Quantization of electromagnetic fields in cavities and spontaneous emission

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The normal modes of electromagnetic fields satisfying three different boundary conditions are constructed systematically. For each of these cases, cavity quantization of the electromagnetic fields is carried out explicitly. It is possible, using normal modes, to perform a unitary transformation of the minimal-coupling Hamiltonian of an atom inside the cavity to a multipolar form. It is shown that the boundary condition plays an important role in the transition rates for the spontaneous emission by the atom. Some explicit calculations are carried out for several different boundary conditions. These results become consistent with those for free space in the large-box limit.

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I. INTRODUCTION

Properties of atoms interacting with electromagnetic fields in cavities have long been the subject of intense investigation [1]. Since the fields in cavities differ from that found in free space, the rate of spontaneous emission by an atom depends on the cavity boundary conditions. The emission rate is increased if the atom is surrounded by a cavity tuned to the transition frequency and is inhibited if the cavity dimensions do not allow the radiation wavelength. Several experiments have verified the enhancement and inhibition of the emission by an atom in a cavity at various frequencies [2–8]. In addition to spontaneous emission, there have been many observable effects of boundary conditions, thanks to recent advances of constructing small cavities. These effects include vacuum Rabi splitting, micromaser and microlaser, quantum collapse and revival, Casimir effect, and atomic-energy shift [1].

In this paper, we shall be primarily concerned with the "infinite-wall" boundary conditions for a single cavity. However, there are also lattices of wells with "finite walls" which produce observable effects, namely, photonic materials (crystals) [9] and quantum dots [10]. They have also attracted considerable attention. Photon waves in a three-dimensionally periodic dielectric structure should be described by band theory, which is analogous to electron waves in a crystal. Many applications of photonic bands are now being pursued in metallic, dielectric, and acoustic structures [9]. The lattice of quantum dots is, in effect, a crystalline layer made of artificial atoms (dots) whose energy levels can be controlled precisely [10]. They will also show band gaps because there are arrays of dots in the lattice. The lattice problem is also concerned with the effect of boundary conditions on electromagnetic fields.

Many authors have studied, classically, quantum mechanically, or field theoretically, spontaneous emission by atoms surrounded by electromagnetic environment [11–22], especially the effect of a flat mirror or two parallel flat mirrors [11–21]. In 1965, Marshall [11] studied the radiation rate of a classical dipole placed in the random zero-point field between two conducting plates and got some reasonable results. Kuhn [12] also considered a similar problem of a molecule using a model based on classical linear harmonic oscillators. Such a classical model has been shown to provide a good quantitative explanation of experimental data. The effect of mirrors has also been obtained by the image method, in which the mirror cavity is replaced by an infinite string of virtual images [13]. Several ideas has been presented to study various surface effects on atoms in different situations [14].

It is quite natural to use normal modes to study cavity quantum electrodynamics, while we usually use plane waves in electrodynamics in free space. In this normal-mode approach, the electromagnetic field is expanded in appropriate mode functions satisfying the boundary conditions imposed by the mirrors [15–21,23–25], and the quantization procedure has been outlined by Milonni [15], by Power and Thirunamachandran [16], and by Glauber and Lewenstein [24]. The quantization of evanescent field in a half-space filled with a homogenous dielectric has been carried out by Carniglia and Mandel, and by others [25].

To analyze electrodynamic level shifts and the natural width of an excited atomic state between parallel mirrors, Barton [17] has presented one solution to the normal modes satisfying the boundary conditions at conducting surfaces. He used the plane waves in two other directions. Using these modes, Philpott [18] has obtained the emission rate for a molecule with different orientations between two mirrors, which reduces to the result of Barton after the average is taken. The effect of the half-infinite dielectric was studied by Arnoldus and George [19] using the plane wave including the Fresnel reflection coefficient. Recently, Loudon and his co-workers [20] discussed spontaneous emission in the vicinity of a dielectric surface and in a dielectric slab in terms of complete spatial

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modes [25]. Moreover, Cetto and Peña [21] have shown that stochastic electrodynamics can also give a satisfactory account of these effects. By introducing the random zero-point field and by using mode functions similar to those used by Barton, they arrived at a similar result.

However, unfortunately, the QED quantization procedure for free space is not applicable to electromagnetic fields in cavities which are affected by boundary conditions. In the case of cavity electrodynamics, we have to obtain first independent mode functions consistent with Maxwell's equations and boundary conditions. As in the case of free electromagnetic waves, there are two independent mode functions corresponding to each wave number. We then apply the quantization procedure on these mode functions. This procedure heavily depends on the geometry of the cavity. Thus the quantization procedures might appear quite different depending on the shape of the cavity. However, they are based on the same physical principle.

In this paper, we carry out the quantization and the calculation of the emission rate for three different forms of cavities. First, we construct, using an orthogonal matrix, the normal modes satisfying various boundary conditions. The first cavity we consider in this paper corresponds to the cavity considered earlier by Barton. The other two cavities have not yet been studied. Second, we derive the multipolar interaction between an atom and the fields in cavities by a unitary transformation. From this, it is possible to derive the dipole interaction. It is possible to extract the transverse part of a physical quantity by making use of the normal modes, which plays an important role in the multipolar expansion. Third, the field-theoretic methods developed as above are applied to spontaneous emission by an atom. By using the first cavity, we will reproduce some of the results which have been obtained earlier by several authors [16,18–21].

In addition, we consider in this paper the effect of additional mirrors on the emission. In the first case, the cavity is closed only along one direction. We can definitely consider the case where the cavity is closed along two different directions as well as all three directions. In order to accommodate all three cases, we develop a quantization procedure with three-dimensional boundary conditions. This quantization procedure allows us to calculate the emission rates with respective boundary conditions. Indeed, these calculated rates can be compared with experimentally observed values.

In Sec. II we obtain the normal modes in a systematic way and carry out quantization of fields for three different cavities. The multipolar expansion of the interaction between an atom and the fields is derived in Sec. III. The spontaneous emission by an atom is treated in Sec. IV. Section V is devoted to concluding remarks.

II. QUANTIZATION OF FIELDS IN CAVITIES

Let us first consider Maxwell's equations for the electric field $\mathbf{E}$ and the magnetic field $\mathbf{B}$ in free space, which are given by

$$\begin{aligned} \operatorname{div}\mathbf{B} &= \mathbf{0}, \qquad \operatorname{curl}\mathbf{E} + \frac{\partial}{\partial t}\mathbf{B} = \mathbf{0}, \\ \operatorname{div}\mathbf{E} &= \mathbf{0}, \qquad \operatorname{curl}\mathbf{B} - \varepsilon_0\mu_0\frac{\partial}{\partial t}\mathbf{E} = \mathbf{0}, \end{aligned} \tag{2.1}$$

where ε_0 and μ_0 are, respectively, the electric permittivity and magnetic permeability for free space ($\varepsilon_0\mu_0 = 1/c^2$). It follows from Eq. (2.1) that

$$\left(\triangle - \varepsilon_0\mu_0\frac{\partial^2}{\partial t^2}\right)\mathbf{E} = \mathbf{0}, \qquad \left(\triangle - \varepsilon_0\mu_0\frac{\partial^2}{\partial t^2}\right)\mathbf{B} = \mathbf{0}, \tag{2.2}$$

where $\triangle$ is the Laplacian operator. We also have, from Eq. (2.1),

$$\begin{aligned} \triangle_T E_x &= \frac{\partial^2}{\partial y\partial t}B_z - \frac{\partial^2}{\partial x\partial z}E_z, \\ \triangle_T E_y &= -\frac{\partial^2}{\partial x\partial t}B_z - \frac{\partial^2}{\partial y\partial z}E_z, \\ \triangle_T B_x &= -\varepsilon_0\mu_0\frac{\partial^2}{\partial y\partial t}E_z - \frac{\partial^2}{\partial x\partial z}B_z, \\ \triangle_T B_y &= \varepsilon_0\mu_0\frac{\partial^2}{\partial x\partial t}E_z - \frac{\partial^2}{\partial y\partial z}B_z, \end{aligned} \tag{2.3}$$

where $\triangle_T = \triangle - \partial^2/\partial z^2$ (see, for example, Chap. 12 of [26]). Assuming that

$$\frac{\partial^2}{\partial t^2}E_i \propto E_i, \qquad \frac{\partial^2}{\partial z^2}E_i \propto E_i, \tag{2.4}$$

and the similar equations for B_i $(i = x, y, z)$, we then get

$$\begin{aligned} \triangle_T E_i &= \left(\varepsilon_0\mu_0\frac{\partial^2}{\partial t^2} - \frac{\partial^2}{\partial z^2}\right)E_i \propto E_i, \\ \triangle_T B_i &= \left(\varepsilon_0\mu_0\frac{\partial^2}{\partial t^2} - \frac{\partial^2}{\partial z^2}\right)B \propto B_i \end{aligned} \tag{2.5}$$

$(i = x, y)$. Therefore, by Eq. (2.3), the x and y components of the fields are described in terms of their z components; it is sufficient to determine E_z and B_z using Eq. (2.1) with boundary conditions.

