n^2k_{1z}}(1+r_p)e^{ik_{2z}z},\quad z\geqslant 0, \tag{6}$$

$$A_{pz}^{(-)}(z)=A_{ipz}^{(-)}(e^{ik_{1z}z}+r_p e^{-ik_{1z}z}),\quad z\leqslant 0$$
$$=A_{ipz}^{(-)}\frac{1}{n^2}(1+r_p)e^{ik_{2z}z},\quad z\geqslant 0, \tag{7}$$

where

$$r_p=\frac{n^2k_{1z}-k_{2z}}{n^2k_{1z}+k_{2z}}. \tag{8}$$

The modes $(-)$ exist in the range $\omega\geqslant ck_\tau$.

Consider now a wave incident at the boundary $z=0$ from inside of the medium 2, that is, $\mathbf{A}^{(+)}$ modes. These modes exist in the range $\omega\geqslant ck_\tau/n$. The solutions are given by

$$\mathbf{A}_s^{(+)}(z)=\mathbf{A}_{is}^{(+)}(e^{-ik_{2z}z}-r_s e^{ik_{2z}z}),\quad z\geqslant 0$$
$$=\mathbf{A}_{is}^{(+)}(1-r_s)e^{-ik_{1z}z},\quad z\leqslant 0, \tag{9}$$

$$\mathbf{A}_{p\tau}^{(+)}(z)=\mathbf{A}_{ip\tau}^{(+)}(e^{-ik_{2z}z}+r_p e^{ik_{2z}z}),\quad z\geqslant 0$$
$$=\mathbf{A}_{ip\tau}^{(+)}\frac{n^2k_{1z}}{k_{2z}}(1-r_p)e^{-ik_{1z}z},\quad z\leqslant 0. \tag{10}$$

$$A_{pz}^{(+)}(z)=A_{ipz}^{(+)}(e^{-ik_{2z}z}-r_p e^{ik_{2z}z}),\quad z\geqslant 0$$
$$=A_{ipz}^{(+)}n^2(1-r_p)e^{-ik_{1z}z},\quad z\leqslant 0. \tag{11}$$

The case of total reflection takes place for $+$ mode when $k_\tau>\omega/c$, implying that the normal component k_{1z} is imaginary and so the waves transmitted in vacuum decay exponentially as a distance from the boundary $z=0$ increases. In this case, r_s and r_p are complex valued, $|r_s|^2=|r_p|^2=1$.

The modes are orthogonal with the piecewise constant weight function $\varepsilon(z)$. In particular, $\varepsilon(z)=1$ for $z\leqslant 0$ and $\varepsilon(z)=n^2$ for $z>0$. The modes with different wave vectors $\mathbf{k}_\tau$ are orthogonal due to periodic boundary conditions in the x-y plane. For vector potentials with the same $\mathbf{k}_\tau$, the orthonormalization condition is given by

$$\int_{-\infty}^{\infty}dz\,\varepsilon(z)\mathbf{A}_{\omega,\mathbf{k}_\tau,\nu,j}(z)\mathbf{A}^*_{\omega',\mathbf{k}_\tau,\nu',j'}(z)$$
$$=\delta_{j,j'}\delta_{\nu,\nu'}\delta(\omega-\omega'). \tag{12}$$

The amplitudes $A_{i\nu}^{(\pm)}$ are determined from Eq. (12) with $\nu,j=\nu',j'$. We use the formula

$$\int_0^{\infty}dz\,e^{\pm ikz}=\pi\delta(k)\pm i\frac{\mathcal{P}}{k}, \tag{13}$$

where k is real, and $\mathcal{P}$ stands for the principal value.

Let us consider $\mathbf{A}_s^{(-)}$ mode. Substituting Eq. (4) into Eq. (12), we found that the total principal-value part equals exactly zero for arbitrary ω and ω'. In the limit $\omega'\to\omega$ this result can be written as the equality

$$k_{1z}|\mathbf{A}_{is}^{(-)}|^2-k_{1z}|\mathbf{A}_{rs}^{(-)}|^2=k_{2z}|\mathbf{A}_{ts}^{(-)}|^2, \tag{14}$$

where $\mathbf{A}_{rs}^{(-)}$ and $\mathbf{A}_{ts}^{(-)}$ are the amplitudes of reflected and transmitted waves, correspondingly. If we now use the fact that for the physical electromagnetic field that is described by the real vector potential $\mathbf{A}_0\exp(i\mathbf{kr}-i\omega t)+\text{c.c.}$, the Poynting vector averaged over time is expressed as

