ATOMIC RADIATION IN AN ANISOTROPIC MEDIUM

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The electromagnetic field in an optically transparent nonmagnetic anisotropic medium is quantized, and a solution is obtained for the problem of atomic radiation. Dipole, magnetic dipole, and quadrupole radiation are considered. The angular distribution of radiation and the total probability of emission per unit time of a quantum with polarization corresponding to either the ordinary or extraordinary wave are determined for the particular case of a uniaxial crystal. The polarization of radiation from a uniaxial ruby laser is discussed.

RADIATION in an anisotropic medium has been considered previously in connection with Cerenkov radiation (see the review [1]) and with the investigation of electromagnetic phenomena in a magnetoactive plasma. [2] Each study was based on classical electrodynamics. We present here a procedure for solving problems concerning radiation from quantum objects. Our formulas will describe particularly Cerenkov radiation in any optically transparent anisotropic medium taking into account the spin and recoil of an emitting particle.

A quantum theory of radiation in a medium was first proposed by Ginzburg [3] and Sokolov [4] in connection with an investigation of light emitted by an electron moving within an isotropic medium. Their phenomenological method of taking the medium into account in quantum electrodynamics will be extended to anisotropic media. For this purpose we shall quantize the macroscopic electromagnetic field within an anisotropic medium in a general form that is suitable for application to various electrodynamic problems and specifically to atomic radiation.

Atomic radiation in an anisotropic medium is of great practical as well as theoretical interest. We know that the emission from some luminescent crystals is partially polarized, [5] and that the emission from some solid state lasers is very highly polarized. For example, the light beam from a ruby laser is completely polarized when the direction of the beam does not coincide with the optic axis of the crystal. [6] A quantum-mechanical study of the radiation from impurity atoms in an anisotropic medium is therefore required as a basis for explaining the foregoing experimental observations.

In connection with emission from an impurity atom it is absolutely necessary to take the surrounding medium into account when a large number of molecules of the medium are contained within a volume equal to about the cube of the emitted wavelength. Since in this case the wavelengths are much greater than the intermolecular spacing the medium can be treated macroscopically, as is frequently done in isotropic cases, [3,4,7-9] by introducing the dielectric constant tensor $\epsilon_{\alpha\beta}$. In the problem of radiation from impurity atoms the important narrow frequency interval is of the order of the radiation width of the atomic line; therefore $\epsilon_{\alpha\beta}$ can be considered constant in this frequency band if the medium is not resonant to the atomic radiation. The dielectric constant at the frequency of the impurity line is taken as the constant value of $\epsilon_{\alpha\beta}$. The magnetic permeability of the medium will be taken as unity in the interest of simplicity.

1. QUANTIZATION OF A FREE ELECTROMAGNETIC FIELD IN AN ANISOTROPIC MEDIUM

In the absence of sources and absorption the macroscopic electromagnetic field in a nonmagnetic anisotropic medium satisfies Maxwell's equations

$$\operatorname{rot}\mathbf{E} = -c^{-1}\dot{\mathbf{H}}, \quad \operatorname{div}\mathbf{H} = 0; \qquad (1)^*$$

$$\operatorname{rot}\mathbf{H} = c^{-1}\dot{\mathbf{D}}, \quad \operatorname{div}\mathbf{D} = 0, \qquad (2)$$

where $D_\alpha = \epsilon_{\alpha\beta}E_\beta$. [1)] The electromagnetic potentials will be introduced in the conventional manner on the basis of (1), and in view of their nonuniqueness will be defined in a gauge where the scalar potential vanishes identically. The electric field

*rot ≡ curl.

[1)]Repeated indices always denote summation from 1 to 3.

E and the magnetic field **H** then depend only on the vector potential **A**:

$$\mathbf{E} = -c^{-1}\dot{\mathbf{A}}, \quad \mathbf{H} = \mathrm{rot}\,\mathbf{A}. \tag{3}$$

Let the medium occupy a limited volume V having the form of a rectangular parallelepiped of much larger linear dimensions than the characteristic wavelengths in the present problem. In this case the free macroscopic electromagnetic field in the medium can be expanded conveniently in running plane waves, so that

$$\mathbf{A}(\mathbf{x}, t) = V^{-1/2}\sum_{\lambda}[a(\mathbf{k}, \lambda; t)\, e^{i\mathbf{kx}} + a^*(\mathbf{k}, \lambda; t)\, e^{-i\mathbf{kx}}]\, \mathbf{l}^{\mathbf{k}\lambda},$$

where the amplitude $a(\mathbf{k}, \lambda; t)$ is time-dependent through the factor $\exp(-i\omega_{\mathbf{k}\lambda}t)$, and the direction of the unit polarization vector $\mathbf{l}^{\mathbf{k}\lambda}$ as well as the dependence of $\omega_{\mathbf{k}\lambda}$ on **k**, in accordance with (2), is determined by solving the algebraic equations

$$[c^2(k_\alpha k_\beta - k^2\delta_{\alpha\beta}) + \omega_{\mathbf{k}\lambda}^2\varepsilon_{\alpha\beta}]\, l_\beta^{\mathbf{k}\lambda} = 0. \tag{4}$$

