Theory of spontaneous and stimulated emission of electromagnetic waves in unidimensionally inhomogeneous media and resonators

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Instead of the Lorentz gauge, which leads to a system of coupled equations, the authors, in solving the Maxwell equations, use another vector-potential gauge that leads to a set of independent Schrödinger-type equations. It is shown that expansion of the field in terms of the damped modes is unjustifiable in the case of low-Q resonators, owing to the incompleteness and nonorthogonality of such a set. A complete set of strictly orthogonal stationary modes is introduced for the unidimensionally inhomogeneous medium—in particular, for the resonator with an arbitrary Q (in the absence of absorption in the mirrors). The electromagnetic field is quantized on the basis of these modes, and the electric and magnetic dipole phototransitions of the radiating center in the medium are considered. A relation between the probabilities of phototransitions in inhomogeneous and unbounded homogeneous media is found. These probabilities differ completely in their magnitudes and their frequency and angular dependences.

1. INTRODUCTION

In the present paper we consider electromagnetic waves in an isotropically polarizing medium of permittivity ϵ that depends only on the x coordinate. To this case also pertain multilayer media described by a step permittivity function $\epsilon(x)$. Alternation of dielectric and metallic layers, i.e., the case of resonators with mirrors and waveguides, is admissible. It is assumed that the wave-frequency dependence of ϵ is sufficiently weak. Instead of the Lorentz condition or the condition div $\mathbf{A} = 0$, we solve the Maxwell equations with the aid of another gauge for the vector potential $\mathbf{A}$ that allows us to reduce the system of Maxwell equations to a single equation of the type of the one-dimensional Schrödinger equation.

To quantize the electromagnetic field and determine the photon emission probabilities, it is necessary, as is well known, to expand $\mathbf{A}(\mathbf{r}, t)$ in terms of a complete set of orthogonal functions $\mathbf{A}_\nu(\mathbf{r})$:

$$\mathbf{A}(\mathbf{r}, t) = \sum_\nu [q_\nu(t)\mathbf{A}_\nu(\mathbf{r}) + q_\nu^*(t)\mathbf{A}_\nu^*(\mathbf{r})]. \quad (1)$$

The expansion can be carried out in terms of any complete set of basis functions $\mathbf{A}_\nu(\mathbf{r})$. In order, however, for the functions $\mathbf{A}_\nu(\mathbf{r})$ to be photon form factors, i.e., to be normal modes in the medium under consideration, the energy of the electromagnetic field must be expressible in terms of the products $q_\nu^* q_{\nu'}$. This happens only if the $\mathbf{A}_\nu(\mathbf{r})$ are orthogonal with respect to integration over $\mathbf{r}$ in a completely defined volume and for a completely defined weight factor under the integral sign (see (19)). In the case of an inhomogeneous medium this requirement is not satisfied by, for example, basis functions of the type $\mathbf{A}_\nu(\mathbf{r}) = Ce^{i\kappa\mathbf{r}}$, which are used in field quantization in a homogeneous medium and which correspond to photons with a definite momentum $\hbar\kappa$. In an inhomogeneous medium the photon form factors will be of different form. The corresponding photons will not be characterized by a definite momentum, may not be transversely polarized, and the probability of emission of such photons by, for example, an impurity center will turn out to be essentially dependent on the macroscopic location of the center, and its frequency and angular dependences will be completely different from those that obtain in the emission by the same center located in a homogeneous medium.

It will be shown that in general the $\mathbf{A}_\nu$ do not possess the requisite orthogonality properties in the case of modes that are damped in time, i.e., in the case of modes with a complex frequency ω. In this connection, let us note that the field quantization and the probability of emission into the resonator are nevertheless considered in many published papers with the aid of damped modes. Thus, for example, in[1] Purcell considers the probability of emission into a resonator with a finite Q. This probability turned out to be Q dependent. The dependence of the probability of emission into a resonator on the resonator's Q factor has also been obtained in other papers[2-11]. For example, to allow for losses in the resonator, Senitzky[10] and Faĭn and Khanin[11] introduce a special system–a thermostat that weakly interacts with the radiation. In the zeroth approximation this interaction is neglected and the field is decomposed in terms of undamped modes and quantized, and then in the first approximation this interaction is introduced as a perturbation and leads to weak damping. Such an approach is correct for high-Q resonators. Other authors (see, for example,[8,9]) decompose the vector potential straightway in terms of weakly damped modes. In this case the above-mentioned necessary orthogonality of the $\mathbf{A}_\nu$ is only slightly violated, and, therefore, hopefully, the resulting errors are small.