$$\mathbf{S}=\frac{\omega}{2\pi}|\mathbf{A}_0|^2\mathbf{k}, \tag{15}$$

we can see that Eq. (14) reflects conservation of the normal component of the energy flow. The remaining part of Eq. (12) is

$$|\mathbf{A}_{is}^{(-)}|^2[(1+r_s r_s')\delta(k_{1z}-k_{1z}') + (1-r_s)(1-r_s')\delta(k_{2z}-k_{2z}')] = \delta(\omega-\omega'), \quad (16)$$

where $r_s' = r_s(\omega')$, $k_{jz}' = k_{jz}(\omega')$, $j=1,2$. From Eq. (16), taking into account that

$$\delta(k_{jz}-k_{jz}') = \frac{\omega k_{jz}}{k_j^2}\delta(\omega-\omega'), \quad (17)$$

we get a sought-for expression for $|\mathbf{A}_{is}^{(-)}|^2$. Normalization of the other modes is carried out analogously. It is convenient to write the vectors $\mathbf{A}_{i\nu}^{(\pm)}$ as $A_0^{(\pm)}\mathbf{e}_\nu^{(\pm)}$, where $\mathbf{e}_\nu^{(\pm)}$ is a unit vector parallel to $\mathbf{A}_{i\nu}^{(\pm)}$. Scalar amplitudes $A_0^{(\pm)}$ are given by

$$|\mathbf{A}_0^{(-)}|^2 = \frac{\omega}{2\pi c^2 k_{1z}}, \quad |\mathbf{A}_0^{(+)}|^2 = \frac{\omega}{2\pi c^2 k_{2z}}. \quad (18)$$

The vectors $\mathbf{e}_\nu^{(\pm)}$ can be taken in the following form:

$$\mathbf{e}_s = \frac{1}{k_\tau}(k_y, -k_x, 0),$$

$$\mathbf{e}_p^{(-)} = \frac{1}{k_1 k_\tau}(k_x k_{1z}, k_y k_{1z}, -k_\tau^2),$$

$$\mathbf{e}_p^{(+)} = -\frac{1}{k_2 k_\tau}(k_x k_{2z}, k_y k_{2z}, k_\tau^2), \quad (19)$$

where $k_1 = \omega/c$ and $k_2 = \omega n/c$; vector $\mathbf{e}_s$ is the same for both $(\pm)$ modes.

Obviously, the solutions that differ by polarization of incident waves are orthogonal to each other due to orthogonality $(\mathbf{e}_\nu^{(\pm)}\mathbf{e}_{\nu'}^{(\pm)}) = \delta_{\nu,\nu'}$. Orthogonality of solutions $\mathbf{A}^{(+)}$ and $\mathbf{A}^{(-)}$ of the same polarization is proved by direct calculations of integral (12).

Thus we have constructed a complete set of orthonormalized vectors $\mathbf{A}_\lambda(\mathbf{r},t)$ by solving the eigenvalue problem of electrodynamics equation (2) in the whole space. Photons are introduced in usual way using the second quantization formalism. In this representation, the vector potential has the form

$$\mathbf{A}(\mathbf{r},t) = \sum_\lambda \sqrt{\frac{2\pi c^2\hbar}{\omega S}}[a_\lambda \mathbf{A}_\lambda(z)e^{i(\mathbf{k}_\tau \mathbf{r}-\omega t)} + \text{c.c.}], \quad (20)$$

where a_λ is a photon annihilation operator at time $t=0$ and Σ_λ denotes the summation over $\mathbf{k}_\tau, j, \nu$ and the integration over ω. The interaction of electrons with a weak electromagnetic field is described by

$$H_{int} = -\frac{i\hbar e}{m^* c}(\mathbf{A}\nabla), \quad (21)$$

where e and m^* are charge and effective mass of electron.

