

CRYSTAL OPTICS WITH ALLOWANCE FOR SPATIAL DISPERSION; EXCITON THEORY. I

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Usp. Fiz. Nauk 76, 643-682 (April, 1962)

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INTRODUCTION

CLASSICAL crystal optics has developed along the following lines. The crystal is characterized by a complex dielectric tensor  $\epsilon_{ij}(\omega)$  which depends on the radiation frequency  $\omega$ . Knowing the tensor  $\epsilon_{ij}(\omega)$ , we can determine from the equations of macroscopic electrodynamics the "normal" electromagnetic waves in the crystal, i.e., waves of the form

$$E = E_0 e^{i(kr - \omega t)}, \quad E_0 = \text{const}, \quad \mathbf{k} = \frac{\omega}{c} \hat{n} \mathbf{s}, \quad s = \frac{k}{k}$$

(we confine ourselves for simplicity to homogeneous plane waves in which the equal-phase planes and equal-amplitude planes coincide). Different normal waves (designated by the index  $l$ ) have different polarizations (the vector  $\mathbf{E}_{0l}$ ) and different values of the complex refractive index  $\hat{n}_l(\omega, \mathbf{s}) = n_l + i\kappa_l$ .

When no boundaries are involved, the tensor  $\epsilon_{ij}(\omega)$  is useful for it replaces the much more complicated quantity  $n_l(\omega, \mathbf{s})$ , which is a function of both  $\omega$  and  $\mathbf{s}$ . Considerations of a general type readily demonstrate the symmetry of the tensor  $\epsilon_{ij}(\omega) = \epsilon'_{ij}(\omega) + i\epsilon''_{ij}(\omega)$  and relate its real and imaginary parts  $\epsilon'_{ij}$  and  $\epsilon''_{ij}$  (dispersion relations). To determine the frequency dependence of  $\epsilon_{ij}$ , we must, however, resort to macroscopic theory, involving the use of a definite model or of various approximations. If a simple model is used, particularly if the symmetry is high, we can just as readily calculate  $\hat{n}_l(\omega, \mathbf{s})$  directly, rather than

$\epsilon_{ij}(\omega)$ . However, even in cubic crystals, when in the final analysis we deal with only one function  $\hat{n}(\omega) = \sqrt{\epsilon(\omega)}$  [here  $\epsilon_{ij}(\omega) = \epsilon(\omega)\delta_{ij}$ ],  $\epsilon(\omega)$  is the simpler of the two quantities; it relates the current induced by the field and the field itself at a single point, and no retarded interaction need be taken into account in its calculation [at the same time, the index  $\hat{n}(\omega)$  is related in obvious fashion with the wave propagation; in addition, a cubic crystal supports three normal waves, counting the longitudinal one, although the degeneracy  $n_1 = n_2 = n$  applies to two of the waves].

Thus, there is no doubt that the main problem of the microscopic theory is in general the determination of the tensor  $\epsilon_{ij}(\omega)$ .

It follows from the symmetry of the kinetic coefficients that in the absence of a macroscopic magnetic field the tensor  $\epsilon_{ij}(\omega)$  is symmetric (see, for example, [1]). Natural optical activity (gyrotropy) is, therefore, beyond the scope of classical crystal optics (in the sense of our definition of the term). As is well known, an analysis of natural optical activity entails the account of some small terms of order  $a/\lambda$ , where  $a$  is the characteristic dimension (lattice constant, dimension of the molecule) and  $\lambda = \lambda_0/n = 2\pi/k$  is the wavelength of light in the medium ( $\lambda_0 = 2\pi c/\omega$  - wavelength in vacuum). Physically this reduces to a situation wherein the current induced by the specified field depends not only on the frequency  $\omega$  but also on the wave vector  $\mathbf{k}$ , i.e., the current varies with the wavelength of the field. Such a dependence is cus-

tomarily called spatial dispersion (frequency dispersion is the dependence of the current on the field frequency  $\omega$ ). In a homogeneous unbounded medium (plasma, liquid), and to good approximation in crystal optics (see Sec. 2a) allowance for spatial dispersion reduces to the use in the field equation of a tensor\*

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon'_{ij}(\omega, \mathbf{k}) + \epsilon''_{ij}(\omega, \mathbf{k}), \quad (1)$$

which depends on  $\omega$  and  $\mathbf{k}$  and satisfies, of course, the condition  $\epsilon_{ij}(\omega, 0) = \epsilon_{ij}(\omega)$ , where  $\epsilon_{ij}(\omega)$  is the tensor considered in classical crystal optics.