When the determinant of (4) is set equal to zero, we obtain for each fixed value of **k** a Fresnel equation that gives $\omega_{\mathbf{k}\lambda}$ as a function of **k**. The solution of the homogeneous equations (4) determines two independent directions of the polarization vector $\mathbf{l}^{\mathbf{k}\lambda}$ corresponding to two possible values of $\omega_{\mathbf{k}\lambda}$ with $\lambda = 1, 2$, It should be noted that in virtue of the chosen gauge the polarization vectors in the general case satisfy the conditions

$$\varepsilon_{\alpha\beta}k_\alpha l_\beta^{\mathbf{k}\lambda} = 0, \quad \varepsilon_{\alpha\beta}l_\alpha^{\mathbf{k}\lambda} l_\beta^{\mathbf{k}\lambda'} = \varepsilon_{\alpha\beta}l_\alpha^{\mathbf{k}\lambda} l_\beta^{\mathbf{k}\lambda}\delta_{\lambda\lambda'} \tag{5}$$

instead of the usual conditions of transversality $\mathbf{k}\cdot\mathbf{l}^{\mathbf{k}\lambda} = 0$ and orthogonality $\mathbf{l}^{\mathbf{k}\lambda}\mathbf{l}^{\mathbf{k}\lambda'} = \delta_{\lambda\lambda'}$ that pertain to isotropic media and the vacuum.[10] Moreover, the Poynting vector of a monochromatic wave is not parallel, as a general rule, to the wave vector **k** in an anisotropic medium.[11]

When $\epsilon_{\alpha\beta}$ is a real tensor it is convenient to write the solution of (4) for the general case in the form

$$\mathbf{l}^{\mathbf{k}\lambda} = \left(\boldsymbol{\tau}^{\mathbf{k}\lambda} - \mathbf{k}\frac{k_\alpha\tau_\beta^{\mathbf{k}\lambda}\varepsilon_{\alpha\beta}}{k_{\alpha'}k_{\beta'}\varepsilon_{\alpha'\beta'}}\right)\frac{1}{g_{\mathbf{k}\lambda}},$$

$$g_{\mathbf{k}\lambda}^2 = 1 + k^2\left(\frac{k_\alpha\tau_\beta^{\mathbf{k}\lambda}\varepsilon_{\alpha\beta}}{k_{\alpha'}k_{\beta'}\varepsilon_{\alpha'\beta'}}\right)^2 = \frac{k^2c^2}{\omega_{\mathbf{k}\lambda}^2\varepsilon_{\alpha\beta}l_\alpha^{\mathbf{k}\lambda}l_\beta^{\mathbf{k}\lambda}},$$

where $\tau^{\mathbf{k}\lambda}$ is a unit vector perpendicular to **k**. The transverse vector $\tau^{\mathbf{k}\lambda}$ is easily determined by transforming to a coordinate system in which the z axis is along **k**. In this system of coordinates $\tau^{\mathbf{k}_1}$ and $\tau^{\mathbf{k}_2}$ are mutually orthogonal and are oriented along the principal axes of the reciprocal two-dimensional dielectric constant tensor $\epsilon^{-1}_{\alpha\beta}$, in which the indices α and β assume two values corresponding to projections on the x and y axes.[11] By means of an inverse coordinate transformation $\tau^{\mathbf{k}\lambda}$ can be obtained in any coordinate system.

The total field energy is represented as a sum of energies belonging to each individual plane wave having a wave vector **k** and polarization $\mathbf{l}^\lambda$:

$$\frac{1}{8\pi}\int(\varepsilon_{\alpha\beta}E_\alpha E_\beta + \mathbf{H}^2)\,dV = \frac{1}{2\pi c^2}\sum_{\mathbf{k}\lambda}\omega_{\mathbf{k}\lambda}^2\varepsilon_{\alpha\beta}l_\alpha^{\mathbf{k}\lambda}l_\beta^{\mathbf{k}\lambda}a(\mathbf{k}, \lambda; t)\,a^*(\mathbf{k}, \lambda; t). \tag{6}$$

We can thus introduce the canonical variables $Q_{\mathbf{k}\lambda}$ and $P_{\mathbf{k}\lambda}$, where $Q_{\mathbf{k}\lambda}$ is the generalized coordinate and $P_{\mathbf{k}\lambda}$ is the generalized momentum:

$$Q_{\mathbf{k}\lambda} = [a(\mathbf{k}, \lambda; t) + a^*(\mathbf{k}, \lambda; t)]\,(\varepsilon_{\alpha\beta}l_\alpha^{\mathbf{k}\lambda}l_\beta^{\mathbf{k}\lambda}/4\pi c^2)^{1/2},$$
$$P_{\mathbf{k}\lambda} = -i\omega_{\mathbf{k}\lambda}[a(\mathbf{k}, \lambda; t) - a^*(\mathbf{k}, \lambda; t)]\,(\varepsilon_{\alpha\beta}l_\alpha^{\mathbf{k}\lambda}l_\beta^{\mathbf{k}\lambda}/4\pi c^2)^{1/2}.$$

