Quantum electrodynamics in the presence of dielectrics and conductors. III. Relations among one-photon transition probabilities in stationary and nonstationary fields, density of states, the field-correlation functions, and surface-dependent response functions

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In this paper the physical entities such as transition probabilities and the density of states are related to appropriate electromagnetic-field correlation functions and to appropriate response functions. Such response functions have already been computed in a previous paper and therefore these can be used to obtain surface-dependent corrections. It is shown how the density of states and hence Planck's law depends on the presence of surfaces. I explicitly calculate the correction terms for the case of a small blackbody bounded by two plane conducting surfaces. An appreciable correction occurs if the linear dimensions of the blackbody are of the order of a wavelength. Next electric-dipole-type transitions in atomic systems are considered and a straightforward perturbation theory is used to obtain one-photon transition probabilities in terms of the surface-dependent response functions. As an illustration of the surface-dependent terms, the transitions in presence of a conducting surface are considered. The transition probabilities show a marked increase or decrease depending on whether the dipole transition is parallel or perpendicular to the surface. Both stationary and nonstationary fields are considered. As a special case of nonstationary fields, the transitions in a coherent field are considered in detail. It is also shown how the coherent radiation field in presence of dielectrics can be realized. It is found that if the radiation field, in arbitrary geometries, is initially in vacuum state then at later times it would be found in a coherent state if perturbed by an external (c-number) electromagnetic field.

I. INTRODUCTION

Paper I discussed how different kinds of response functions can be used to calculate the electromagnetic field fluctuations, and in Paper II these were used to obtain dispersion forces between microscopic and macroscopic bodies.[1] This paper continues with this response-function formalism to study the impact of dielectric and conducting surfaces on the density of states and one-photon transition probabilities. One-photon transitions, in the presence of surfaces, have been studied both experimentally and theoretically by Carniglia, Mandel, and Drexhage[2] and by Carniglia and Mandel.[3] In the theoretical work[3] they used the explicit quantization of the field. Their results are valid only in a specialized situation. As mentioned in Paper I, I would like to avoid the explicit quantization of the field because of problems inherent in such a procedure. It is for these reasons that the above effects are studied using response functions. Results are presented both for fields in thermal equilibrium as well as fields which need not be in thermal equilibrium.

In Sec. II are calculated the corrections to the density of states and Planck's law using the appropriate response functions. Explicit results are presented for electromagnetic fields in the domain bounded by two plane conducting surfaces. In Sec. III the interaction between the radiation field and the atomic systems is considered. It is restricted only to the calculation of one-photon transition probabilities. A straightforward perturbation theory is used to obtain the one-photon transition probabilities in terms of the correlation functions of the radiation field. I first consider stationary radiation fields in thermal equilibrium and use the fluctuation-dissipation theorem to express the transition probabilities in terms of response functions like χ_{ijEE}, χ_{ijEH}, etc. I explicitly consider the change in transition probabilities due to the presence of a conductor which gives rise to interesting coherence and anticoherence effects depending on the orientation of the dipole moment. Next, transitions in nonstationary fields initially in a coherent state are treated. Finally, I consider how such coherent states can be experimentally realized. In a future publication the formalism of this paper and Paper I will be used to discuss lifetime and Lamb shifts of excited states in presence of surfaces. In later papers we hope to discuss the anomalous magnetic moment of the electron, multiphoton transition probabilities, optical nutation, etc.

II. SURFACE-DEPENDENT CORRECTIONS TO THE DENSITY OF STATES AND PLANCK'S LAW

It is known from Bose statistics that the average energy per unit volume between frequencies ω and $\omega + d\omega$ is given by

$$U(\omega)\,d\omega = \frac{\hbar\omega}{e^{\beta\hbar\omega}-1}\,\rho_H(k_0)\,\frac{dk_0}{V}\,, \tag{2.1}$$

where $\rho_H(k_0)\,dk_0$ is the density of states, i.e., the number of modes lying in the interval k_0 to k_0+dk_0. For the electromagnetic field in a *vacuum* one has

$$\rho_H(k_0)\,dk_0 = V(k_0^2/\pi^2)\,dk_0\,, \tag{2.2}$$

if the volume V occupied by the field is very large (going to infinity) and if the *periodic* boundary conditions are imposed at the surface bounding the volume V. On combining (2.1) and (2.2), one finds that for the entire free space

$$U(\omega)\,d\omega = \frac{\hbar\omega^3}{e^{\beta\hbar\omega}-1}\,\frac{d\omega}{\pi^2c^3}\,. \tag{2.3}$$

If we are studying fields in a *finite* domain such as the electromagnetic field between two conducting plates, then (2.2) no longer holds. There would be a size-dependent correction to (2.2) and accordingly a correction to Planck's law (2.3). In this section we consider such correction terms and discuss the conditions under which such terms could make a significant contribution. The usual eigenfunction expansion of the Green's function leads to the following relation between the density of eigenvalues and its imaginary part[4]:

$$\rho_G(k_0)\,dk_0 = (k_0/2\pi^2)\int_V d^3r\ \mathrm{Im}\sum G_{ii}(\vec{r},\vec{r},k_0)\,dk_0\,, \tag{2.4a}$$

where the Green's dyadic $\vec{\vec{G}}$ (with elements G_{ij}) satisfies

$$\nabla\times\nabla\times\vec{\vec{G}} - k_0^2\,\vec{\vec{G}} = 4\pi\vec{\vec{I}}\,\delta(\vec{r}-\vec{r}')\,, \tag{2.4b}$$

and where $\vec{\vec{I}}$ is the unit dyadic. The density function ρ_G depends on the type of boundary conditions imposed on $\vec{\vec{G}}$. On comparing (2.4b) with (I.4.2) we find that $\vec{\vec{G}}_{ij}$ satisfies same equation as χ_{ijEE}/k_0^2, χ_{ijHH}/k_0^2. The density-of-states factor ρ_H, appearing in Planck's law, is with respect to the eigenvalues of the Hamiltonian. It is clear that there is a relation between ρ_H and the density of eigenvalues for (2.4b). It is easy to obtain such a relation for the electromagnetic field in free but bounded space. We show that

$$\rho_H(k_0)\,dk_0 = \frac{dk_0}{4\pi^2k_0}\int_V d^3r\,\mathrm{Im}\sum_i\left[\chi_{iiEE}(\vec{r},\vec{r},\omega) + \chi_{iiHH}(\vec{r},\vec{r},\omega)\right] \tag{2.5}$$

$$= \tfrac{1}{2}\left[\rho_{EE}(k_0)\,dk_0 + \rho_{HH}(k_0)\,dk_0\right]\,, \tag{2.6}$$

where $\rho_{EE}(k_0)\,dk_0$ $[\rho_{HH}(k_0)\,dk_0]$ is given by (2.4) with G_{ij} replaced by χ_{iiEE} (χ_{iiHH}). We know that the electromagnetic field in free space is characterized by the Hamiltonian

$$\langle H\rangle = \langle{:}\int[(E^2+H^2)/8\pi]\,d^3r\,{:}\rangle\,, \tag{2.7a}$$

where the pair of colons denotes the *normal* ordering. On using the results of Sec. III of Paper I, (2.7a) becomes

$$\langle H\rangle = \frac{1}{4\pi}\int d^3r\sum_i\left[\mathcal{E}_{ii}^{(N)}(\vec{r},\vec{r},0)+\mathcal{H}_{ii}^{(N)}(\vec{r},\vec{r},0)\right], \tag{2.7b}$$

where $\mathcal{E}_{ii}^{(N)}$ and $\mathcal{H}_{ii}^{(N)}$ are defined by (I.3.3) and (I.3.6), respectively. On writing (2.7b) in terms of response functions [(I.3.13), (I.3.14)], we obtain

$$\langle H\rangle = \frac{1}{8\pi^2}\int_{-\infty}^{+\infty}d\omega\int d^3r\sum_i\left[\mathcal{E}_{ii}^{(N)}(\vec{r},\vec{r},\omega) + \mathcal{H}_{ii}^{(N)}(\vec{r},\vec{r},\omega)\right]$$

$$= \frac{\hbar}{4\pi^2}\int_{-\infty}^{+\infty}d\omega\int d^3r\,\eta(\omega)\,\frac{e^{-\beta\omega\hbar}}{e^{-\beta\omega\hbar}-1} \times\mathrm{Im}\sum_i\left[\chi_{iiEE}(\vec{r},\vec{r},-\omega)+\chi_{iiHH}(\vec{r},\vec{r},-\omega)\right],$$

which on using the symmetry property of χ can be written as

$$\langle H\rangle = V\int_0^\infty d\omega\,h(\omega)(e^{\beta\omega\hbar}-1)^{-1} \equiv V\int_0^\infty U(\omega)\,d\omega, \tag{2.8}$$

with

$$h(\omega) = \frac{\hbar}{4\pi^2V}\int d^3r\,\mathrm{Im}\sum_i\left[\chi_{iiEE}(\vec{r},\vec{r},\omega) + \chi_{iiHH}(\vec{r},\vec{r},\omega)\right]. \tag{2.9}$$