III. NEAR-SURFACE EMISSION OF LIGHT

We first focus on electron transitions between discrete energy levels. Let us consider a rectangular quantum dot (QD) of the dimensions $L_x \times L_y \times L_z$ placed at a finite distance $z_0 \geqslant L_z/2$ from the surface $z=0$. The potential barrier outside of the dot is assumed to be infinite. With this assumption we can separate the electron motion in all three spatial directions. In this model, the proximity of the boundary $z=0$ does not disturb the electron states, and the effective-mass electron wave function and the energy are given by

$$\psi_{n_x n_y n_z} = \left(\frac{8}{L_x L_y L_z}\right)^{3/2} \sin\frac{n_x \pi x}{L_x} \sin\frac{n_y \pi y}{L_y} \sin\frac{n_z \pi(z-z_0+L_z/2)}{L_z}, \quad (22)$$

$$E_{n_x n_y n_z} = \frac{\pi^2\hbar^2}{2m^*}\left(\frac{n_x^2}{L_x^2}+\frac{n_y^2}{L_y^2}+\frac{n_z^2}{L_z^2}\right), \quad (23)$$

where n_x, n_y, n_z are positive integers.

Let us consider transitions between the first excited state and the ground state due to interaction of electron with above obtained full set of photon modes. For the ground state (state 1), the set $(n_x n_y n_z)$ equals (111). For the first excited state, the quantum numbers $(n_x n_y n_z)$ equal (211), (121), or (112) depending on a relationship between the sizes L_x, L_y, and L_z. We will denote these states as $2x$, $2y$ or $2z$, respectively. The rate of photon emission associated with the single-electron transition is calculated in the first order of perturbation using the Fermi golden rule. We get

$$\nu_\alpha = \frac{2\pi}{\hbar}\sum_\lambda |M_\alpha(\lambda)|^2[N(\hbar\omega/k_B T)+1]\delta(E_{2\alpha}-E_1-\hbar\omega). \quad (24)$$

Here $M_\alpha(\lambda)$ is the matrix element of the transition, $\alpha = (x,y,z)$, the summation runs over all quantum numbers of photons, N is the Planck distribution function. Energy-conservation law in Eq. (24) shows that the emitted photons have large wavelengths compared to the QD size. So in the matrix elements the functions $\mathbf{A}_\lambda(z)$ can be replaced with $\mathbf{A}_\lambda(z_0)$ (dipole approximation). For total contribution of all modes with fixed ω and $\mathbf{k}_\tau$ in the range $\omega \geqslant c k_\tau$ we get

$$|M_x|^2 = M_{0x}^2[e_{sx}^2 + e_{px}^{(+)2} + (e_{px}^{(+)2} r_p - e_{sx}^2 r_s)\cos Z], \quad (25)$$

$$|M_z|^2 = M_{0z}^2 e_{pz}^{(+)2}[1 - r_p \cos Z], \quad (26)$$

where

$$M_{0\alpha}^2=\frac{2^7\hbar^3e^2}{9L_\alpha^2Sm^2c^2k_{2z}},\quad Z=2k_{2z}z_0. \tag{27}$$

In the range $ck_\tau/n\leqslant\omega\leqslant ck_\tau$, where the total reflection occurs, the only $(+)$ modes are emitted. We get

$$|M_x|^2=M_{0x}^2(e_{sx}^2+e_{px}^{(+)2})+\frac{M_{0x}^2}{2}[\cos Z(e_{px}^{(+)2}P^+-e_{sx}^2S^+)$$
$$+i\sin Z(e_{px}^{(+)2}P^--e_{sx}^2S^-)], \tag{28}$$

$$|M_z|^2=M_{0z}^2e_{pz}^{(+)2}\left(1-\frac{\cos Z}{2}P^+-\frac{i\sin Z}{2}P^-\right), \tag{29}$$

where

$$P^\pm=r_p\pm r_p^*,\quad S^\pm=r_s\pm r_s^*.$$

For all ω, $|M_y(\omega)|^2$ is obtained from the corresponding $|M_x(\omega)|^2$ by the change $x\rightarrow y$.

We see that the matrix elements depend on a distance between QD and a crystal surface. On comparing M_x and M_z we see that this dependence appears to be different for the electron transition from the states of different symmetry. For QD in the bulk ($z_0\rightarrow\infty$), we can omit fast oscillating terms in the above obtained matrix elements. To calculate the bulk emission rate ν_α^b we replace the summation over $\mathbf{k}_\tau$ in Eq. (24) by the integration in a polar coordinate system. Upon integration we get

$$\nu_\alpha^b=\frac{2^8}{27L_\alpha^2}\frac{\hbar\omega e^2n}{m^2c^3}(N+1), \tag{30}$$

where now $\omega=(E_{2\alpha}-E_1)/\hbar$.