When the total energy (6) is expressed in terms of the generalized coordinates $Q_{\mathbf{k}\lambda}$ and generalized momenta $P_{\mathbf{k}\lambda}$ we obtain the Hamiltonian of a macroscopic field in an anisotropic medium:

$$H_\gamma = \sum_{\mathbf{k}\lambda}{}^1\!/_2(P_{\mathbf{k}\lambda}^2 + \omega_{\mathbf{k}\lambda}^2Q_{\mathbf{k}\lambda}^2),$$

and Maxwell's equations (1) and (2) become Hamilton's equations of motion:

$$\dot{Q}_{\mathbf{k}\lambda} = \partial H_\gamma/\partial P_{\mathbf{k}\lambda} = P_{\mathbf{k}\lambda},$$
$$\dot{P}_{\mathbf{k}\lambda} = -\partial H_\gamma/\partial Q_{\mathbf{k}\lambda} = -\omega^2_{\mathbf{k}\lambda}Q_{\mathbf{k}\lambda}.$$

The field is now quantized conventionally by introducing the commutation relations

$$Q_{\mathbf{k}\lambda}Q_{\mathbf{k}'\lambda'} - Q_{\mathbf{k}'\lambda'}Q_{\mathbf{k}\lambda} \equiv [Q_{\mathbf{k}\lambda}, Q_{\mathbf{k}'\lambda'}] = 0,$$
$$[P_{\mathbf{k}\lambda}, P_{\mathbf{k}'\lambda'}] = 0, \quad [Q_{\mathbf{k}\lambda}, P_{\mathbf{k}'\lambda'}] = i\hbar\delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}.$$

The creation operator $c^+_{\mathbf{k}\lambda}$ and absorption operator $c_{\mathbf{k}\lambda}$ of a quantum with the wave vector **k** and polarization $\mathbf{l}^\lambda$ are defined as follows:

$$c^+_{\mathbf{k}\lambda} = -i(P_{\mathbf{k}\lambda} + i\omega_{\mathbf{k}\lambda}Q_{\mathbf{k}\lambda})\,/\,(2\hbar\omega_{\mathbf{k}\lambda})^{1/2},$$
$$c_{\mathbf{k}\lambda} = i(P_{\mathbf{k}\lambda} - i\omega_{\mathbf{k}\lambda}Q_{\mathbf{k}\lambda})\,/\,(2\hbar\omega_{\mathbf{k}\lambda})^{1/2},$$
$$[c_{\mathbf{k}\lambda}, c^+_{\mathbf{k}'\lambda'}] = \delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}.$$

As a result, the Hamiltonian H_γ of a macroscopic free electromagnetic field in an anisotropic medium and the vector potential operator **A**(**x**) in the Schrödinger representation become

$$H_\gamma = \sum_{\mathbf{k}\lambda}\hbar\omega_{\mathbf{k}\lambda}(c^+_{\mathbf{k}\lambda}c_{\mathbf{k}\lambda} + {}^1\!/_2), \tag{7}$$

$$\mathbf{A}(\mathbf{x}) = \sum_{\mathbf{k}\lambda}(2\pi\hbar c^2/\omega_{\mathbf{k}\lambda}\varepsilon_{\alpha\beta}l_\alpha^{\mathbf{k}\lambda}l_\beta^{\mathbf{k}\lambda}V)^{1/2}(c_{\mathbf{k}\lambda}e^{i\mathbf{kx}} + c^+_{\mathbf{k}\lambda}e^{-i\mathbf{kx}})\,\mathbf{l}^{\mathbf{k}\lambda}, \tag{8}$$

where the direction $\mathbf{l}^{\mathbf{k}\lambda}$ for a given **k** and the dependence of $\omega_{\mathbf{k}\lambda}$ on **k** are determined by solving the algebraic equations (4).

In quantizing the macroscopic free field in a medium no special assumption was made regarding the constant tensor $\epsilon_{\alpha\beta}$. Therefore all of the foregoing procedure can easily be extended to the case of a nonabsorbing gyrotropic medium in which the dielectric constant tensor $\epsilon_{\alpha\beta}$ is Hermitian: $\epsilon_{\alpha\beta} = \epsilon^*_{\beta\alpha}$. Also, a nondispersive medium has thus far been assumed for the sake of simplicity. However, the final equations can also be used for a dispersive medium if we remember the conclusion reached in several investigations [10,12,13] that for a transparent medium dispersion can be ignored in intermediate calculations and a frequency-dependent refractive index can be assumed in the final result. This procedure yields the correct result, agreeing with that obtained when dispersion is taken into account rigorously.

The interaction Hamiltonian of any quantum system with a macroscopic field (8) is based on the laws of quantum mechanics. Thus (7) and (8) enable us to solve all problems regarding the interaction of an electromagnetic field with quantum objects in an optically transparent nonmagnetic anisotropic medium, with the possible exception of questions (such as radiation corrections) that are associated with the covariant formulation of field quantization in a medium.