Finally (2.5) and (2.6) follow if we compare (2.9) with (2.1). If $\chi^{(0)}$ represents the translationally invariant response (Sec. IV of Paper I), then integration over $\vec{r}$ gives V and

$$\mathrm{Im}\chi^{(0)}_{iiEE}(\vec{r},\vec{r},\omega) = \mathrm{Im}\chi^{(0)}_{iiHH}(\vec{r},\vec{r},\omega) = \left(k_0^2+\frac{\partial^2}{\partial x_i^2}\right)\frac{\sin k_0|\vec{r}-\vec{r}'|}{|\vec{r}-\vec{r}'|}\bigg|_{\vec{r}'=\vec{r}}$$

$$= \tfrac{2}{3}k_0^3\,,$$

and on substituting these results in (2.6) we obtain (2.2).

We now consider the following situation: we have the electromagnetic radiation in the region $-d\le z\le 0$ which is supposed to be vacuum and which is bounded by two conducting plane surfaces at $z=0$ and at $z=-d$. The surface-dependent response functions are to be obtained from (I.5.33)–(I.5.37). We have first from (I.5.35) and (I.5.36)

$$\chi^{(1)}_{33EE}(\vec{r},\vec{r}_0,\omega)=\frac{-i}{2\pi}\int\int\frac{du\,dv}{w_0}D_0^{-1}\,k_\parallel^2\left\{e^{i\vec{K}_0\cdot\vec{r}}(e^{-i\vec{K}_0'\cdot\vec{r}_0}+e^{-i\vec{K}_0\cdot\vec{r}_0})+e^{i\vec{K}_0'\cdot\vec{r}}(e^{-i\vec{K}_0'\cdot\vec{r}_0}+e^{-i\vec{K}_0\cdot\vec{r}_0-2iw_0d})\right\}, \tag{2.10}$$

$$\begin{aligned}\chi^{(1)}_{11EE}(\vec{r},\vec{r}_0,\omega)=\frac{i}{2\pi}\int\int\frac{du\,dv}{w_0k_\parallel^2}D_0^{-1}\{&e^{i\vec{K}_0\cdot\vec{r}}[u^2w_0^2e^{-i\vec{K}_0'\cdot\vec{r}_0}-u^2w_0^2e^{-i\vec{K}_0\cdot\vec{r}_0}-v^2k_0^2e^{-i\vec{K}_0\cdot\vec{r}_0}+v^2k_0^2e^{-i\vec{K}_0'\cdot\vec{r}_0}]\\&+e^{i\vec{K}_0'\cdot\vec{r}}[-u^2w_0^2e^{-i\vec{K}_0'\cdot\vec{r}_0}+u^2w_0^2e^{-i\vec{K}_0\cdot\vec{r}_0-2iw_0d}+k_0^2v^2e^{-2iw_0d-i\vec{K}_0\cdot\vec{r}_0}\\&-k_0^2v^2e^{-i\vec{K}_0'\cdot\vec{r}_0}]\}.\end{aligned} \tag{2.11}$$

$\chi^{(1)}_{22EE}(\vec{r},\vec{r}_0,\omega)$ is obtained from (2.11) if the replacement $u\rightarrow v$ in the terms in square brackets is made. Hence

$$\begin{aligned}\chi^{(1)}_{11EE}(\vec{r},\vec{r}_0,\omega)+\chi^{(1)}_{22EE}(\vec{r},\vec{r}_0,\omega)=\frac{i}{2\pi}\int\frac{du\,dv}{w_0}D_0^{-1}(w_0^2+k_0^2)[&e^{i\vec{K}_0\cdot\vec{r}}(e^{-i\vec{K}_0'\cdot\vec{r}_0}-e^{-i\vec{K}_0\cdot\vec{r}_0})\\&+e^{i\vec{K}_0'\cdot\vec{r}}(e^{-i\vec{K}_0\cdot\vec{r}_0-2iw_0d}-e^{-i\vec{K}_0'\cdot\vec{r}_0})],\end{aligned} \tag{2.12}$$

and therefore letting $r\rightarrow r_0$,

$$\chi^{(1)}_{11EE}(\vec{r},\vec{r},\omega)+\chi^{(1)}_{22EE}(\vec{r},\vec{r},\omega)=\frac{i}{2\pi}\int\int\frac{du\,dv}{w_0}D_0^{-1}(w_0^2+k_0^2)(e^{2iw_0z}+e^{-2iw_0d-2iw_0z}-2). \tag{2.13}$$

Similarly in (2.10) letting $\vec{r}\rightarrow\vec{r}_0$, we find that

$$\chi^{(1)}_{33EE}(\vec{r},\vec{r},\omega)=\frac{-i}{2\pi}\int\int\frac{du\,dv}{w_0}k_\parallel^2D_0^{-1}(e^{2iw_0z}+e^{-2iw_0d-2iw_0z}+2). \tag{2.14}$$

On adding (2.13) and (2.14) we find that the trace $\overleftrightarrow{\chi}$ is given by

$$\sum\chi^{(1)}_{iiEE}(\vec{r},\vec{r},\omega)=\frac{i}{\pi}\int\int\frac{du\,dv}{w_0}D_0^{-1}[w_0^2(e^{2iw_0z}+e^{-2iw_0z-2iw_0d})-2k_0^2], \tag{2.15}$$

which on changing the variable of integration becomes

$$\sum\chi^{(1)}_{iiEE}(\vec{r},\vec{r},\omega)=2ik_0^3\int_0^\infty\frac{\kappa d\kappa}{w_0}(1-e^{-2iw_0k_0d})^{-1}[w_0^2(e^{2iw_0zk_0}+e^{-2iw_0zk_0-2iw_0k_0d})-2], \tag{2.16a}$$

where now

$$w_0^2=1-\kappa^2. \tag{2.16b}$$

When $\kappa>1$, w_0 is pure imaginary and therefore

$$\begin{aligned}\mathrm{Im}\sum\chi^{(1)}_{iiEE}(\vec{r},\vec{r},\omega)&=2k_0^3\mathrm{Re}\int_0^1\frac{\kappa d\kappa}{w_0}(1-e^{-2iw_0k_0d})^{-1}[w_0^2(e^{2iw_0zk_0}+e^{-2iw_0zk_0-2iw_0k_0d})-2]\\&=2k_0^3\mathrm{Re}\int_0^1d\alpha\,(1-e^{-2ik_0d\alpha})^{-1}[\alpha^2(e^{2i\alpha k_0z}+e^{-2i\alpha k_0z-2ik_0d})-2].\end{aligned} \tag{2.17}$$

Because of the singularity in the integrand, we have to specify how to go around the singularity. Equation (2.17) should be interpreted in the following manner: Replace α by $\alpha+i\epsilon$, and after the integral has been done, take the limit $\epsilon\rightarrow 0$. This limit is motivated from the fact that if we move both the conductors to infinity then (2.17) should vanish. To see this write (2.17) as

$$\mathrm{Im}\sum\chi^{(1)}_{iiEE}(\vec{r},\vec{r},\omega)=2k_0^3\mathrm{Re}\int_0^1d\alpha\,(1-e^{-2ik_0d(\alpha+i\epsilon)})^{-1}\{2\alpha^2e^{-ik_0d(\alpha+i\epsilon)}\cos(2k_0x_0\alpha)-2\},$$

$$x_0=\tfrac{1}{2}d+z. \tag{2.18}$$

Now it is easily seen from (2.18) that if we let $d \to \infty$ keeping x_0 fixed, then (2.18) gives zero. It is now clear from (2.17) that

$$\int_{-d}^{0} \mathrm{Im} \sum \chi^{(1)}_{iiEE}(\vec{r}, \vec{r}, \omega)\, dz = -4k_0^3\, dI, \tag{2.19}$$

where

$$\begin{aligned} I &= \mathrm{Re} \int_0^1 d\alpha\, (1 - e^{-2idk_0\alpha + 2dk_0\epsilon})^{-1} \\ &= -\frac{1}{2} \int_0^1 d\alpha \left(e^{2idk_0\alpha - 2dk_0\epsilon} \sum_0^\infty e^{2indk_0\alpha - 2dk_0\epsilon n} \right. \\ &\qquad \left. + e^{-2idk_0\alpha - 2dk_0\epsilon} \sum_0^\infty e^{-2indk_0\alpha - 2dk_0\epsilon n} \right) \\ &= -\frac{1}{2} \int_0^1 d\alpha \sum_{-\infty}^{+\infty} e^{2indk_0\alpha + \frac{1}{2}}. \end{aligned} \tag{2.20}$$

