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RADIATION THEORY IN CRYSTALS

By Wojciech Królikowski

Institute of Theoretical Physics, University of Warsaw and Institute of Physics of the Polish Academy of Sciences, Warsaw

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This paper contains the radiation theory for the atom in a crystalline medium. In the first part we obtain equations for that class of problems using the formalism of quantum electrodynamics. An apparent mass of the photon appears in that formalism. In the second part we define (with certain simplifying assumptions) the formula for the apparent mass of the photon and solve the equations of the radiation field in a crystal. The solution obtained is consistent with Ewald's classical result. In the third part we give an estimate of the emission probability in crystals.

1. *Introduction*

We shall consider an atom interacting with radiation in a crystalline medium, i. e. a system composed of a radiating atom, the field of radiation $A_\mu(x)$ and the field of electrons $\psi(x)$ forming the medium. We assume that the electrons move in a given (non quantized) periodic field $A_\mu^{ex}(x)$. The electrons can be bound to nuclei or not.

The equations of motion of the considered system have in the Heisenberg picture the following form

$$\left.\begin{aligned} &\text{a)}\ \Box \underset{H}{A_\mu}(x) = -\frac{1}{c}(\underset{H}{j_\mu}(x) + \underset{H}{j_\mu^A}(x)),\\ &\text{b)}\ \left[\gamma_\mu\left(\frac{\partial}{\partial x_\mu} - \frac{ie}{\hbar c}A_\mu^{ex}(x)\right) + \varkappa\right]\underset{H}{\psi}(x) = \frac{ie}{\hbar c}\gamma_\mu \underset{H}{A_\mu}(x)\,\underset{H}{\psi}(x),\\ &\text{c) } \textit{the equations of motion of the atom perturbed by the fields } \underset{H}{A_\mu}(x) \textit{ and } A_\mu^{ex}(x)\\ &\text{d)}\ i\hbar c\frac{\delta \Psi_H}{\delta\sigma(x)} = 0,\ \frac{\partial \underset{H}{A_\mu}(x)}{\partial x_\mu}\Psi_H = 0, \end{aligned}\right\} \quad (1)$$

where $\underset{H}{j_\mu}(x) = \mathrm{iec} : \underset{H}{\bar{\psi}}\gamma_\mu \underset{H}{\psi}(x):$ and $\underset{H}{j_\mu^A}(x)$ are respectively currents of electrons of the medium (we take the antisymmetrized current) and of the atom. Operators $\underset{H}{A_\mu}(x)$,t

$\psi(x)$ have here the common commutation or anticommutation properties. The symbol $\underset{H}{:}(\ldots)\underset{H}{:}$ denotes an ordered product where all annihilation operators are on the right of the creation operators [Wick 1950]. We use a notation in which $x = (x_\mu) = (\vec{x}, \mathrm{ict})$, $(\gamma_\mu) = (-i\vec{\beta}\vec{a}, \beta)$, $\bar{\psi}(x) = \psi^*(x)\,\beta$.

By means of the unitary transformation:

$$\underset{H}{0}(x) = S[\sigma]\,\underset{H}{0}(x)\,S^{-1}[\sigma], \qquad \Psi[\sigma] = S[\ ]\,\Psi_H,$$

where

$$i\hbar c\,\frac{\delta S[\sigma]}{\delta\sigma(x)} = -\frac{1}{c}\,j^A_\mu(x)\,A_\mu(x)\,S[\sigma],\quad S[-\infty] = 1,$$

we get a new picture, in which the equations of motion take the form

$$\left.\begin{aligned}
&\text{a)}\ \Box A_\mu(x) = -\frac{1}{c}\,j_\mu(x),\\
&\text{b)}\ \left[\gamma_\mu\left(\frac{\partial}{\partial x_\mu} - \frac{ie}{\hbar c}\,A^{ex}_\mu(x)\right) + \varkappa\right]\psi(x) = \frac{ie}{\hbar c}\,\gamma_\mu A_\mu(x)\,\psi(x),\\
&\text{c) } \textit{the eqations of motion of the atom perturbed only by the field } A^{ex}_\mu(x),\\
&\text{d)}\ ihc\,\frac{\delta\Psi[\sigma]}{\delta\sigma(x)} = -\frac{1}{c}\,j^A_\mu(x)\,A_\mu(x)\,\Psi[\sigma],\quad \frac{\partial A_\mu(x)}{\partial x_\mu}\,\Psi[\sigma]\ (x\ \textit{on}\ \sigma),
\end{aligned}\right\} \tag{2}$$

where $j_\mu(x) = iec\colon\bar{\psi}\gamma_\mu\psi(x)\colon$. Of course this is not an interaction picture.

We shall solve equations (2) using a unitary transformation leading to the picture, for which the forms of the electron and electromagnetic operators are known.

The interaction picture has such properties but it is adapted to the case of photons in vacum. We shall use here instead a new picture, better fitted for the description of photons in a material medium. It can be expected that in a material medium photons acquire (from a macroscopic point of view) an apparent mass depending on their direction, frequency, and on the structure of the medium. We shall determine this mass later. Now we introduce it formally, using the following unitary transformation.

$$O^\circ(x) = U[\ ]\,O(x)\,U^{-1}[\ ], \qquad \Psi^\circ[\ ] = U[\ ]\,\psi[\sigma], \tag{3}$$

where

$$i\hbar c\,\frac{\delta U[\sigma]}{\delta\sigma(x)} = H^\circ(x)\,U[\sigma],\quad U[-\infty] = 1 \tag{4}$$

and

$$H^\circ(x) = -\left(\frac{1}{c}\,j^\circ_\mu(x)\,A^\circ_\mu(x) + \frac{1}{2}\,\mu^2\,A^\circ_\mu(x)\,A^\circ_\mu(x)\right),$$

