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310

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# *ELECTROMAGNETIC WAVES IN ISOTROPIC AND CRYSTALLINE MEDIA CHARACTERIZED BY DIELECTRIC PERMITTIVITY WITH SPATIAL DISPERSION*

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The dielectric permittivity tensor $\epsilon_{ik}$ is usually taken to be a function of frequency alone, i.e. one neglects spatial dispersion — the dependence of $\epsilon_{ik}$ on the wavelength. However, even in non-gyrotropic media spatial dispersion must be considered in cases of weak absorption, when the refractive index increases rapidly and becomes infinite if dispersion and absorption are not taken into account. Spatial dispersion is also important in the analysis of longitudinal (plasma) waves which propagate in an isotropic medium or along the principal dielectric axes in crystals. It is also shown that spatial dispersion leads to a weak optical anisotropy in cubic crystals. In addition to the above, an analysis is made of the collective (discrete) energy losses in solids.

## 1. INTRODUCTION

In analyzing the propagation of light and electromagnetic waves of longer wavelengths in a medium, one usually uses the local relation

$$D_i = \varepsilon_{ik}(\omega)E_k, \tag{1.1}$$

where **D** and **E** are taken at $\omega$, the frequency of the Fourier components of the electric induction and field intensity at the point **r**. If there is absorption, the tensor $\epsilon_{ik}$ becomes complex and **D** must be replaced by $\mathbf{D} - i(4\pi/\omega)\mathbf{j}$ where $\mathbf{j}$ is the density of the conduction current. In order to simplify the analysis this substitution is implied below, but not carried out explicitly.

The relation in (1.1) does not reflect the nature of the field variation in space, that is to say, it applies only if we neglect spatial dispersion — the dependence of the tensor $\epsilon_{ik}$ on the wavelength. The spatial dispersion can be characterized by the parameter $a/\lambda = an/\lambda_0$, where $a$ is a characteristic length for a given medium (molecular dimensions, lattice constants, Debye radius, etc.), $\lambda_0 = 2\pi c/\omega$ is the wavelength in vacuum, $\lambda = \lambda_0/n$ is the wavelength in the medium and $n$ is the index of refraction. In condensed media in the optical region usually $a/\lambda_0 \sim 1$ to $3 \times 10^{-3}$ so that spatial dispersion is negligibly small in most cases.* This, however, is not the case if we are interested in effects associated with spatial inhomogeneities of the field. A well-known example of this type is natural optical activity — an effect which is of order $a/\lambda$. It will be shown below that taking terms of order $(a/\lambda)^2$ into account leads to an additional effect — weak optical anisotropy in cubic crystals.

*The time dispersion, which leads to a dependence of $\varepsilon_{ik}$ on $\omega$ may be large under these same conditions because it is characterized by the parameter $\omega/\omega_j$, where $\omega_j$ is a characteristic frequency of the medium.

It is also necessary to go beyond the local relation (1.1), as is well-known, when one considers longitudinal (plasma) waves in a medium (ionized gas, solids). Finally, in cases of weak absorption, it is necessary to take account of terms of order $(a/\lambda)^2$ at frequencies close to characteristic frequencies of the medium, i.e., in regions in which the refractive index n becomes very large. This situation is completely understandable because when $n \to \infty$, the parameter $an/\lambda_0$ also increases without limit. This case has been already considered in an analysis of waves in a magneto-active plasma close to resonance[1] and in absorption of light by excitons in crystals.[2] Although calculations involving spatial dispersion have already been carried out in individual cases, it would appear that there are still a number of points which are not entirely clear. For this reason certain aspects of this problem are considered below.

If small, the effect of spatial dispersion can be taken into account by writing the relation between **D** and **E** in the form

$$D_i = \varepsilon_{ik}(\omega) E_k + \gamma_{ikl}(\omega) \frac{\partial E_k}{\partial x_l} + \delta_{iklm}(\omega) \frac{\partial^2 E_k}{\partial x_l \partial x_m}. \quad (1.2)$$

The term containing $\gamma_{ikl}$ is responsible for optical activity (cf. reference 3, §83) and vanishes if there is a center of aymmetry in the body. Below we shall assume with one important exception, that $\gamma_{ikl} = 0$. Considering plane waves, for which the field is proportional to the factor $\exp\{i(\omega t - \mathbf{k}\cdot\mathbf{r}\} = \exp\{i(\omega t - \omega \hat{n}\mathbf{s}\cdot\mathbf{r}/c)\}$ we write (1.2) in the form (we consider only plane waves for which the planes of constant phase coincide with planes of constant amplitude):

$$D_i = \hat{\varepsilon}_{ik} E_k, \quad \hat{\varepsilon}_{ik} = \varepsilon_{ik} - \alpha_{iklm} s_l s_m \hat{n}^2, \quad \alpha_{iklm} = (\omega / c)^2 \delta_{iklm}, \quad (1.3)$$

where **s** is the unit vector normal to the wave, $\hat{n} = n - i\kappa$ is the complex refractive index (n is the index of refraction, $\kappa$ is the absorption factor). The coefficient $\delta$ is of the order of the square of the characteristic length a and $|\alpha_{iklm}| \sim (a/\lambda_0)^2$. It is apparent that the tensor $\alpha_{iklm}$ can always be assumed symmetric with respect to the indices $l$ and m. Using the principal of symmetry for the kinetic coefficients it can be shown (reference 3, §83) that the tensor $\alpha_{iklm}$ is also symmetric with respect to the indices i and k (it is assumed that there is no external field). In the absence of absorption, this tensor is real. Further simplification of the tensor derives from the symmetry of the medium.