2. ATOMIC RADIATION IN AN ANISOTROPIC MEDIUM

Let us consider long-wave atomic radiation as an example. The Hamiltonian H of the system in the Schrödinger representation is

$$H = H_0 + H_\gamma + H'; \tag{9}$$

$$H_0 = \int \psi^+(\mathbf{x}) \left[\frac{\mathbf{p}^2}{2m} + U(\mathbf{x})\right] \psi(\mathbf{x})\, dV, \tag{9$'$}$$

$$H' = -\frac{e}{2mc} \int \psi^+(\mathbf{x})\, (\mathbf{pA} + \mathbf{Ap} + 2s\,\mathrm{rot}\,\mathbf{A})\, \psi(\mathbf{x})\, dV, \tag{9$''$}$$

where H_0 and H_γ are the Hamiltonians of an impurity atom and of the macroscopic free field (7), respectively, and H' is the interaction operator of the given quantum-mechanical systems. $U(\mathbf{x})$ is the external unquantized potential field in which an atomic electron moves with charge e, mass m, and spin s. This potential field includes the self-consistent field of the impurity atom and the potential field of the medium (such as a crystalline lattice). Because of the chosen gauge the momentum operator p of the electron does not commute with **A**. In (9$''$) the quadratic term in the vector potential (8) has been dropped, because it makes no contribution in the present problem. The operators of the electronic field are written as a superposition of terms pertaining to stationary states of the impurity atom, characterized by a complete set of quantum numbers n and energies $\mathscr{E}_n$:

$$\psi(\mathbf{x}) = \sum_n c_n \psi_n(\mathbf{x}), \qquad \psi^+(\mathbf{x}) = \sum_n c_n^+ \psi_n^+(\mathbf{x}),$$
$$[\mathbf{p}^2/2m + U(\mathbf{x})]\psi_n(\mathbf{x}) = \mathscr{E}_n \psi_n(\mathbf{x}).$$

Here c_n^+ and c_n are the creation and annihilation operators of an electron in the n-th atomic state:

$$c_n c_{n'}^+ + c_{n'}^+ c_n = \delta_{nn'}.$$

The temporal evolution of the system is described by an S matrix, which is formulated according to familiar methods of quantum electrodynamics. [14] In the interaction representation the S matrix element describing an atomic transition from an excited state n_2, $\mathscr{E}_{n_2}$ to a lower state n_1, $\mathscr{E}_{n_1}$ accompanied by the simultaneous emission of a quantum with the wave vector **k** and polarization $\mathbf{l}^{\mathbf{k}}$, is written

$$S_{\mathbf{k}\lambda n_1;\, 0 n_2} = (2\pi / \hbar\omega_{\mathbf{k}\lambda}\varepsilon_{\alpha\beta} l_\alpha^{\mathbf{k}\lambda} l_\beta^{\mathbf{k}\lambda} V)^{1/2} \times (\mathbf{l}^{\mathbf{k}\lambda}\mathbf{M}_{12}) \cdot 2\pi\delta(\omega_{\mathbf{k}\lambda} - \omega_{21}). \tag{10}$$

Here $\mathbf{M}_{12}$ is the matrix element of the atomic transition (n_2, $\mathscr{E}_{n_2} \to n_1$, $\mathscr{E}_{n_1}$):

$$M_{\alpha 12} = \omega_{21} d_\alpha + c[\boldsymbol{\mu}\mathbf{k}]_\alpha - i\omega_{21} Q_{\alpha\beta} k_\beta / 2,$$

where **d**, μ, and $Q_{\alpha\beta}$ are, respectively, the matrix elements of the dipole, magnetic-dipole, and quadrupole moments of the atom, and $\omega_{21} = (\mathscr{E}_{n_2} - \mathscr{E}_{n_1})/\hbar$.

We note that, unlike the case of radiation in an isotropic medium, the trace of the tensor $Q_{\alpha\beta}$ representing the atomic quadrupole moment,

$$Q_{\alpha\beta} = \int \psi_{n_1}^+(\mathbf{x})\, x_\alpha x_\beta \psi_{n_2}(\mathbf{x})\, dV$$

does not vanish. This is associated with the fact that in an anisotropic medium div $\mathbf{A} \neq 0$; this follows from (4) and (5).

We can in principle replace $Q_{\alpha\beta}$ by a different tensor for the atomic quadrupole moment:

$$Q_{\alpha\beta}' = \int \psi_{n_1}^+(\mathbf{x}) \left(3x_\alpha x_\beta - \frac{3x^2\varepsilon_{\alpha\beta}}{\varepsilon_{\gamma\gamma}}\right) \psi_{n_2}(\mathbf{x})\, dV,$$

which in an isotropic medium assumes a canonical form [15] with zero trace. There is no accompanying change of the matrix element (10), in accordance with (5). However, the tensor $Q'_{\alpha\beta}$ contains $\epsilon_{\alpha\beta}$, which is a material property of the medium, so that it is introduced somewhat artificially, especially for a highly rarefied medium in which neighboring molecules do not influence the motion of impurity-atom electrons, and the emitted wavelength is, as previously, larger than the intermolecular spacing. Therefore we shall hereafter take $Q_{\alpha\beta}$ to be the quadrupole moment tensor of an impurity atom.