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$j^\circ_\mu(x) = iec{:}\,\bar{\psi}^\circ\gamma_\mu\psi^\circ(x){:}$. μ is here a c-number equal to the apparent mass multiplied by $c/\hbar$. The equations of motion in the new picture have the form

$$\left.\begin{aligned} &\text{a)}\ (\Box - \mu^2)\, A^\circ_\mu(x) = 0,\\ &\text{b)}\ \left[\gamma_\mu\left(\frac{\partial}{\partial x_\mu} - \frac{ie}{\hbar c} A^{ex}_\mu(x)\right) + \varkappa\right]\psi^\circ(x) = 0,\\ &\text{c) } \textit{the equations of motion of the atom perturbed by the field } A^{ex}_\mu(x),\\ &\text{d)}\ i\hbar c\,\frac{\delta\Psi^\circ[\sigma]}{\delta\sigma(x)} = -\left[\frac{1}{c}\,(j^{A^\circ}_\mu(x) + j^\circ_\mu(x)\, A^\circ_\mu(x) + \frac{1}{2}\,\mu^2 A^\circ_\mu(x)\, A^\circ_\mu(x)\right]\Psi^\circ[\sigma]. \end{aligned}\right\} \quad (5)$$

The operator $A^\circ_\mu(x)$ satisfies here the following commutation rules:

$$[A^\circ_\mu(x), A^\circ_\nu(x')] = i\hbar c\,\delta_{\mu\nu}\,\Delta(x - x'), \quad (6)$$

where $\Delta(x - x)$ is an ordinary Jordan-Pauli function for mass μ. Our new picture becomes the interaction picture for $\mu = 0$. Note that, independently of the value of μ, the operator $\psi^\circ(x)$ is allways identical with the electron operator in the interaction picture.

The relation between the electromagnetic tensor and vector does not change after the unitary transformations given by our operators $S([\sigma]$ and $U[\sigma]$. Therefore we have $f^\circ_{\mu\nu} = A^\circ_{\nu\mu} - A^\circ_{\mu\nu}$. Thus we can gauge A°_μ in different manners, especially so that $A^\circ_4 = 0$. We obtain it by replacing A°_μ by $A^\circ_\mu - \chi_{,\mu}$, where $\chi_{,4} = A^\circ_4$. We are entitled to do so because the first equation (5) does not change when $(\Box - \mu^2)\,\chi = 0$ (if the initial condition for χ is appropriate).

It follows from (3) and (4) [cf. Dyson 1949, Wick 1950] that

$$O(x) = U^{-1}[\infty] \sum_{n=0}^{\infty} \frac{1}{n!}\left(-\frac{i}{\hbar c}\right)^n \int_{-\infty}^{+\infty} dx_1 \ldots dx_n T(O^\circ(x)\, H^\circ(x_1) \ldots H^\circ(x_n)). \quad (7)$$

Here the symbol $T(\ldots)$ is the chronological product as defined by Wick. Further calculations consist in splitting the chronological product $T(\ldots)$ into ordered products with suitable contractions which lead to the Feynman [1949] functions: $S^{ex}_+(x,x')$ and $\Delta_+(x - x')$ [Wieck 1950]:

$$\psi^{\circ\cdot}_\alpha(x)\,\psi^{\circ\cdot}_\beta(x') = S^{\circ\varkappa}_{+\alpha\beta}(x, x),\ A^{\circ\cdot}_\mu(x)\, A^{\circ\cdot}_\nu(x') = \hbar c\,\delta_{\mu\nu}\, D_+(x - x')$$

(the dot over operators repeated twice denotes contraction of both operators). These functions satisfy the following equations:

$$\left[\gamma_\mu\left(\frac{\partial}{\partial x_\mu} - \frac{ie}{\hbar c} A^{ex}_\mu(x)\right) + \varkappa\right] S^{ex}_+(x, x') = -i\,\delta(x - x'),$$

and

$$(\Box - \mu^2)\,\Delta_+(x - x') = i\delta(x - x').$$

Assuming the potential to be independent of time $A^{ex}_\mu(x) = A^{ex}_\mu(\vec{x})$, we have:

$$S^{ex}_{+\alpha\beta}(x, x') = \begin{cases} \sum_r^+ \psi_{r\alpha}(x)\,\bar{\psi}_{r\beta}(x') & (t > t') \\ -\sum_r^- \psi_{r\alpha}(x)\,\bar{\psi}_{r\beta}(x') & (t < t'), \end{cases} \quad (8)$$

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where $\psi_r(x) = u_r(\vec{x})\, exp\,(-iE_r t/\hbar)$ are eigensolutions of Dirac equations with potential $A_\mu^{ex}(x)$. The sums $\Sigma^{\pm}$ extend over states of positive and negative energy respectively.

Inserting $O^\circ(x) = A_\mu^\circ(x)$ in (7), we would obtain the perturbation solution for $A_\mu(x)$ if we knew μ. That would be true, e.g., if we used the interaction picture ($\mu = 0$). When μ is not known, we shall proceed in a way which will enable us to find it.