The expansion given in (1.2) and (1.3) is not always possible. For example, if one of the components $\epsilon_{ab}$ of the tensor $\epsilon_{ik}$ tends toward infinity, terms of order $(a/\lambda_0)^2 n^2$ can always be neglected as compared with it. On the other hand, the quantity $1/\epsilon_{ab}$ becomes small and the last term may become important in the expansion

$$1/\hat{\varepsilon}_{ab} = 1/\varepsilon_{ab} + \beta_{ablm} s_l s_m \hat{n}^2$$

Thus, the tensor $\hat{\epsilon}_{ik}$ is sometimes replaced by $\hat{\epsilon}_{ik}^{-1}$:

$$E_i = \hat{\varepsilon}_{ik}^{-1} D_k, \quad \hat{\varepsilon}_{ik}^{-1} = \varepsilon_{ik}^{-1} + \beta_{iklm} s_l s_m \hat{n}^2. \quad (1.4)$$

The symmetry properties of this tensor are the same as those of $\epsilon_{ik}$. The introduction of two tensors $\hat{\epsilon}_{ik}$ and $\hat{\epsilon}_{ik}^{-1}$ should not occasion surprise since the vectors **D** and **E** are of equal rank. In order of magnitude terms the coefficients $\alpha$ and $\beta$ are generally the same. The choice of one or the other of the expansions (1.3) or (1.4) is dictated by the nature of the individual problem and will be clarified below.

The propagation of plane waves is determined by the field equations

$$\mathbf{H} = \hat{n}[\mathbf{s}\times\mathbf{E}], \quad \mathbf{D} = -\hat{n}[\mathbf{s}\times\mathbf{H}], \quad (1.5)$$

whence

$$\mathbf{D} = \hat{n}^2 \{\mathbf{E} - \mathbf{s}(\mathbf{s}\cdot\mathbf{E})\}. \quad (1.6)$$

Here **H** is the magnetic field associated with the wave, the magnetic permeability $\mu$ is set equal to unity and the medium is assumed homogeneous (the same applies in (1.3), (1.4), and below).

Substituting in Eq. (1.6) the relation given in (1.1), we obtain the well-known equation for $\hat{n}^2$:

$$(\varepsilon_x s_x^2 + \varepsilon_y s_y^2 + \varepsilon_z s_z^2)\hat{n}^4 - [\varepsilon_x(\varepsilon_y + \varepsilon_z) s_x^2 + \varepsilon_y(\varepsilon_x + \varepsilon_z) s_y^2 + \varepsilon_z(\varepsilon_x + \varepsilon_y) s_z^2]\hat{n}^2 + \varepsilon_x\varepsilon_y\varepsilon_z = 0, \quad (1.7)$$

where $\epsilon_x$, $\epsilon_y$ and $\epsilon_z$ are the principal values of the tensor $\epsilon_{ik}$. It is characteristic that Eq. (1.7) is quadratic in $\hat{n}^2$ whereas the system in (1.6) consists of three homogeneous equations and, in the general case, leads to a cubic equation in $\hat{n}^2$. The coefficient for $\hat{n}^6$ can be set equal to zero only when the local relation in (1.1) applies; if, on the other hand, (1.3) applies, the equation in $\hat{n}^2$ is of the third degree. This fact is responsible for the appearance of the additional "proper" wave. These considerations indicate the need for taking spatial dispersion into account under certain conditions.

## 2. ISOTROPIC MEDIUM

In an isotropic medium the tensors $\hat{\epsilon}_{ik}$, $\epsilon_{ik}$, $\alpha_{iklm}$ and $\beta_{iklm}$ become scalars and from (1.3) we have*

$$D = \hat{\varepsilon}E, \quad \hat{\varepsilon} = \varepsilon - \alpha\hat{n}^2. \tag{2.1}$$

For transverse waves, in which $s \cdot E = 0$, substitution of (2.1) in (1.6) leads to the double root $\hat{n}_{1,2}^2 = \epsilon/(1+\alpha)$ which, because $\alpha$ is so small, virtually coincides with the value $\hat{n}^2 = \epsilon$ which is obtained when spatial dispersion is neglected. For longitudinal waves, in which $s(s \cdot E) = E$, we have:

$$\hat{n}_3^2 = \varepsilon(\omega)/\alpha. \tag{2.2}$$

In a longitudinal wave $D = 0$, as is clear from the condition $\operatorname{div} D = -(i\omega/c)\, n\, s \cdot D = 0$ or, better still, from Eq. (1.6). However, the field $E$ associated with this wave may be different from zero since the expression in (2.2) is equivalent to $\hat{\epsilon} = 0$, which, according to Eq. (2.1), is compatible with the conditions $D = 0$ and $E \neq 0$. If spatial dispersion is neglected, when $\hat{\epsilon} = \epsilon$ a longitudinal wave is possible only if

$$\varepsilon(\omega) = 0. \tag{2.3}$$

This condition is also clear from Eq. (2.2) since $\hat{n}_3^2$ remains finite as $\alpha \to 0$ only if $\epsilon \to 0$.

The existence condition for longitudinal waves in the form given in (2.3) was given a long time ago (cf. for example reference 4, §4) but has been a subject of discussion until very recently.[5,6]

If spatial dispersion is neglected the longitudinal waves are characterized by a vanishing group velocity $u = d\omega/dk$ and are associated only with discrete frequencies $\omega_l$ which satisfy (2.3). When $\alpha \neq 0$ the longitudinal waves assume the same importance as the proper waves in the medium. Longitudinal waves are well-known in plasmas: if collisions are neglected,

$$\varepsilon(\omega) = 1 - 4\pi e^2 N/m\omega^2 = 1 - \omega_0^2/\omega^2$$

and, if a Maxwellian velocity distribution obtains and the quasi-hydrodynamic approximation is used $\alpha = \kappa T/mc^2$. On the other hand, if the calculation is carried out using the kinetic equation, $\alpha = 3\kappa T/mc^2$ where $\kappa$ is the Boltzmann constant and $T$ is the temperature. In a plasma longitudinal waves are not strongly attenuated close to the frequency $\omega_0$ only when the index $n_3$ is small (plasma waves are considered in detail in reference 1). In the general case, in a plasma we have $\alpha \sim (v_0/c)^2$ where $v_0$ is some characteristic velocity (in a degenerate gas $v_0$ is the order of the velocity at the Fermi limit). This result for $\alpha$ is found to be in agreement with an estimate made earlier $\alpha \sim (a/\lambda_0)^2 = (\omega a/2\pi c)^2$ since, for a gas in a high frequency field the characteristic length $a \sim 2\pi v_0/\omega$, that is, the path traversed by the particle during one oscillation period (in order of magnitude terms $2\pi v_0/\omega_0$ is the shielding radius or Debye radius).