A. Case 1

The cavity we treat first is enclosed by rectangular walls having sides L_1, L_2, and L_3 in the x, y, and z directions, respectively, i.e., $0 < x < L_1$, $0 < y < L_2$, and $0 < z < L_3$. The walls in the z direction are assumed to be perfectly conducting and the tangential component of the electric field $\mathbf{E}|_{\text{tan}}$ and the normal component of the magnetic field $\mathbf{B}|_{\text{norm}}$ must accordingly vanish at the cavity boundaries in this direction. To remove the effect of the walls in the x and y directions, we assume $L_1, L_2 \gg L_3$ and take the periodic boundary conditions in these directions; finally we must take the limit $L_1, L_2 \to \infty$.

Set $E_z \sim X(x)Y(y)Z(z)$ and $B_z \sim \bar{X}(x)\bar{Y}(y)\bar{Z}(z)$ as their spacial components. Equation (2.3) and the boundary condition in the z direction give $E_x = E_y = B_z = 0$ at $z = 0, L_3$, which leads to $dZ/dz|_{z=0,L_3} = \bar{Z}|_{z=0,L_3} = 0$. The periodic boundary conditions in the x and y directions determine the other functions X, Y, $\bar{X}$, and $\bar{Y}$. We thus have

$$Z(z) = \cos k_{\ell_z} z, \quad \bar{Z}(z) = \sin k_{\ell_z} z \tag{2.6}$$

and

$$X(x), \bar{X}(x) = e^{ik_{\ell_x} x}, \quad Y(y), \bar{Y}(y) = e^{ik_{\ell_y} y}, \tag{2.7}$$

where

$$\mathbf{k}_\ell = (k_{\ell_x}, k_{\ell_y}, k_{\ell_z}) = \left(\frac{2\pi\ell_1}{L_1}, \frac{2\pi\ell_2}{L_2}, \frac{\pi\ell_3}{L_3} \right), \tag{2.8}$$

with $\ell_1, \ell_2 = 0, \pm1, \pm2, \ldots$ and $\ell_3 = 0, 1, 2, \ldots$. By Eq. (2.3), we can obtain the components of the fields:

$$\begin{aligned} &E_x, E_y, B_z \sim e^{ik_{\ell_x} x} e^{ik_{\ell_y} y} \sin k_{\ell_z} z, \\ &B_x, B_y, E_z \sim e^{ik_{\ell_x} x} e^{ik_{\ell_y} y} \cos k_{\ell_z} z. \end{aligned} \tag{2.9}$$

Let us define the normal modes to expand the electric field:

$$\begin{aligned} u_{\ell,x} = u_{\ell,y} &= \sqrt{\frac{2}{V}}\, e^{ik_{\ell_x} x} e^{ik_{\ell_y} y}\, i \sin k_{\ell_z} z, \\ u_{\ell,z} &= \sqrt{\frac{2}{V}}\, e^{ik_{\ell_x} x} e^{ik_{\ell_y} y} \cos k_{\ell_z} z, \end{aligned} \tag{2.10}$$

where V is the volume of the cavity ($V = L_1 L_2 L_3$) and $\ell = (\ell_1, \ell_2, \ell_3)$. When $\ell_3 = 0$, the mode $u_{\ell,z}$ is changed to

$$u_{\ell,z}|_{\ell_3=0} = \sqrt{\frac{1}{V}}\, e^{ik_{\ell_x} x}\, e^{ik_{\ell_y} y}. \tag{2.11}$$

Then the above normal modes (2.10) and (2.11) are orthonormal. The electric field can be expanded as follows:

$$\mathbf{E}(\mathbf{r}, t) = i \sum_{\ell,i} \sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}}\, \mathbf{e}_i [b_{\ell,i}(t) u_{\ell,i}(\mathbf{r}) - b^\dagger_{\ell,i}(t) u^*_{\ell,i}(\mathbf{r})], \tag{2.12}$$

where $\omega_\ell = c|\mathbf{k}_\ell|$ and $\mathbf{e}_i$ is the unit vector in the ith direction ($i = x, y$, or z). The coefficient $\sqrt{\hbar\omega_\ell/2\varepsilon_0}$ is introduced for normalization purposes.

The transversality condition for the coefficient functions (operators in quantum mechanics) $b_{\ell,i}$ and $b^\dagger_{\ell,i}$

$$\begin{aligned} &\mathbf{k}_\ell \cdot (b_{\ell,x}\mathbf{e}_x + b_{\ell,y}\mathbf{e}_y + b_{\ell,z}\mathbf{e}_z) \\ &\qquad = \mathbf{k}_\ell \cdot (b^\dagger_{\ell,x}\mathbf{e}_x + b^\dagger_{\ell,y}\mathbf{e}_y + b^\dagger_{\ell,z}\mathbf{e}_z) = 0 \end{aligned} \tag{2.13}$$

is derived from $\mathrm{div}\mathbf{E} = 0$ and the relations $\partial u_{\ell,x}/\partial x, \partial u_{\ell,y}/\partial y, \partial u_{\ell,z}/\partial z \propto u_{\ell,x}$. It is convenient to define a unit vector in the direction $\mathbf{k}_\ell$ as $\bar{\mathbf{e}}_{\ell 3}$ and unit vectors $\bar{\mathbf{e}}_{\ell\sigma}$ ($\sigma = 1, 2$) being perpendicular to $\bar{\mathbf{e}}_{\ell 3}$; for example, we take $\bar{\mathbf{e}}_{\ell\sigma} = \sum_i O^{(\ell)}_{\sigma i} \mathbf{e}_i$ ($\sigma = 1, 2, 3; i = x, y, z$), where $O^{(\ell)}$ is an orthogonal matrix

$$O^{(\ell)} = \begin{pmatrix} \cos\theta_\ell \cos\phi_\ell & \cos\theta_\ell \sin\phi_\ell & -\sin\theta_\ell \\ -\sin\phi_\ell & \cos\phi_\ell & 0 \\ \sin\theta_\ell \cos\phi_\ell & \sin\theta_\ell \sin\phi_\ell & \cos\theta_\ell \end{pmatrix}, \tag{2.14}$$

with $\mathbf{k}_\ell = |\mathbf{k}_\ell|(\sin\theta_\ell \cos\phi_\ell, \sin\theta_\ell \sin\phi_\ell, \cos\theta_\ell)$. Using the new functions (operators) defined by

$$a_{\ell\sigma} = \sum_i O^{(\ell)}_{\sigma i} b_{\ell,i}, \quad a^\dagger_{\ell\sigma} = \sum_i O^{(\ell)}_{\sigma i} b^\dagger_{\ell,i}, \tag{2.15}$$

the electric field becomes

$$\mathbf{E} = i \sum_{\ell\sigma} \sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}} \left(a_{\ell\sigma} \bar{\mathbf{u}}_{\ell\sigma} - a^\dagger_{\ell\sigma} \bar{\mathbf{u}}^*_{\ell\sigma} \right), \tag{2.16}$$

where

$$\bar{\mathbf{u}}_{\ell\sigma} = \sum_i \mathbf{e}_i O^{(\ell)}_{\sigma i} u_{\ell,i}. \tag{2.17}$$

Note that $\bar{\mathbf{u}}_{\bar{\ell}\sigma} = \bar{\mathbf{u}}^*_{\ell\sigma}$, where $\bar{\ell} = (-\ell_1, -\ell_2, \ell_3)$ if $\ell = (\ell_1, \ell_2, \ell_3)$; this follows from the fact that the orthogonal matrix $O^{(\bar{\ell})}$ is derived from $O^{(\ell)}$ by changing ϕ_ℓ to $\phi_\ell + \pi$. It should also be noted that the sum over σ in Eq. (2.16) denotes $\sigma = 1, 2$ because the transversality condition (2.13) gives $a_{\ell 3} = 0$. Substituting Eq. (2.16) into the wave equation (2.2), we then have, for each $\ell\sigma$,

$$\triangle \bar{\mathbf{u}}_{\ell\sigma} + \mathbf{k}^2_\ell \bar{\mathbf{u}}_{\ell\sigma} = \mathbf{0}, \quad \frac{d^2}{dt^2} a_{\ell\sigma} + \omega^2_\ell\, a_{\ell\sigma} = 0. \tag{2.18}$$