The nonrigorousness of the results obtained when nonstationary modes are used has been pointed out by Faĭn and Khanin (see[11]).

Any of the above-mentioned methods of allowing for damping cannot be justifiably used if the Q factor of the resonator is not very high. We shall, however, be interested in the cases of the often used (see, for example,[12,13]) thinly laminated resonators, when, as a result of the small intermirror distance, the Q factor of the resonator is roughly equal to unity (i.e., $Q \sim 1$). In this case the damped modes are already greatly nonorthogonal, and the expansion of $\mathbf{A}(\mathbf{r}, t)$ in terms of them is inadmissible. Therefore, below, independent of the value of Q, we shall use undamped basis functions

with strictly real frequencies—the modes of a "super-resonator," which arises upon the inclusion of an infinite empty space in the orthonormalization volume.

Using such basis functions, we shall compute the probabilities of emission of light by impurity atoms located in an optically inhomogeneous medium, and compare them with the emission probabilities for the same atoms located in a homogeneous medium. As an example, we consider emission into a resonator and a waveguide consisting of a multilayer system located between two metallic mirrors, one of which is perfectly reflecting and the other semitransparent. The modes of one such resonator have been computed in detail in[14].

2. THE REDUCTION OF THE SYSEM OF MAXWELL EQUATIONS TO A ONE-DIMENSIONAL SCHRÖDINGER-TYPE EQUATION. THE ORTHOGONALITY OF THE MODES

The magnetic permeability of the medium is assumed to be equal to unity. Assuming

$$\mathbf{H}=\operatorname{rot}\mathbf{A}, \tag{2}$$

we satisfy the equation div H = 0. Since the coefficients of the maxwell equations do not depend on y, z, and t, the solution can be sought in the form

$$\begin{aligned}&\mathbf{E},\ \mathbf{H}\sim\exp\{i(\mathbf{k}_\tau\mathbf{r}-\omega t)\},\\ &\mathbf{A}(\mathbf{r},t)=\mathbf{f}(x)\exp\{i(\mathbf{k}_\tau\mathbf{r}-\omega t)\},\end{aligned} \tag{3}$$

where $\mathbf{k}_\tau$ is a two-dimensional vector with components k_y, k_z; $\mathbf{k}_\tau\cdot\mathbf{r}=k_y y+k_z z$. On other vectors, the index τ will indicate the component in the direction $\boldsymbol{\tau}=\mathbf{k}_\tau/|\mathbf{k}_\tau|$. Generalizing the term used in the case of multilayer media, we shall call the plane passing through $\mathbf{k}_\tau$ and the x axis the plane of incidence.

The remaining Maxwell equations reduce to three equations:

$$\operatorname{rot}(\mathbf{E}-i\omega\mathbf{A}/c)=0, \tag{4}$$

$$\operatorname{rot}\operatorname{rot}\mathbf{A}=-i\omega\mathbf{D}/c, \tag{5}$$

$$\mathbf{D}=\varepsilon(x)\mathbf{E}, \tag{6}$$

for the equation

$$\operatorname{div}\mathbf{D}=0 \tag{7}$$

follows from (5). As usual, $\epsilon(x)$ includes the term with the high-frequency electrical conductivity of the medium.