Let us assume that μ^2 is of the second order in the coupling constant. This asumption will be shown further to be consistent with the results. Inserting $O^\circ(x) = j_\mu^\circ(x)$ in (7) and using Wick's method we have

$$j_\mu(x) = U^{-1}[\infty]\left\{ j_\mu^\circ(x) - \frac{ie^2}{\hbar}\int_{-\infty}^{+\infty} dx' \,[:\bar{\psi}^\circ(x)\gamma_\mu S_+^{ex}(x,x')\,\gamma_\nu\psi^\circ(x'): + \right.$$

$$\left. : \bar{\psi}^\circ(x')\gamma_\nu S_+^{ex}(x',x)\,\gamma_\mu\psi(x): - \mathrm{tr}\,(\gamma_\mu S_+^{ex}(x,x')\,\gamma_\nu S_+^{ex}(x',x))]\, A_\nu^\circ(x') + \ldots \right\}. \quad (9)$$

Further terms in $\{\}$ brackets are of higher order in the coupling constant.

Operator $U[\sigma]$ given by (4) describes processes in the medium (independent of rhe considered atom) resulting from interaction between electrons of the medium and the radiation field. Let us assume that in the medium processes of the first order do not occur. Then, for all states $\Psi[\sigma]$, we have

$$U[\infty]\,\Psi[\sigma] = \Psi[\sigma]$$

(for terms up to the first order). After this assumption formula (9) takes the form

$$j_\mu(x) = j_\mu^\circ(x) - \frac{ie^2}{\hbar}\int_{+\infty}^{+\infty} dx' \,[:\bar{\psi}^\circ(x)\,\gamma_\mu S_+^{ex}(x,x')\,\gamma_\nu\,\psi^\circ(x'): + : \bar{\psi}^\circ(x')\gamma_\nu S_+^{ex}(x',x)\,\gamma_\mu\psi^\circ(x):$$

$$- \mathrm{tr}\,(\gamma_\mu S_+^{ex}(x,x')\,\gamma_\nu S_+^{ex}(x',x))]\, A_\nu^\circ(x')\,, \quad (10)$$

where we have taken into account terms up to the second order only.

If we put (10) in the first equation (2), and insert afterwards the thus obtained $A_\mu(x)$ in the fourth equation (2), the latter will describe processes following from interaction between the atom and the radiation field in the medium.

In order to find, e.g., the probability of one-quantum (spontaneous) emission we consider the transition from the initial state $\Psi_i = \Psi[-\infty]$ without photons in the field, to the state Ψ_f with one photon. Assuming an adiabatic switching on of interaction at the initial moment $t = -\infty$ and switching it off by the measurement at the final moment t, we obtain

$$\Psi_i = \Psi_i^A\,\Psi_i^R\,\Psi_i^e, \quad \Psi_f = \Psi_f^A\,\Psi_f^R\,\Psi_f^e,$$

where Ψ^A, Ψ^R, Ψ^{el} are state vectors of the atom, radiation field, and electrons of the medium respectively. Then the probability amplitude of one-quantum emission in the ime interval $(-\infty, t)$ will be equal to:

$$\langle \Psi_f\,\Psi[\sigma]\rangle = \frac{i}{\hbar c}\int_{-\infty}^{\sigma} dx\,\langle \Psi_f^A\, j_\mu^A(x)\,\Psi_i^A\rangle\langle \Psi_f^R\,\langle A_\mu(x)\rangle\,\Psi_i^R\rangle, \quad (11)$$

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where

$$\langle A_\mu(x)\rangle = \langle \Psi_f^{el} A_\mu(x) \Psi_i^{el}\rangle. \tag{12}$$

We assume also that the electrons of the medium are in equilibrium with the radiation field, i.e. that

$$\Psi_i^{el} = \Psi_f^{el}.$$

When the first and fourth equations (2) are solved the term $j_\mu^\circ(x)$ in formula (10) gives the interaction of the electrons of the medium with the atom. In the first step of the method of successive approximation this term leads to diagram a. in Fig. 1. Using a suitable unitary transformation, we can eliminate this interaction from the equations of the radiation field, inserting it simultaneously in the Hamiltonian of the atom. The eigenstates of the atom will then be only slightly modified. There appears namely (according to Bethe and others) a splitting of the energy levels depending on crystal structure and resulting from electrostatic interaction (a kind of Stark effect). References relating to that subject are given by Rubinowicz [1949]. We will not take into account the above modification of the states when comparing probabilities of emission in crystals and in vacuum.

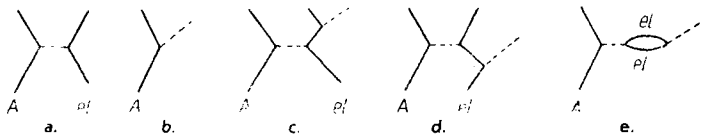

Fig. 1

Three remaining terms in formula (10) give additional terms to the probabilities of transitions, e.g. to the probability of one-quantum transition. In the first step of the successive approximation method (formula(11)) they lead for that emission to diagrams c., d., e. of Fig. 1. Diagram e. describes the self-mass of the emitted photon in the medium. Besides, diagram b. will also appear.

From the first equation (2) and formula (10) we get the following equation describing in the considered approximation the radiation field in the given medium

$$\Box A_\mu(x) = \int_{-\infty}^{+\infty} dx'\, K_{\mu\nu}(x,x')\, A_\nu^\circ(x'), \tag{13}$$

where

$$K_{\mu\nu}(x,x') = \frac{ie^2}{\hbar c}\,(:\bar{\psi}^\circ(x)\,\gamma_\mu\, S_+^{ex}(x,x')\,\gamma_\nu\,\psi(x'):+:\bar{\psi}^\circ(x')\,\gamma_\nu\, S_+^{ex}(x',x)\,\gamma_\mu\,\psi^\circ(x):).$$

We dropped here terms leading to diagrams a. and e. in Fig. 1. (14)

2. *Ewald's solution*

Let us assume that the electrons of the medium interacting with radiation are assembled in the vertices of a crystal lattice and that the latters can be considered as points compared to the radiation wave lengths. This assumption is used with good

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results in classical crystal optics (as well as in X — ray optics — Ewald 1912, 1918). We shall solve equation (13) with that assumption and find μ at the same time. The solution obtained will correspond with Ewald's solution in clasical crystal optics.

