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ELECTOMAGNETIC WAVES IN A MEDIUM WITH A CONTINUOUS ENERGY SPECTRUM. I.

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The dependence of the polarization vector on the electric field strength is found for a medium with a continuous energy spectrum. An integral expression which defines the polarizability is derived. Dispersion relationships are found between the Hermitian and anti-Hermitian parts of the kernel.

THE wave functions of states of a continuous spectrum extend widely in space. Therefore when such states are excited by electromagnetic waves, the polarization at any point of the medium will depend on the values of the field throughout the entire medium; i.e., there will be a spatial dispersion. Under these conditions the concept of polarizability, defined as the coefficient of proportionality between the polarization vector and the field strength, loses its meaning; one can speak only of a "polarizability kernel" which enters into the corresponding integral expression. The effect of spatial dispersion has been studied by Pekar[1] for exciton states. We consider here the case of arbitrary currentless states.

1. PERTURBATION THEORY FOR A CONTINUOUS SPECTRUM

We use a variant of the method of Weisskopf and Wigner, similar to the one used by Born and Kun Huang[2] to investigate the influence of anharmonicity on the dispersion. The Hamiltonian of the system is

$$\hat{H} = \hat{H}^0 + \hat{H}'_d + \hat{H}',$$

where $\hat{H}_0$ is the unperturbed Hamiltonian, $\hat{H}'_d$ is the dissipative perturbation, and $\hat{H}'$ is the electromagnetic perturbation. The corresponding wave function in the medium (the field is considered classical) is

$$\Psi = \Psi^0 + \Psi' + \Psi'_d,$$

where

$$\Psi^0 = a_0 \exp(-i\omega_0 t)\,\psi_0,$$

describes the ground state,

$$\Psi' = \sum_n a_n \exp(-i\omega_n t)\,\psi_n$$

describes the states into which direct transitions from Ψ^0 can be induced by $\hat{H}'$, and

$$\Psi'_d = \sum_s a_s \exp(-i\omega_s t)\,\psi_s$$

are the states of the quasi-continuous spectrum, into which transitions from Ψ' are possible as a result of $\hat{H}'_d$. For electronic states n, the operator $\hat{H}'_d$ can be found from the interaction of the electrons with the lattice vibrations; for the phonon states, it can be found from the anharmonicity. In the expressions given above, the ψ_m $(m = 0, n, s)$ do not depend on time, and are normalized to unity; $\omega_m = E_m/\hbar$, E_m being the energy of the unperturbed state.

Assuming that at the initial moment of time the system is in the ground state, neglecting interference of the energy dissipations of different n-states and the width of the ground state, and postulating a harmonic electromagnetic perturbation of the form

$$\hat{H}' = \hat{h}^{(+)} \exp(-i\omega t) + \hat{h}^{(-)} \exp(i\omega t), \tag{1}$$

we obtain the stationary solution

$$\Psi' = -\frac{1}{\hbar}\left\{\exp[-i(\omega_0+\omega)t]\sum_n \frac{(n\,|\,\hat{h}^{(+)}\,|\,0)}{\omega_{n0}-\omega-i\gamma_n(\omega)}\,\psi_n + \exp[-i(\omega_0-\omega)t]\sum_n \frac{(n\,|\,\hat{h}^{(-)}\,|\,0)}{\omega_{n0}+\omega-i\gamma_n(-\omega)}\,\psi_n\right\}$$

$$(\omega_{n0} = \omega_n - \omega_0),$$

$$\gamma_n(\omega) = \frac{\pi}{\hbar^2}\lim_{\Delta\omega\to 0}\sum_{s_n}^{\omega,\,\omega+\Delta\omega} |(s_n\,|\,\hat{H}'_d\,|\,n)|^2,$$

in which the summation is carried out over the states s_n which produce dissipation in the states n, the frequencies of which lie in the interval from $\omega_0 + \omega$ to $\omega_0 + \omega + \Delta\omega$.