Equation (10) leads to the unusual conclusion that the quadrupole transition 0 − 0 is allowed in an anisotropic medium. The probability of this transition is, of course, reduced by the additional factor

$$k^2(k_\alpha \tau_\beta^{\mathbf{k}\lambda}\varepsilon_{\alpha\beta} / k_{\alpha'}k_{\beta'}\varepsilon_{\alpha'\beta'})^2,$$

which is, as a rule, considerably smaller than unity (see below) and vanishes in an isotropic medium. The possibility of the quadrupole transition 0 − 0 results from the fact that a quantum of a macroscopic electromagnetic field in an anisotropic medium can exist in a state with zero total moment, unlike the case of an isotropic medium or vacuum. The existence of the 0 − 0 transition indicates that the long-wave emission from an impurity atom is accompanied by a macroscopic quantum. More exactly, the impurity atom interacts with the zero-point vibrations of the macroscopic field and macroscopic emission occurs even in the virtual quantum stage preceding real emission. In other words, the long-wave radiation from an impurity atom is macroscopic even before the emitted field has reached the wave zone.

Utilizing (10), we obtain the probability $dW_{\mathbf{k}\lambda}$ that in unit time a quantum with the wave vector **k** and polarization $\mathbf{l}^{\mathbf{k}\lambda}$ is emitted within the solid angle $d\Omega$ by an atom located in an optically transparent nonmagnetic anisotropic medium:

$$dW_{\mathbf{k}\lambda} = \frac{|\mathbf{l}^{\mathbf{k}\lambda}\mathbf{M}_{12}|^2 k^3\, d\Omega}{2\pi\hbar\omega_{21}^2\varepsilon_{\alpha\beta}\, l_\alpha^{\mathbf{k}\lambda} l_\beta^{\mathbf{k}\lambda}} \tag{11}$$

where the numerical value of **k** is obtained in terms of ω_{21} from the energy conservation law $\omega_{\mathbf{k}\lambda}(k; \mathbf{k}/k) = \omega_{21}$.

Let us consider the angular distribution of atomic radiation. We shall assume for simplicity that the initial and final atomic states are unpolarized; therefore all spatial polarizations of the complex vectors **d** and $\boldsymbol{\mu}$ and of the $Q_{\alpha\beta}$ axes are equally probable. Then, averaging the radiation probability (11) over all directions of **d** and $\boldsymbol{\mu}$ and of the axes of $Q_{\alpha\beta}$, for the general case in an anisotropic medium we obtain

$$\begin{aligned} dW_{\mathbf{k}\lambda} = {} & [\omega_{21}^2|\mathbf{d}|^2 + c^2(k^2 - (\mathbf{k}\mathbf{l}^{\mathbf{k}\lambda})^2)|\boldsymbol{\mu}|^2 \\ & + \omega_{21}^2(k^2 + 2(\mathbf{k}\mathbf{l}^{\mathbf{k}\lambda})^2)|Q_{\alpha\beta}|^2/20]k^3 d\Omega \\ & \times [6\pi\hbar\omega_{21}^2\varepsilon_{\alpha\beta} l_\alpha^{\mathbf{k}\lambda} l_\beta^{\mathbf{k}\lambda}]^{-1}. \end{aligned} \tag{12}$$

3. UNIAXIAL CRYSTALS

We shall now apply the derived relations to atomic radiation in a uniaxial crystal having its optic axis parallel to the z axis. Then, $\epsilon_{11} = \epsilon_{22} = \epsilon_\perp$ and $\epsilon_{33} = \epsilon_\parallel$. The solution of (4) for a given **k** yields two values of the polarization vector $\mathbf{l}^{\mathbf{k}\lambda}$; for the ordinary wave (λ = 1) in the crystal we have

$$\mathbf{l}^{\mathbf{k}1} = [\mathbf{i}_3\mathbf{k}] / k\sin\theta, \qquad \omega^2_{\mathbf{k}1} = k^2c^2/\varepsilon_\perp \tag{13*}$$

and for the extraordinary wave (λ = 2) we have

$$\begin{aligned} \mathbf{l}^{\mathbf{k}2} = {} & \left(\frac{[[\mathbf{i}_3\mathbf{k}]\mathbf{k}]}{k^2\sin\theta} + \frac{\mathbf{k}}{k}\frac{(\varepsilon_\parallel - \varepsilon_\perp)\sin\theta\cos\theta}{\varepsilon_\perp + (\varepsilon_\parallel - \varepsilon_\perp)\cos^2\theta}\right) \\ & \times \frac{\varepsilon_\perp + (\varepsilon_\parallel - \varepsilon_\perp)\cos^2\theta}{[\varepsilon_\perp^2 + (\varepsilon_\parallel^2 - \varepsilon_\perp^2)\cos^2\theta]^{1/2}} \\ & \times \omega_{\mathbf{k}2}^2 = c^2[(k_1^2 + k_2^2)\varepsilon_\perp + k_3^2\varepsilon_\parallel] / \varepsilon_\parallel\varepsilon_\perp, \end{aligned} \tag{14}$$

where θ is the angle between **k** and the unit vector $\mathbf{i}_3$ of the z axis which is along the optic axis. The foregoing formulas show that the electric vector of the ordinary wave emitted by impurity atoms is always perpendicular to the optic axis and to the direction of wave propagation, whereas the electric vector of the extraordinary wave always lies in the plane defined by the optic axis and **k**. When the direction of **k** coincides with the optic axis, the difference between the ordinary and extraordinary waves disappears, the aforementioned orientation of the electric vector is completely disrupted, and waves propagating in the direction of the optic axis are depolarized. The polarization vectors (13) and (14) are mutually orthogonal and satisfy (5).