Since $O^{(\ell)}$ is an orthogonal matrix, it follows from Eqs. (2.10) and (2.11) that the new modes (2.17) have the orthonormal property

$$\begin{aligned} &\int_c dv\, \bar{\mathbf{u}}^*_{\ell\sigma} \cdot \bar{\mathbf{u}}_{\ell'\sigma'} = \delta_{\ell\ell'} \delta_{\sigma\sigma'}, \\ &\int_c dv\, \bar{\mathbf{u}}_{\ell\sigma} \cdot \bar{\mathbf{u}}_{\ell'\sigma'} = \delta_{\ell\bar{\ell}'} \delta_{\sigma\sigma'}, \end{aligned} \tag{2.19}$$

where $\int_c dv = \int_{\text{cavity}} dxdydz$. They also satisfy

$$\mathrm{div}\, \bar{\mathbf{u}}_{\ell\sigma} = 0, \tag{2.20}$$

because $\sum_i k_{\ell_i} O^{(\ell)}_{\sigma i} = |\mathbf{k}_\ell| \delta_{\sigma 3}$ ($\sigma = 1, 2$), and satisfy

$$\bar{\mathbf{u}}_{\ell\sigma}|_{\tan} = \mathbf{0}, \qquad \mathrm{curl}\, \bar{\mathbf{u}}_{\ell\sigma}|_{\text{norm}} = \mathbf{0} \tag{2.21}$$

on the walls in the z direction. It should be noted that we can use only the normal modes $\bar{\mathbf{u}}_{\ell\sigma}$ to obtain creation and annihilation operators as expansion coefficients. The old modes $\mathbf{u}_{\ell,i}$ are merely means to obtain the real ones $\bar{\mathbf{u}}_{\ell\sigma}$.

Taking into account Eqs. (2.1), (2.2), and (2.16), we can derive the magnetic fields expanded in terms of the new modes:

$$\mathbf{B} = \sum_{\ell\sigma} \sqrt{\frac{\hbar}{2\varepsilon_0 \omega_\ell}} \left(a_{\ell\sigma} \mathrm{curl}\, \bar{\mathbf{u}}_{\ell\sigma} + a^\dagger_{\ell\sigma} \mathrm{curl}\, \bar{\mathbf{u}}^*_{\ell\sigma} \right). \tag{2.22}$$

In Eq. (2.22) we have used the relation $da_{\ell\sigma}/dt = -i\omega_\ell a_{\ell\sigma}$ [see Eq. (2.18)], which makes $a_{\ell\sigma}$ an annihilation operator. If we use another possibility $da_{\ell\sigma}/dt = i\omega_\ell a_{\ell\sigma}$, then $a_{\ell\sigma}$ becomes a creation operator. We can also obtain the expression (2.22) for the magnetic field directly from Eq. (2.9) using $db_{\ell,i}/dt = -i\omega_\ell b_{\ell,i}$. It is easy to see that the fields (2.16) and (2.22) indeed satisfy the boundary conditions. By Eq. (2.22), we get the vector potential

$$\mathbf{A} = \sum_{\ell\sigma} \sqrt{\frac{\hbar}{2\varepsilon_0 \omega_\ell}} \left(a_{\ell\sigma} \bar{\mathbf{u}}_{\ell\sigma} + a^\dagger_{\ell\sigma} \bar{\mathbf{u}}^*_{\ell\sigma} \right) - \mathrm{grad}\, f, \quad (2.23)$$

where f is a function. It follows from Eqs. (2.16) and (2.23) that $\mathbf{E} = -\partial \mathbf{A}/\partial t - \mathrm{grad}\,(\partial f/\partial t)$, so that we have $\mathrm{grad}\, \phi = \mathrm{grad}\,(\partial f/\partial t)$, where ϕ is a scalar potential. Throughout this paper, we shall use the Coulomb gauge by ignoring the second term of Eq. (2.23).

Using the equality

$$\int_c dv \, \mathrm{curl}\, \bar{\mathbf{u}}^*_{\ell\sigma} \cdot \mathrm{curl}\, \bar{\mathbf{u}}_{\ell'\sigma'} = \mathbf{k}_\ell^2 \int_c dv \bar{\mathbf{u}}^*_{\ell\sigma} \cdot \bar{\mathbf{u}}_{\ell'\sigma'}, \quad (2.24)$$

and Eqs. (2.16) and (2.22), the electromagnetic Hamiltonian is derived as follows:

$$\begin{aligned} H_R &= \sum_{\ell\sigma} \hbar\omega_\ell \frac{1}{2} \left(a^\dagger_{\ell\sigma} a_{\ell\sigma} + a_{\ell\sigma} a^\dagger_{\ell\sigma} \right) \\ &= \sum_{\ell\sigma} \hbar\omega_\ell \left(a^\dagger_{\ell\sigma} a_{\ell\sigma} + \frac{1}{2} \right), \end{aligned} \quad (2.25)$$

where we have introduced the canonical quantization for $a_{\ell\sigma}$ and $a^\dagger_{\ell\sigma}$:

$$[a_{\ell\sigma}, a^\dagger_{\ell'\sigma'}] = \delta_{\ell\ell'} \delta_{\sigma\sigma'}. \quad (2.26)$$

From Eqs. (2.25) and (2.26), we get $a_{\ell\sigma}(t) = a_{\ell\sigma}(0) \exp(-i\omega_\ell t)$.

B. Case 2

Next, we consider the second cavity, which is also enclosed by rectangular walls having sides L_1, L_2, and L_3 in the x, y, and z directions, respectively: $0 < x < L_1$, $0 < y < L_2$, and $0 < z < L_3$. We assume that the walls in the y and z directions are perfectly conducting, while the periodic boundary condition will be taken in the x direction.

As in the first case, the boundary condition in the y direction gives $Y|_{y=0,L_2} = d\bar{Y}/dy|_{y=0,L_2} = 0$, while the condition in the z direction gives $dZ/dz|_{z=0,L_3} = \bar{Z}|_{z=0,L_3} = 0$. Thus, by Eq. (2.3), the electric fields satisfying the boundaries can be derived as follows:

$$\begin{aligned} E_x &\sim e^{ik_{\ell_x} x} \sin k_{\ell_y} y \, \sin k_{\ell_z} z, \\ E_y &\sim e^{ik_{\ell_x} x} \cos k_{\ell_y} y \, \sin k_{\ell_z} z, \\ E_z &\sim e^{ik_{\ell_x} x} \sin k_{\ell_y} y \, \cos k_{\ell_z} z, \end{aligned} \quad (2.27)$$

where

$$\mathbf{k}_\ell = (k_{\ell_x}, k_{\ell_y}, k_{\ell_z}) = \left(\frac{2\pi\ell_1}{L_1}, \frac{\pi\ell_2}{L_2}, \frac{\pi\ell_3}{L_3} \right) \quad (2.28)$$

with $\ell_1 = 0, \pm 1, \pm 2, \ldots$ and $\ell_2, \ell_3 = 0, 1, 2, \ldots$. Let us introduce the normal modes

$$\begin{aligned} v_{\ell,x} &= \frac{2}{\sqrt{V}} e^{ik_{\ell_x} x} \, i \sin k_{\ell_y} y \, \sin k_{\ell_z} z, \\ v_{\ell,y} &= \frac{2}{\sqrt{V}} e^{ik_{\ell_x} x} \cos k_{\ell_y} y \, \sin k_{\ell_z} z, \\ v_{\ell,z} &= \frac{2}{\sqrt{V}} e^{ik_{\ell_x} x} \sin k_{\ell_y} y \, \cos k_{\ell_z} z. \end{aligned} \quad (2.29)$$

In the case of ℓ_2 or ℓ_3 is zero, we have to change $v_{\ell,y}$ and $v_{\ell,z}$ to

$$\begin{aligned} v_{\ell,y}|_{\ell_2=0} &= \sqrt{\frac{2}{V}} e^{ik_{\ell_x} x} \sin k_{\ell_z} z, \\ v_{\ell,z}|_{\ell_3=0} &= \sqrt{\frac{2}{V}} e^{ik_{\ell_x} x} \sin k_{\ell_y} y, \end{aligned} \quad (2.30)$$

respectively.