Ψ' is of the same order of smallness as $\hat{H}'$; Ψ'_d, obviously, is of the order of $\hat{H}'\hat{H}'_d$. Hence the principal (of the order of $\hat{H}'$) time-dependent part of the operator $\hat{f}$ is

$$\bar{f}(t) = -\frac{1}{\hbar}\sum_n\left\{\frac{(0\,|\,\hat{f}\,|\,n)\,(n\,|\,\hat{h}^{(+)}\,|\,0)}{\omega_{n0}-\omega-i\gamma_n(\omega)} + \frac{(n\,|\,\hat{f}\,|\,0)\,(n\,|\,\hat{h}^{(-)}\,|\,0)^*}{\omega_{n0}+\omega+i\gamma_n(-\omega)}\right\}\exp(-i\omega t) + \text{c.c.} \tag{2}$$

2. DEPENDENCE OF THE POLARIZATION ON THE ELECTRIC FIELD

Let us consider the penetration of a transverse field into the medium. If a Coulomb calibration is used, the Coulomb interaction reduces to the portion $\hat{H}^0 + \hat{H}'_d$ of the Hamiltonian, and $\hat{H}'$ is determined by the solenoidal field only;

$$\hat{H}' = -\frac{1}{c}\sum_i (e_i/m_i)\mathbf{A}(\mathbf{r}_i, t)\hat{\mathbf{p}}_i, \qquad \operatorname{div}\mathbf{A}(\mathbf{r}, t) = 0, \quad (3)$$

where $\mathbf{r}_i$, e_i, m_i, and $\hat{\mathbf{p}}_i$ are the coordinates, charge, mass, and momentum operator of the i-th particle, respectively, $\mathbf{A}(\mathbf{r}, t)$ is the vector potential of the field, and c is the velocity of light in vacuum. In the case of harmonic time dependence,

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}(\mathbf{r})\exp(-i\omega t) + \text{c.c} \qquad (4)$$

Substituting (4) into (3) and comparing with (1), we find

$$\hat{h}^{(+)} = -\frac{1}{c}\sum_i (e_i/m_i)\,\mathbf{A}(\mathbf{r}_i)\hat{\mathbf{p}}_i, \qquad \hat{h}^{(-)} = \hat{h}^{(+)+}.$$

For the matrix elements appearing in (2) it is easy to obtain

$$(n|\hat{h}^{(+)}|0) = -\frac{1}{c}\int \mathbf{A}(\mathbf{r})\boldsymbol{\Pi}_n(\mathbf{r})\,d\mathbf{r}, \qquad (5)$$

where

$$\begin{aligned}\boldsymbol{\Pi}_n(\mathbf{r}) = \sum_i (e_i/m_i)\int \psi_n(\mathbf{r}_1, \ldots, \mathbf{r}_{i-1}, \mathbf{r}, \mathbf{r}_{i+1}, \ldots, \mathbf{r}_N)\\ \times \hat{\mathbf{p}}\psi_0(\mathbf{r}_1, \ldots, \mathbf{r}_{i-1}, \mathbf{r}, \mathbf{r}_{i+1}, \ldots, \mathbf{r}_N)\\ \times d\mathbf{r}_1 \ldots d\mathbf{r}_{i-1}\,d\mathbf{r}_{i+1} \ldots d\mathbf{r}_N. \qquad (6)\end{aligned}$$

The intensity of the solenoidal field is

$$\begin{aligned}\mathbf{E}^{(\perp)}(\mathbf{r}, t) &= -\frac{1}{c}\frac{\partial \mathbf{A}(\mathbf{r}, t)}{\partial t} = \frac{i\omega}{c}\mathbf{A}(\mathbf{r})\exp(-i\omega t) + \text{c.c.}\\ &= \mathbf{E}^{(\perp)}(\mathbf{r})\exp(-i\omega t) + \text{c.c.}\end{aligned}$$

Expressing $\mathbf{A}(\mathbf{r})$ in terms of $\mathbf{E}(\mathbf{r})$ in (5) and substituting the result and the analogous expressions for $(n|\hat{h}^{(-)}|0)$ into (2), we obtain

$$\bar{f}(t) = \exp(-i\omega t)\int \mathbf{K}_f(\mathbf{r}; \omega)\,\mathbf{E}^{(\perp)}(\mathbf{r})\,d\mathbf{r} + \text{c.c.}, \qquad (7)$$

$$\mathbf{K}_f(\mathbf{r}, \omega) = -\frac{i}{\hbar\omega}\sum_n \left\{\frac{(0|\hat{f}|n)\,\boldsymbol{\Pi}_n(\mathbf{r})}{\omega_{n0} - \omega - i\gamma_n(\omega)} + \frac{(n|\hat{f}|0)\,\boldsymbol{\Pi}^*_n(\mathbf{r})}{\omega_{n0} + \omega + i\gamma_n(-\omega)}\right\}. \qquad (8)$$