Inserting in the right hand of (13) a plane wave satisfying the first equation (5)

$$A_{\mu}^{\circ\pm}(x) = A_{\mu}^{\circ\pm}(\vec{x})\, e^{\mp i\omega t} \qquad \left(k^2 + \mu^2 = \frac{\omega^2}{c^2}\right) \tag{15}$$

and using formula (12), we obtain

$$\Box \langle A_{\mu}^{\pm}(x) \rangle = \int_{-\infty}^{+\infty} dx' \langle K_{\mu\nu}(x, x') \rangle A_{\nu}^{\circ\pm}(x') e^{\mp i\omega t'}, \tag{16}$$

where

$$\langle K_{\mu\nu}(x,x') \rangle = \langle \Psi_i^{el} K_{\mu\nu}(x, x') \Psi_i^{el} \rangle.$$

We will take into consideration the one-particle model for the electrons of the medium. Then the wave-function of electrons in the position representation

$$\Psi_{a_1 \ldots a_N}^{el}(x_1, \ldots, x_N) = \langle x_1 \alpha_1, \ldots, x_N \alpha_N \mid \Psi^{el} \rangle$$

is a product of one-electron wave-functions.

$$\Psi_{a_1 \ldots a_N}^{el}(x_1, \ldots, x_N) = \Psi_{a_1}^{el}(x_1) \ldots \Psi_{a_N}^{el}(x_N)$$

($\alpha_1, \ldots, \alpha_N$ — spinor labels of N electrons). It can be proved then that

$$\langle \Psi_i^{el}\, \psi_a^{\circ *}(x)\, \psi_{a'}^{\circ}(x')\, \Psi_i^{el} \rangle = N \Psi_{ia'}^{el *}(x) \Psi_{ia}^{el}(x') \tag{17}$$

N — number of medium electrons in state Ψ_i^{el}), where

$$\left[\gamma_{\mu}\left(\frac{\partial}{\partial x_{\mu}} - \frac{ie}{\hbar c} A_{\mu}^{ex}(\vec{x})\right) + \varkappa\right] \Psi_i^{el}(x) = 0.$$

We expand the configuration one-electron wave function (in state Ψ_i^{el}) in eigensolutions $\psi_r(x) = u_r(\vec{x})\, exp\,(-iE_r t/\hbar)$ of the Dirac equation with potential $A_{\mu}^{ex}(\vec{x})$:

$$\Psi_i^{el}(x) = \sum_r c_r \psi_r(x). \tag{18}$$

The coefficients c_r give the initial distribution of electrons in the medium. With our former assumption of equilibrium this distribution is preserved constantly in the medium.

Using succesively (17), (18) and (8), we find after some calculations (cf. Rzewuski 1952), that formula (16) takes the form

$$\Box \langle A_{\mu}^{\pm}(x) \rangle = \int d_3 x'\, K_{\mu\nu}^{\pm}(\vec{x}, \vec{x}'; \omega)\, A_{\nu}^{\circ\pm}(\vec{x}') e^{\mp i\omega t}, \tag{19}$$

where

$$K_{\mu\nu}^{\pm}(\vec{x}, \vec{x}'; \omega) = \frac{e^2}{\hbar} N \sum_{r,r'} P_r \left[\frac{(u_r^*(\vec{x}) \alpha_{\mu} u_{r'}(\vec{x}))\, (u_{r'}^*(\vec{x}') \alpha_{\nu} u_r(\vec{x}'))}{\omega_r - \omega_{r'} \pm \omega} + \right.$$

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$$+ \frac{(u_r^*(\vec{x})\,\alpha_\mu u_{r'}(\vec{x}))^*\,(u_{r'}^*(\vec{x}')\alpha_\nu u_r(\vec{x}'))^*}{\omega_r - \omega_{r'} \mp \omega}\Bigg], \tag{20}$$

here $(\alpha_\mu) = (\vec{\alpha}, i)$, $E_r = \hbar\omega_r$, and $P_r = |c_r|^2$ is the distribution function of electrons of the medium in state Ψ_i^{el} over r states. The operator $\langle A_\mu^\pm(x)\rangle$ has been averaged above over the initial states (phases) of the electrons, using the assumption of a random distribution:

$$\overline{c_r^* c_{r'}} = |c_r|^2 \delta_{rr'}.$$

Moreover, in the above calculations, we have excluded succesive transitions through intermediate states r of the medium electron. These transitions are characterized by the energy conservation law in the intermediate states: $\omega_r - \omega_{r'} \pm \omega = 0$. In reality these transitions do appear for resonance frequencies. In that case the Weisskopf-Wigner method must be used for calculations. In order to exclude from our considerations the case of resonance frequencies, we shall assume that the radiating atom differs from the medium atoms.