We shall call the crystal-optics theory based on the use of the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  crystal optics with allowance for spatial dispersion. Although such a crystal optics is evidently broader than the classical theory, it is in turn confined to a region of not too short waves  $\lambda_0 = 2\pi c/\omega$ , so that the parameter  $a/\lambda_0$  is small. The lattice constant is  $a \sim 10^{-8} - 10^{-7}$  cm, in the optical region we have  $\lambda_0 \gtrsim 10^{-5}$  cm, and consequently  $a/\lambda_0 < 10^{-2} - 10^{-3}$ . On the other hand, in optics usually, even near the absorption lines,  $n \lesssim 10$ , so that the following inequality is also satisfied

$$\frac{a}{\lambda} = \frac{an}{\lambda_0} \ll 1. \quad (2)$$

Condition (2) denotes that the spatial dispersion is weak and this is precisely the assumption (which in the optical region corresponds to all known cases) that we shall use below.

As already stated, terms of order  $a/\lambda$  must be taken into account in the analysis of gyrotropy. Centrosymmetric crystals, as well as a few others, have no terms proportional to  $(a/\lambda)$ , and the terms of order of  $(a/\lambda)^2$  are usually quite small. Nonetheless, allowance for the terms proportional to  $(a/\lambda)^2$  is in some cases essential even for a qualitative understanding of the phenomena. Thus, the propagation of longitudinal waves in any medium, and particularly in crystals is completely determined by spatial dispersion (disregard of spatial dispersion yields zero group velocity for the longitudinal waves; see, for example, [2] and below). Further, as long ago as in 1878 Lorentz [3] pointed out that cubic crystals become anisotropic if terms proportional to  $(a/\lambda)^2$  are taken into account (see also [4,5]). Optical anisotropy of cubic crystals has recently been experimentally established. [6] Finally, inclusion of the terms proportional to  $a/\lambda$  or, in the absence of gyrotropy, of terms proportional to  $(a/\lambda)^2$ , may prove necessary in the region of anomalous dispersion, i.e., near the absorption lines, where the refractive index, and consequently the parameter  $a/\lambda = an/\lambda_0$ , increases. Therefore, as is well demonstrated by the example of a magnetoactive plasma, [2] spatial dispersion in the absorption-line region (near the natural frequency) can appreciably change the

course of the dispersion curves, and in particular, lead to the appearance of new waves (new values of  $n_l$  at a given frequency  $\omega$ ). The appearance of new waves is also possible in principle, for example, near the lines of the so-called exciton absorption of light in non-gyrotropic crystals [7] [this effect was regarded in [7] as some new phenomenon; in this connection it was shown in [5] that the situation involves a particular case of allowance for spatial dispersion, and that in non-gyrotropic crystals it is sufficient to take into account terms proportional to  $(a/\lambda)^2$ ]. The observation of new waves in non-gyrotropic crystals is complicated considerably by the influence of absorption. In the case of gyrotropic crystals, a new wave should appear near the absorption line with properties characterized by the parameter  $a/\lambda$ , making the conditions for its observation more favorable. [5]

Thus, there are many problems, the analysis of which lies in the field of crystal optics with account of spatial dispersion. The task of the theory is first to establish a connection between  $\hat{n}_l(\omega, \mathbf{s})$  and  $\epsilon_{ij}(\omega, \mathbf{k})$  and to use the corresponding formulas to reduce the experimental data. The experiments yield the complex refractive index  $\hat{n}_l(\omega, \mathbf{s}) = n_l + ik_l$ , and failure to use crystal optics would necessitate measurements for a very large number of directions of  $\mathbf{s}$ . On the other hand, if  $\hat{n}_l(\omega, \mathbf{s})$  is expressed in terms of  $\epsilon_{ij}(\omega, \mathbf{k})$ , it is sufficient to measure  $n_l$  for several directions only. If spatial dispersion is neglected, this is evident at once, since the symmetrical complex tensor  $\epsilon_{ij}(\omega)$  is characterized at a given frequency by at most six numbers (recognizing that  $\epsilon'_{ij}$  and  $\epsilon''_{ij}$  have already been reduced to the principal axes). Account of weak spatial dispersion complicates the situation, but the measurements need be made only in a few directions, by virtue of the simple dependence of  $\epsilon_{ij}(\omega, \mathbf{k})$  on  $\mathbf{k}$ .

If  $\epsilon_{ij}(\omega, \mathbf{k})$  is known in some approximation, all the normal waves in the crystal corresponding to this approximation can be regarded as known [in particular, the dispersion law  $\omega_l = \omega_l(\mathbf{k})$  is equivalent to specifying the functions  $\hat{n}_l(\omega, \mathbf{s}) = ck/\omega_l(\mathbf{k})$ ]. In addition, the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  determines the energy lost when the particles move in the medium, the molecular forces between the bodies, the fluctuations of the electromagnetic field, and provides in general a fairly complete description of the medium (crystal). [1,8-10]

The principal purpose of the present article is to present the fundamentals of crystal optics with allowance for spatial dispersion (we shall make considerable use of [5,8]). We also consider it advisable to dwell here on the connection between crystal optics and exciton theory and on certain related problems.