In solids the longitudinal waves are similar to plasma waves and, as is well known, are described by "plasmons" when quantized.[6] Just as in the plasma case, the attenuation of these longitudinal waves can be small only in the neighborhood of frequencies $\omega_l$, at which $\epsilon(\omega_l) = 0$. We may note that in principal $\alpha$ can also be negative, as in optical vibrations of a solid lattice. When $\alpha < 0$ obviously $\hat{n}_3^2 > 0$ for $\epsilon(\omega) < 0$.

For transverse waves, in the region $\hat{n}_{1,2}^2 = \epsilon \to \infty$ the expansion in (2.1) is not affected although one would expect strong effects due to spatial dispersion (cf. Introduction). However, because the quantity $\epsilon^{-1}$ is so small it is necessary to use an expansion of form (1.4) which applies for an isotropic medium (it is assumed that $s \cdot D = 0$)

$$E = D/\hat{\varepsilon}, \quad 1/\hat{\varepsilon} = 1/\varepsilon + \beta\hat{n}^2. \tag{2.4}$$

In the case of longitudinal waves, substituting Eq. (2.4) in Eq. (1.6) we obtain the condition $\hat{\epsilon} = 0$, that is to say, when $\epsilon \neq 0$ we have $\hat{n}_3^2 = \infty$. In other words, in the resonance region, as well as at frequencies far from $\omega_l$, it is impossible for longitudinal waves to exist in an isotropic medium (keeping in mind the fact that macroscopic waves exist only when $n \ll \lambda_0/a$; (cf. below). For transverse waves, from Eqs. (2.4) and (1.6) we obtain the equation $\beta\hat{n}^4 + \hat{n}^2/\epsilon - 1 = 0$, whence

$$\hat{n}^2 = -1/2\varepsilon\beta \pm \sqrt{(1/2\varepsilon\beta)^2 + 1/\beta}. \tag{2.5}$$

Having in mind the frequency region close to an absorption line we may use the following expression for $\epsilon$:

$$\varepsilon(\omega) \equiv \varepsilon_1 - i\varepsilon_2 = \varepsilon_0 + \frac{4\pi e^2 N_{\text{eff}}/m}{\omega_j^2 - \omega^2 + i\omega\nu}$$

$$\approx \varepsilon_0 - \frac{A\xi}{\xi^2 + \delta^2} - i\frac{A\delta}{\xi^2 + \delta^2}, \tag{2.6}$$

$$\xi = (\omega - \omega_j)/\omega_j, \quad \delta = \nu/2\omega_j, \quad A = 2\pi e^2 N_{\text{eff}}/m\omega_j^2.$$

In the absence of absorption, in which case $\delta = 0$,

$$\varepsilon(\omega) = \varepsilon_1(\omega) = \hat{n}_0^2 = \varepsilon_0 - A/\xi, \tag{2.7}$$

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*In an isotropic medium a tensor of the 4-th rank has two independent components which, in (1.3) correspond to the expansion

$$D = \varepsilon E + \delta_1 \Delta E + \delta_2 \operatorname{grad} \operatorname{div} E = (\varepsilon - \alpha_1\hat{n}^2)E - \alpha_2 s(sE)\hat{n}^2.$$

Thus, for transverse waves, in Eq. (2.1) $\alpha = \alpha_1$; for longitudinal waves $\alpha = \alpha_1 + \alpha_2$. In (1.4), because of the condition $\operatorname{div} D = 0$ in an isotropic medium, there is only one coefficient $\beta$, i.e., $\beta_{iklm}$ can be replaced by a scalar unconditionally.

where $\hat{n}_0$ is the refractive index when both absorption and spatial dispersion are neglected. From Eq. (2.5) it is clear that when

$$\varepsilon^2|\beta| \ll 1, \tag{2.8}$$

$$\hat{n}_2^1 = \varepsilon(1 - \varepsilon^2\beta + \cdots), \quad \hat{n}_2^2 = -1/\varepsilon\beta - \varepsilon + \cdots. \tag{2.9}$$

With $\beta \sim 10^{-6}$, the condition given in (2.8) becomes $\hat{n}_0^2 \ll 10^3$, and when $\epsilon_0 \sim 1$ and $A \sim 1$ [cf. Eq. (2.7)] we find $|\xi| = |\omega - \omega_j|/\omega_j \gg 10^{-3}$; in the optical region, with $\lambda_j = 2\pi c/\omega_j \sim 5 \times 10^{-5}$ cm this means that the expressions in (2.9) apply at distances $\Delta\lambda \gg 10^{-3}\lambda_j \sim 5$ A from the center of the absorption line. In this region it is obvious that $\hat{n}_1^2 = \hat{n}_0^2$ since, if absorption is present, the root $n_1^2$ is approximately equal to $\hat{n}^2$ which corresponds to the case $\beta = 0$. However, the root $\hat{n}_2^2$ is very large and, when $\epsilon \sim 1$, as can be the case far from the line, $|\hat{n}_2^2| \sim 1/|\beta| \sim 10^6$. In this case $\lambda = \lambda_0/n_2 \sim 5 \times 10^{-8}$ and Eqs. (1.3), (1.4), (2.1), and (2.4) no longer apply, so that macroscopic analysis of the problem is no longer meaningful. The new root of the dispersion equation $n_2$ is real only close to the line, in the region where $\lambda = \lambda_0/n_2 \sim \sqrt{|\beta|}\,\lambda_0 \gg a \sim 3 \times 10^{-8}$, i.e., as long as $n_2 \ll \lambda_0/a$. It will be assumed below that this condition is satisfied.