We introduce in place of the Lorentz condition the following gauge A:

$$\operatorname{div}(\mathbf{E}-i\omega\mathbf{A}/c)=0. \tag{8}$$

From this equation and (4) follows the relation

$$\mathbf{E}=i\omega\mathbf{A}/c. \tag{9}$$

From (5), (6), and (9) we obtain the equation determining A:

$$\Delta\mathbf{A}-\nabla(\nabla\mathbf{A})+\varepsilon(x)\omega^2\mathbf{A}/c^2=0. \tag{10}$$

The Polarization p = 1 (A Perpendicular to the Plane of Incidence)

One of the solutions to (10) possesses the polarization $\mathbf{A}\parallel\mathbf{s}$, where s is a unit vector perpendicular to the x axis and the vector $\mathbf{k}_\tau$. Indeed, in this case it follows from (7) that div A = 0 (the wave is a transverse wave), and the three scalar equations that follow from (10) upon allowance for (3) reduce to a single scalar equation of the Schrödinger type:

$$\frac{\partial^2 f_s}{\partial x^2}+\frac{2m}{\hbar^2}[\mathscr{E}-u(x)]f_s=0, \tag{11}$$

where

$$\mathscr{E}=-\hbar^2\mathbf{k}_\tau^2/2m,\qquad u(x)=-\hbar^2\omega^2\varepsilon(x)/2mc^2. \tag{12}$$

The finite solutions to (10) correspond to real k_τ, i.e., to $\mathscr{E}<0$. Besides Eq. (11), $f_S(x)$ and df_S/dx should satisfy the finiteness and continuity requirements in the same way as the wave function does in quantum mechanics. In solving (11), we can use all the various methods that have been worked out in quantum mechanics, as well as the theorems on the properties of the solutions. The function $\epsilon(x)$ is assumed to be real.

If the function u(x) tends to a constant value u_1 as $x\to-\infty$ and we have $u(x)\to u_2<u_1$ as $x\to\infty$, then, as in quantum mechanics, we have the following possibilities:

1) For $\mathscr{E}<u_2$, $f_S(x)$ attenuates exponentially as $x\to\pm\infty$ (total internal reflection), and the spectrum of the eigenvalues $\mathscr{E}$ is discrete. This corresponds to wave-guide modes (the wave propagates in the direction $\mathbf{k}_\tau$) for which there exist at fixed ω a number of selected discrete values of $\mathbf{k}_{\tau j}^2$, i.e., for which there exist a number of dispersion branches

$$\mathbf{k}_{\tau j}^2=\mathbf{k}_{\tau j}^2(\omega); \tag{13}$$

2) For $u_1>\mathscr{E}>u_2$ the function $f_S(x)$ attenuates exponentially only when $x\to-\infty$: it oscillates undamped in the region $x\to\infty$. At fixed ω the spectrum of $\mathscr{E}$ or k_τ^2 is continuous, i.e., there is no functional relationship between ω and k_τ^2. Such solutions represent a wave that is a running wave in the direction $\mathbf{k}_\tau$, but a standing wave in the x direction.

3) For $\mathscr{E}>u_1$ the function $f_S(x)$ oscillates undamped as $x\to\infty$, as well as when $x\to-\infty$. The spectrum of $\mathbf{k}_\tau^2$ is continuous at fixed ω. The wave can be a running wave in the x direction as well. In contrast to the two preceding cases, the component of the Poynting vector in the x direction can be different from zero.

The Polarization p = 2 (A Lies in the Plane of Incidence)

We shall seek the second type of solutions to (10) under the assumption that $\mathbf{A}\perp\mathbf{s}$, i.e., that A lies in the plane of incidence. Then Eq. (10) then reduces to a system of two equations for A_x and A_τ. But the second of these equations can be replaced by Eq. (7), which follows from the two equations. Using (6), (9), and (3), we can rewrite Eq. (7) in the form

$$\frac{1}{\varepsilon}\frac{d\varepsilon}{dx}f_x+\frac{df_\tau}{dx}+ik_\tau f_\tau=0. \tag{14}$$

Now div $\mathbf{A}\neq0$, i.e., the wave is not transverse.