In order to obtain the material Maxwell equation connecting the polarization with the field, we must take for $\hat{f}$ the operator of the electric moment per unit volume

$$\mathbf{G}(\mathbf{r}) = \frac{1}{\Delta v}\sum_i \gamma_{\Delta v}(\mathbf{r}_i)\,e_i(\mathbf{r}_i - \mathbf{r}),$$

where Δv is a physically infinitesimal element of volume around the point $\mathbf{r}$ and

$$\gamma_{\Delta v}(\mathbf{r}) = \begin{matrix} 1, & \mathbf{r} \text{ inside } \Delta v \\ 0, & \mathbf{r} \text{ outside } \Delta v \end{matrix}.$$

In characterizing the electrical state of the material by a polarization vector, we are limiting ourselves to the dipole approximation. The same limitation therefore applies in (5) and (6). The corresponding calculations yield

$$\boldsymbol{\Pi}_n(\mathbf{r}) = (n|\dot{\mathbf{G}}(\mathbf{r})|0) = i\omega_{n0}(n|\mathbf{G}(\mathbf{r})|0). \qquad (9)$$

Substituting (9) and (8) into (7), we obtain

$$\mathbf{P}(\mathbf{r}, t) \equiv \{\overline{\mathbf{G}}(\mathbf{r})\}(t) = \mathbf{P}(\mathbf{r})\exp(-i\omega t) + \text{c.c.}, \qquad (10)$$

$$P_{x'}(\mathbf{r}) = \sum_{y'}\int K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega)\,E^{(\perp)}_{y'}(\mathbf{r}_0)\,d\mathbf{r}_0, \qquad (11)$$

where the polarizability kernel is

$$\begin{aligned}K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) = \frac{1}{\hbar\omega}\sum_n \omega_{n0}\left\{\frac{(0|G_{x'}(\mathbf{r})|n)(n|G_{y'}(\mathbf{r}_0)|0)}{\omega_{n0} - \omega - i\gamma_n(\omega)}\right.\\ \left. - \frac{(n|G_{x'}(\mathbf{r})|0)(0|G_{y'}(\mathbf{r}_0)|n)}{\omega_{n0} + \omega + i\gamma_n(-\omega)}\right\}. \qquad (12)\end{aligned}$$

We shall write the polarizability kernel in the form

$$K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) = Q_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) + R_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega), \qquad (13)$$

where

$$\begin{aligned}Q_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) = \frac{1}{\hbar\omega}\sum_n \omega_{n0}\left\{\frac{(\omega_{n0} - \omega)(0|G_{x'}(\mathbf{r})|n)(n|G_{y'}(\mathbf{r}_0)|0)}{(\omega_{n0} - \omega)^2 + \gamma_n^2(\omega)}\right.\\ \left. - \frac{(\omega_{n0} + \omega)(n|G_{x'}(\mathbf{r})|0)(0|G_{y'}(\mathbf{r}_0)|n)}{(\omega_{n0} + \omega)^2 + \gamma_n^2(-\omega)}\right\},\end{aligned}$$

$$\begin{aligned}R_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) = \frac{i}{\hbar\omega}\sum_n \omega_{n0}\left\{\frac{\gamma_n(\omega)(0|G_{x'}(\mathbf{r})|n)(n|G_{y'}(\mathbf{r}_0)|0)}{(\omega_{n0} - \omega)^2 + \gamma_n^2(\omega)}\right.\\ \left. + \frac{\gamma_n(-\omega)(n|G_{x'}(\mathbf{r})|0)(0|G_{y'}(\mathbf{r}_0)|n)}{(\omega_{n0} + \omega)^2 + \gamma_n^2(-\omega)}\right\}.\end{aligned}$$