On using Poisson's summation formula[5]

$$\sum_{-\infty}^{+\infty} e^{2indk_0\alpha} = \frac{\pi}{dk_0} \sum_{-\infty}^{+\infty} \delta\left(\alpha - \frac{\pi n}{dk_0}\right), \tag{2.21}$$

(2.20) reduces to

$$\begin{aligned} I &= \frac{1}{2} - \frac{\pi}{2dk_0} \int_0^1 d\alpha \sum_{-\infty}^{+\infty} \delta\left(\alpha - \frac{\pi n}{dk_0}\right) \\ &= \frac{1}{2} - \frac{\pi}{2dk_0} (\mathfrak{N} + \tfrac{1}{2}), \end{aligned} \tag{2.22}$$

where $\mathfrak{N}$ is the largest integer less than dk_0/π. In obtaining (2.22) the normalization condition

$$\int_0^1 \delta(\alpha)\, d\alpha = \tfrac{1}{2}$$

has been used. Hence on combining (2.19) and (2.22), we have

$$\mathrm{Im} \int_{-d}^{0} dz \sum \chi^{(1)}_{iiEE}(\vec{r}, \vec{r}, \omega) = -2k_0^3 d + 2\pi k_0^2 (\mathfrak{N} + \tfrac{1}{2}). \tag{2.23}$$

One also has

$$\mathrm{Im} \int_{-d}^{0} dz \sum \chi^{(0)}_{iiEE}(\vec{r}, \vec{r}, \omega) = 2k_0^3 d, \tag{2.24}$$

and hence

$$\begin{aligned} \mathrm{Im} \int_{-d}^{0} dz \sum \chi_{iiEE}(\vec{r}, \vec{r}, \omega) &= 2\pi k_0^2 (\mathfrak{N} + \tfrac{1}{2}) \\ &= 2k_0^3 d \left\{ 1 - \frac{\pi}{k_0 d} \left[\left(\frac{k_0 d}{\pi} - \mathfrak{N} \right) - \frac{1}{2} \right] \right\}. \end{aligned} \tag{2.25}$$

A similar analysis for the magnetic field response functions using (I.5.38)–(I.5.41) shows that

$$\mathrm{Im} \int_{-d}^{0} dz \sum \chi_{iiHH}(\vec{r}, \vec{r}, \omega) = 2\pi k_0^2 (\mathfrak{N} + \tfrac{1}{2}). \tag{2.26}$$

Note that $(k_0 d/\pi - \mathfrak{N})$ varies from 0 to 1 and hence $(k_0 d/\pi - \mathfrak{N} - \frac{1}{2})$ varies from $-\frac{1}{2}$ to $+\frac{1}{2}$, and therefore it is clear from (2.25) that the correction term $(\pi/k_0 d)(k_0 d/\pi - \mathfrak{N} - \frac{1}{2})$ can be appreciable only for $k_0 d \sim \pi$, or for distances of the order of a wavelength. The final expression for the density of states, on using (2.25), (2.26), and (2.5), is

$$\rho_H(k_0)\, dk_0 = \frac{V k_0^2}{\pi^2} dk_0 \left[1 - \frac{\pi}{k_0 d} \left(\frac{k_0 d}{\pi} - \mathfrak{N} - \frac{1}{2} \right) \right], \tag{2.27a}$$

and Planck's law becomes

$$U(\omega)\, d\omega = \frac{d\omega}{\pi^2 c^3} \frac{\hbar \omega^3}{(e^{\beta\hbar\omega} - 1)} \left[1 - \frac{\pi}{k_0 d} \left(\frac{k_0 d}{\pi} - \mathfrak{N} - \frac{1}{2} \right) \right]. \tag{2.27b}$$

It should be noted that (2.27a) and (2.27b) are exact results.[6] If $k_0 d/\pi$ is equal to an integer $\mathfrak{M}$, then the $\mathfrak{N} + \frac{1}{2}$ factor of (2.22) should be replaced by $\mathfrak{M}$ leading to $I = 0$, and therefore for such values of $k_0 d$ the density of states and the Planck's law are unaffected.

The result (2.27b) should be compared with the result obtained by Balian and Bloch[4] that the *lowest order* size correction to a spherical blackbody is given by

$$U(\omega)\, d\omega \cong \frac{d\omega}{\pi^2 c^3} \frac{\hbar \omega^3}{(e^{\beta\hbar\omega} - 1)} \left[1 - 2 \left(\frac{c}{\omega R} \right)^2 \right], \tag{2.28}$$

which again show that the correction terms become appreciable only when the radius is of the order of the wavelength.

III. RELATION BETWEEN ONE-PHOTON TRANSITION PROBABILITIES AND THE ELECTROMAGNETIC FIELD CORRELATION FUNCTIONS

We now consider the one-photon transition in an atomic system and show how the transition probabilities are related to electromagnetic field correlation functions, which are known in terms of the response functions. The entire calculation will be done in $\vec{r}$ space. As already emphasized several times, we would like to avoid the mode expansion of the electric field operator as far as possible. In this way size- or surface-dependent corrections to transition probabilities are obtained from the surface-dependent response functions.

As discussed by Power and Zineau,[7] the interaction Hamiltonian between a bound electron and the second quantized electric field, in dipole approximation, can be written as

$$H = H_0 - \int \vec{P}_A(\vec{r})\cdot\vec{E}(\vec{r})\, d^3r + 2\pi \int |\vec{P}_A^{\perp}(r)|^2\, d^3r\ . \tag{3.1}$$

Here $\vec{P}_A(\vec{r})$ is the polarization operator corresponding to the atomic system and the last term in (3.1) is important only for self-interactions and will be ignored. The density matrix ρ of the combined atomic-plus-field system satisfies

$$\partial\rho/\partial t = (-i/\hbar)[H_1(t), \rho]\,, \tag{3.2}$$

where $H_1(t)$ is the interaction Hamiltonian in the interaction picture. We will take the initial state of the system as

$$\rho(t_0) = |\psi_i\rangle\langle\psi_i|\rho_F(t_0)\,, \tag{3.3}$$

where $|\psi_i\rangle$ denotes the pure state of the atomic system and $\rho_F(t_0)$ the initial state of the radiation field. On integrating (3.2) we obtain, in the lowest order (Born approximation),

$$\rho(t_0+T) \approx \rho(t_0) - \frac{i}{\hbar}\int_{t_0}^{t_0+T} dt_1[H_1(t_1),\ \rho(t_0)] - \frac{1}{\hbar^2} \times \int_{t_0}^{t_0+T} dt_1 \int_{t_0}^{t_1} dt_2\, [H_1(t_1), [H_1(t_2), \rho(t_0)]]\,. \tag{3.4}$$

Let $p_{fi}(t_0+T)$ be the probability that the atomic system will be found in the state $|\psi_f\rangle$ after the one-photon transition has taken place. It is related to ρ by

$$p_{fi}(t_0+T) = \mathrm{Tr}_F\langle\psi_f|\rho(t_0+T)|\psi_f\rangle\,, \tag{3.5}$$

where Tr_F denotes the trace over the radiation-field variables. On using (3.4) and (3.5) and the orthogonality of $|\psi_i\rangle$ and $|\psi_f\rangle$, we find that

$$p_{fi}(t_0+T) = -\frac{1}{\hbar^2}\int_{t_0}^{t_0+T} dt_1 \int_{t_0}^{t_1} dt_2 \times \mathrm{Tr}_F\langle\psi_f|[H_1(t_1),\ [H_1(t_2), \rho(t_0)]]|\psi_f\rangle\,. \tag{3.6}$$