The rate given by Eq. (30) coincides with the rate of emission of conventional photons that are introduced by the help of the periodic boundary conditions. In this representation

$$\mathbf{A}(\mathbf{r},t)=\sum_{\nu,\mathbf{k}}\sqrt{\frac{2\pi c^2\hbar}{\omega n^2V}}[a_\mathbf{k}\mathbf{e}_\nu(\mathbf{k})e^{i(\mathbf{kr}-\omega t)}+\text{c.c.}], \tag{31}$$

where V is the normalization volume, $\mathbf{k}=\{\mathbf{k}_\tau,k_{2z}\}$, the unit vectors of polarization may be chosen, e.g., as $\mathbf{e}_s$ and $-\mathbf{e}_p^{(+)}$. In Eq. (24) now $\lambda=\{\nu,\mathbf{k}\}$. For photons of the both polarizations, we get

$$|M_\alpha|^2=\frac{2^7\pi\hbar^3e^2}{9L_\alpha^2Vm^2nck}\left(1-\frac{k_\alpha^2}{k^2}\right). \tag{32}$$

Substituting Eq. (32) into Eq. (24) and calculating the sum over $\mathbf{k}$ by the integration in a spherical coordinate system, we obtain result of Eq. (30).

To illustrate an influence of the crystal surface, we have calculated the rates of photon emission for interlevel electron transitions in QDs. The calculations were carried out for the transitions to the ground state from a nondegenerate state (QD with $L_x\neq L_y\neq L_z$), from twofold-degenerate first excited state for a parallelepiped-shaped QD ($L_x=L_y\neq L_z$), and from the threefold-degenerate first excited state (a cubical QD of dimension L^3). The emission rates normalized to the corresponding bulk rates are shown in Fig. 2. For GaAs QD with $L_z=100$ Å and $m=0.067m_0$, the distance $z_0>L_z/2$ corresponds to the dimensionless coordinate $2\omega z_0/c>6\times10^{-3}$. We see that influence of the surface on emission of light has long-distance behavior. Comparison of the curves 1 and 3 shows that the surface effect, as well as the magnetic dipole transitions,[7] depends on the symmetry of the electron excited states, i.e., on the orientation of QD with respect to the crystal surface.

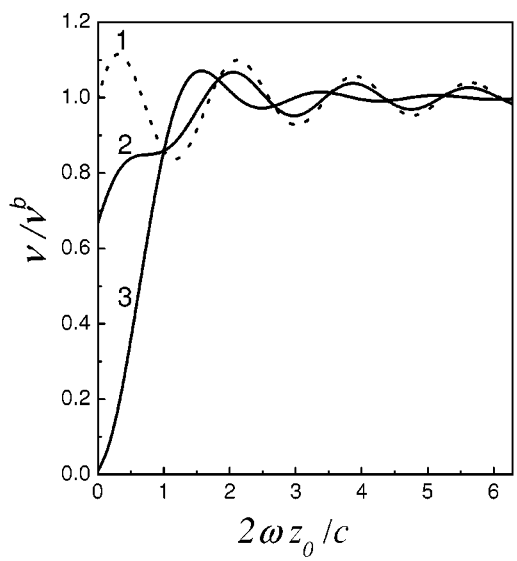

FIG. 2. Ratio of photon emission rate to the bulk values ($z_0\rightarrow\infty$) versus the distance z_0 between quantum dot and the surface ($\varepsilon=13$). Curve 1 represents results for the transition $2x\rightarrow1$ and also for the transitions from twofold-degenerate first excited state of a parallelepiped-shaped QD ($L_x=L_y\neq L_z$). Curve 2—the transitions from threefold-degenerate first excited state of cubic QD. Curve 3—the transition $2z\rightarrow1$.

In this paper, the details of near-surface spontaneous emission of light have been presented for QD's. The obtained results reflect also the peculiarities of the light emission in nanostructures of other types, which provide electron confinement in the direction normal to the crystal-air interface. To demonstrate it, we calculated the rate of photon emission for transitions between electron subbands in an infinite deep rectangular quantum well. In Fig. 2, curve 3 also exhibits the surface influence on the electron radiative transitions from the second subband to the ground subband (z_0 is the distance between the center of the quantum well and the surface).

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