In formula (15) operator $A_\mu^{\circ\pm}(\vec{x})$ has the form

$$A_\mu^{\circ\pm}(\vec{x}) = \sqrt{\frac{\hbar c^2}{2V\omega}}\, e_\mu e^{\pm i\vec{k}\vec{x}} a^\pm, \tag{21}$$

where $k^2 + \mu^2 = \frac{\omega^2}{c^2}$, $e_\mu^2 = 1$ $\left((k_\mu) = \left(\vec{k}, i\frac{\omega}{c}\right), (e_\mu) = (\vec{e}, e_4)\right)$, $V = L^3$ is the volume of the normalization cube ($Re\vec{k} = \frac{2\pi}{L}\vec{m}$, $\vec{m}$ — vector with integral numbers as components), and $a^\pm$ are photon absorption and emission operators respectively. The normalization factor appears in consequence of the commutation relation

$$\left[A_\mu^\circ(\vec{x}, t), \frac{1}{c}\frac{\partial A_\nu^\circ(\vec{x}', t)}{\partial t}\right] = i\hbar c\, \delta_{\mu\nu}\, \delta(\vec{x} - \vec{x}').$$

We can now put equation (19) into the form

$$\Box \langle A_\mu^\pm(x)\rangle = (\overset{\pm}{\mu}{}^2(\vec{x}, \vec{k}))_{\mu\nu} A_\nu^{\circ\pm}(x), \tag{22}$$

where

$$(\overset{\pm}{\mu}{}^2(\vec{x}, \vec{k}))_{\mu\nu} = \int d_3 x' e^{\mp i\vec{k}\vec{x}} K_{\mu\nu}^\pm(\vec{x}, \vec{x}'; \omega) e^{\pm i\vec{k}\vec{x}'} \tag{23}$$

describes microscopically in the considered approximation the apparent mass of the photon (without self-mass) in the given crystalline medium. Averaging over a crystal of volume G, we obtain

$$(\overset{\pm}{\mu}{}^2(\vec{k}))_{\mu\nu} = \frac{1}{G}\int_G d_3 x' (\overset{\pm}{\mu}{}^2(\vec{x}, \vec{k}))_{\mu\nu}. \tag{24}$$

The coefficient μ in formulae (5) is now so adjusted that $A_\mu^\circ(x)$ represents „macroscopically" the plane wave propagating in the given medium, i. e. that

$$\Box A_\mu^\circ(x) = (\overset{\pm}{\mu}{}^2(\vec{k}))_{\mu\nu} A_\nu^\circ(x).$$

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Inserting here formulae (15) and (21), we obtain the expression for the macroscopic apparent mass of the photon $\overset{\pm}{\mu}$ (without self-mass)

$$\overset{\pm}{\mu}{}^2 = e_\mu (\overset{\pm}{\mu}{}^2(\vec{k}))_{\mu\nu} e_\nu.$$

Let us notice that we can write

$$k = \sqrt{\frac{\omega^2}{c^2} - \overset{\pm}{\mu}{}^2} = \frac{\overset{\pm}{n}\omega}{c},$$

where ω is a real number, k a complex number und

$$\overset{\pm}{n}{}^2 = 1 - \frac{c^2}{\omega^2}\overset{\pm}{\mu}{}^2 = 1 - \frac{c^2}{\omega^2} e_\mu(\overset{\pm}{\mu}{}^2(\vec{k}))_{\mu\nu} e_\nu. \tag{25}$$

Formula (25) determines in the considered approximation the macroscopic (complex) refractive index. For all directions of propagation $\vec{k}$ and polarization $\vec{e}$, there exists a certain value of the refractive index $\overset{\pm}{n}(\vec{k}, \vec{e})$, which depends besides on the magnitude $Re\, k = 2\pi/\lambda$ (double refraction and dispersion).

We remind that we can without loss of generality gauge $A^\circ_\mu(x)$ so that $A^\circ_4(x) = 0$. Then $e_4 = 0$, thus $\vec{e}^2 = 1$ and from the Lorentz condition $\vec{e}\vec{k} = 0$.

In further calculations we shall assume (for simplicity) that the lattice is cubic. Generalizations do not lead to any essential difficulties. We shall denote by a the edge length of an elementary cell.

We shall now use the assumption that electrons in the medium are assembled in vertices of the crystal lattice. From that assumption it follows that function $K^\pm_{\mu\nu}(\vec{x}, \vec{x}', ;\omega)$ given by formula (20) has near the lattice-points $\vec{a} = \vec{m}a$ ($\vec{m}$ – vector with integers as components) the character of $\delta(\vec{x} - \vec{a})$ and $\delta(\vec{x}' - \vec{a})$, and in other points it is equal to zero:

$$K^\pm_{\mu\nu}(\vec{x}, \vec{x}';\omega)\, e^{\mp i\vec{k}(\vec{x}-\vec{x}')} = \sum_{\vec{a}}\sum_{\vec{a}'} \delta(\vec{x} - \vec{a})\, \delta(\vec{x}' - \vec{a}') e^{\mp i\vec{k}(\vec{a}-\vec{a}')} \int_{A_{\vec{a}}} d_3x \int_{A_{\vec{a}'}} dx' K^\pm_{\mu\nu}(\vec{x}, \vec{x}';\omega),$$

where the integrals expand over regions $A_{\vec{a}}$ and $A_{\vec{a}'}$ of atoms in the lattice-points $\vec{a}$ and $\vec{a}'$.

From formula (23) we obtain

$$\overset{\pm}{(\mu}{}^2(\vec{x}, \vec{k}))_{\mu\nu} = \sum_{\vec{a}}\sum_{\vec{a}'} \delta(\vec{x} - \vec{a}) e^{\pm i\vec{k}(\vec{a}-\vec{a}')} \int_{A_{\vec{a}}} d_3x \int_{A_{\vec{a}'}} d_3x'\, K^\pm_{\mu\nu}(\vec{x}, \vec{x}';\omega). \tag{23'}$$

We can see that $\overset{\pm}{(\mu}{}^2(\vec{x}, \vec{k}))_{\mu\nu}$ is a periodic function with period of the lattice (we assume here that the crystal has infinite extention and all atoms in the lattice-points are identical). From (24) and (23') we obtain further

$$\overset{\pm}{(\mu}{}^2(\vec{k}))_{\mu\nu} = \frac{1}{G}\sum_{\vec{a}}\sum_{\vec{a}'} e^{\mp i\vec{k}(\vec{a}-\vec{a}')} \int_{A_{\vec{a}}} d_3x \int_{A_{\vec{a}'}} d_3x'\, K^\pm_{\mu\nu}(\vec{x}, \vec{x}';\omega). \tag{24'}$$

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Now summing over $\vec{a}$ extends to all lattice-points of the domain G, over $\vec{a}'$ to all lattice-points of the whole medium.