As in Eq. (2.12), the electric field can be expanded in terms of the normal modes, and they can be written as

$$\mathbf{E} = i \sum_{\ell,i} \sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}} \mathbf{e}_i \left(b_{\ell,i} v_{\ell,i} - b^\dagger_{\ell,i} v^*_{\ell,i} \right). \quad (2.31)$$

Equations $\mathrm{div}\mathbf{E} = 0$ and $\partial v_{\ell,x}/\partial x$, $\partial v_{\ell,y}/\partial y$, $\partial v_{\ell,z}/\partial z \propto v_{\ell,x}$ lead to the transversality condition given by Eq. (2.13). Using the orthogonal matrix (2.14) and new functions (operators) (2.15), the electric field (2.31) becomes

$$\mathbf{E} = i \sum_{\ell\sigma} \sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}} \left(a_{\ell\sigma} \bar{\mathbf{v}}_{\ell\sigma} - a^\dagger_{\ell\sigma} \bar{\mathbf{v}}^*_{\ell\sigma} \right), \quad (2.32)$$

where

$$\bar{\mathbf{v}}_{\ell\sigma} = \sum_i \mathbf{e}_i O^{(\ell)}_{\sigma i} v_{\ell,i}. \quad (2.33)$$

Note here that $\bar{\mathbf{v}}_{\bar{\ell}\sigma} = \epsilon_\sigma \bar{\mathbf{v}}^*_{\ell\sigma}$, where $\bar{\ell} = (-\ell_1, \ell_2, \ell_3)$ and $\epsilon_1 = -\epsilon_2 = 1$ because $O^{(\bar{\ell})}$ is obtained from $O^{(\ell)}$ by changing θ_ℓ to $\pi - \theta_\ell$. The new modes $\bar{\mathbf{v}}_{\ell\sigma}$ and $a_{\ell\sigma}$ satisfy the same equations as (2.18). Since $a_{\ell 3} = 0$ as in the case 1, the sum over σ in Eq. (2.32) is $\sigma = 1, 2$.

The new modes $\bar{\mathbf{v}}_{\ell\sigma}$ satisfy the orthonormality condition

$$\begin{aligned} &\int_c dv \, \bar{\mathbf{v}}^*_{\ell\sigma} \cdot \bar{\mathbf{v}}_{\ell'\sigma'} = \delta_{\ell\ell'} \delta_{\sigma\sigma'}, \\ &\int_c dv \, \bar{\mathbf{v}}_{\ell\sigma} \cdot \bar{\mathbf{v}}_{\ell'\sigma'} = \epsilon_\sigma \delta_{\ell\bar{\ell}'} \delta_{\sigma\sigma'}. \end{aligned} \quad (2.34)$$

Since $\sum_i k_{\ell_i} O^{(\ell)}_{\sigma i} = |\mathbf{k}_\ell| \delta_{\sigma 3} = 0$, we have the condition

$$\mathrm{div}\, \bar{\mathbf{v}}_{\ell\sigma} = 0. \quad (2.35)$$

Furthermore, it is easy to check that the modes $\bar{\mathbf{v}}_{\ell\sigma}$ satisfy the boundary conditions on the walls in the y and z directions:

$$\bar{\mathbf{v}}_{\ell\sigma}|_{\tan} = \mathbf{0}, \qquad \mathrm{curl}\, \bar{\mathbf{v}}_{\ell\sigma}|_{\mathrm{norm}} = \mathbf{0}. \quad (2.36)$$

In a way similar to case 1, the magnetic field and the vector potential can be obtained as follows:

$$\mathbf{B} = \sum_{\ell\sigma} \sqrt{\frac{\hbar}{2\varepsilon_0\omega_\ell}} \left(a_{\ell\sigma} \mathrm{curl}\, \bar{\mathbf{v}}_{\ell\sigma} + a^\dagger_{\ell\sigma} \mathrm{curl}\, \bar{\mathbf{v}}^*_{\ell\sigma} \right),$$

$$\mathbf{A} = \sum_{\ell\sigma} \sqrt{\frac{\hbar}{2\varepsilon_0\omega_\ell}} \left(a_{\ell\sigma} \bar{\mathbf{v}}_{\ell\sigma} + a^\dagger_{\ell\sigma} \bar{\mathbf{v}}^*_{\ell\sigma} \right). \tag{2.37}$$

Using Eq. (2.34) and the equality such as Eq. (2.24) for $\bar{\mathbf{v}}_{\ell\sigma}$, we find the Hamiltonian

$$H_R = \sum_{\ell\sigma} \hbar\omega_\ell \left(a^\dagger_{\ell\sigma} a_{\ell\sigma} + \frac{1}{2} \right). \tag{2.38}$$

C. Case 3

Finally, we consider a rectangular cavity with perfectly conducting walls on all three sides. Using Eq. (2.3) and the boundary conditions, we get $X|_{x=0,L_1} = d\bar{X}/dx|_{x=0,L_1} = 0$, $Y|_{y=0,L_2} = d\bar{Y}/dy|_{y=0,L_2} = 0$, and $dZ/dz|_{z=0,L_3} = \bar{Z}|_{z=0,L_3} = 0$. The electric field satisfying the boundary conditions can therefore be written as

$$\begin{aligned} E_x &\sim \cos k_{\ell_x} x \, \sin k_{\ell_y} y \, \sin k_{\ell_z} z, \\ E_y &\sim \sin k_{\ell_x} x \, \cos k_{\ell_y} y \, \sin k_{\ell_z} z, \\ E_z &\sim \sin k_{\ell_x} x \, \sin k_{\ell_y} y \, \cos k_{\ell_z} z, \end{aligned} \tag{2.39}$$

where

$$\mathbf{k}_\ell = (k_{\ell_x}, k_{\ell_y}, k_{\ell_z}) = \left(\frac{\pi\ell_1}{L_1}, \frac{\pi\ell_2}{L_2}, \frac{\pi\ell_3}{L_3} \right), \tag{2.40}$$

with $\ell_i = 0,1,2,\ldots$ $(i = x,y,z)$. We set the normal modes as

$$\begin{aligned} w_{\ell,x} &= \sqrt{\frac{8}{V}} \cos k_{\ell_x} x \, \sin k_{\ell_y} y \, \sin k_{\ell_z} z, \\ w_{\ell,y} &= \sqrt{\frac{8}{V}} \sin k_{\ell_x} x \, \cos k_{\ell_y} y \, \sin k_{\ell_z} z, \\ w_{\ell,z} &= \sqrt{\frac{8}{V}} \sin k_{\ell_x} x \, \sin k_{\ell_y} y \, \cos k_{\ell_z} z \end{aligned} \tag{2.41}$$

and

$$\begin{aligned} w_{\ell,x}|_{\ell_1=0} &= \frac{2}{\sqrt{V}} \sin k_{\ell_y} y \, \sin k_{\ell_z} z, \\ w_{\ell,y}|_{\ell_2=0} &= \frac{2}{\sqrt{V}} \sin k_{\ell_x} x \, \sin k_{\ell_z} z, \\ w_{\ell,z}|_{\ell_3=0} &= \frac{2}{\sqrt{V}} \sin k_{\ell_x} x \, \sin k_{\ell_y} y. \end{aligned} \tag{2.42}$$

The electric field is expanded in terms of $w_{\ell,i}$ as

$$\mathbf{E} = i \sum_{\ell,i} \sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}} \mathbf{e}_i \left(b_{\ell,i} - b^\dagger_{\ell,i} \right) w_{\ell,i}. \tag{2.43}$$

Taking into account

$$\frac{\partial w_{\ell,x}}{\partial x}, \; \frac{\partial w_{\ell,y}}{\partial y}, \; \frac{\partial w_{\ell,z}}{\partial z} \propto \sin k_{\ell_x} x \, \sin k_{\ell_y} y \, \sin k_{\ell_z} z, \tag{2.44}$$

we get the trasversality condition corresponding to Eq. (2.13). Thus using $O^{(\ell)}$ and $a_{\ell\sigma}$, which are given by Eqs. (2.14) and (2.15), respectively, we arrive at

$$\mathbf{E} = i \sum_{\ell\sigma} \sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}} \left(a_{\ell\sigma} - a^\dagger_{\ell\sigma} \right) \bar{\mathbf{w}}_{\ell\sigma}, \tag{2.45}$$

where

$$\bar{\mathbf{w}}_{\ell\sigma} = \sum_i \mathbf{e}_i O^{(\ell)}_{\sigma i} w_{\ell,i}, \tag{2.46}$$

which satisfies the orthonormality condition

$$\int_c dv \, \bar{\mathbf{w}}_{\ell\sigma} \cdot \bar{\mathbf{w}}_{\ell'\sigma'} = \delta_{\ell\ell'} \delta_{\sigma\sigma'}. \tag{2.47}$$

We also get

$$\mathrm{div}\, \bar{\mathbf{w}}_{\ell\sigma} = 0 \tag{2.48}$$

and at all boundaries

$$\bar{\mathbf{w}}_{\ell\sigma}|_{\tan} = \mathbf{0}, \qquad \mathrm{curl}\, \bar{\mathbf{w}}_{\ell\sigma}|_{\mathrm{norm}} = \mathbf{0}. \tag{2.49}$$

The magnetic field, the vector potential, and the Hamiltonian are determined as in the first and second cases.