As is clear from Figs. 1 and 2, close to the absorption line spatial dispersion has a qualitative effect on the $\hat{n}^2(\omega)$ curves. Both of the curves refer to the case $A = 1$ and $\delta = 0$ but in Fig. 1 the value $\beta = 10^{-5}$ has been taken while in Fig. 2 $\beta = -10^{-5}$. The dashed curves in both cases represent the limiting curve (2.7), also for $A = 1$ (generally $\epsilon_0 \sim 1$ and since we are interested in the region $|\epsilon| \gg 1$ we may set $\epsilon_0 = 0$ everywhere). We may note that if there is no absorption and if $\hat{n}^2$ is real with $\hat{n}^2 > 0$ it is obvious that $\hat{n}^2 = n^2$; for $\hat{n}^2 < 0$ we have $\hat{n}^2 = -\kappa^2$, i.e., there is total internal reflection from the medium. For $\beta = 0$ and $\beta > 0$ we are dealing with cases of this kind since $\hat{n}^2$ is real. If, however $\beta < 0$ in the region $|\epsilon| > 1/2\sqrt{|\beta|}$ the values $(\hat{n}^2 = n - i\kappa)^2$ are complex (in this region of Fig. 2 there are no solid curves).

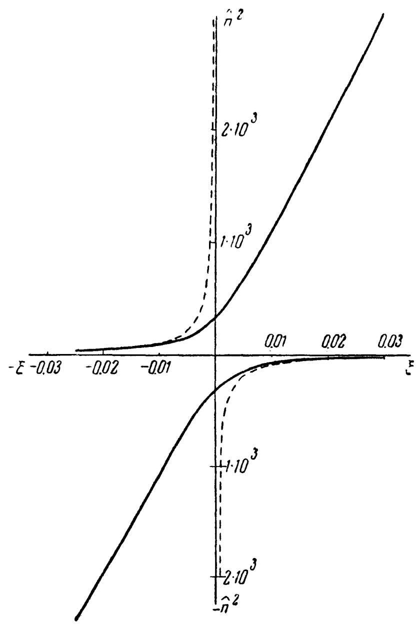


FIG. 1

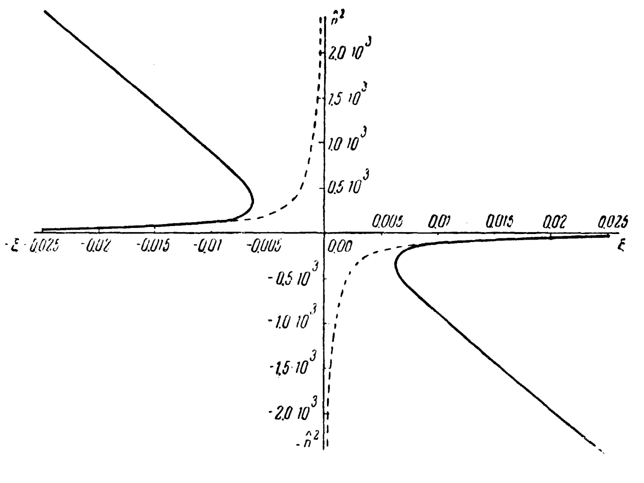


FIG. 2

Thus, when spatial dispersion is taken into account, near a resonance (absorption line), for a given value of $\omega$ we find a new root for $\hat{n}^2$ (more precisely double roots, because of the two independent directions of polarization). The peculiar behavior of the $\hat{n}^2$-curves close to resonance, shown in Figs. 1 and 2, was known earlier from work on magneto-active plasmas;[1] in the crystal case an expression such as (2.5) has been obtained recently by Pekar[2] starting from a concrete model.*

To a considerable degree the possibility of observing the new wave close to resonance depends on the amount of absorption. In the absence of absorption the effect of spatial dispersion is large in the region in which $4\epsilon_1^2|\beta| \sim 1$, i.e., when $|\xi| \sim \xi_k = 2A\sqrt{|\beta|}$. Furthermore, if

$$\delta \ll \xi_k = 2A\sqrt{|\beta|}, \tag{2.10}$$

at a frequency such that $|\xi| = |\omega - \omega_j|/\omega_j \geq \xi_k$ the absorption has only a small effect on the quantity $\epsilon_1 = \mathrm{Re}\,\epsilon$ while $\epsilon_2 = \mathrm{Im}\,\epsilon \ll |\epsilon_1|$. Similarly the curves $n^2(\omega)$ and $\kappa^2(\omega)$ are not changed if we neglect the effect of attenuation in the region which is completely transparent for $\delta = 0$. Suppose, for example, the condition in (2.10) is satisfied, $\beta > 0$, and we consider a frequency for which $4\epsilon_1^2|\beta| = 1$. Then, with $\xi < 0$ we have $n \approx 0.6\,\beta^{-1/4}$

*The fact that the results obtained by Pekar[2] correspond to an expansion such as that given in (2.4) was noted still earlier by L. D. Landau in conversation with the author.