Equation (10) for f_x with allowance for (14) and (3) has the form

$$\frac{d^2f_x}{dx^2}-k_\tau^2f_x+\frac{d}{dx}\left(\frac{1}{\varepsilon}\frac{d\varepsilon}{dx}f_x\right)+\frac{\omega^2}{c^2}\varepsilon f_x=0. \tag{15}$$

Setting

$$f_x=X(x)/\varepsilon^{1/2}(x), \tag{16}$$

we obtain for the new unknown function X the equation

$$\frac{d^2X}{dx^2}+\frac{2m}{\hbar^2}[\mathscr{E}-v(x)]X=0, \tag{17}$$

where $\mathscr{E}$ is given, as before, by the formula (12), while

$$v(x)=-\frac{\hbar^2}{2m}\left[\frac{1}{2\varepsilon}\frac{d^2\varepsilon}{dx^2}-\frac{3}{4\varepsilon^2}\left(\frac{d\varepsilon}{dx}\right)^2+\frac{\omega^2}{c^2}\varepsilon\right]; \tag{18}$$

X and dX/dX should be finite and continuous. In spite of the difference between the expressions for v(x) and u(x), their limiting values as $x\to\pm\infty$ coincide if $\epsilon(x)$ tends to constant limits as $x\to\pm\infty$. Therefore, the con-

ditions for the above-considered three types of solutions 1), 2), and 3) are the same for both polarizations

After determining X and f_X from (17) and (16), we can derive f_τ from (14).

We shall denote the obtained solutions (modes) by $\mathbf{A}_\nu(\mathbf{r})$, where ν is a multicomponent index comprising of ω, $\mathbf{k}_\tau$, and p. The general solution to (10) is a linear combination of the $\mathbf{A}_\nu(\mathbf{r})$ with the same value of ω^2, but with all possible values of $\mathbf{k}_\tau$ and p.

Proceeding to the consideration of the orthogonality of the modes, we note that the energy of the electromagnetic field will be expressible in terms of $q^+_\nu q_{\nu'}$ only if the orthogonality relation has the following form:

$$\int\int\int \mathbf{A}_{\nu'}^{*}\mathbf{A}_\nu \varepsilon(x)\,dx\,dy\,dz = \text{const}\cdot\delta_{\nu'\nu}. \tag{19}$$

Here the integration should be carried out over the basic cyclicity region with the period L, in which the energy of the electromagnetic field will be computed below. Below we shall, for convenience, normalize $\mathbf{A}_\nu$ so that the constant in (19) is equal to

$$2\pi c^2\hbar/\omega. \tag{20}$$

The modes with different $\mathbf{k}_\tau$ are necessarily orthogonal, owing to the orthogonality of the Fourier exponential functions (see (3)). For coincident $\mathbf{k}_\tau$, but different p, the modes are orthogonal, owing to the perpendicularity of $\mathbf{A}^*_{\nu'}$ and $\mathbf{A}_\nu$.

It remains to show the orthogonality of the modes with different ω. Using the usual procedure, we can derive (19) from (10), provided the operator $\nabla^2 - \nabla(\nabla\ldots)$ is self-adjoint, a condition which is satisfied only in the case of a class of functions which, together with their first derivatives with respect to x, have the same values at the points x_1yz and x_2yz, where x_1 and x_2 are the limits of the x integration in (19). If the $\mathbf{A}_\nu$ belong to this class of functions, then the x component of the Poynting vector also has the same value at the points x_1 and x_2. This means that the energy of the electromagnetic field does not flow out from the basic region. Consequently, for real $\epsilon(\mathbf{x})$, when the damping in time of a mode can be due only to energy leakage through the walls of the resonator, the damped modes are nonorthogonal.

For a complex $\epsilon(\mathbf{x})$, however, the orthogonality relations (19) cannot, in general, be satisfied, since it follows from (19) that

$$\int\int\int (\varepsilon^*-\varepsilon)\mathbf{A}_{\nu'}^{*}\mathbf{A}_\nu\,dx\,dy\,dz = 0$$

for any ν and ν', which implies that $\epsilon^*(\mathbf{x}) - \epsilon(\mathbf{x}) = 0$. Thus, the modes that for any reason attenuate or intensify in time (i.e., that have complex ω) do not satisfy (19).

3. THE PROBABILITY OF RADIATION EMISSION BY MOLECULE LOCATED IN AN INHOMOGENEOUS MEDIUM

Using (1), (19), and (20), we can, in the same way as is done in the case of homogeneous media, derive for the electromagnetic-radiation energy operator the expression

$$\mathcal{H} = \sum_\nu \hbar\omega_\nu q_\nu^+ q_\nu, \tag{21}$$

where q_ν and q^+_ν are the standard photon annihilation and creation operators for the mode ν. The eigenvalues n_ν of the operator $q^+_\nu q_\nu$ is the number of photons in this mode.