It is easy to see that

$$\begin{aligned}Q_{y'x'}(\mathbf{r}_0, \mathbf{r}; \omega) &= Q^*_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega),\\ R_{y'x'}(\mathbf{r}_0, \mathbf{r}; \omega) &= -R^*_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega),\\ Q_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) &= Q^*_{x'y'}(\mathbf{r}, \mathbf{r}_0; -\omega),\\ R_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) &= \bar{R}^*_{x'y'}(\mathbf{r}, \mathbf{r}_0; -\omega).\end{aligned}$$

Here the first two identities show that Q and R are the Hermitian and anti-Hermitian parts of K respectively. The last two identities express the fact that (10) is real.

For a system with a continuous spectrum, the matrix elements $(0|\mathbf{G}(\mathbf{r})|n)$ differ from zero for a wide range of values of $\mathbf{r}$. Hence the portion of (12) which results from these states leads to a considerable spatial dispersion in (11).

3. DISPERSION RELATIONSHIPS FOR THE POLARIZABILITY KERNEL

Taking into account the spatial dispersion, the polarization, which depends causally on the strength of the solenoidal field, can be represented in the form

$$P_{x'}(\mathbf{r}, t) = \sum_{y'} \iint_0^\infty K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \tau)\, E_{y'}^{(\perp)}(\mathbf{r}_0, t - \tau)\, d\tau\, d\mathbf{r}_0, \quad (14)$$

where $K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \tau)$ is real. This is in full agreement with the results obtained above. At the same time, for the case of harmonic time-dependence we obtain, from (11),

$$P_{x'}(\mathbf{r}, t) = \sum_{y'} \int K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) \exp(-i\omega t)\, E_{y'}^{(\perp)}(\mathbf{r}_0)\, d\mathbf{r}_0 + \text{c.c.} \quad (15)$$

For the case of an arbitrary time-dependence of the field,

$$\mathbf{E}^{(\perp)}(\mathbf{r}, t) = \int_0^\infty \exp(-i\omega t)\, \mathbf{E}^{(\perp)}(\mathbf{r}, \omega)\, d\omega + \text{c.c.} \quad (16)$$

and (15) leads to

$$P_{x'}(\mathbf{r}, t) = \sum_{y'} \iint_0^\infty K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) \exp(-i\omega t)\, E_{y'}^{(\perp)}(\mathbf{r}_0, \omega)\, d\omega\, d\mathbf{r}_0 + \text{c.c.} \quad (17)$$

On the other hand, by substituting (16) into (14) we also obtain (17), since

$$K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) = \int_0^\infty \exp(i\omega\tau)\, K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \tau)\, d\tau.$$

As a result of its finite relaxation time, the function $K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \tau)$ in the case of a dielectric tends to zero as $\tau \to \infty$, being finite for all values of τ. Hence, if ω is taken as the complex variable, the function $K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega)$ will have no singularities in the entire upper half-plane (including the real axis). As $\omega \to \infty$ along any path in the upper half-plane, $K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) \to 0$. From the foregoing properties of this function, we obtain in the usual way[3]

$$\int_{-\infty}^{+\infty} \frac{K_{x'y'}(\mathbf{r}, \mathbf{r}_0; x)}{x - \omega}\, dx - i\pi K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) = 0.$$

Inserting (13) here and separating the Hermitian and anti-Hermitian parts of the equation so obtained, we obtain the dispersion relationships that connect the Hermitian and anti-Hermitian parts of the polarizability kernel:

$$R_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) = -\frac{i}{\pi} \int_{-\infty}^{+\infty} \frac{Q_{x'y'}(\mathbf{r}, \mathbf{r}_0; x)}{x - \omega}\, dx,$$

$$Q_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) = -\frac{i}{\pi} \int_{-\infty}^{+\infty} \frac{R_{x'y'}(\mathbf{r}, \mathbf{r}_0; x)}{x - \omega}\, dx. \quad (18)$$

These relationships represent a generalization of the Kramers-Kronig formulas to the case when spatial dispersion is taken into account.