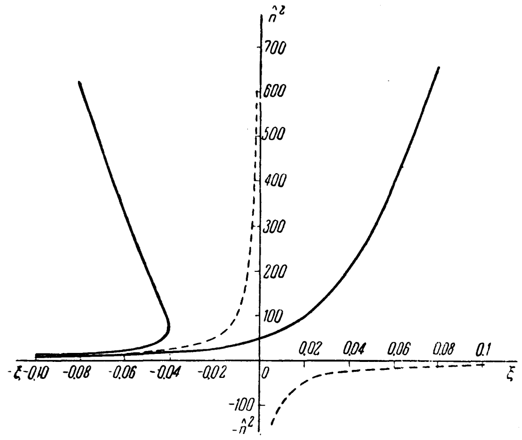


FIG. 3

and $\kappa \approx 0.2(\delta/\xi_k)\beta^{-1/4}$, which, for $\xi_k \sim 10^{-3}$, $\beta \sim 10^{-6}$ and $\delta \sim 10^{-7}$ yields $n \sim 20$ and $\kappa \sim 6 \times 10^{-4}$. Since the radiation intensity falls off in accordance with the relation $I = I_0 e^{-2\omega\kappa z/c} = I_0 e^{-\mu z}$, this means that $\mu = (2\omega/c)n \sim 150\ \text{cm}^{-1}$ (for $\lambda_0 \sim 5 \times 10^{-5}$ cm). For the same values but with $\beta < 0$:

$$n = 1/\sqrt{2\varepsilon_1|\beta|} = |\beta|^{-1/4} \approx 30, \quad \varkappa = \sqrt{\varepsilon_2/\varepsilon_1}/2(2\varepsilon_1|\beta|)^{1/4} = \sqrt{\delta/\xi_k}/2|\beta|^{1/4} \approx 0.15,$$

i.e., $\mu \sim 3 \times 10^4\ \text{cm}^{-1}$. Thus the intensity is reduced by a factor of e in a distance $\lambda_0/1.8 = 3 \times 10^{-5}$ cm whereas $\lambda = \lambda_0/n \approx 2 \times 10^{-6}$ cm. Hence, even in the second case, the attenuation in one wavelength in the medium is relatively small although it is enormous in terms of macroscopic distances. With $\beta < 0$ and $4\epsilon_1^2|\beta| < 1$ the absorption is less and in order of magnitude terms is approximately the same as that for $\beta > 0$. On the other hand, in actual crystals it is probably always true that $\delta \gg 10^{-7}$. From these examples it is clear that the observation of spatial dispersion close to absorption lines is very much complicated by absorption effects and would require very special and favorable conditions (assuming ordinary optical methods of observation).

Because of the small spatial dispersion in a non-gyrotropic medium, it is of special interest to consider the resonance region in naturally active materials in which case the effect of interest is of order $a/\lambda$. For optically active isotropic solids and cubic crystals far from resonance (cf. reference 3, §83)

$$\mathbf{D} = \varepsilon\mathbf{E} - if[\mathbf{s}\times\mathbf{E}]\hat{n}. \tag{2.11}$$

Assuming large values of $\epsilon$ it is necessary to use the related expression

$$\mathbf{E} = \mathbf{D}/\varepsilon + ig[\mathbf{s}\times\mathbf{D}]n. \tag{2.12}$$

Substituting this expression in (1.6), for transverse waves we have:

$$g^2\hat{n}^6 - (\hat{n}^2/\varepsilon - 1)^2 = 0. \tag{2.13}$$

With $gn_0^3 \ll 1$, $n_0^2 = \epsilon$ it is easily shown that

$$\hat{n}_{1,2}^2 = n_0^2(1 \pm gn_0^3), \quad \hat{n}_3^2 = 1/\varepsilon^2 g^2. \tag{2.14}$$

The curves for real values of $\hat{n}_{1,2,3}^2$ are shown in Fig. 3 for $\epsilon = -1/\xi$, $g^2 = 10^{-5}$. We may note that the multiple root corresponds to the values

$$\varepsilon_m = 2^{2/3}/3g^{2/3}, \quad \hat{n}_m^2 = (2/g)^{2/3}, \quad \hat{n}^2 = \frac{1}{4}(2/g)^{2/3}$$

whence, with $g \sim 3 \times 10^{-3}$ we have: $\epsilon_m = -1/\xi_m \sim 25$ and $\xi_m = |\Delta\omega|/\omega_j \sim 4 \times 10^{-2}$ or, in the optical region, $\Delta\lambda \sim 100$ to 200 A. It is also apparent that in the case of a gyrotropic medium the absorption will not be as effective in masking the effect of spatial dispersion simply because the latter effect is so much stronger.

## 3. CRYSTALLINE MEDIUM

In rhombic, tetragonal, and cubic crystals the principle axes of the tensor $\epsilon_{ik}$ coincide with the symmetry axes (these will also be used as the coordinate axes x, y and z below). Under these conditions the tensor $\alpha_{iklm}$ is simplified and in rhombic, tetragonal, and cubic crystals has 12, 7 and 3 independent components respectively. Thus, in the tetragonal case the nonvanishing components are

$$\alpha_{xxxx} = \alpha_{yyyy}, \quad \alpha_{zzzz}, \quad \alpha_{xxyy} = \alpha_{yyxx}, \quad \alpha_{xxzz} = \alpha_{yyzz}, \quad \alpha_{zzxx} = \alpha_{zzyy}, \quad \alpha_{xzxz} = \alpha_{yzyz} \text{ and } \alpha_{xyxy},$$

where the z axis is the axis of 4-th order symmetry. In a cubic crystal the non-vanishing components are

$$\alpha_1 = \alpha_{xxxx} = \alpha_{yyyy} = \alpha_{zzzz}, \quad \alpha_2 = \alpha_{xxyy} = \alpha_{yyxx} = \alpha_{xxzz} = \alpha_{zzxx} = \alpha_{yyzz} = \alpha_{zzyy} \text{ and } \alpha_3 = \alpha_{xyxy} = \alpha_{xzxz} = \alpha_{yzyz}.$$