The interaction-energy operator for the interaction between the radiation and the electrons of the emitting complex, which consists of the impurity molecule and the molecules of the dielectric that surround and substantially interact with it, has the form

$$\mathcal{H}_{int} = -\frac{e}{mc}\sum_i \left[\, \mathbf{A}(\mathbf{r}_i)\,\mathbf{p}_i + \frac{\hbar}{2}\boldsymbol{\sigma}_i \operatorname{rot}\mathbf{A}(\mathbf{r}_i) \,\right]. \tag{22}$$

Here e and m are the electron charge and mass; $\mathbf{p}_i$, $\mathbf{r}_i$, and $\boldsymbol{\sigma}_i$ are the momentum, coordinate, and dimensionless-spin operators of the i-th electron.

Let $|a\rangle$ and $|b\rangle$ be the stationary vibronic states of the complex in the absence of an electromagnetic field. The matrix element of the phototransition $|b\rangle \to |a\rangle$ accompanied by the emission of a photon into the mode ν is equal to

$$\langle \mathcal{H}_{int}\rangle \equiv \langle a, n_\nu+1\,|\,\mathcal{H}_{int}\,|\,b, n_\nu\rangle = (n_\nu+1)^{1/2}\hat{\mathcal{L}}(\mathbf{R})A_\nu(\mathbf{R}), \tag{23}$$

where R is the coordinate of the center of the complex, while

$$\mathcal{L}(\mathbf{R}) \equiv -\frac{e}{mc}\sum_i \left\{ \langle a|\mathbf{p}_i|b\rangle + \langle a|\,(\mathbf{r}_i-\mathbf{R},\,\nabla_{\mathbf{R}})\,\mathbf{p}_i|b\rangle + \frac{\hbar}{2}\langle a|\boldsymbol{\sigma}_i|b\rangle \operatorname{rot}_{\mathbf{R}} \right\}. \tag{24}$$

Here the first term represents the dipole, the second the quadrupole, and the third the magnetic-dipole contributions to the matrix element. It is assumed that the wavelength of the electromagnetic wave is much greater than the dimensions of the complex.

The linear dimensions of the complex is usually of the order of several lattice constants. Let us assume that $\epsilon(\mathbf{x})$ and the other properties of the medium do not change appreciably over such a distance. Then the quantum-mechanical states $|a\rangle$ and $|b\rangle$ of the complex and the operator (24) are the same as in an infinite homogeneous dielectric with $\epsilon = \epsilon(\mathbf{R})$. In the case of a multilayer medium the thickness of the layer in which the vibronic complex is located should be much greater than the dimensions of the complex. The matrix element (23) of the phototransition will then differ from that of the case of the infinite homogeneous medium only in the value of the factor $\mathbf{A}_\nu(\mathbf{R})$. The latter fact will be extremely important below, and will allow us to relate the probabilities of phototransitions in inhomogeneous and homogeneous media.

The probability per second of the $|b\rangle \to |a\rangle$ phototransition accompanied by the emission of a photon into a definite mode ν is equal to

$$P_\nu = \frac{2\pi}{\hbar}\overline{\rho_{Ea}|\langle \mathcal{H}_{int}\rangle|^2}, \tag{25}$$

where ρ_{Ea} is the number of final states of the vibronic complex per unit energy interval. The emission of a given photon $\hbar\omega_\nu$ is possible from the set of different initial states $|b\rangle$. It is assumed that prior to the phototransition there is a definite probability for the complex to be in each of the initial states $|b\rangle$, a probability which depends only on the energy E_b of the state in question and which is determined by equilibrium statistics. The point is that each of the states $|b\rangle$ is assumed to consist of one definite nonequilibrium electronic state combined with different equilibrium vibrational states.