III. MULTIPOLAR INTERACTIONS BETWEEN AN ATOM AND FIELDS

We now proceed to consider the interaction of the electromagnetic fields with a neutral atom in the cavities considered in the preceding section. The atom has a massive nucleus essentially stationary at a position $\mathbf{R}$ and Z_e electrons at $\mathbf{r}_a$ $(a = 1,2,\ldots,Z_e)$. The charge and the mass of each electron are $-e$ and m, respectively.

The total Hamiltonian H in the Coulomb gauge is given by

$$H = \frac{1}{2m} \sum_{a=1}^{Z_e} [\mathbf{p}_a + e\mathbf{A}(\mathbf{r}_a)]^2 + \frac{1}{2} \int_c dv \, \rho(\mathbf{r})\phi(\mathbf{r}) + \frac{1}{2} \int_c dv \left(\varepsilon_0 \mathbf{E}^2 + \frac{1}{\mu_0} \mathbf{B}^2 \right), \tag{3.1}$$

where $\mathbf{p}_a$ is the momentum of the ath electron, $\rho(\mathbf{r})$ the charge density of the atom, and $\phi(\mathbf{r})$ the scalar potential. The Hamiltonian (3.1) is converted to a more convenient form by a unitary transformation [16,27,28]. Define a unitary operator

$$U = \exp\left[\frac{i}{\hbar} \int_c dv \, \mathbf{P}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) \right], \tag{3.2}$$

where $\mathbf{P}$ is the polarization associated with the atomic charges:

$$\mathbf{P}(\mathbf{r}) = -e \sum_a (\mathbf{r}_a - \mathbf{R}) \int_0^1 d\lambda \, \delta(\mathbf{r} - \mathbf{R} - \lambda(\mathbf{r}_a - \mathbf{R})). \tag{3.3}$$

Note that

$$\mathrm{div}\mathbf{P}(\mathbf{r}) = e\sum_a \delta(\mathbf{r}-\mathbf{r}_a) - eZ_a\delta(\mathbf{r}-\mathbf{R}) = -\rho(\mathbf{r}). \quad (3.4)$$

The transformation of the first term of the Hamiltonian (3.1) is derived in a usual way (see, for example, [27]). Equations (3.2) and (3.3) lead to

$$U^\dagger \mathbf{p}_a U = \mathbf{p}_a - e\boldsymbol{\nabla}_a \times \int_0^1 d\lambda\,(\mathbf{r}_a-\mathbf{R})\cdot\mathbf{A}(\mathbf{R}+\lambda(\mathbf{r}_a-\mathbf{R})), \quad (3.5)$$

where $\boldsymbol{\nabla}_a$ is the gradient operator in coordinates $\mathbf{r}_a$. The vector potential can be expressed as the integral

$$\mathbf{A}(\mathbf{r}_a) = \int_0^1 d\lambda\,[1+(\mathbf{r}_a-\mathbf{R})\cdot\boldsymbol{\nabla}_a]\,\mathbf{A}(\mathbf{R}+\lambda(\mathbf{r}_a-\mathbf{R})), \quad (3.6)$$

which is verified by the Taylor expansion of $\mathbf{A}(\mathbf{R}+\lambda(\mathbf{r}_a-\mathbf{R}))$. Using Eqs. (3.5) and (3.6) we arrive at

$$U^\dagger[\mathbf{p}_a+e\mathbf{A}(\mathbf{r}_a)]U = \mathbf{p}_a - e\int_0^1 d\lambda\,(\mathbf{r}_a-\mathbf{R})\times\lambda\mathbf{B}(\mathbf{R}+\lambda(\mathbf{r}_a-\mathbf{R})), \quad (3.7)$$

where we have used $\boldsymbol{\nabla}_a\times\mathbf{A}(\mathbf{R}+\lambda(\mathbf{r}_a-\mathbf{R})) = \lambda\mathbf{B}(\mathbf{R}+\lambda(\mathbf{r}_a-\mathbf{R}))$.

Since the second term of Eq. (3.1) is unaffected by the transformation, let us turn to the third term. First note that

$$\left[\mathbf{E}(\mathbf{r}), \int_c dv'\mathbf{A}(\mathbf{r}')\cdot\mathbf{P}(\mathbf{r}')\right] = \frac{i\hbar}{\varepsilon_0}\tilde{\mathbf{P}}(\mathbf{r}), \quad (3.8)$$

where $\tilde{\mathbf{P}}$ is the transverse part of $\mathbf{P}$ defined by

$$\tilde{\mathbf{P}}(\mathbf{r}) = \frac{1}{2}\sum_{\ell\sigma}\left[\bar{\mathbf{u}}_{\ell\sigma}(\mathbf{r})\int_c dv'\bar{\mathbf{u}}^*_{\ell\sigma}(\mathbf{r}')\cdot\mathbf{P}(\mathbf{r}') + \text{c.c.}\right] \quad (3.9)$$

for case 1 in Sec. II. The modes $\bar{\mathbf{u}}_{\ell\sigma}$ are changed to $\bar{\mathbf{v}}_{\ell\sigma}$ in case 2, while in case 3 they are changed to $\bar{\mathbf{w}}_{\ell\sigma} = \bar{\mathbf{w}}^*_{\ell\sigma}$. Note that $\mathrm{div}\tilde{\mathbf{P}} = 0$. By Eq. (3.8),

$$U^\dagger\mathbf{E}(\mathbf{r})U = \mathbf{E}(\mathbf{r}) - \frac{1}{\varepsilon_0}\tilde{\mathbf{P}}(\mathbf{r}). \quad (3.10)$$

Using the following equalities:

$$\begin{aligned}
\int_c dv\,\mathbf{E}(\mathbf{r})\cdot\bar{\mathbf{u}}_{\ell\sigma}(\mathbf{r}) &= i\sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}}\left(a_{\bar{\ell}\sigma} - a^\dagger_{\ell\sigma}\right) \quad \text{for case 1,}\\
\int_c dv\,\mathbf{E}(\mathbf{r})\cdot\bar{\mathbf{v}}_{\ell\sigma}(\mathbf{r}) &= i\sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}}\left(\varepsilon_\sigma a_{\bar{\ell}\sigma} - a^\dagger_{\ell\sigma}\right) \quad \text{for case 2,}\\
\int_c dv\,\mathbf{E}(\mathbf{r})\cdot\bar{\mathbf{w}}_{\ell\sigma}(\mathbf{r}) &= i\sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}}\left(a_{\ell\sigma} - a^\dagger_{\ell\sigma}\right) \quad \text{for case 3,}
\end{aligned} \quad (3.11)$$

where $\bar{\ell} = (-\ell_1,-\ell_2,\ell_3)$ in case 1 and $\bar{\ell} = (-\ell_1,\ell_2,\ell_3)$ in case 2, we get

$$\int_c dv\,\mathbf{E}(\mathbf{r})\cdot\tilde{\mathbf{P}}(\mathbf{r}) = \int_c dv\,\mathbf{E}(\mathbf{r})\cdot\mathbf{P}(\mathbf{r}). \quad (3.12)$$