Whence it is clear that a weak optical anisotropy should be observed in cubic crystals where $\epsilon_{ik} = \epsilon\delta_{ik}$. Far from frequencies at which $\epsilon$ is zero or infinite, to a first approximation $\hat{n}^2 = n_0^2 = \epsilon$ (it is assumed that $\epsilon > 0$ and that absorption can be neglected). To take account of the weak anisotropy is obviously necessary to substitute this value $\hat{n}^2 = n_0^2$ for $\hat{\epsilon}_{ik}$ in (1.3). The resulting tensor $\hat{\epsilon}_{ik}$ is of the form:

$$\hat{\varepsilon}_{xx} = \varepsilon[1 + \alpha_1 s_x^2 + \alpha_2(s_x^2 + s_z^2)],$$
$$\hat{\varepsilon}_{yy} = \varepsilon[1 + \alpha_1 s_y^2 + \alpha_2(s_x^2 + s_z^2)], \tag{3.1}$$
$$\hat{\varepsilon}_{zz} = \varepsilon[1 + \alpha_1 s_z^2 + \alpha_2(s_x^2 + s_y^2)], \quad \hat{\varepsilon}_{xy} = 2\varepsilon\alpha_3 s_x s_y,$$
$$\hat{\varepsilon}_{xz} = 2\varepsilon\alpha_3 s_x s_z, \quad \hat{\varepsilon}_{yz} = 2\varepsilon\alpha_3 s_y s_z.$$

The principle axes of the tensor $\hat{\epsilon}_{ik}$ depend on the direction of the normal and in the general case will not coincide with the axes of 4-th order symmetry x, y and z. To detect the weak birefringence it is probably easiest to observe the ellipticity of the light transmitted through the crystal. The ellipticity is defined by the difference in the refractive indices for waves with different polarization $\Delta n \sim n_0\alpha$, where $\alpha$ is a suitable combination of the coefficients $\alpha_i$ and $s_i$. With $n_0 \sim 1$ and $\alpha \sim 10^{-6}$ the phase shift $\Delta\varphi = (2\pi/\lambda_0)\Delta n l \sim 0.1 l$ where $l$ is the path in cm traversed by the light in the crystal. An effect of this order of magnitude is detectable. On the other hand it should be kept in mind that the values of $\alpha$ may be somewhat smaller than those which have been assumed and that the observation may be complicated by possible directional effects and other factors.

When the light propagates along a cubic axis, there is no birefringence and when $\epsilon \to 0$ or $\epsilon \to \infty$, one should observe the features discussed above for the isotropic case. For other directions of $\mathbf{s}$ and $\epsilon = 0$ all the roots $\hat{n}^2_{1,2,3} = 0$ and in this region one of the waves is like a plasma wave while the others are like transverse waves. In other words, the picture is similar to that which obtains for the isotropic case, being only slightly complicated by the weak anisotropy. The same considerations apply to the region $\epsilon \to \infty$ in which it is necessary to make use of the expansion in (1.4).

In non-cubic crystals, in which the tensor $\epsilon_{ik}$ is not completely degenerate, in the general case the situation is entirely different. For a uniaxial crystal, to which we shall limit ourselves for simplicity, with $\alpha_{iklm} = 0$:

$$\hat{n}_1^2 = \varepsilon_\perp, \quad 1/\hat{n}_2^2 = \sin^2\theta/\varepsilon_\parallel + \cos^2\theta/\varepsilon_\perp, \tag{3.2}$$

where

$$\sin\theta = s_z, \quad \cos\theta = s_x, \quad s_y = 0, \quad \varepsilon_\parallel = \varepsilon_z, \quad \varepsilon_\perp = \varepsilon_x = \varepsilon_y.$$

Suppose that

$$\theta \neq 0, \quad \theta \neq \pi/2, \quad \varepsilon_\parallel \neq \varepsilon_\perp. \tag{3.3}$$

In this case the refractive index for the extraordinary wave $n_2$ does not approach infinity as either $\epsilon_\parallel$ or $\epsilon_\perp$ approach infinity. If, however, the values of $\epsilon_\parallel$ and $\epsilon_\perp$ are finite but of different sign, $\hat{n}_2^2 = \infty$ when

$$\varepsilon_\perp \sin^2\theta + \varepsilon_\parallel \cos^2\theta = 0. \tag{3.4}$$

If either $\epsilon_\parallel$ or $\epsilon_\perp$ is zero, $\hat{n}_2^2 = 0$. The cases $\theta = 0$ and $\theta = \pi/2$ are degenerate; wave 2 becomes transverse and there is a longitudinal wave whose frequency, for $\alpha = 0$, is determined by the conditions $\epsilon_\parallel(\omega) = 0$ or $\epsilon_\perp(\omega) = 0$. Another special case, which is not realizable, occurs if $\epsilon_\parallel = \epsilon_\perp = 0$ or $\epsilon_\parallel = \epsilon_\perp = \infty$ at the same frequency (in a biaxial crystal this refers to all three principal values $\epsilon_x$, $\epsilon_y$ and $\epsilon_z$). In essence, at this frequency the crystal degenerates into a cubic crystal, with all the features characteristic of the latter. In particular, only with $\epsilon_x = \epsilon_y = \epsilon_z$ for any direction will there be a wave (longitudinal) characterized by $\mathbf{D} = 0$. In the case indicated by (3.3) $\mathbf{D} \neq 0$, but if (3.4) is satisfied wave 2 becomes longitudinal. This is due to the fact that as $\hat{n}^2$ increases, for a given $\mathbf{D}$ the angle between $\mathbf{s}$ and $\mathbf{E}$ is reduced and when $n^2 \to \infty$, $\mathbf{s}\cdot\mathbf{E} \to 0$ [cf. Eq. (1.6)]. Hence, in non-cubic crystals, with $\alpha_{iklm} = 0$ in general there is no new wave analogous to the longitudinal wave 3 in an isotropic medium. The limiting transiton to a cubic crystal is not trivial. If $\epsilon_\parallel \to 0$ and $\epsilon_\perp > 0$ the tensor ellipsoid

$$(x^2 + y^2)/\varepsilon_\perp + z^2/\varepsilon_\parallel = 1$$

degenerates into a plane disc; if $\epsilon_\perp \to 0$ and $\epsilon_\parallel > 0$, it becomes a line. In other words, as one of the principal values $\epsilon_i$ approaches zero the crystal becomes anisotropic in the limit even if the other principal values are small. Different signs for $\epsilon_\parallel$ and $\epsilon_\perp$ means that we do not have a tensor ellipsoid but a tensor hyperboloid, with (3.4) as the asymptote. As one of the principle values approaches zero in the limit the anisotropy is also large in this case. If $\epsilon_x = \epsilon_y = \epsilon_z = 0$ the tensor ellipsoid or hyperboloid degenerates to a point.