Writing $H_1(t)$ as

$$H_1(t) = -\int \vec{P}_A(\vec{r}, t)\cdot\vec{E}(\vec{r}, t)\, d^3r\,, \tag{3.7}$$

with $\vec{E}(\vec{r}, t)$, $\vec{P}_A(\vec{r}, t)$, etc. as operators in the interaction picture, we find that the term $H_1(t_1)\rho(t_0)H_1(t_2)$ can be written as

$$\mathrm{Tr}_F\langle\psi_f|H_1(t_1)\rho(t_0)H_1(t_2)|\psi_f\rangle = \sum_{mn}\int\int d^3r_1\, d^3r_2 \langle\psi_f|\vec{P}_{Am}(\vec{r}_1, t_1)|\psi_i\rangle\langle\psi_i|\vec{P}_{An}(\vec{r}_2, t_2)|\psi_f\rangle\langle E_n(\vec{r}_2, t_2)E_m(\vec{r}_1, t_1)\rangle\,. \tag{3.8}$$

The terms like $H_1(t_1)H_1(t_2)\rho(t_0)$, $\rho(t_0)H_1(t_1)H_1(t_2)$ vanish due to the orthogonality of $|\psi_i\rangle$ and $|\psi_f\rangle$. On combining (3.6) and (3.8), we obtain

$$p_{fi}(t_0+T) = \frac{1}{\hbar^2}\sum_{mn}\int\int d^3r_1\, d^3r_2 \int_{t_0}^{t_0+T} dt_1 \int_{t_0}^{t_1} dt_2 \langle E_m(\vec{r}_1, t_1)E_n(\vec{r}_2, t_2)\rangle\langle\psi_i|P_{Am}(\vec{r}_1, t_1)|\psi_f\rangle \times\langle\psi_f|P_{An}(\vec{r}_2, t_2)|\psi_i\rangle + \text{c.c.},$$

which by a change of variables becomes

$$p_{fi}(t_0+T) = \frac{1}{\hbar^2}\sum_{mn}\int\int d^3r_1\, d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1, 0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2, 0)|\psi_i\rangle \times\int_0^T dt_1\int_0^{t_1} dt_2 \langle E_m(\vec{r}_1, t_1+t_0)E_n(\vec{r}_2, t_2+t_0)\rangle \exp[(i/\hbar)(E_i - E_f)(t_1 - t_2)] + \text{c.c.}\,, \tag{3.9}$$

where E_i and E_f are respectively the unperturbed energies of the initial and final state. Equation (3.9) can also be written as

$$p_{fi}(t_0+T) = \frac{1}{\hbar^2}\sum_{mn}\int\int d^3r_1\, d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1, 0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2, 0)|\psi_i\rangle\, I_{mn}(\vec{r}_1, \vec{r}_2, t_0), \tag{3.10}$$

with

$$I_{mn}(\vec{r}_1, \vec{r}_2, t_0) = \int_0^T dt_1\int_0^{t_1} dt_2 \{\langle E_m(\vec{r}_1, t_1+t_0)E_n(\vec{r}_2, t_2+t_0)\rangle\, e^{(i/\hbar)(E_i - E_f)(t_1 - t_2)} + (\text{the term with } t_1 \rightleftarrows t_2)\}\,. \tag{3.11}$$

Further simplification of (3.11) depends on the nature of the electromagnetic field. We first consider transitions in *stationary* electromagnetic fields for which the correlation functions are invariant under time translation. Denoting by ω_{fi} the energy difference

$$\hbar\omega_{fi} = E_f - E_i \,, \tag{3.12}$$

and on using stationarity, (3.11) becomes

$$I_{mn}(\vec{r}_1, \vec{r}_2) = \int_0^T dt_1 \int_0^{t_1} dt_2 [\langle E_m(\vec{r}_1, t_2) E_n(\vec{r}_2, 0)\rangle e^{-i\omega_{fi}t_2} + \langle E_m(\vec{r}_1, -t_2) E_n(\vec{r}_2, 0)\rangle e^{i\omega_{fi}t_2}]$$

$$= \int_0^T dt_1 \int_{-t_1}^{+t_1} dt_2 \langle E_m(\vec{r}_1, t_2) E_n(\vec{r}_2, 0)\rangle e^{-i\omega_{fi}t_2} \,. \tag{3.13}$$

Equation (3.13) in the usual long-time approximation leads to

$$I_{mn}(\vec{r}_1, \vec{r}_2) = T \int_{-\infty}^{+\infty} d\tau \langle E_m(\vec{r}_1, \tau) E_n(\vec{r}_2, 0)\rangle e^{-i\omega_{fi}\tau} \,, \tag{3.14}$$

which on using Eqs. (I.2.5), (I.2.8), and (I.2.9) becomes

$$I_{mn}(\vec{r}_1, \vec{r}_2) = T[\hbar\chi''_{mnEE}(\vec{r}_1, \vec{r}_2, \omega_{if}) + \mathcal{G}^{(S)}_{mn}(\vec{r}_1, \vec{r}_2, \omega_{if})] \,. \tag{3.15}$$

On combining (3.10) and (3.15), we find that the one-photon transition probability γ_{fi} for unit time from the state $|\psi_i\rangle$ to final state $|\psi_f\rangle$ in stationary fields is given by

$$\gamma_{fi} = \frac{p_{fi}}{T} = \frac{1}{\hbar^2} \sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1, 0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2, 0)|\psi_i\rangle [\hbar\chi''_{mnEE}(\vec{r}_1, \vec{r}_2, \omega_{if}) + \mathcal{G}^{(S)}_{mn}(\vec{r}_1, \vec{r}_2, \omega_{if})] \,, \tag{3.16}$$

where $\mathcal{G}^{(S)}$ is the symmetrized correlation function defined by (I.2.16) and χ'' the imaginary part of the response function defined by (I.2.12). From Eqs. (I.3.11) and (I.3.21) it is clear that (3.16) can also be written as

$$\gamma_{fi} = \frac{1}{\hbar^2} \sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1, 0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2, 0)|\psi_i\rangle \, \mathcal{G}^{(N)}_{mn}(\vec{r}_1, \vec{r}_2, \omega_{if}) \quad \text{if } \omega_{if} < 0 \,, \tag{3.17}$$

$$= \frac{1}{\hbar^2} \sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1, 0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2, 0)|\psi_i\rangle \, \mathcal{G}^{(A)}_{mn}(\vec{r}_1, \vec{r}_2, \omega_{if}) \quad \text{if } \omega_{if} > 0 \,. \tag{3.18}$$

Hence in a photon absorption process ($\omega_{if} < 0$) [one photon emission processes ($\omega_{if} > 0$)] the transition probability is determined from the normally ordered correlation function [antinormally ordered correlation function] of the electromagnetic field. This is, of course, what one expects on physical grounds from the interpretation of the positive and negative frequency parts of the field operator as corresponding to photon absorption and emission, respectively. The results (3.16)–(3.18) are valid for any kind of electromagnetic fields, *not* necessarily one in thermal equilibrium. We now specialize to thermal fields; and then, writing symmetrized correlation functions in terms of the response function, (3.16) becomes

$$\gamma_{fi} = \frac{1}{\hbar} \sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1, 0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2, 0)|\psi_i\rangle [1 + \coth(\beta\omega_{if}\hbar/2)] \, \mathrm{Im}\chi_{mnEE}(\vec{r}_1, \vec{r}_2, \omega_{if}) \,. \tag{3.19}$$

It is obvious from (3.19) that at zero temperature

$$\gamma_{fi} = 0, \quad \text{if } \omega_{if} < 0 \,; \tag{3.20a}$$

i.e., there is no possibility of spontaneous absorption. The spontaneous emission transition probability is equal to

$$\gamma^{(\text{spon})}_{fi} = \frac{2}{\hbar} \sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1, 0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2, 0)|\psi_i\rangle \, \mathrm{Im}\chi_{mnEE}(\vec{r}_1, \vec{r}_2, \omega_{if}), \quad \omega_{if} > 0. \tag{3.20b}$$