This equality plays an important role in obtaining a multipolar-interaction Hamiltonian. With the help of Eqs. (3.10) and (3.12), we can transform the third term with respect to the electric field in the Hamiltonian to

$$\int_c dv\,U^\dagger\mathbf{E}^2(\mathbf{r})U = \int_c dv\,\mathbf{E}^2(\mathbf{r}) - \int_c dv\,\mathbf{E}(\mathbf{r})\cdot\mathbf{P}(\mathbf{r}) + \frac{1}{4}\int_c dv\,\tilde{\mathbf{P}}^2(\mathbf{r}). \quad (3.13)$$

The complete transformed Hamiltonian obtained from Eq. (3.1) with the help of Eqs. (3.7) and (3.13) is

$$U^\dagger HU = H_A + H_R + H_E + H_M + H_N + H_F, \quad (3.14)$$

where

$$\begin{aligned}
H_A &= \frac{1}{2m}\sum_a \mathbf{p}_a^2 + \frac{1}{2}\int_c dv\,\sigma(\mathbf{r})\phi(\mathbf{r}),\\
H_R &= \frac{1}{2}\int_c dv\left(\varepsilon_0\mathbf{E}^2(\mathbf{r}) + \frac{1}{\mu_0}\mathbf{B}^2(\mathbf{r})\right),\\
H_E &= -\int_c dv\,\mathbf{E}(\mathbf{r})\cdot\mathbf{P}(\mathbf{r}),\\
H_M &= -\frac{e}{2m}\sum_a\int_0^1 d\lambda\,\{\mathbf{p}_a\cdot[(\mathbf{r}_a-\mathbf{R})\times\lambda\mathbf{B}(\mathbf{R}+\lambda(\mathbf{r}_a-\mathbf{R}))] + \text{H.c.}\},\\
H_N &= \frac{e^2}{2m}\sum_a\left[\int_0^1 d\lambda\,(\mathbf{r}_a-\mathbf{R})\times\lambda\mathbf{B}(\mathbf{R}+\lambda(\mathbf{r}_a-\mathbf{R}))\right]^2,\\
H_F &= \frac{1}{2\varepsilon_0}\int_c dv\,\tilde{\mathbf{P}}^2(\mathbf{r}).
\end{aligned} \quad (3.15)$$

The Hamiltonian H_A is for the isolated atom; H_R is the electromagnetic (radiation-field) Hamiltonian discussed for the three cases in Sec. II. The interaction of the atom with the fields is given by $H_E + H_M + H_N$; H_E and H_M correspond to the electric and magnetic multipolar interactions, respectively, while H_N is nonlinear. The last term H_F of the transformed Hamiltonian is only a function of the atomic variables.

From the definition (3.3) of $\mathbf{P}$, the Hamiltonian H_E is expressed in terms of the multipole moments of the atomic charge distribution:

$$H_E = e\sum_{n=0}^{\infty}\sum_{a=1}^{Z_a}\frac{1}{(n+1)!}[(\mathbf{r}_a-\mathbf{R})\cdot\boldsymbol{\nabla}]^n \times(\mathbf{r}_a-\mathbf{R})\cdot\mathbf{E}(\mathbf{r})|_{\mathbf{r}=\mathbf{R}}, \quad (3.16)$$

where $\boldsymbol{\nabla}$ is the gradient operator applicable to $\mathbf{r}$. Similarly, the Hamiltonians H_M and H_N are, respectively, expanded as follows:

$$H_M = \frac{e}{2m}\sum_{n=0}^{\infty}\sum_a \frac{n+1}{(n+1)!}\{[(\mathbf{r}_a-\mathbf{R})\cdot\boldsymbol{\nabla}]^n \times[(\mathbf{r}_a-\mathbf{R})\times\mathbf{p}_a]\cdot\mathbf{B}(\mathbf{r})+\text{H.c.}\}|_{\mathbf{r}=\mathbf{R}},$$

$$H_N = \frac{e^2}{2m}\left[\sum_{n=0}^{\infty}\sum_a \frac{n+1}{(n+2)!}[(\mathbf{r}_a-\mathbf{R}) \times[(\mathbf{r}_a-\mathbf{R})\cdot\boldsymbol{\nabla}]^n\mathbf{B}(\mathbf{r})]|_{\mathbf{r}=\mathbf{R}}\right]^2. \quad (3.17)$$

The first term of Eq. (3.16), which is the largest interaction, is called the electric dipole interaction and takes the form

$$H_{ED} = e\,\mathbf{D}\cdot\mathbf{E}(\mathbf{R}), \quad (3.18)$$

where $-e\mathbf{D} = -e\sum_a(\mathbf{r}_a-\mathbf{R})$ is the total electric dipole moment of the atom.

In the following section, we will obtain the spontaneous emission rate per second using the electric dipole interaction H_{ED}.

IV. SPONTANEOUS EMISSION

In this section, we consider transition rates of the atom in the cavities (cases 1–3) discussed in Sec. II, using, for simplicity, the dipole approximation. The Hamiltonian for the atom and the fields is

$$U^\dagger HU = H_A + H_R + H_I, \quad (4.1)$$

where H_A is the Hamiltonian for the free atom, H_R for the fields in the absence of the atom, and the interaction $H_I = H_{ED}$ is in the electric dipole approximation, which are given by Eqs. (3.15) and (3.18), respectively. We ignore the other terms in Eq. (3.15).

The free atom and the fields have the energy eigenstates $|s\rangle$ and $|n_{\ell\sigma}, n_{\ell'\sigma'},\ldots\rangle \equiv |\{n_{\ell\sigma}\}\rangle$, respectively, i.e.,

$$H_A|s\rangle = E_s|s\rangle, \quad H_R\,|\{n_{\ell\sigma}\}\rangle = \sum_{\ell\sigma}\hbar\omega_\ell n_{\ell\sigma}\,|\{n_{\ell\sigma}\}\rangle\,, \quad (4.2)$$

where the states $|s\rangle$ ($|\{n_{\ell\sigma}\}\rangle$) are assumed to be complete orthonormal for describing the atom (fields).

At $t=0$, the atom is assumed to be in a state $|s_0\rangle$ and the density matrix of the fields to be $\rho_R(0)$. According to perturbation theory, the probabilty per second of finding the atom in a state $|s\rangle$ at time t is given by [see Chap. 1, Eq. (1.21.27a), of Ref. [23]]

$$w = \frac{1}{\hbar^2}\frac{d}{dt}\int_0^t\int_0^t dt_1dt_2e^{i\omega_0(t_2-t_1)}\mathrm{Tr}\Big[\rho_R(0)U_R^\dagger(t_2) \times\langle s_0|H_I|s\rangle U_R(t_2)U_R^\dagger(t_1)\langle s|H_I|s_0\rangle U_R(t_1)\Big], \quad (4.3)$$

where

$$U_R(t) = \exp\left(-\frac{i}{\hbar}H_Rt\right), \quad \omega_0 = \frac{1}{\hbar}(E_{s_0}-E_s). \quad (4.4)$$

Note that ω_0 is positive in spontaneous emissions, because the state $|s_0\rangle$ is an excited one ($E_{s_0} > E_s$). It is sufficient to consider $\rho_R(0) = |\{0\}\rangle\langle\{0\}|$ as the denssity matrix for fields in the case of spontaneous emission.

Let us first consider the cavity for the first case (see Sec. II A). Using the Hamiltonian H_R of Eq. (2.25), we find

$$U_R^\dagger(t)\langle s_0|H_I|s\rangle U_R(t) = i\sum_{\ell\sigma}\sqrt{\frac{\hbar\omega_\ell}{2\varepsilon_0}}\Big[a_{\ell\sigma}e^{-i\omega_\ell t}\bar{\mathbf{u}}_{\ell\sigma}(\mathbf{R}) -a_{\ell\sigma}^\dagger e^{i\omega_\ell t}\bar{\mathbf{u}}_{\ell\sigma}^*(\mathbf{R})\Big]e\langle s_0|\mathbf{D}|s\rangle. \quad (4.5)$$

From

$$\mathrm{Tr}\,\rho_R(0)a_{\ell\sigma}a_{\ell'\sigma'}^\dagger = \delta_{\ell\ell'}\delta_{\sigma\sigma'}, \quad (4.6)$$

the transition rate w becomes

$$w = \frac{e^2\pi}{\varepsilon_0\hbar}\sum_{\ell\sigma}{}'\,|\bar{\mathbf{u}}_{\ell\sigma}(\mathbf{R})\cdot\langle s_0|\mathbf{D}|s\rangle|^2\,\omega_\ell\,g(\omega_\ell-\omega_0,t), \quad (4.7)$$

where $\sum'$ denotes that there is a possibility that we multiply it by 1/2 when $\ell_i = 0$, which comes from Eq. (2.11), and

$$g(x,t) = \frac{\sin xt}{\pi x}. \quad (4.8)$$

To show various results explicitly, we take $t\to\infty$ and thus $g(x,t)\to\delta(x)$.