To solve equation (22) we seek a solution of the form

$$\langle A_\mu^\pm(x)\rangle = A_\mu^\pm(\vec{x})e^{\mp i\omega t},\quad A_\mu^\pm(\vec{x}) = f^\pm(\vec{x})\sqrt{\frac{\hbar c^2}{2V\omega}}\,e_\mu e^{\pm i\vec{k}\vec{x}}\left(k = \frac{\overset{\pm}{n}\omega}{c},\ e_\mu^2 = 1\right), \tag{26}$$

where $f^\pm(\vec{x})$ is a periodic function with period of the lattice, i.e.

$$f^\pm(\vec{x}) = \sum_{\vec{q}} f^\pm(\vec{q})e^{i\vec{q}\vec{x}},\ \vec{q} = \frac{2\pi}{a}\vec{m} \tag{27}$$

($\vec{m}$ — vector with integers as components). From (22) and (26) we obtain

$$\left(\triangle \pm 2i\vec{k}\nabla = \frac{\omega^2}{c^2} - k^2\right) f^\pm(x) = e_\mu\,(\overset{\pm}{\mu}{}^2(\vec{x},\vec{k}))_{\mu\nu}\,e_\nu\,a^\pm. \tag{28}$$

We shall solve equation (28) by applying Green's theorem (cf. Born 1933) to the domain g of the elementary cell of the crystal

$$\int_g d_3x\,(u\triangle v - v\triangle u) = \int_f d_2\sigma\left(u\frac{\partial v}{\partial n} - v\frac{\partial u}{\partial n}\right),$$

substituting there $u = exp\,(-i\vec{q}\vec{x})$, $v = f^\pm(\vec{x})$. We obtain then (in consequence of the fact that the surface integral vanishes due to the periodicits of the function $f^\pm(\vec{x})$):

$$f^\pm(\vec{q}) = -\frac{1}{(\vec{q}\pm\vec{k})^2 - k^{\circ 2}}\frac{1}{g}\int_g d_3x e_\mu(\overset{\pm}{\mu}{}^2\,(\vec{x},\vec{k}))_{\mu\nu}e_\nu\ a^\pm e^{-i\vec{q}\vec{x}}, \tag{29}$$

where $k^\circ = \omega/c$, $g = a^3$. We can replace the integral $(1/g)\int_g d_3x$ in formula (29) by $(1/G)\int_G d_3x$, where G is a domain of the crystal containing an integral number of elementary cells (this is allowed, because the function under the integral sign is periodic).

From the fact that function $\overset{\pm}{\mu}{}^2\,(\vec{x},\vec{k})_{\mu\nu}$ has the character of $\delta\,(\vec{x}-\vec{a})$ near the lattice-points $\vec{a} = \vec{ma}$ and vanishes elsewhere in obtain

$$\frac{1}{G}\int_G d_3x e_\mu(\overset{\pm}{\mu}{}^2\,(\vec{x},\vec{k}))_{\mu\nu}e_\nu e^{-i\vec{q}\vec{x}} = \frac{1}{G}\sum_{\vec{a}} e^{-i\vec{q}\vec{a}}\int_g d_3x e_\mu(\overset{\pm}{\mu}{}^2(\vec{x},\vec{k}))_{\mu\nu}e_\nu$$

$$= \frac{1}{G}\int_G d_3x e_\mu(\overset{\pm}{\mu}{}^2(\vec{x},\vec{k}))_{\mu\nu}e_\nu = e_\mu(\overset{\pm}{\mu}{}^2\,(\vec{k}))_{\mu\nu}e_\nu = \frac{\omega^2}{c^2}(1-\overset{\pm}{n}{}^2),$$

for $\vec{q}\vec{a} = \frac{2\pi}{a}\vec{m}\vec{a}\vec{m}' = 2\pi\vec{m}\vec{m}'$, and $exp\,(-i\vec{q}\vec{a}) = 1$. Finally from (26,) (27), and (29) we get

$$A_\mu^\pm(\vec{x}) = \sqrt{\frac{\hbar c^2}{2V\omega}}\,e_\mu\frac{\omega^2}{c^2}(\overset{\pm}{n}{}^2 - 1)\sum_{\vec{q}}\frac{e^{i\vec{q}\vec{x}}}{(\vec{q}\pm\vec{k})^2 - k^{\circ 2}}e^{\pm i\vec{k}\vec{x}}\,a^\pm\left(k^\circ = \frac{\omega}{c},\ k = \frac{\overset{\pm}{n}\omega}{c}\right). \tag{30}$$

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This solution corresponds with Ewald's solution (1912) from classical crystal optics.

The function

$$\sum_{\vec{q}} \frac{e^{i\vec{q}\vec{x}}}{(\vec{q} \pm \vec{k})^2 - k^{\circ 2}} e^{\pm i\vec{k}\vec{x}} \; ; \left(\vec{q} = \frac{2\pi}{a}\vec{m}\right) \tag{31}$$

can be represented (Ewald 1912) as a superposition of spherical waves outgoing from the lattice-points

$$\frac{g}{4\pi} \sum_{\vec{a}} \frac{e^{ik^\circ|\vec{x}-\vec{a}|}}{|\vec{x}-\vec{a}|} e^{\pm i\vec{k}\vec{a}} \; ; (\vec{a} = a\vec{m}\, g = a^3). \tag{31'}$$

To obtain the convergence of these series (in points $\vec{x} \neq \vec{a}$) we must assume (the medium is infinite) that $Im\, k^\circ > |Im\, k|$, and after summing up we must find the limit for $Im\, k^\circ \to 0$, as $k^\circ = \omega/c$ is a real number.