It is also possible for a given frequency ω_ν to be emitted in transitions from a $|b\rangle$ state to several different $|a\rangle$ states. Therefore, to obtain the total prob-

ability of emission of a ν photon, the quantity (25) is averaged over the initial $|b\rangle$ states and summed over the final $|a\rangle$ states for which $E_b - E_a = \hbar\omega_\nu$. The combined averaging and summation operation is denoted in (25) by the top bar. Notice that a formula of the type (25) for the transition probability can be proved only if the final (or initial) state belongs to the continuous energy spectrum, and if ρ and $|\langle\mathscr{H}_{int}\rangle|^2$ are sufficiently smooth functions of the quantum numbers of the final states. Usually, in radiation theory, in the case of a homogeneous medium, the states $|a\rangle$ and $|b\rangle$ of the vibronic subsystem are exactly specified, and the continuity of the spectrum of the photon subsystem is used. Accordingly, the photon-level density figures in a formula of the type (25). However, in the case of an optically inhomogeneous medium this traditional approach is, generally speaking, inapplicable. The point is that an inhomogeneous medium can form a small optical "resonator" of dimensions comparable to the wavelength of light. In this case the energy spectrum of the photons becomes substantially discrete if the "resonator" walls are perfect reflectors. If, on the other hand, the "resonator" walls are weakly penetrable by light, then the photon energy spectrum remains continuous, but the amplitude $\mathbf{A}_\nu(\mathbf{r})$ of the field inside the "resonator" turns out to be not a smooth but a spiked function of ω_ν[13,14]. In this case $|\langle H_{int}\rangle|^2$ also ceases to be a smooth function of ω_ν, and we cannot, using the continuity of the photon spectrum, prove a formula like (25).

For this reason, in deriving the formula (25) in the present paper, we specify instead the emitted-photon index ν (the mode index) exactly and use the continuity of the energy spectrum of the vibronic subsystem. Accordingly, in (25) figures the level density ρ_{Ea} of this subsystem and not the photon density. It is assumed that the energy spectrum of the vibronic subsystem is quasi-continuous, owing to the quasi-continuity of the vibrational spectrum.

Even if the medium is a multilayer medium not only optically, but also acoustically, and if the thickness of a layer is of the order of the wavelength of light, then this thickness still remains much greater than the wavelength of the actual phonons. Therefore, the energy spectrum and the wave functions of the vibrations in this layer can be assumed to be the same as in an unbounded homogeneous medium.

The spectrum of the mechanical (vibronic) subsystem is continuous in many cases–in particular, in condensed phases, in which an optically inhomogeneous medium is usually realized. There are, however, cases in which this spectrum is substantially discrete, e.g., in the case of an atomic gas or the very popular "two-level model" that has been considered in many papers. A distinctive feature of such mechanical systems is the impossibility of heat release and relaxation in them after a phototransition. In these cases the formula (25) is inapplicable: the density ρ_{Ea} figuring in it has no meaning. The subsequent results of the present paper are also inadmissible: we cannot, in particular, introduce the concept of phototransition probability per unit time in the case of an absolutely perfect resonator[3].

Returning to the consideration of our case, we note that the operator (24) will, after being integrated over $\mathbf{r}_1$, cease to depend on $\mathbf{R}$: it will contain only derivatives with respect to R_x, R_y, and R_z. If the dipole transition is allowed, then we can retain only the first term in (24). Then, using (23)–(25), we can represent P_ν as a bilinear form in $\mathbf{A}_{\nu l}(l = x, y, z)$:

$$P_\nu = (n_\nu+1)\sum_{l,k=x,y,z} D_{lk}(\mathbf{R})A_{\nu l}^*(\mathbf{R})A_{\nu k}(\mathbf{R}), \tag{26}$$

where

$$D_{lk} \equiv \frac{2\pi}{\hbar}\left(\frac{e}{mc}\right)^2 \overline{\rho_{Ea}\left\langle a\left|\sum_i p_{il}\right|b\right\rangle\left\langle a\left|\sum_i p_{ik}\right|b\right\rangle^*}.$$

If the dipole transition is forbidden, then the second and third terms in (24) must be retained, and P_ν can be represented as a bilinear form in $\partial\mathbf{A}_{\nu l}/\partial R_k$:

$$P_\nu = (n_\nu+1)\sum_{jlkm} D_{jlkm}(\mathbf{R})\frac{\partial A_{\nu l}^*}{\partial R_j}\frac{\partial A_{\nu m}}{\partial R_k}. \tag{27}$$