Let us assume that the dipole moment is along the z direction. Then the transition rate $w = w_{1z}$ becomes

$$w_{1z} = \frac{e^2|\langle s_0|\mathbf{D}|s\rangle|^2}{\varepsilon_0\hbar}\frac{2\pi}{V}\sum_\ell{}'\left(1-\frac{k_{\ell_z}^2}{\mathbf{k}_\ell^2}\right) \times\omega_\ell\cos^2(k_{\ell_z}Z)\,\delta(\omega_\ell-\omega_0), \quad (4.9)$$

where Z is the z component of the position $\mathbf{R}$ of the atom: $\mathbf{R} = (X,Y,Z)$. In the above expression, we have used the equality

$$\sum_{\sigma=1}^{2}O_{\sigma3}^{(\ell)}O_{\sigma3}^{(\ell)} = 1 - O_{33}^{(\ell)}O_{33}^{(\ell)} = 1-\frac{k_{\ell_z}^2}{\mathbf{k}_\ell^2}. \quad (4.10)$$

We shall now see whether the influence of the cavity walls diappears in the the x and y directions when the lengths L_1 and L_2 become infinity. We are interested in whether the problem reduces to the case of periodic boundary condition (with infinite cavity size) in this limit.

Taking into account

$$\lim_{L_1,L_2\to\infty}\sum_{\ell_1\ell_2} = \frac{1}{4\pi^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}dk_xdk_y = \frac{1}{2\pi c^2}\int_{ck_{\ell_z}}^{\infty}d\omega_\ell\,\omega_\ell, \quad (4.11)$$

we arrive at

$$\frac{w_{1z}}{w_0} = \frac{3}{\xi_3}\sum_{0\le\ell_3<\xi_3}{}'\left(1-\frac{\ell_3^2}{\xi_3^2}\right)\cos^2\left(\frac{\pi\ell_3Z}{L_3}\right), \quad (4.12)$$

where w_0 is the transition rate in free space

$$w_0 = \frac{e^2|\langle s_0|\mathbf{D}|s\rangle|^2\omega_0^3}{3\pi\hbar\varepsilon_0 c^3} \tag{4.13}$$

and $\xi_3 = \omega_0 L_3/\pi c$. Note that we must multiply Eq. (4.12) by 1/2 when $\ell_3 = 0$ [see Eq. (2.11)].

If $\xi_3 \le 1$, then the sum over ℓ in Eq. (4.12) is the only $\ell_3 = 0$ term; thus we get

$$\frac{w_{1z}}{w_0} = \frac{3}{2\xi_3}, \tag{4.14}$$

where 1/2 came from Eq. (2.11). The result (4.14) shows that the transition rate w_{1z} approaches infnity as $\xi_3 \to 0$, i.e., enhanced spontaneous emission.

Here we take $L_3 \to \infty$. The sum over ℓ_3 changes to the integral with respect to k_z:

$$\frac{1}{L_3}\sum_{0\le\ell_3<\xi_3}{}' \to \frac{1}{\pi}\int_0^{\omega_0/c} dk_z. \tag{4.15}$$

Thus Eq. (4.12) leads to the well-known result (see, for example, Ref. [18])

$$\frac{w_{1z}}{w_0} = 1 - 3\left(\frac{\cos a_z}{a_z^2} - \frac{\sin a_z}{a_z^3}\right), \tag{4.16}$$

where $a_z = 2\omega_0 Z/c$. This result shows the influence of the wall (mirror). As is expected, w_{1z}/w_0 approaches 1 as the distance between the atom and the wall becomes larger. On the other hand, if the atom approaches the wall, then $w_{1z}/w_0 \to 2$.

Next consider the case where the dipole moment is along the x direction. Similarly, the tarnsition rate w_{1x} is given by

$$\frac{w_{1x}}{w_0} = \frac{3}{2\xi_3}\sum_{1\le\ell_3<\xi_3}\left(1+\frac{\ell_3^2}{\xi_3^2}\right)\sin^2\left(\frac{\pi\ell_3 Z}{L_3}\right). \tag{4.17}$$

If $\xi_3 \le 1$, then $\ell_3 = 0$ and thus we have $w_{1x} = 0$; the spontaneous emission is forbidden. Taking $L_3 \to \infty$, we recover, after the integration, the following (see, for example, [18]):

$$\frac{w_{1x}}{w_0} = 1 - \frac{3}{2}\left(\frac{\sin a_z}{a_z} + \frac{\cos a_z}{a_z^2} - \frac{\sin a_z}{a_z^3}\right). \tag{4.18}$$

This shows that $w_{1x}/w_0 \to 1$ $(a_z \to \infty)$ and $w_{1x}/w_0 \to a_z^2/5$ $(a_z \to 0)$.

Let us turn to the cavity of case 2 and assume that the dipole moment has only the z component. By Eqs. (2.29) and (2.30), the transition rate (4.9) changes to

$$w_{2z} = \frac{e^2|\langle s_0|\mathbf{D}|s\rangle|^2}{\varepsilon_0\hbar}\frac{4\pi}{V}\sum_\ell{}'\left(1-\frac{k_{\ell_z}^2}{\mathbf{k}_\ell^2}\right) \times \omega_\ell \sin^2(k_{\ell_y}Y)\cos^2(k_{\ell_z}Z)\,\delta(\omega_\ell-\omega_0). \tag{4.19}$$

When $L_1 \to \infty$, taking into account the change from sum to integral

$$\frac{1}{L_1}\sum \to \frac{1}{2\pi}\int_{-\infty}^{\infty} dk_x = \frac{1}{\pi c}\int_{c(k_{\ell_y}^2+k_{\ell_z}^2)^{1/2}}^{\infty} d\omega_\ell \times \frac{\omega_\ell}{\sqrt{\omega_\ell^2 - c^2(k_{\ell_y}^2+k_{\ell_z}^2)}}, \tag{4.20}$$

we find

$$\frac{w_{2z}}{w_0} = \frac{12\pi c^2}{\omega_0 L_2 L_3}\sum_{\ell_2\ell_3}{}'\frac{(1-c^2k_{\ell_z}^2/\omega_0^2)}{\sqrt{\omega_0^2-c^2(k_{\ell_y}^2+k_{\ell_z}^2)}} \times \sin^2(k_{\ell_y}Y)\cos^2(k_{\ell_z}Z), \tag{4.21}$$

where ℓ_2 and ℓ_3 have to satisfy $\sqrt{k_{\ell_y}^2+k_{\ell_z}^2} < \omega_0/c$, i.e.,

$$\frac{\ell_2^2}{\xi_2^2}+\frac{\ell_3^2}{\xi_3^2} < 1, \tag{4.22}$$

with $\xi_i = \omega_0 L_i/\pi c$ $(i = 1,2)$. Note that we multiply Eq. (4.21) by 1/2 when $\ell_3 = 0$.

Here let us assume that $\xi_3 \le 1$; then only the $\ell_3 = 0$ term is allowed in the sum over ℓ_3 in Eq. (4.21). Thus Eq. (4.21) becomes

$$\frac{w_{2z}}{w_0} = \frac{6\pi c^2}{\omega_0 L_2 L_3}\sum_{1\le\ell_2<\xi_2}\frac{\sin^2(k_{\ell_y}Y)}{\sqrt{\omega_0^2-c^2k_{\ell_y}^2}}. \tag{4.23}$$

To obtain the result corresponding to Eq. (4.14), take $L_2 \to \infty$. Then Eq. (4.23) changes into

$$\frac{w_{2z}}{w_0} = \frac{6}{\pi\xi_3}\int_0^{\frac{\pi}{2}} d\theta\,\sin^2\left(\frac{1}{2}a_y\sin\theta\right) = \frac{3}{2\xi_3}\left[1-J_0(a_y)\right], \tag{4.24}$$

where J_0 is the zeroth Bessel function and $a_y = 2\omega_0 Y/c$. Equation (4.24) indicates a side-mirror effect explicitly, because it depends on the distance Y between the atom and the side mirror lying at the $y = 0$ plane. As a_y becomes very large, Eq. (4.24) approaches $3/2\xi_3$, which is the same as Eq. (4.14). On the other hand, it becomes

$$\frac{w_{2z}}{w_0} = \frac{3}{2\xi_3}\left(\frac{a_y^2}{4}\right), \tag{4.25}$$

as a_y approaches zero, where we have used $J_0(a_y) = 1 - a_y^2/4 + a_y^4/64 - \cdots$.