The multiplication of the tensor $\epsilon_{\alpha\beta}$ by the polarization vectors in a uniaxial crystal yields the following result for the ordinary wave (λ = 1):

$$\varepsilon_{\alpha\beta} l_\alpha^{\mathbf{k}1} l_\beta^{\mathbf{k}1} = \varepsilon_\perp$$

and for the extraordinary wave (λ = 2):

$$\frac{1}{\varepsilon_{\alpha\beta} l_\alpha^{\mathbf{k}2} l_\beta^{\mathbf{k}2}} = \frac{1}{\varepsilon_\perp\varepsilon_\parallel}\,\frac{\varepsilon_\perp^2 + (\varepsilon_\parallel^2 - \varepsilon_\perp^2)\cos^2\theta}{\varepsilon_\perp + (\varepsilon_\parallel - \varepsilon_\perp)\cos^2\theta}$$

As a result, the total probability (integrated over the angles of **k**) that a quantum is emitted in unit time with the polarization $\mathbf{l}^{\mathbf{k}1}$ corresponding to an ordinary wave will coincide with the probability that a quantum is emitted with a fixed polarization in an isotropic medium having the dielectric constant $\epsilon_\perp$. At the same time, the probability that in unit time a quantum is emitted with the polarization $\mathbf{l}^{\mathbf{k}2}$ corresponding to an extraordinary wave is (with W_{2d} for dipole, $W_{2\mu}$ for magnetic-dipole, and W_{2Q} for quadrupole radiation)

$$W_{2d} = W_d^0(\varepsilon_\parallel + 2\varepsilon_\perp)/6\varepsilon_\perp^{1/2}, \qquad W_{2\mu} = W_\mu^0\varepsilon_\parallel\varepsilon_\perp^{1/2}/2,$$
$$W_{2Q} = W_Q^0(2\varepsilon_\parallel^2 + \varepsilon_\parallel\varepsilon_\perp + 2\varepsilon_\perp^2)/10\varepsilon_\perp^{1/2},$$

where W_d^0, W_μ^0, and W_Q^0 are the total probabilities of dipole, magnetic-dipole, and quadrupole radiation, respectively, per unit time, of quanta having both polarizations in a vacuum:

$$W_d^0 = 4\omega_{21}^3|\mathbf{d}|^2/3\hbar c^3, \qquad W_\mu^0 = 4\omega_{21}^3|\boldsymbol{\mu}|^2/3\hbar c^3,$$
$$W_Q^0 = \omega_{21}^5|Q_{\alpha\beta}|^2/15\hbar c^5.$$

*$[\mathbf{i}_3\mathbf{k}] \equiv \mathbf{i}_3 \times \mathbf{k}$.

Here the numerical coefficient preceding $|Q_{\alpha\beta}|^2$ differs from the customary coefficient because the tensor $Q_{\alpha\beta}$ of the atomic quadrupole moment has not been reduced to a canonical form with a vanishing trace.

The total probabilities W_d (dipole), W_μ (magnetic-dipole), and W_Q (quadrupole) of radiation per unit time of quanta having both polarizations in an anisotropic medium are, from (11),

$$W_d = W_d{}^0(\varepsilon_\parallel + 5\varepsilon_\perp) / 6\varepsilon_\perp{}^{1/2}, \quad W_\mu = W_\mu{}^0(\varepsilon_\parallel + \varepsilon_\perp)\varepsilon_\perp{}^{1/2} / 2,$$
$$W_Q = W_Q{}^0(2\varepsilon_\parallel{}^2 + \varepsilon_\parallel\varepsilon_\perp + 7\varepsilon_\perp{}^2) / 10\varepsilon_\perp{}^{1/2}.$$

One or more of the quantities W_d^0, W_μ^0, and W_Q^0 may vanish because of the selection rules.