Let us emphasize again that the tensors $D_{lk}(\mathbf{R})$ and $D_{jlkm}(\mathbf{R})$ are the same as in the case of a homogeneous medium with the same specific properties that the inhomogeneous medium possesses at the point $\mathbf{R}$; they do not depend on the form of the inhomogeneity of the medium, the parameters of the resonator, the form of the modes, etc. Therefore, we can take their values from experiments on spontaneous luminescence in homogeneous, unbounded media.

In the case of complexes that are, on the average, optically isotropic, or that possess cubic symmetry, $D_{lk} = \delta_{lk}D(\mathbf{R})$, and (26) assumes the form

$$P_\nu = (n_\nu+1)D(\mathbf{R})|\mathbf{A}_\nu(\mathbf{R})|^2. \tag{28}$$

It is convenient to write the experimental expression for the probability per second of spontaneous emission of a photon of any polarization and any direction of emission in the frequency range $d\omega$ for a homogeneous medium in the form

$$dW = \varphi(\omega)d\omega/\tau. \tag{29}$$

If for $\varphi(\omega)$ we use the normalization relation

$$\int_0^\infty \varphi(\omega)d\omega = 1,$$

then τ is the mean lifetime of an excited complex in an infinite homogeneous dielectric. At the same time, this same quantity dW can be derived from the formula (28) (for $n_\nu = 0$), in which the A_ν's should, in the case of a homogeneous medium, be replaced by a plane waves normalized according to (19):

$$dW = 2d\omega\int d\Omega P_\nu\rho^\infty(\omega) = \frac{2\hbar\omega D}{\pi c}\varepsilon^{1/2}(\mathbf{R})d\omega. \tag{30}$$

Here $\rho^\infty(\omega)d\omega d\Omega$ is the number of modes in the frequency range $d\omega$ that have the same polarization and the direction of whose wave vector lies inside the solid angle $d\Omega$ in the homogeneous medium. Equating the right-hand sides of (30) and (29), we find

$$D = \pi c\varphi(\omega)/2\hbar\omega\tau\varepsilon^{1/2}(\mathbf{R}). \tag{31}$$

Now, knowing D, we can use the formula (28) for any inhomogeneous medium (or resonator), substituting into it only the $\mathbf{A}_\nu(\mathbf{R})$ computed for this medium.

Let us consider the example of a multilayer system bounded on one side by a perfectly reflecting mirror and, on the other, by an infinite vacuum (the case 2 of the preceding section). Let the emitting complex be located in one of the layers with ϵ = const > 0. The quantity $\mathbf{A}_\nu(\mathbf{r})$ in this layer, as well as in a vacuum, has the form

$$\mathbf{A}_{\nu s} = \exp\{i\mathbf{k}_\tau\mathbf{r}\}\mathbf{B}\sin(kx+\delta_1'), \quad p=1;$$

$$A_{\nu x}=\exp\{i\mathbf{k}_\tau\mathbf{r}\}B_x\cos(kx+\delta_2'),$$
$$|\mathbf{B}|^2=|B_x|^2(1+k^2/k_\tau^2),\quad p=2. \tag{32}$$

For the expression for $A_{\nu\tau}$, see (14). Of course, the values of the constants B, δ', and k in the dielectric layer are different from the values in a vacuum (see[11]).

To use Eq. (28), it is sufficient to know the ratio of the wave amplitude B_{di} in the indicated layer to the amplitude B_{vac} in a vacuum:

$$|B_{di}|^2|B_{vac}|^2\equiv\Phi(\omega,\vartheta). \tag{33}$$

Here ϑ is the angle of incidence of the ray in the vacuum:

$$\vartheta=\operatorname{arctg}(k_\tau/k)_{vac},\quad 0\leqslant\vartheta\leqslant\pi/2.$$