The formulas (18), which apply to the polarizability kernel, will in general not be valid for the complex refractive index, which has been introduced formally and measured experimentally. This fact has been observed in experiments.[4]

4. OUTLINE OF THE SOLUTION OF MAXWELL'S EQUATIONS

The total strength of the macroscopic electric field is

$$\mathbf{E}(\mathbf{r}, t) = \exp(-i\omega t)\, \mathbf{E}(\mathbf{r}) + \text{c.c.},$$

in which $\mathbf{E}(\mathbf{r})$ can be divided into solenoidal and irrotational parts:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{(\perp)}(\mathbf{r}) + \mathbf{E}^{(\parallel)}(\mathbf{r}), \quad \operatorname{div} \mathbf{E}^{(\perp)}(\mathbf{r}) = 0, \quad \operatorname{curl} \mathbf{E}^{(\parallel)}(\mathbf{r}) = 0.$$

Similarly, we put

$$\mathbf{P}(\mathbf{r}) = \mathbf{P}^{(\perp)}(\mathbf{r}) + \mathbf{P}^{(\parallel)}(\mathbf{r}).$$

This corresponds to the separation

$$K_{x'y'}(\mathbf{r}, \mathbf{r}_0; \omega) = K_{x'y'}^{(\perp)}(\mathbf{r}, \mathbf{r}_0; \omega) + K_{x'y'}^{(\parallel)}(\mathbf{r}, \mathbf{r}_0; \omega),$$
$$\operatorname{div}_{\mathbf{r}} \mathbf{K}_{y'}^{(\perp)}(\mathbf{r}, \mathbf{r}_0; \omega) = 0, \quad \operatorname{curl}_{\mathbf{r}} \mathbf{K}_{y'}^{(\parallel)}(\mathbf{r}, \mathbf{r}_0; \omega) = 0,$$

where $(\mathbf{K}_{y'})_{x'} = K_{x'y'}$. In this case

$$P_{x'}^{(\perp)}(\mathbf{r}) = \sum_{y'} \int K_{x'y'}^{(\perp)}(\mathbf{r}, \mathbf{r}_0; \omega)\, E_{y'}^{(\perp)}(\mathbf{r}_0)\, d\mathbf{r}_0, \quad (19)$$

$$P_{x'}^{(\parallel)}(\mathbf{r}) = \sum_{y'} \int K_{x'y'}^{(\parallel)}(\mathbf{r}, \mathbf{r}_0; \omega)\, E_{y'}^{(\perp)}(\mathbf{r}_0)\, d\mathbf{r}_0. \quad (20)$$

From Maxwell's equations we find

$$\mathbf{E}^{(\parallel)}(\mathbf{r}) + 4\pi \mathbf{P}^{(\parallel)}(\mathbf{r}) = 0 \quad (21)$$

and, taking (19) into account,

$$\Delta_{\mathbf{r}}^2 E_{x'}^{(\perp)}(\mathbf{r}) + (\mu\omega^2/c^2)\, E_{x'}^{(\perp)}(\mathbf{r}) + (4\pi\mu\omega^2/c^2) \sum_{y'} \int K_{x'y'}^{(\perp)}(\mathbf{r}, \mathbf{r}_0; \omega)\, E_{y'}^{(\perp)}(\mathbf{r}_0)\, d\mathbf{r}_0 = 0, \quad (22)$$

where μ is the magnetic permeability, which for simplicity will be considered to be a scalar. (22) is a system of equations for the determination of $\mathbf{E}^{(\perp)}(\mathbf{r})$. When $\mathbf{E}^{(\perp)}(\mathbf{r})$ has been found it is then possible, according to (20) and (21), to find

$$E_{x'}^{\parallel}(\mathbf{r}) = -4\pi \sum_{y'} \int K_{x'y'}^{(\parallel)}(\mathbf{r}, \mathbf{r}_0; \omega)\, E_{y'}^{(\perp)}(\mathbf{r}_0)\, d\mathbf{r}_0.$$

Note that the expression $\exp(i\mathbf{k} \cdot \mathbf{r})$ is not, generally speaking, a solution of (22), since the spatial dispersion leads to complications in the spatial behavior of monochromatic waves.

The application of the theory developed here to the case of exciton states in a crystal will be given later.

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171