The application of (12) to atomic radiation in a uniaxial crystal shows that quanta with the polarization 1^{k1} have the same angular distribution as in an isotropic medium. However, the angular distribution of quanta with the polarization 1^{k2} corresponding to an extraordinary wave has the complicated form

$$dW_{2d} = \frac{W_d{}^0(\varepsilon_\parallel\varepsilon_\perp)^{1/2}}{8\pi}\frac{\varepsilon_\perp{}^2 + (\varepsilon_\parallel{}^2 - \varepsilon_\perp{}^2)\cos^2\theta}{[\varepsilon_\parallel + (\varepsilon_\parallel - \varepsilon_\perp)\cos^2\theta]^{5/2}}d\Omega, \quad (15)$$

$$dW_{2\mu} = \frac{W^0{}_\mu(\varepsilon_\parallel\varepsilon_\perp)^{3/2}}{8\pi}\frac{d\Omega}{[\varepsilon_\parallel + (\varepsilon_\parallel - \varepsilon_\perp)\cos^2\theta]^{3/2}}, \quad (16)$$

$$dW_{2Q} = \frac{W_Q{}^0(\varepsilon_\parallel\varepsilon_\perp)^{3/2}}{8\pi}\Big[\frac{\varepsilon_\perp{}^2}{[\varepsilon_\parallel + (\varepsilon_\parallel - \varepsilon_\perp)\cos^2\theta]^{7/2}}$$
$$+ \frac{(3\varepsilon_\parallel - \varepsilon_\perp)(\varepsilon_\parallel - \varepsilon_\perp)\cos^2\theta - 2(\varepsilon_\parallel - \varepsilon_\perp)^2\cos^4\theta}{[\varepsilon_\parallel + (\varepsilon_\parallel - \varepsilon_\perp)\cos^2\theta]^{7/2}}\Big]d\Omega, \quad (17)$$

where dW_{2d}, $dW_{2\mu}$, and dW_{2Q} are the respective probabilities of dipole, magnetic-dipole, and quadrupole emission of a quantum with the wave vector **k** and polarization 1^{k2} in a solid angle $d\Omega$.

Equations (15)—(17) show that anisotropy of the extraordinary wave in a uniaxial crystal exists even when the orientations of **d**, μ, and the axes of $Q_{\alpha\beta}$ are not specified, so that (15)—(17) are averaged over all these directions. We also emphasize that the angular distribution of radiation is correct everywhere only for the given anisotropic medium. Upon emerging from this medium the rays will be refracted in accordance with the laws of geometric optics; this refraction can be determined easily for any given direction.

If, in accordance with external conditions, **d**, μ, and the axes of the impurity-atom quadrupole moment $Q_{\alpha\beta}$ have definite orientations with respect to the optic axis of the crystal, both the intensities and angular distributions of the ordinary and extraordinary waves will be strongly dependent on the orientations of **d**, μ, and the axes of $Q_{\alpha\beta}$. Indeed, if the selection rules permit only dipole radiation and this should be emitted with no change ($\Delta m = 0$) in the projection m of the total atomic mechanical moment, then **d** could be assumed to be real (here m is the projection of the moment in some chosen direction).

Let us assume that **d** forms the angle θ_d with the optic axis and that all azimuthal angles are equally probable, thus permitting us to average over the latter. Then, in accordance with (11), for the ordinary wave the probability per unit time that a quantum is emitted within the solid angle $d\Omega = 2\pi\sin\theta\, d\theta$ is

$$dW_{1d} = 3W_d{}^0\varepsilon_\perp{}^{1/2}\sin^2\theta_d d\Omega / 16\pi, \quad (18)$$

while the angular dependence for the extraordinary wave is much more complicated in general. However, if we assume $(\epsilon_\parallel - \epsilon_\perp)^2/\epsilon_\perp^2 \ll 1$ and drop terms of the order $(\epsilon_\parallel - \epsilon_\perp)^2/\epsilon_\perp^2$, then in the case of the extraordinary wave we obtain

$$dW_{2d} = 3W_d{}^0\varepsilon_\perp{}^{1/2}(\sin^2\theta_d\cos^2\theta + 2\cos^2\theta_d\sin^2\theta)d\Omega / 16\pi. \quad (19)$$

The total probability of dipole radiation, W_{1d} for the ordinary wave and W_{2d} for the extraordinary wave, depends strongly on the angle between **d** and the optic axis:

$$W_{1d} = {}^3/_4 W_d{}^0\varepsilon_\perp{}^{1/2}\sin^2\theta_d, \quad (20)$$
$$W_{2d} = {}^1/_4 W_d{}^0\varepsilon_\perp{}^{1/2}(1 + 3\cos^2\theta_d). \quad (21)$$

Specifically, at small values of θ_d the intensity of the ordinary dipole wave is considerably reduced compared with the extraordinary wave. Therefore the dipole emission is partially polarized in this case. This is one reason for the experimental observation of polarized light from some luminescent crystals.

For magnetic-dipole radiation the substitutions $\mathbf{d} \to \mu$ and $\epsilon^{1/2} \to \epsilon^{3/2}$ in (18)—(21) along with an exchange of the indices for the ordinary and extraordinary waves are required. The character of the polarization is then the opposite of the dipole case.

It is easy to derive the explicit dependences of atomic radiation intensity and angular distribution on the orientations of **d**, μ, and the axes of $Q_{\alpha\beta}$ in all other cases. By investigating the angular distribution and polarization we can arrive at definite conclusions concerning the character of optical atomic transitions and the orientations of matrix elements for **d**, μ, and $Q_{\alpha\beta}$ transitions in the case of an impurity atom within an anisotropic medium. This investigation of radiation from impurity atoms can also obtain additional information regarding the dielectric properties of crystals, particularly the dielectric constant in specified directions.