The stimulated emission transition probability is equal to

$$\gamma_{fi}^{(\mathrm{stim})} = \frac{2}{\hbar} \sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1,0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2,0)|\psi_i\rangle (e^{\beta\hbar\omega_{if}}-1)^{-1} \mathrm{Im}\chi_{mnEE}(\vec{r}_1,\vec{r}_2,\omega_{if}) . \tag{3.20c}$$

The absorption probability is equal to

$$\gamma_{fi}^{(\mathrm{abs})} = -\frac{2}{\hbar} \sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1,0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2,0)|\psi_i\rangle (e^{\beta\hbar|\omega_{if}|}-1)^{-1} \mathrm{Im}\chi_{mnEE}(\vec{r}_1,\vec{r}_2,-|\omega_{if}|),$$

which on using the symmetry property

$$\chi''_{mnEE}(\vec{r}_1,\vec{r}_2,\omega) = \chi''_{nmEE}(\vec{r}_2,\vec{r}_1,\omega) = -\chi''_{mnEE}(\vec{r}_1,\vec{r}_2,-\omega) , \tag{3.21}$$

becomes

$$\gamma_{fi}^{(\mathrm{abs})} = \frac{2}{\hbar} \sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{An}(\vec{r}_2,0)|\psi_f\rangle\langle\psi_f|P_{Am}(\vec{r}_1,0)|\psi_i\rangle (e^{\beta\hbar|\omega_{if}|}-1)^{-1} \mathrm{Im}\chi_{mnEE}(\vec{r}_1,\vec{r}_2,|\omega_{if}|) . \tag{3.22}$$

It is clear from (3.20c) and (3.22) that

$$\gamma_{fi}^{(\mathrm{abs})} = \gamma_{if}^{(\mathrm{stim})} . \tag{3.23a}$$

Similarly from (3.19) one has

$$\frac{\gamma_{fi}}{\gamma_{if}} = -\frac{1+\coth(\beta\omega_{if}\hbar/2)}{1-\coth(\beta\omega_{if}\hbar/2)} = e^{\beta\hbar\omega_{if}} . \tag{3.23b}$$

Relations (3.23a) and (3.23b) yield the right equilibrium value.

We have thus shown how physical quantities like transition probabilities are related to the electromagnetic field correlations functions and the response functions. The surface-dependent corrections, corresponding to a given geometrical arrangement, to the transition probabilities are now obtained by substituting the appropriate response functions in (3.19); we have already computed a number of such response functions in Paper I. Let $\gamma_{fi}^{(0)}$ be the transition probability in the radiation field in the absence of dielectric, and let $\gamma_{fi}^{(s)}$ be the transition probability in the presence of surfaces. Then it follows from (3.19) that

$$\frac{\gamma_{fi}^{(s)}-\gamma_{fi}^{(0)}}{\gamma_{fi}^{(0)}} \equiv \frac{\Gamma^{(s)}-\Gamma^{(0)}}{\Gamma^{(0)}} = \sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1,0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2,0)|\psi_i\rangle \mathrm{Im}\chi^{(1)}_{mnEE}(\vec{r}_1,\vec{r}_2,\omega_{if}) \Big/$$
$$\sum_{mn} \int\int d^3r_1 d^3r_2 \langle\psi_i|P_{Am}(\vec{r}_1,0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2,0)|\psi_i\rangle \mathrm{Im}\chi^{(0)}_{mnEE}(\vec{r}_1,\vec{r}_2,\omega_{if}), \tag{3.24}$$

where $\chi^{(0)}$ is the usual translationally invariant response function and $\chi^{(1)}$ denotes the surface-dependent contribution to the total response function. Thus the relative change in the transition probability depends essentially on the ratio $\chi^{(1)}/\chi^{(0)}$. In particular for a *two-level* atom located at $\vec{r}=\vec{r}_0$ we have

$$\frac{\Gamma^{(s)}-\Gamma^{(0)}}{\Gamma^{(0)}} = \frac{\sum d_m d_n \mathrm{Im}\chi^{(1)}_{mnEE}(\vec{r}_0,\vec{r}_0,\omega_{if})}{\sum d_m d_n \mathrm{Im}\chi^{(0)}_{mnEE}(\vec{r}_0,\vec{r}_0,\omega_{if})} , \tag{3.25}$$

where $\vec{d}$ is the dipole matrix element between the two states of the atom. To have some idea of the change, we consider the simplified situation: we have the radiation field in the semi-infinite domain $\infty \geq z \geq 0$ and bounded by a conducting surface at $z=0$ and the atom is located at $\vec{r}_0 = b$ along the z axis. The ratio still depends on the orientation of the dipole moment. We discuss the two cases separately: (i) the dipole moment has a random orientation in x-y plane; (ii) the dipole moment is parallel to z axis. We denote the transition rates in the two cases by $\Gamma_{\parallel}$ and $\Gamma_{\perp}$ respectively:

$$\left(\frac{\Gamma^{(s)}-\Gamma^{(0)}}{\Gamma^{(0)}}\right)_{\parallel} = \frac{\mathrm{Im}\sum_{i=1}^{2}\chi^{(1)}_{iiEE}(\vec{r}_0,\vec{r}_0,\omega_{if})}{\mathrm{Im}\sum_{i=1}^{2}\chi^{(0)}_{iiEE}(\vec{r}_0,\vec{r}_0,\omega_{if})} , \tag{3.26a}$$

$$\left(\frac{\Gamma^{(s)}-\Gamma^{(0)}}{\Gamma^{(0)}}\right)_{\perp} = \frac{\mathrm{Im}\,\chi^{(1)}_{33EE}(\vec{r}_0,\vec{r}_0,\omega_{if})}{\mathrm{Im}\,\chi^{(0)}_{33EE}(\vec{r}_0,\vec{r}_0,\omega_{if})} . \tag{3.26b}$$

The response functions for the geometry under consideration are given by

$$\chi^{(0)}_{ijEE}(\vec{r},\vec{r}',\omega)=\left(k_0^2\delta_{ij}+\frac{\partial^2}{\partial x_i\partial x_j}\right)\frac{e^{ik_0|\vec{r}-\vec{r}'|}}{|\vec{r}-\vec{r}'|}, \tag{3.27a}$$

$$\chi^{(1)}_{ijEE}(\vec{r},\vec{r}',\omega)=\left(k_0^2\delta_{ij}(2\delta_{i3}-1)+\frac{\partial^2}{\partial x_i\partial x'_j}\right)\times\frac{e^{ik_0|\vec{r}-\vec{r}'_i|}}{|\vec{r}-\vec{r}'_i|}, \tag{3.27b}$$

$$\vec{r}'_i\equiv(x',\,y',\,-z'). \tag{3.27c}$$

It is obvious from (3.27a) that

$$\mathrm{Im}\,\chi^{(0)}_{iiEE}(\vec{r}_0,\vec{r}_0,\omega)=\tfrac{2}{3}\omega^3/c^3. \tag{3.28}$$

Similarly,

$$\mathrm{Im}\chi^{(1)}_{xxEE}(\vec{r}_0,\vec{r}_0,\omega)=\mathrm{Im}\,\chi^{(1)}_{yyEE}(\vec{r}_0,\vec{r}_0,\omega)=-k_0^3\left[\left(\frac{1}{\alpha}-\frac{1}{\alpha^3}\right)\sin\alpha+\frac{\cos\alpha}{\alpha^2}\right],\qquad \alpha=2k_0z_0, \tag{3.29}$$

$$\mathrm{Im}\chi^{(1)}_{zzEE}(\vec{r}_0,\vec{r}_0,\omega)=2k_0^3\left(\frac{\sin\alpha}{\alpha^3}-\frac{\cos\alpha}{\alpha^2}\right). \tag{3.30}$$

Hence on combining (3.26) and (3.28)–(3.30), we obtain

$$\left(\frac{\Gamma^{(s)}-\Gamma^{(0)}}{\Gamma^{(0)}}\right)_{\parallel}=-\frac{3}{2}\left[\left(\frac{1}{\alpha}-\frac{1}{\alpha^3}\right)\sin\alpha+\frac{\cos\alpha}{\alpha^2}\right], \tag{3.31}$$

$$\left(\frac{\Gamma^{(s)}-\Gamma^{(0)}}{\Gamma^{(0)}}\right)_{\perp}=+3\left(\frac{\sin\alpha}{\alpha^3}-\frac{\cos\alpha}{\alpha^2}\right). \tag{3.32}$$

These relations show how the transition probabilities are altered as a function of the distance of the atom from the conducting surface. $\Gamma^{(s)}$ becomes a damped oscillatory function of α. The surface shows a marked effect on these transition probabilities provided the distance of the atom from the surface is smaller than or of the order of a wavelength. On carrying out expansions for small α, we obtain

$$\left(\frac{\Gamma^{(s)}-\Gamma^{(0)}}{\Gamma^{(0)}}\right)_{\parallel}\approx-1+\frac{\alpha^2}{5}, \tag{3.33}$$

$$\left(\frac{\Gamma^{(s)}-\Gamma^{(0)}}{\Gamma^{(0)}}\right)_{\perp}\approx 1-\frac{\alpha^2}{10}. \tag{3.34}$$

Equation (3.33) shows that the transition probability goes down if the atom is placed in the vicinity of a conductor and if the dipole moment is oriented parallel to the surface. This is a type of anti-coherence effect. In the case of the dipole moment perpendicular to the surface, the transition probability is increased, leading to a positive coherence effect. These properties are the reflections of the coherence properties of the black-body fluctuations in finite geometries which we discussed in Paper I. Other characteristics will be discussed in a future paper.