The dipole moment is now assumed in the x direction. Then, by Eq. (4.3), the transition rate w_{2x} is given by

$$w_{2x} = \frac{e^2|\langle s_0|\mathbf{D}|s\rangle|^2}{\hbar\varepsilon_0}\frac{4\pi}{V}\sum_\ell{}'\left(1-\frac{k_{\ell_x}^2}{\mathbf{k}_\ell^2}\right)^2 \times \omega_\ell\sin^2(k_{\ell_y}Y)\sin^2(k_{\ell_z}Z)\,\delta(\omega_\ell-\omega_0). \tag{4.26}$$

When $L_1 \to \infty$, then the above transition rate becomes

$$\frac{w_{2x}}{w_0} = \frac{12\pi c^2}{\omega_0 L_2 L_3} \sum_{\ell_2\ell_3} \frac{c^2(k_{\ell_y}^2 + k_{\ell_z}^2)}{\omega_0^2\sqrt{\omega_0^2 - c^2(k_{\ell_y}^2 + k_{\ell_z}^2)}} \times \sin^2(k_{\ell_y}Y)\,\sin^2(k_{\ell_z}Z), \quad (4.27)$$

where ℓ_2 and ℓ_3 are restricted by the condition (4.22). The transition is thus prohibited if $\xi_2 \leq 1$ or $\xi_3 \leq 1$ is satisfied.

Finally let us proceed with the case-3 cavity, which is more general than cases 1 and 2. Assume that the dipole moment is pointing along the z direction. Then, by Eq. (4.3), the transition rate w_{3z} is given by

$$w_{3z} = \frac{e^2|\langle s_0|\mathbf{D}|s\rangle|^2}{\varepsilon_0\hbar}\frac{8\pi}{V}\sum_{\ell}{}' \left(1 - \frac{k_{\ell_z}^2}{\mathbf{k}_\ell^2}\right)\omega_\ell \times \sin^2(k_{\ell_x}X)\,\sin^2(k_{\ell_y}Y)\,\cos^2(k_{\ell_z}Z)\,\delta(\omega_\ell - \omega_0). \quad (4.28)$$

After taking $L_1 \to \infty$ and integrating with respect to k_x, we obtain

$$\frac{w_{3z}}{w_0} = \frac{24\pi c^2}{\omega_0 L_2 L_3}\sum_{\ell_2\ell_3}{}' \frac{(1 - c^2k_{\ell_z}^2/\omega_0^2)}{\sqrt{\omega_0^2 - c^2(k_{\ell_y}^2 + k_{\ell_z}^2)}} \times \sin^2\left(\frac{1}{c}\sqrt{\omega_0^2 - c^2(k_{\ell_y}^2 + k_{\ell_z}^2)}X\right) \times \sin^2(k_{\ell_y}Y)\,\cos^2(k_{\ell_z}Z), \quad (4.29)$$

where the condition for ℓ_2 and ℓ_3 is given by Eq. (4.22).

We set here $\xi_3 \leq 1$ in order to get a side-mirror effect explicitly. Since $\ell_3 = 0$, taking into account an additional factor 1/2, we have

$$\frac{w_{3z}}{w_0} = \frac{12\pi c^2}{\omega_0 L_2 L_3} \times \sum_{1\leq\ell_2<\xi_2} \frac{\sin^2(\sqrt{\omega_0^2 - c^2k_{\ell_y}^2}X/c)\sin^2(k_{\ell_y}Y)}{\sqrt{\omega_0^2 - c^2k_{\ell_y}^2}}. \quad (4.30)$$

If we take $L_2 \to \infty$, then Eq. (4.30) becomes

$$\frac{w_{3z}}{w_0} = \frac{12}{\pi\xi_3}\int_0^{\frac{\pi}{2}} d\theta\,\sin^2\left(\frac{1}{2}a_y\sin\theta\right)\,\sin^2\left(\frac{1}{2}a_x\cos\theta\right) = \frac{3}{2\xi_3}\left[1 - J_0(a_x) - J_0(a_y) + J_0\left(\sqrt{a_x^2 + a_y^2}\right)\right], \quad (4.31)$$

where $a_x = 2\omega_0 X/c$. This result depends on the coordinates X and Y of the atom, which are distances between the atom and the side mirrors. If $a_x \to \infty$ ($X \to \infty$), then Eq. (4.31) becomes that of case 2 given by Eq. (4.24). As is also expected, Eq. (4.31) approaches Eq. (4.14) if $a_x, a_y \to \infty$; the side-mirror effect disappears in this case. On the other hand, if $a_x \ll 1$ and $a_y \ll 1$, then we find

$$\frac{w_{3z}}{w_0} = \frac{3}{2\xi_3}\left(\frac{a_x^2 a_y^2}{32}\right). \quad (4.32)$$

V. CONCLUDING REMARKS

From the theoretical point of view, we have developed a quantization procedure with three-dimensional boundary conditions. In this paper, we have applied this procedure to three different boundary conditions in the rectangular coordinate system. We hope to discuss the same problem in the cylindrical coordinate system in the future using the same quantization procedure.

We have considered three kinds of cavities in Sec. II. The first one is enclosed by perfect conducting walls in the z direction, while the second is enclosed by the same kind of walls in the y and z directions. The third cavity we have treated is completely enclosed by perfectly conducting walls. In the first case, our result is consistent with those existing in the literature [16–21]. We are presenting new results in the second and third cases.

In each case, we have introduced explicitly the normal modes $\bar{\mathbf{u}}_{\ell\sigma}$, $\bar{\mathbf{v}}_{\ell\sigma}$, and $\bar{\mathbf{w}}_{\ell\sigma}$ defined by Eqs. (2.17), (2.33), and (2.46), respectively. Using them, we have carried out canonical quantization of the fields in the cavities. The normal modes have played important roles Throughout the process of quantization.

The multipolar expansion of the interaction between an atom and the fields in the cavities has been obtained in Sec. III. The normal modes play also an important role in this case, on the basis of which the transverse part of the polarization $\tilde{\mathbf{P}}$ was introduced. With the help of the transverse polarization, it is possible to carry out the multipolar expansion in a succinct manner.

The spontaneous emission by an atom in the cavities has been studied using the field-theoretic methods developed in Sec. II. In particular, we have obtained the transition rates per second in three types of cavities when $\xi_3 = \omega_0 L_3/\pi c \leq 1$ (L_1, $L_2 \to \infty$). The resuts are given by Eqs. (4.14), (4.24), and (4.31); they show a very interesting side-mirror effect. That is, the result (4.24) or (4.31) depends on the coordinate X or Y of the atom, which are the distances between the atom and the side mirrors (walls). Equation (4.31) approaches Eqs. (4.24) and (4.14) as X or Y becomes very large. On the contrary, if X or Y is very small, the X or Y dependence of the transition rate becomes dominant as has been shown in Eq. (4.32).

Let us finally consider Eq. (4.24) more explicitly. The first few roots of $J_0(a_y) = 0$ are found to be [29]

$$a_y = 2.405, 5.520, 8.654; \quad (5.1)$$

the transition rate w_{2z}/w_0 becomes $w_{1z}/w_0 = 3/2\xi_3$ ($\xi_3 \leq 1$), i.e., the side-mirror effect disappears at these points. On the other hand, the first two roots of $dJ_0/da_y = 0$ are given by [29]

$$a_y = 3.832, 7.016; \quad (5.2)$$

the transition rate w_{2z}/w_0 has its maximum at $a_y = 3.832$ and its (relative) minimum at $a_y = 7.016$. That is, the side-mirror effect becomes dominant at these points. The rate w_{2z}/w_0 oscillates and approaches w_{1z}/w_0 as a_y becomes very large.

Setting $\omega_0 = 10^{13}$ Hz, we have $Y \simeq 5.7\times10^{-5}$ m as the distance between the atom and the side mirror. It follows from $\xi_3 \leq 1$ that $L_3 \leq \pi c/\omega_0 \simeq 9.4 \times 10^{-5}$ m. Thus we may observe experimentally the oscillation phenomenon of the transition rate.

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