4. DISCUSSION OF THE POLARIZATION OF RUBY RADIATION

It has been found experimentally [6] that the R_1 line of ruby fluorescence consists of two components, corresponding to the ordinary and extraordinary waves of the crystal; the latter is relatively weak, so that the radiation is partially polarized as a whole. When a ruby is used as the working material of a laser, the R_1 line is generated under certain specific conditions. The electric vector of the generated light beam is perpendicular to the optic axis of the ruby crystal and to the direction of the beam. The beam is completely depolarized if its direction coincides with the optic axis. [6] According to (3), (8), (13), and (14), this indicates that a uniaxial ruby laser emits only the ordinary wave. The evident explanation is that the wave vector of standing waves in the cavity resonator is perpendicular to the two parallel reflecting mirrors. However, the direction of energy flow in the extraordinary wave does not coincide with the direction of the wave vector; therefore some angle is formed with the cavity axis that is perpendicular to the two mirrors. Consequently, extraordinary wave quanta moving at some angle to the cavity axis must escape through the sides of the latter following a few reflections from the parallel mirrors. On the other hand, the energy flow vector of the ordinary wave coincides with the wave vector and is therefore perpendicular to the two cavity mirrors. Successive reflections of the ordinary wave from these parallel mirrors do not disrupt the parallelism between the wave vector and the cavity axis; therefore ordinary wave quanta are confined within the volume between the mirrors. Thus the cavity has a higher Q for the electromagnetic oscillations of the ordinary wave and an induced coherent process is set up for the ordinary wave, which suffers considerably smaller energy loss.

The Poynting vector **S** of a monochromatic extraordinary wave is, according to (14), proportional to a vector given by

$$\mathbf{S} \sim \frac{\mathbf{k}}{k} + \mathbf{i}_3 \frac{\varepsilon_\parallel - \varepsilon_\perp}{\varepsilon_\perp} \cos\theta, \qquad (22)$$

where θ is the angle between **k** and the unit vector $\mathbf{i}_3$ along the z axis, which is oriented along the optic axis of a uniaxial crystal. This equation is used to determine the tangent of the angle θ_S between **S** and **k**:

$$\tan\theta_s = \frac{(\varepsilon_\parallel - \varepsilon_\perp)\cos\theta\sin\theta}{\varepsilon_\perp + (\varepsilon_\parallel - \varepsilon_\perp)\cos^2\theta}. \qquad (23)$$

If the wave vector **k** of the monochromatic extraordinary wave coincides with the cavity axis, then in each specific case (23) enables the calculation of the energy loss resulting from the escape of the extraordinary wave through the sides of the cavity following reflections from the parallel mirrors.

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[1] B. M. Bolotovskiĭ, UFN **62**, 201 (1957) and **75**, 295 (1961), Soviet Phys. Uspekhi **4**, 781 (1962).

[2] V. L. Ginzburg, Rasprostranenie élektromagnitnykh voln v plazme (The Propagation of Electromagnetic Waves in Plasmas), Fizmatgiz, 1960, p. 108.

[3] V. L. Ginzburg, JETP **10**, 589 (1940).

[4] A. A. Sokolov, DAN SSSR **28**, 415 (1940).

[5] P. P. Feofilov, Polyarizovannaya lyuminestsentsiya atomov, molekul i kristallov (Polarized Luminescence of Atoms, Molecules, and Crystals), Fizmatgiz, 1959.

[6] D. F. Nelson and R. J. Collins, The Polarization of the Output from a Ruby Optical Maser, in Advances in Quantum Electronics, edited by J. R. Singer, Columbia U. Press, New York, 1961, p. 79.

[7] M. I. Ryazanov, JETP **32**, 1244 (1957), Soviet Phys. JETP **5**, 1013 (1957).

[8] V. L. Bonch-Bruevich and S. V. Tyablikov, Metod funktsiĭ Grina v statisticheskoĭ mekhanike (Green's Function Methods in Statistical Mechanics), Fizmatgiz, 1961, p. 280.

[9] Abrikosov, Gor'kov, and Dzyaloshinskiĭ, Metody kvantovoĭ teorii polya v statisticheskoĭ fizike (Methods of Quantum Field Theory in Statistical Physics), Fizmatgiz, 1962, p. 325.

[10] V. L. Ginzburg, JETP **10**, 601 (1940).

[11] L. D. Landau and E. M. Lifshitz, Élektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Fizmatgiz, 1959, p. 395.

[12] V. L. Ginzburg and V. Ya. Éĭdman, JETP **36**, 1823 (1959) and **43**, 1865 (1962), Soviet Phys. JETP **9**, 1300 (1959) and **16**, 1316 (1963).

[13] Yu. A. Ryzhov, Izv. VUZov, Radiofizika (News of the Universities, Radio Physics) **2**, 869 (1959).

[14] A. I. Akhiezer and V. B. Berestetskiĭ, Kvantovaya elektrodinamika (Quantum Electrodynamics), Fizmatgiz, 1959, p. 232.

[15] L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Fizmatgiz, 1963, p. 312.

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