IV. ONE-PHOTON TRANSITIONS IN NONSTATIONARY FIELDS

We have so far considered the *stationary* electromagnetic fields in *thermal* equilibrium. For such fields using the theory of Sec. II of Paper I, it was possible to compute all the necessary correlation functions in terms of the response functions. The case of nonstationary fields is extremely difficult because for such fields there is, in general, no fluctuation-dissipation theorem, and nice results like (I.3.11) and (I.3.21) break down. However for electromagnetic fields the commutator of the fields at two different space-time points is still a c number, and hence the response functions which we have obtained in Paper I give us the commutator of the fields at two space-time points irrespective of whether the field is stationary or not. The real problem arises in the computation of the field correlation functions. Before we discuss such a computation, we would like to describe how the transition probability is related to the field correlation functions.

The one-photon transition probability is given by (3.10) and (3.11). On using the identity

$$\int_0^T\int dt_1\,dt_2 f(t_1,t_2)=\int_0^T dt_1\times\int_0^{t_1}dt_2[f(t_1,t_2)+f(t_2,t_1)], \tag{4.1}$$

(3.11) becomes

$$I_{mn}(\vec{r}_1,\vec{r}_2,t_0)=\int_0^T dt_1\int_0^T dt_2\times\langle E_m(\vec{r}_1,t_1+t_0)E_n(\vec{r}_2,t_2+t_0)\rangle\times\exp[i\omega_{if}(t_1-t_2)]. \tag{4.2}$$

Further simplification of (4.2) depends on the nature of the process, i.e., whether it is an absorption type or emission type. We first treat the absorption processes. We express the fields in terms of positive and negative frequency parts and retain only the *slowly* varying contributions; then (4.2) reduces to

$$I_{mn}(\vec{r}_1, \vec{r}_2, t_0) = \int_0^T dt_1 \int_0^T dt_2 \langle E_m^{(-)}(\vec{r}_1, t_1 + t_0) \times E_n^{(+)}(\vec{r}_2, t_2 + t_0) \rangle e^{i\omega_{if}(t_1 - t_2)}. \tag{4.3}$$

We moreover assume that initially the field is in a *coherent* state so that the correlation appearing in (4.3) is equal to

$$\langle E_m^{(-)}(\vec{r}_1, t_1 + t_0) E_n^{(+)}(\vec{r}_2, t_2 + t_0) \rangle = V_m^*(\vec{r}_1, t_1 + t_0) V_n(\vec{r}_2, t_2 + t_0). \tag{4.4}$$

(The construction of such a coherent state in which the electric field has an amplitude V_m will be discussed in Sec. V.) On substituting (4.3) and (4.4) into (3.10), we obtain

$$p_{fi}(t_0 + T) = \left| \frac{1}{\hbar} \sum_n \int d^3 r_2 \langle \psi_f | P_{An}(\vec{r}_2, 0) | \psi_i \rangle \times \int_0^T dt_2 \, V_n(\vec{r}_2, t_2 + t_0) e^{i\omega_{fi} t_2} \right|^2. \tag{4.5}$$

If we write V_n as

$$V_n(\vec{r}_2, t_2 + t_0) = \int_0^\infty \upsilon_n(\vec{r}_2, \omega) e^{-i\omega t_2} \, d\omega, \tag{4.6}$$

then (4.5) becomes

$$p_{fi}^{(\mathrm{abs})}(t_0 + T) = \left| \frac{1}{\hbar} \sum_n \int d^3 r_2 \langle \psi_f | P_{An}(\vec{r}_2, 0) | \psi_i \rangle \int_0^\infty d\omega \, \upsilon_n(\vec{r}_2, \omega) \frac{\sin[\frac{1}{2}(\omega_{fi} - \omega)T]}{\frac{1}{2}(\omega_{fi} - \omega) e^{-i(\omega_{fi} - \omega)T/2}} \right|^2. \tag{4.7}$$

If the usual condition that the time interval T be small compared with the reciprocal frequency spread of υ_n is satisfied, then (4.7) reduces to

$$p_{fi}^{(\mathrm{abs})}(t_0 + T) = \left| \frac{1}{\hbar} \sum_n \int d^3 r_2 \langle \psi_f | P_{An}(\vec{r}_2, 0) | \psi_i \rangle \, V_n(\vec{r}_2, t_0 + T/2) \frac{\sin[(\omega_{fi} - \omega_0)T/2]}{\frac{1}{2}(\omega_{fi} - \omega_0)} \right|^2, \tag{4.8}$$

where ω_0 is the frequency at which υ_n is centered. If (4.8) is summed over all the final states, then (4.8), under the usual condition of slowly varying density of states and the matrix element, reduces

$$p_{fi}^{(\mathrm{abs})}(t_0 + T) = \frac{2\pi}{\hbar^2} T \left| \sum_n \int d^3 r_2 \langle \psi(E_f = E_i + \hbar\omega_0) | P_{An}(\vec{r}_2, 0) | \psi_i \rangle \, V_n(\vec{r}_2, t_0 + T/2) \right|^2 \sigma(E_i + \hbar\omega_0), \tag{4.9}$$

where σ denotes the density of the final electronic states. The structure of (4.9) is similar to that of the formula derived by Carniglia and Mandel[3] using the mode expansion of field operators.

The case of emission in nonstationary fields could be treated similarly. In place of (4.3) we would have

$$I_{mn}(\vec{r}_1, \vec{r}_2, t_0) = \int_0^T dt_1 \int_0^T dt_2 \langle E_m^{(+)}(\vec{r}_1, t_1 + t_0) E_n^{(-)}(\vec{r}_2, t_2 + t_0) \rangle e^{i\omega_{if}(t_1 - t_2)} \tag{4.10}$$

$$= I_{mn}^{(\mathrm{stim})}(\vec{r}_1, \vec{r}_2, t_0) + I_{mn}^{(\mathrm{spon})}(\vec{r}_1, \vec{r}_2, t_0), \tag{4.11}$$

where

$$I_{mn}^{(\mathrm{stim})}(\vec{r}_1, \vec{r}_2, t_0) = \int_0^T dt_1 \int_0^T dt_2 \langle E_n^{(-)}(\vec{r}_2, t_1 + t_0) E_m^{(+)}(\vec{r}_1, t_2 + t_0) \rangle e^{i\omega_{if}(t_2 - t_1)}, \tag{4.12}$$

$$I_{mn}^{(\mathrm{spon})}(\vec{r}_1, \vec{r}_2, t_0) = \int_0^T dt_1 \int_0^T dt_2 \langle [E_m^{(+)}(\vec{r}_1, t_1 + t_0), E_n^{(-)}(\vec{r}_2, t_2 + t_0)] \rangle e^{i\omega_{if}(t_1 - t_2)}, \tag{4.13}$$