3. *The emission in a crystal*

We shall now investigate the probability of (spontaneous) one-quantum emission in a crystal.

The electrons of the radiating atom will be treated configurationally. We have

$$j_\mu^A(x) = e^{\frac{i}{\hbar}H^A t} j_\mu^A(\vec{x})\, e^{-\frac{i}{\hbar}H^A t}, \; j_\mu^A(\vec{x}) = ec \sum_{i=1}^{Z} \alpha_{\mu i}\, \delta(\vec{x} - \vec{x}_i) \tag{32}$$

(H^A is here the Hamiltonian of the radiating atom, Z — the number of electrons in the atom interacting with radiation).

Next we gauge $A_\mu^\circ(x)$ so that $A_4^\circ(x) = 0$. Then $e_4 = 0$, and from (30) we get $A_4(x) = 0$.

From (11), (32) and (26) we obtain (for a flat hypersurface σ)

$$\langle \Psi_f \Psi(t) \rangle = \frac{e}{\hbar} \langle B | \sum_{i=1}^{Z} \vec{\alpha}_i \vec{A}_{\vec{k}}^{-}(\vec{x}_i) | A \rangle \frac{e^{i(\omega_B - \omega_A + \omega)t} - 1}{\omega_B - \omega_A - \omega},$$

where $\vec{A}_{\vec{k}}^{-}(\vec{x})$ is obtained from $\vec{A}^{-}(\vec{x})$ defined by formula (30) by dropping the emission operator a^{-}, and $|A\rangle = \Psi_i^A$, $|B\rangle = \Psi_f^A$ are the initial and final states of the radiating atom with energies $E_A = \hbar\omega_A$ and $E_B = \hbar\omega_B$ respectively.

The probability per unit time and unit solid angle of photon emission (in the transition $A \to B$) in the direction (ϑ, φ) and with the polarization $\vec{e}$ is given by the well known formula

$$w_{A \to B}(\vartheta, \varphi, \vec{e}) = \frac{2\pi}{\hbar} \frac{e^2}{\hbar} \left(\varrho(\omega) |\langle B| \sum_{i=1}^{Z} \vec{\alpha}_i \vec{A}_{\vec{k}}(\vec{x}_i) |A\rangle|^2 \right)_{\omega = \omega_A - \omega_B} \tag{33}$$

in which

$$\varrho(\omega) = \varrho(Re\, k) \frac{d(Re\, k)}{d\omega} = \frac{V(Re\, k)^2}{(2\pi)^3} \frac{d(Re\, k)}{d\omega},$$

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and $k = \frac{n\omega}{c} = \sqrt{\frac{\omega^2}{c^2} - \mu^2}$, $\mu = \bar{\mu}$, $n = \bar{n}$. Here the apparent mass of the photon $\mu^2 = e_\mu(\mu^2)_{\mu\nu}e_\nu$ depends slightly on ω, because we have excluded from our considerations the resonance frequencies $\omega = |\omega_r - \omega_{r'}|$ (cf. (20)). Therefore we have approximately: $dk/d\omega = \omega/c^2k = 1/nc$, and

$$\varrho(\omega) = \frac{(Re\, n)^3}{|n|^2} \frac{V\omega^2}{(2\pi c)^3} . \tag{34}$$

From (33), (34) and (30) we obtain

$$\underset{A \to B}{\omega(\vartheta, \varphi, \vec{e})} = \alpha \left(\frac{(Re\, n)^3 \omega}{2\pi |n|^2} \left| \langle B | \sum_{i=1}^{Z} \vec{\alpha}_i \vec{e} \frac{\omega^2}{c^2} (n^2 - 1) \sum_{\vec{q}} \frac{e^{i(\vec{q}-\vec{k})\vec{x}_i}}{(\vec{q} - \vec{k})^2 - \frac{\omega^2}{c^2}} |A\rangle \right|^2 \right)_{\omega = \omega_A - \omega_B} \tag{35}$$

where $\alpha = e^2/4\pi\hbar c$ is the fine-structure constant and $k = \frac{n\omega}{c}$.

If the radiating atom is placed in one of the lattice-points of the crystal, e.g. in the point $\vec{a} = 0$, then in equation (35) we must replace function (31) (which is equal to (31') with the lower signs) by the function

$$\frac{g}{4\pi} \sum_{\vec{a} \neq 0} \frac{e^{ik^\circ|\vec{x}-\vec{a}|}}{|\vec{x} - \vec{a}|} e^{\pm i\vec{k}\vec{a}} \tag{36}$$

of the radiation field externally interacting with the atom. This function is equal to

$$\sum_{\vec{q}} \frac{e^{i(\vec{q}\pm\vec{k})\vec{x}}}{(\vec{q} \pm \vec{k})^2 - k^{\circ 2}} - \frac{g}{4\pi} \int d_3q \frac{e^{i(\vec{q}\pm\vec{k})\vec{x}}}{(\vec{q} \pm \vec{k})^2 - k^{\circ 2}} \tag{36'}$$

$\left(\text{in the integral } \vec{q} \text{ changes continuously, in the series } \vec{q} = \frac{2\pi}{a}\vec{m}\right)$. In the above integral the path of integration in the complex plane passes over the pole $-k^\circ$ and under the pole k°.