In the case given by (3.3), which we also consider, if spatial dispersion is taken into account and the condition in (3.4) is not satisfied there is a non-real third wave with a very high value of $\hat{n}_3^2$. Only in the region of angles and frequencies for which the quantity $\epsilon_\perp \sin^2\theta + \epsilon_\parallel \cos^2\theta$ is very small does spatial dispersion lead to an interesting result.* If the condition in (3.4) is observed the values of $\epsilon_\parallel$ and $\epsilon_\perp$ are different from zero and finite. In this case the expansions in (1.3) and (1.4) are equivalent. In the general case the expressions which are obtained are extremely complicated. In order to illustrate the qualitative aspects of the problem we make a further far-reaching assumption: namely,

$$\hat{\varepsilon}_\parallel = \varepsilon_\parallel - \alpha\hat{n}^2, \quad \hat{\varepsilon}_\perp = \varepsilon_\perp - \alpha\hat{n}^2. \tag{3.5}$$

The equation for $\hat{n}^2$ assumes the form:

*The fact that in general the third wave in an anisotropic medium has a real value only in the region where $n_2 \to \infty$ was noted by the author earlier in the case of a magneto-active plasma (cf. reference 7, § 75).

$$(\varepsilon_\perp - \hat{n}^2)\{\alpha\hat{n}^4 - [\varepsilon_\perp \sin^2\theta + \varepsilon_\parallel \cos^2\theta + (\varepsilon_\perp + \varepsilon_\parallel)\alpha]\hat{n}^2 + \varepsilon_\parallel\varepsilon_\perp\} = 0, \quad (3.6)$$

where small terms have been neglected.

With $\alpha = 0$ Eq. (3.6) becomes Eq. (1.7) with $\epsilon_x = \epsilon_y = \epsilon_\perp$ and $\epsilon_z = \epsilon_\parallel$ and its solution is given by (3.2). When

$$\hat{n}_1^2 = \varepsilon_\perp, \quad \hat{n}_{2,3}^2 = \mu/2\alpha \pm \sqrt{(\mu/2\alpha)^2 - \varepsilon_\perp\varepsilon_\parallel/\alpha}, \quad \mu = \varepsilon_\perp \sin^2\theta + \varepsilon_\parallel \cos^2\theta + (\varepsilon_\parallel + \varepsilon_\perp)\alpha. \quad (3.7)$$

The dependence of $\hat{n}_{2,3}^2$ on angle $\theta$ or frequency $\omega$ close to the point $\mu = 0$ is completely analogous to the dependence on $\omega$ in (2.5) close to the point $\epsilon = \infty$ (cf. Figs. 1 and 2). With $\mu^2 \gg |4\alpha\epsilon_\perp\epsilon_\parallel|$, it is apparent that $\hat{n}_2^2$ is determined by (3.2) and $\hat{n}_3^2 \approx \mu/\alpha$. When $\theta \to 0$ and $\epsilon_\perp \to 0$: $\hat{n}_2^2 = \epsilon_\perp$, $\hat{n}_3^2 = \epsilon_\parallel$ as is to be expected. When $\theta \to \pi/2$ and $\epsilon_\perp \to 0$, similarly $\hat{n}_2^2 = \epsilon_\parallel$ and $n_3^2 = \epsilon_\perp/\alpha$. Moreover, when $\theta = 0$ and $\theta = \pi/2$, $\hat{n}_2^2 \to \infty$ correspondingly for $\epsilon_\perp \to \infty$ and $\epsilon_\parallel \to \infty$ ($\hat{n}_1^2 \to \infty$ for $\epsilon_\perp \to \infty$ at all angles for wave 1). In these cases spatial dispersion is taken into account through the use of an expansion such as that given in (2.4).

As in an isotropic medium and cubic crystals in non-cubic crystals, spatial dispersion is considerably stronger when the medium is gyrotropic (cf. Sec. 2). However, we shall not stop here to consider this problem nor a number of other problems which require special investigation (the behavior of the rays and the group velocity, gyrotropic effects under the influence of an external magnetic field, magnetic media such as ferrites, biaxial crystals, conical refraction, axial dispersion, dispersion relations, artificial anisotropy induced by electric fields, in which case it is possible to realize the limiting transition to isotropy, a more detailed analysis of absorption, etc.).

We have considered above only waves in an unbounded medium. Any observation of these effects would always require an examination of the boundary situation (for example, a first step would be to solve the problem of transmission of light through a plane slab). Thus boundary conditions must be considered. The ordinary conditions

$$D_{2n} = D_{1n}, \; H_{2n} = H_{1n}, \; E_{2t} = E_{1t}, \; H_{1t} = H_{2t},$$

are valid but not adequate if one is to take account of the new waves. Within the framework of the phenomenological approach employed here, additional boundary conditions can be obtained only by introducing certain assumptions.

For example, we may consider the boundary between a medium and vacuum and write the vector **D** in the form

$$D_i = \varepsilon_{ik}E_k + D_i' = E_i + (\varepsilon_{ik} - \delta_{ik})E_k + D_i' = E_i + D_i''. \quad (3.8)$$

In vacuum $\mathbf{D}'' = 0$ and if the boundary is not sharp $\mathbf{D}''$ approaches zero smoothly in the transition layer. On this basis, one would write at a sharp boundary:

$$\mathbf{D}'' = \mathbf{D} - \mathbf{E} = 4\pi\mathbf{P} = 0. \quad (3.9)$$

On the other hand, if spatial dispersion is neglected completely, and $\mathbf{D}' = 0$, (3.9) is no longer correct (in this case, from the condition $D_{2n} = D_{1n} = E_{1n}$ it is clear that only $P_n = 0$). Hence, a more reasonable boundary condition is

$$\mathbf{D}' = 0. \quad (3.10)$$

In his paper Pekar[2] used a boundary condition which was an "average" of (3.9) and (3.10) while the vector **P** was specifically set equal to zero except for an indefinite part of local origin. Since the new waves generally appear only under very special circumstances, further refinement of the analysis of boundary conditions will probably result from the solution of specific problems.