$I_{mn}^{(\mathrm{spon})}(\vec{r}_1, \vec{r}_2, t_0)$ is independent of t_0. On substituting (4.12) and (4.13) in (3.10), we find that

$$p_{fi}(t_0 + T) = p_{fi}^{(\mathrm{stim})}(t_0 + T) + p_{fi}^{(\mathrm{spon})}(t_0 + T), \tag{4.14}$$

where

$$p_{fi}^{(\mathrm{spon})}(t_0 + T) = \frac{1}{\hbar^2} \sum_{mn} \int\int d^3 r_1 \, d^3 r_2 \langle \psi_i | P_{Am}(r_1, 0) | \psi_f \rangle \langle \psi_f | P_{An}(\vec{r}_2, 0) | \psi_i \rangle \times \int_0^T \int dt_1 \, dt_2 \langle [E_m^{(+)}(\vec{r}_1, t_1 - t_2), E_n^{(-)}(\vec{r}_2, 0)] \rangle e^{i\omega_{if}(t_1 - t_2)}, \tag{4.15}$$

$$p_{fi}^{(\mathrm{stim})}(t_0+T)=\frac{1}{\hbar^2}\sum_{mn}\int\int d^3r_1\,d^3r_2\langle\psi_i|P_{Am}(\vec{r}_1,0)|\psi_f\rangle\langle\psi_f|P_{An}(\vec{r}_2,0)|\psi_i\rangle$$

$$\times\int_0^T\int dt_1\,dt_2\langle E_n^{(-)}(\vec{r}_2,t_1+t_0)E_m^{(+)}(\vec{r}_1,t_2+t_0)\rangle\, e^{i\omega_{if}(t_2-t_1)}\,. \tag{4.16}$$

It is easily shown that $p_{fi}^{(\mathrm{spon})}$ as given by (4.15) is identical to $T\gamma_{fi}^{(\mathrm{spon})}$ calculated in Sec. III [Eq. (3.20b)]. For a field in the coherent state, $p_{fi}^{(\mathrm{stim})}$ becomes

$$p_{fi}^{(\mathrm{stim})}(t_0+T)=\left|\frac{1}{\hbar}\sum_m\int d^3r_1\langle\psi_i|P_{Am}(\vec{r}_1,0)|\psi_f\rangle\int_0^T dt_2\,V_m(\vec{r}_1,t_2+t_0)e^{i\omega_{if}t_2}\right|^2, \tag{4.17}$$

which on comparison with (4.5) shows that

$$p_{fi}^{(\mathrm{abs})}(t_0+T)=p_{if}^{(\mathrm{stim})}(t_0+T)\,. \tag{4.18}$$

It should be noted that $p_{fi}^{(\mathrm{abs})}$ ($p_{fi}^{(\mathrm{stim})}$) is nothing but the Einstein B coefficient generalized to *nonstationary* fields. In next section we discuss how the coherent states of the radiation field in the presence of dielectrics and conductors can be produced.

V. EXPERIMENTAL EXCITATION OF THE RADIATION FIELD IN PRESENCE OF DIELECTRIC AND CONDUCTING SURFACES TO A COHERENT STATE

In Sec. IV we treated one-photon transitions in nonstationary fields and in particular the case when the initial state of the field was a coherent state.[8] In this section we describe how the radiation field can be produced in such a state. It is well known that a harmonic oscillator perturbed by an interaction of the form $f(t)a+f^*(t)a^\dagger$ ends up in a coherent state (with time-dependent amplitude) if the oscillator was initially in the ground state. In our realization of coherent states we will be guided by this fact. Let us assume that our electromagnetic field is perturbed by an external polarization $\vec{\mathcal{P}}$ and magnetization $\vec{\mathfrak{M}}$; i.e., the perturbing Hamiltonian is

$$H_{\mathrm{ext}}=-\int d^3r[\vec{\mathcal{P}}(\vec{r},t)\cdot\vec{E}(\vec{r},t)+\vec{\mathfrak{M}}(\vec{r},t)\cdot\vec{H}(\vec{r},t)], \tag{5.1}$$

where $\vec{E}$ and $\vec{H}$ are second quantized operators evolving according to the unperturbed Hamiltonian. In our notation, the linear response of $\vec{E}$ and $\vec{H}$ would be

$$\delta\langle E_i(\vec{r},\omega)\rangle=\sum_j\int d^3r'[\chi_{ijEE}(\vec{r},\vec{r}',\omega)\mathcal{P}_j(\vec{r}',\omega)+\chi_{ijEH}(\vec{r},\vec{r}',\omega)\mathfrak{M}_j(\vec{r}',\omega)], \tag{5.2}$$

$$\delta\langle H_i(\vec{r},\omega)\rangle=\sum_j\int d^3r'[\chi_{ijHE}(\vec{r},\vec{r}',\omega)\mathcal{P}_j(\vec{r}',\omega)+\chi_{ijHH}(\vec{r},\vec{r}',\omega)\mathfrak{M}_j(\vec{r}',\omega)]. \tag{5.3}$$

The Hamiltonian (5.1) has the special feature that one can study the exact time evolution of the system. The time-evolution operator is

$$U(t,t_0)=T\exp\left(-\frac{i}{\hbar}\int_{t_0}^t H_{\mathrm{ext}}(\tau)\,d\tau\right), \tag{5.4}$$

where T is the time-ordering operator. It is easily seen that $[H_{\mathrm{ext}}(t),H_{\mathrm{ext}}(\tau)]$ is a c number, and this fact enables one to evaluate (5.4) exactly using a generalization of Baker-Hausdorff identity.[9] Using this, (5.4) becomes

$$U(t,t_0)=\exp\left(-\frac{i}{\hbar}\int_{t_0}^t H_{\mathrm{ext}}(\tau)\,d\tau+\varphi(t)\right), \tag{5.5a}$$

$$\varphi(t)=-\frac{1}{2\hbar^2}\int_{t_0}^t dt_1\int_{t_0}^{t_1}dt_2[H_{\mathrm{ext}}(t_1),H_{\mathrm{ext}}(t_2)]$$

$$=-\varphi^*(t)\,. \tag{5.5b}$$

If initially the field was in vacuum state $|0\rangle$, then at time t it will be in a state

$$|\psi(t)\rangle=\exp\left(\frac{-i}{\hbar}\int_{t_0}^t H_{\mathrm{ext}}(\tau)\,d\tau+\varphi(t)\right)|0\rangle\,. \tag{5.6}$$

We now show that the state $|\psi\rangle$ is a coherent state. To see this we calculate

$$E_l^{(+)}(\vec{R},t)\,|\psi\rangle=[E_l^{(+)}(\vec{R},t),U(t,t_0)]\,|0\rangle\,, \tag{5.7}$$

since $E_l^{(+)}$ acting on vacuum gives zero. Next the identity[10]

$$[A,e^{-B}]=-\int_0^1 dy\,e^{-(1-y)B}[A,B]e^{-yB}, \tag{5.8}$$

leads to

$$[A,e^{-B}]=-[A,B]e^{-B}, \tag{5.9}$$

if $[A, B]$ is a c number and hence (5.7) reduces to

$$E_i^{(+)}(\vec{R}, t)|\psi\rangle = (i/\hbar)[E_i^{(+)}(\vec{R}, t), \int_{t_0}^{t} H_{\text{ext}}(\tau)\, d\tau]|\psi\rangle , \tag{5.10}$$

which we would write as

$$E_i^{(+)}(\vec{R}, t)|\psi\rangle = V_i^{(E)}(\vec{R}, t)|\psi\rangle . \tag{5.11}$$

Equations (5.10), (5.11) show that $|\psi\rangle$ as defined by (5.6) is an eigenstate of $E_i^{(+)}$ and hence a coherent state. Similarly, one can show

$$H_i^{(+)}(\vec{R}, t)|\psi\rangle = V_i^{(H)}(\vec{R}, t)|\psi\rangle , \tag{5.12}$$

where

$$V_i^{(H)}(\vec{R}, t) = (i/\hbar)[H_i^{(+)}(\vec{R}, t), \int_{t_0}^{t} H_{\text{ext}}(\tau)\, d\tau] . \tag{5.13}$$