Let us observe that this integral does not depend on $\vec{k}$. Thus the divergence of this integral at the point $\vec{x} = 0$ is compensated by the divergence of the series at the point $\vec{x} = 0$ independently of the value of $\vec{k}$. We shall use this fact when estimating the probability of emission.

If the wave length $\lambda = 2\pi/Re\, k$ is greater than the double edge length of the crystal cell a, then in functions (31) or (36') the term with $\vec{m} = 0$ is much greater than the following ones and we can approximate these functions by the expression

$$\frac{e^{\pm i\vec{k}\vec{x}}}{\frac{\omega^2}{c^2}(n^2 - 1)} .$$

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Equation (35) takes now form

$$w_{A\to B}(\vartheta, \varphi, \vec{e}) = \alpha \left(\frac{(Re\, n)^3 \omega}{2\pi |n|^2} |\langle B| \sum_{i=1}^{Z} \vec{\alpha}_i \vec{e}\, e^{-i\vec{k}\vec{x}_i} |A\rangle|^2 \right)_{\omega = \omega_A - \omega_B}. \tag{35'}$$

Formula (35') differs from the corresponding emission formula in vacuum only by the fact that in (35') the refractive index n appears in the coefficient and in the exponent. The difference in the exponent will not appear for electric dipole radiation (where we put $exp\,(-i\vec{k}\vec{x}) = 1$) but only for magnetic dipole and electric quadripole radiation (where we put $exp\,(-i\vec{k}\vec{x}) = 1 - i\vec{k}\vec{x}$). The probability of emission for electric dipole radiation will be multiplied in comparison to the probability in vacum by the factor $(Re\, n)^3 |n|^2$, for magnetic dipole and electric quadripole radiation — by the factor $(Re\, n)^3$.

If the wave length λ is smaller than the double edge length of the elementary cell (but greater according to our assumption than the domain of the lattice where electrons interacting with radiation are assembled), then besides the term with $\vec{m} = 0$, then terms for which the value of $\vec{q}$ is near to $\mp 2\vec{k}$ (and $\lambda \approx 2a\,|m|$) will play an important part. For simplicity let us assume that $Re\vec{k}$ has the form $\frac{2\pi}{a}\vec{m}_0$ ($\vec{m}_0$ — a vector with integers as components). Then, using only terms with $\vec{m} = 0$ and $\vec{m} = \mp 2m_0$, we approximate the function (31) or (36') by

$$\frac{e^{\pm i\vec{k}\vec{x}} + e^{\mp i\vec{k}\vec{x}}}{\frac{\omega^2}{c^2}(n^2 - 1)}.$$

Formula (35) will then take the form

$$w_{A\to B}(\vartheta, \varphi, \vec{e}) = \alpha \left(\frac{(Re\, n)^3 \omega}{2\pi |n|^2} \langle B| \sum_{i=1}^{Z} \vec{\alpha}_i \vec{e} (e^{-i\vec{k}\vec{x}_i} + e^{i\vec{k}\vec{x}_i}) |A\rangle|^2 \right)_{\omega = \omega_B - \omega_A} \tag{35''}$$

where $Re\vec{k} = \frac{2\pi}{a}\vec{m}_0$. Thus we can see that for electric dipole radiation the probability of emission of a photon with the wave vector $Re\,\vec{k} = \frac{2\pi}{a}\vec{m}_0$ distinctly increases in comparison with vacuum ($4(Re\, n)^3 / |n|^2$ times in our estimation), whereas for magnetic dipole and alectric quadripole radiation the probability of emission strongly decreases (it is equal to zero in our estimation). For other wave vectors $Re\,\vec{k}$ the approximate value of the emission probabilty is contained between the values (35') and (35'').

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4. *Final Remarks*

The considerations contained in this paper can be transposed to the case of a radiating nucleus in the crystalline medium, if only the wave lengths of the γ-radiation are greater than the dimensions of the nuclei of the medium, for only then the nuclei can be treated as points. But then we must neglect the interaction between γ-radiation and electrons of the medium, because the clouds of electrons diffuse the points of the crystal lattice.

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КРАТКОЕ СОДЕРЖАНИЕ

Круликовский, *Теория излучения в кристалле*

Настоящая работа содержит теорию излучения атома погруженного в кристаллической среде. В первой части автор выводит уравнения рассматриваемого класса проблем из формализма квантовой электродинамики. Появляется при этом в формализме кажущаяся масса фотона. Во второй части автор рассматривает, при некоторых упрощающих предположениях, определение кажущейся массы фотона и решение уравнений поля излучения в кристалле. Полученное решение согласуется с классическим результатом Эвальда. Третья часть содержит оценку вероятности эмиссии излучения в кристалле. Для длин волны вида $\lambda=2a|\vec{m}|$ (a — длина ребра элементарной ячейки, $\vec{m}$ — вектор, которого компоненты — целые числа) возрастает вероятность эмиссии для дипольного электрического излучения, а уменьшается — для дипольного магнитного и квадрупольного электрического.

REFERENCES

Born, M., *Optik*, p. 331, Berlin 1933.

Dyson, F. J., *Phys. Rev.*, **75**, 486 (1949).

Ewald, P. P., *Dissertation*, München 1912; *Ann. Phys.* [Leipzig], **49**, 1, 117 (1916); *Habilitationsschrift*, München 1918.

Feynman, R., *Phys. Rev.*, **76**, 749, 769 (1949).

Rubinowicz, A., *Rep. Progr. Phys.*, **12**. 233 (1949).

Rzewuski, J., *Acta phys. Polon.*, **11**, 179 (1952).

Wick, G. C., *Phys. Rev.*, **80**, 268 (1950).