One might also be interested in finding the electromagnetic field and radiated energy for sources located in a medium with spatial dispersion. In the isotropic case this problem can be solved by an extension of classical methods; in crystals, however, it is more feasible to use the Hamiltonian method. This approach was used in reference 8, in which spatial dispersion was not taken into account.

## 4. REMARKS ON COLLECTIVE ENERGY LOSSES AND THE CERENKOV EFFECT

The discrete energy losses of fast electrons or other charged particles which move in a medium are related to the excitation of longitudinal waves (plasmons).[6]

If spatial dispersion is not taken into account, the longitudinal oscillations take place at frequencies $\omega_l$ such that $\epsilon(\omega_l) = 0$ and the corresponding energy losses are associated with the so-called polarization or Bohr energy loss in the medium (cf. reference 9). In the presence of spatial dispersion the polarization loss becomes the Cerenkov effect and is associated with the emission of a longitudinal wave 3 for which $n_3^2 = \epsilon(\omega)/\alpha$ [cf. Eq. (2.2)], assuming an isotropic medium. In this case it is apparent that the following condition must be satisfied:

$$\cos\theta = c/vn(\omega), \quad (4.1)$$

where $\theta$ is the angle between the normal **s** and the particle velocity **v**, $n = n_3$ and the variation in particle energy is neglected (this last condition assumes that $v \gg v_0$ – the velocity at the Fermi limit).

Since $\alpha \sim (v_0/c)^2 \sim 10^{-5}$, even with $\Delta\omega/\omega_l = (\omega - \omega_l)/\omega_l \sim 0.03$ to $0.1$ a macroscopic analysis becomes unfeasible because $n_3 \sim 10^2$, $\epsilon \sim 0.1$ and $\lambda = \lambda_0/n_3 \sim 10^{-7}$. As a result the intensity of the Cerenkov radiation falls off sharply and the absorption increases. Hence the radiated longitudinal waves (plasmons) are characterized by a relatively narrow spectrum with $\Delta\omega \leq 0.1\,\omega_l$. Transverse Cerenkov radiation occurs if the condition

$$\varepsilon = n_{1,2}^2 > (c/v)^2 \tag{4.2}$$

is satisfied but usually the radiation is characterized by a wide spectrum, i.e., non-discrete losses. An exception might be a medium with weak absorption in which the condition in (4.2) would be satisfied only over a relatively narrow frequency range, for example close to an absorption line.

In cubic crystals, in the first approximation the discrete (collective) losses are of the same nature as those in an isotropic medium. The only distinction is due to the spatial dispersion which leads to the weak optical anisotropy for cubic crystals (cf. Sec. 3). This anisotropy is especially marked in metals because for a cubic structure the Fermi surface is not a sphere and the corresponding velocity $v_0$ depends on direction. Inasmuch as the width of the longitudinal Cerenkov radiation in an isotropic medium is not small in absolute magnitude the weak anisotropy which is indicated in the discrete losses is not of great importance.*

The situation in non-cubic crystals is different because in the general case there is no specific individual branch for the longitudinal waves. Even if spatial dispersion is neglected, there are no polarization losses. Hence, all the collective losses are to be associated with the Cerenkov effect (cf. also reference 9). On the other hand, in non-cubic crystals even when $\epsilon_i < 1$ there are frequencies for which the refractive index becomes rather large. This is the case particularly in a region which corresponds to the condition in (3.4) or, for an arbitrary crystal, to the condition

$$\varepsilon_x(\omega_{ls})\, s_x^2 + \varepsilon_y(\omega_{ls})\, s_y^2 + \varepsilon_z(\omega_{ls})\, s_z^2 = 0. \tag{4.3}$$

Under these conditions, as has already been pointed out, wave 1 or 2 is longitudinal and it is important to take account of spatial dispersion. In solids, particularly in metals, at frequencies of the order of $10^{16}$ (energies of approximately 10 ev) the Cerenkov condition (4.1) may be satisfied only for $\omega = \omega_{ls}$ [cf. Eq. (4.3)]. As a result the collective losses are again of discrete nature although, in addition to spatial dispersion there is one other effect which tends to broaden the spectrum. This effect is a result of the fact that the frequency $\omega_{ls}$, as is clear from Eq. (4.3), depends on the direction of wave propagation in the crystal. If the vector associated with the normal $\mathbf{s}$ approaches the direction of the principal axis i Eq. (4.3) assumes the form $\epsilon_i(\omega_{li}) = 0$, which is the same as the existence condition for longitudinal waves along the i axis. The additional broadening of the discrete line is then due simply to the fact that $\omega_{lx} \neq \omega_{ly} \neq \omega_{lz}$, or, in a uniaxial crystal the fact that $\omega_{l\perp} \neq \omega_{l\parallel}$.

In conclusion we may note that the use of the Cerenkov effect would seem to offer the best approach to the problem of exciting "spatial dispersion" waves although problems such as those mentioned at the end of Sec. 3 would arise.

*In this connection the anisotropy in the optical properties of cubic crystals, which results from spatial dispersion, may be important in analyzing the interaction of plasmons with an external transverse electromagnetic field and may lead, for example, to the excitation of quasi-longitudinal plasmons when light is incident on the crystal boundary. It is likely that taking account of spatial dispersion in the macroscopic behavior is, in some degree, equivalent to the quasi-microscopic analysis of plasmons as waves in a degenerate electron gas of inhomogeneous density.[10]

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311