Thus $|\psi\rangle$ is a coherent state in which the electric and magnetic fields have the amplitudes $V_i^{(E)}$ and $V_i^{(H)}$, respectively. From the definition (I.3.1) of positive and negative frequency parts, we have

$$E_i^{(+)}(\vec{R}, t) = (1/2\pi)\int E_i(\vec{R}, t')\delta_+(t' - t)\, dt', \tag{5.14}$$

which can be used to express the commutator of $E_i^{(+)}$ in terms of response as

$$\begin{aligned}[E_i^{(+)}(\vec{R}, t),\ & E_j(\vec{r}, \tau)] \\ &= (1/2\pi)\int [E_i(\vec{R}, t'), E_j(\vec{r}, \tau)]\, \delta_+(t' - t)\, dt' \\ &= (\hbar/\pi)\int \chi''_{ijEE}(\vec{R}, \vec{r}, t' - \tau)\delta_+(t' - t)\, dt' \\ &= (\hbar/\pi)\int \chi''_{ijEE}(\vec{R}, \vec{r}, t')\delta_+(t' + \tau - t)\, dt'\end{aligned}$$

and therefore

$$\int [E_i^{(+)}(\vec{R}, t), E_j(\vec{r}, \tau)]\, e^{i\omega(t-\tau)} d(t - \tau) = 2\hbar\chi''_{ijEE}(\vec{R}, \vec{r}, \omega)\eta(\omega) . \tag{5.15}$$

Consider now the amplitude $\mathcal{V}_i^{(E)}$ defined by

$$\begin{aligned}\mathcal{V}_i^{(E)}(\vec{R}, t) &= V_i^{(E)}(\vec{R}, t) + \text{c.c.} \\ &= (i/\hbar)[E_i(\vec{R}, t), \int_{t_0}^{t} H_{\text{ext}}(\tau)\, d\tau] .\end{aligned} \tag{5.16}$$

The Fourier transform of it is given by

$$\mathcal{V}_i^{(E)}(\vec{R}, \omega) = -\sum_j \int d^3r' [\chi_{ijEE}(\vec{R}, \vec{r}', \omega)\mathcal{P}_j(\vec{r}', \omega) + \chi_{ijEH}(\vec{R}, \vec{r}', \omega)\mathfrak{M}_j(\vec{r}', \omega)] , \tag{5.17}$$

where the definitions (I.2.4) and (I.2.5) of the response functions have been used. Comparing (5.2) and (5.17), we find that

$$\mathcal{V}_i^{(E)}(\vec{R}, \omega) = -\delta\langle E_i(\vec{R}, \omega)\rangle ; \tag{5.18}$$

i.e., the amplitude $\mathcal{V}_i^{(E)}$ of the coherent state is precisely the linear response of the variable E_i. Finally expressing $V_i^{(E)}$ (which is an analytic signal[11]) in terms of $\mathcal{V}_i^{(E)}$, we have

$$V_i^{(E)}(\vec{R}, t) = -(1/2\pi)\int_0^\infty d\omega\, e^{-i\omega t}\delta\langle E_i(\vec{R}, \omega)\rangle . \tag{5.19}$$

Similarly, the eigenvalue of $H^{(+)}$ is given by

$$V_i^{(H)}(\vec{R}, t) = -(1/2\pi)\int_0^\infty d\omega\, e^{-i\omega t}\delta\langle H_i(\vec{R}, \omega)\rangle . \tag{5.20}$$

Equations (5.19) and (5.20) are very basic relations relating the linear response to the amplitudes of the coherent states. $\delta\langle E_i\rangle$ represents the response to an applied polarization and magnetization. Experimentally, it is of course more convenient to use an electromagnetic field $\vec{e}$, $\vec{h}$ itself as an external probe, and hence we introduce the response function

$$\frac{\delta\langle E_i(\vec{r}, \omega)\rangle}{\delta e_j(\vec{r}', \omega)} = \chi_{ijEe}(\vec{r}, \vec{r}', \omega). \tag{5.21}$$

The external field can be taken as equivalent to

$$\begin{aligned}\vec{\mathcal{P}}(\vec{r}, \omega) &= -(1/4\pi)e(\vec{r}, \omega) \\ \vec{\mathfrak{M}}(\vec{r}, \omega) &= (1/4\pi ik_0)\nabla \times \vec{e}(\vec{r}, \omega), \quad k_0 = \omega/c .\end{aligned} \tag{5.22}$$

Equations (5.22) can be used to obtain the relation between $\chi_{ijEE}\chi_{ijEH}$ and χ_{ijEe}. One has

$$\frac{\delta\langle E_i(\vec{r}, \omega)\rangle}{\delta e_j(\vec{r}', \omega)} = \sum_i \int d^3r'' \left(\frac{\delta\langle\langle E_i(\vec{r}, \omega)\rangle\rangle}{\delta\mathcal{P}_i(\vec{r}'', \omega)} \frac{\delta\mathcal{P}_i(\vec{r}'', \omega)}{\delta e_j(\vec{r}', \omega)} + \frac{\delta\langle E_i(\vec{r}, \omega)\rangle}{\delta\mathfrak{M}_i(\vec{r}'', \omega)} \frac{\delta\mathfrak{M}_i(\vec{r}'', \omega)}{\delta e_j(\vec{r}', \omega)} \right), \tag{5.23}$$

which on simplification leads to

$$\chi_{ijEe}(\vec{r}, \vec{r}', \omega) = -(1/4\pi)\chi_{ijEE}(\vec{r}, \vec{r}', \omega) - \frac{1}{4\pi ik_0}\sum_{im}\epsilon_{imj}\frac{\partial}{\partial x'_m}\chi_{iiEH}(\vec{r}, \vec{r}', \omega) . \tag{5.24}$$

In view of (5.17), (5.22), and (5.24), (5.19) becomes

$$V_i^{(E)}(\vec{R}, t) = -\frac{1}{2\pi}\int_0^\infty e^{-i\omega t}\int \sum_j \chi_{ijEe}(\vec{R}, \vec{r}', \omega) \times e_j(\vec{r}', \omega)\, d^3r'\, d\omega, \tag{5.25}$$

which shows how the amplitude of the coherent state is related to the incident field $\tilde{e}(\vec{r}, \omega)$. In particular, for an incident monochromatic field

$$\tilde{e}(\vec{r}, \omega) = 2\pi e(\vec{r})\delta(\omega - \omega_0) + 2\pi e^*(\vec{r})\delta(\omega + \omega_0), \tag{5.26}$$

the amplitude of the coherent state is

$$V_l^{(E)}(\vec{R}, t) = -e^{-i\omega_0 t} \int \sum_j \chi_{lj Ee}(\vec{R}, \vec{r}', \omega_0) e_j(\vec{r}')\, d^3r'. \tag{5.27}$$

It is moreover obvious that (5.27) (apart from a minus sign) is nothing but an alternative way of writing the Fresnel formulas. This amplitude could now be substituted into (4.9) and (4.17) to obtain the transition rates.

Note added in proof. Since this work was done we have learned of the very interesting work of Baltes and co-workers on the corrections to Planck's law of radiation in finite cavities [H. P. Baltes and F. K. Kneubuhl, Helv. Phys. Acta 45, 481 (1972); H. P. Baltes, Phys. Rev. A 6, 2252 (1972); H. P. Baltes and E. R. Hilf, *Spectra of Finite Systems* (Bibliographical Institute, Mannheim, West Germany, 1974)]. The approach of Baltes and co-workers is, however, entirely different. I would like to thank Professor H. P. Baltes for sending many of his reprints and preprints on this subject.

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[1]G. S. Agarwal, first preceding paper, Phys. Rev. A 10, 230 (1974); second preceding paper, 10, 243 (1974); these papers will be referred to as I and II, respectively. Equations referring to these papers will be preceded by I and II, respectively, e.g., Eq. (I. 2.10).

[2]C. K. Carniglia, L. Mandel, and K. H. Drexhage, J. Opt. Soc. Am. 62, 479 (1972).

[3]C. K. Carniglia and L. Mandel, Phys. Rev. D 3, 280 (1971).

[4]Cf. R. Balian and C. Bloch, Ann. Phys. (N.Y.) 64, 271 (1971); see also T. H. Boyer, Phys. Rev. 174, 1764 (1968) and K. M. Case and S. C. Chiu, Phys. Rev. A 1, 1170 (1970).

[5]Cf. P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953), Vol. I, p. 466.

[6]I would like to thank Professor E. Wolf, who several years back had drawn my attention to this problem.

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[8]For a treatment of coherent states see, for example, J. R. Klauder and E. C. G. Sudarshan, *Fundamentals of Quantum Optics* (Benjamin, New York, 1968); R. J. Glauber, Phys. Rev. 131, 2766 (1963).

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[10]See, e.g., R. M. Wilcox, J. Math. Phys. 8, 962 (1967).

[11]See, e.g., M. Born and E. Wolf, *Principles of Optics* (Pergamon, London, 1970), Chap. X.