In fact, we find from (19) that

$$|B_{vac}|^2=4\pi c^2\hbar/\omega L^3, \tag{34}$$

where L is the period of the fundamental cyclicity region. Substituting (14) and (31)–(34) into (28), we finally obtain

$$P_\nu=\frac{\pi^2c^3\varphi(\omega)\Phi(\omega,\vartheta)}{\omega^2\tau\varepsilon^{1/2}L^3}[1+G\cos 2(kR_x+\delta')](n_\nu+1); \tag{35}$$

where

$$G=1\text{ for }p=1,\quad G=G_0\equiv 2\varepsilon^{-1}\sin^2\vartheta-1\text{ for }p=2. \tag{36}$$

The number of modes of the type 2) with a given polarization and with a wave vector (k_{vac}, k_y, k_z) whose direction lies within the solid angle $d\Omega$ is equal to

$$\rho(\omega)\,d\omega\,d\Omega=\frac{L^3\omega^2}{4\pi^3c^3}\,d\omega\,d\Omega. \tag{37}$$

Using (35) and (37), we obtain the probability per second of spontaneous emission of a photon of given polarization in the frequency range $d\omega$ and in the directions $d\Omega$:

$$P(\omega,\vartheta)\,d\omega\,d\Omega=P_\nu\rho(\omega)\,d\omega\,d\Omega=\frac{\varphi(\omega)\Phi(\omega,\vartheta)}{4\pi\tau\varepsilon^{1/2}}[1+G\cos 2(kR_x+\delta')]\,d\omega\,d\Omega. \tag{38}$$

If we average (35) and (38) over the various equally probable positions **R** of the emitting complex in the dielectric layer, then the term with the cosine vanishes, and we obtain the important formulas (20) and (21) of the paper[13], where they are given without proof.

The detailed derivation of the function $\Phi(\omega,\vartheta)$ is given in[14] for the case of an infinite dielectric layer located between two metallic mirrors, one of which is perfectly reflecting and the other is semitransparent. In this case, as well as in the other cases when the multilayer system constitutes some high-Q resonator, Φ as a function of ω and ϑ has a pinnacled character[13, 14]: in the neighborhood of the chosen discrete values of ω (or ϑ), $\Phi\gg 1$, i.e., $|B_{di}|\gg|B_{vac}|$. The corresponding photons are localized predominantly in the dielectric layer and, according to (35), are emitted (absorbed) with a probability much greater than when the same is located in an infinite homogeneous dielectric. For the other values of ω (or ϑ), $\Phi(\omega,\vartheta)\ll 1$, and the corresponding photons almost do not penetrate the dielectric layer and are emitted (absorbed) with a probability much smaller than when the complex is located in an infinite homogeneous dielectric.

Comparison of the formulas (38) and (29) shows what enormous anisotropy can be introduced into the emission probability by the location of layers with a different ϵ next to the layer under consideration, i.e., the layer containing the emitting complexes, and how significantly this can change the frequency dependence of the emission probability.

If the phototransition is forbidden in the dipole approximation, and, say, the magnetic dipole contribution to the transition matrix element predominates, then for emitting complexes that are isotropic on the average, or that possess cubic symmetry, (27) reduces to

$$P_\nu=D'\operatorname{rot}\mathbf{A}_\nu^*\operatorname{rot}\mathbf{A}_\nu, \tag{39}$$

i.e., the tensor D_{jklm} reduces to the single constant D', which can also be determined by equating the expression obtained from (39) for dW for the case of the infinite homogeneous medium to the experimental expression (29). We find as a result that

$$D'=\pi c^3\varphi(\omega)/2\varepsilon^{3/2}\hbar\omega^3\tau. \tag{40}$$

For the above-mentioned multilayer system that is bounded on one side by a perfectly reflecting mirror and, on the other, by an infinite vacuum, we can with the aid of (14), (32), (34), (37), (39), and (40) in the case of magnetic-dipole transitions, rederive the formulas (35) and (38), but with different values of G: instead of (36) we now have $G = G_0$ for $p = 1$ and $G = 1$ for $p = 2$. After being averaged over **R**, the terms with G vanish, and we again obtain the formulas (20) and (21) of[13], this time for magnetic-dipole transitions.

The probability of photon emission into modes of the type 1) and 3) should be computed with the same values of D and D' (see the preceding section of the paper).

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