Since there is no universally accepted terminology, we shall define excitons as "elementary excitations" in crystals, obeying Bose statistics. This definition obviously classifies as excitons all normal electro-

\*In the present article we refer only to the linear theory (the superposition principle holds) and disregard the Raman effect. Concerning the latter see, for example, [9].

magnetic waves in a crystal, which in quantum mechanical language are none other than "photons in the medium" (including longitudinal photons in the medium—plasmons).<sup>\*</sup> By the same token, the general exciton theory embraces, on the one hand, crystal optics with allowance for spatial dispersion, and on the other all the theoretical constructions aimed at calculating the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$ .

The transition from problems in terminology to the essence of the matter occurs when one begins to discuss methods for the calculation of  $\epsilon_{ij}(\omega, \mathbf{k})$  and the character of various possible approximations. The latter are dictated primarily by the type of crystal and by the nature of the excitations studied. Thus, in ionic crystals in the infrared region the optical modes of lattice vibration are particularly important.<sup>[12]</sup> However, in the same ionic crystals but at higher frequencies, and especially in molecular crystals and some semiconductors, the electronic excitations play the principal role.<sup>[13]</sup> These excitations can be visualized as an excited state of a molecule proceeding from site to site (molecular crystal), or as a moving bound electron-hole pair (semiconductors). At the same time, by virtue of the translational symmetry of the crystal, the eigenfunctions corresponding to the excitations encompass the entire crystal and have the character of modulated plane waves with wave vector  $\mathbf{k}$ .<sup>†</sup> If we confine ourselves for simplicity to an ideal immobile lattice, then the wave function of the excitation can be written in the form<sup>[13]</sup>

$$\Psi_{\mathbf{k}, l} = e^{i\mathbf{k}\mathbf{R}} U_{\mathbf{k}, l}(\mathbf{R}, \mathbf{r}_i - \mathbf{r}_j), \quad (3)$$

where  $\mathbf{R} = \sum_j \mathbf{r}_j / NV$  is the radius vector of the centroid of all the  $NV$  electrons (with radius vectors  $\mathbf{r}_j$ ), the function  $U_{\mathbf{k}, l}$  is periodic (with the period of the lattice) with respect to  $\mathbf{R}$ , and the index  $l$  corresponds to quantum numbers that do not reduce to  $\mathbf{k}$ . Confining ourselves in (3) to an account of the particle coordinates only, we imply by the same token a mechanical problem, with an analysis of the Coulomb interaction only. This immediately raises the question of relating the "mechanical" excitations (excitons) thus obtained to the real excitons, and of their role from the point of view of calculating  $\epsilon_{ij}(\omega, \mathbf{k})$ .

Before touching upon this problem, let us make more precise the concept of the "mechanical exciton." It is not rational to define as a "mechanical exciton" any solution of the Coulomb problem. In fact, among such

<sup>\*</sup>Concerning the use of the concept of photons in a medium (with energy  $\hbar\omega$  and momentum  $\hbar\omega n/c$ ) as applied to other problems in radiation theory, see<sup>[11]</sup>. We note that the definition given in the text for excitons includes also acoustic waves, for which, however, it is advantageous to retain the universal term "phonon."

<sup>†</sup>This is why a localized electron-hole pair or an excited molecule (which can be considered as a pair with small radius) is described in terms of a wave packet. In some case, however, the consideration of packets is fully justified and can even serve for quantitative calculations.

excitations there are included such real excitons as longitudinal normal waves (plasmons), in which the electric field is potential and there are no magnetic fields. In addition, the solutions of the Coulomb problem include waves which we call "fictitious" longitudinal waves (see Sec. 1b). In such waves (with small values of  $\mathbf{k}$ ), as in longitudinal waves, there exists a non-vanishing long-wave potential electric field  $\mathbf{E} = \mathbf{E}_{||}$  (the field  $\mathbf{E}_{||}$  is non-vanishing whenever  $\text{div } \mathbf{P} \neq 0$ , where  $\mathbf{P}$  is the electric polarization<sup>\*</sup>).

In addition to having polarization of longitudinal character ( $\text{curl } \mathbf{E}_{||} = 0$ ), the macroscopic Coulomb field  $\mathbf{E}_{||}$  does not differ in any respect from an arbitrary macroscopic field (having, of course, the same values of  $\omega$  and  $\mathbf{k}$ ). In addition, the distinction between the longitudinal and transverse fields is in the general case of an anisotropic medium and an arbitrary wave-vector direction by no means natural, since in the corresponding normal waves the field  $\mathbf{E}$  is neither transverse nor longitudinal. Finally, if we deal with an account of the role of the long-wave field, this account (even if the normal waves do separate into longitudinal and transverse) is obtained in a unified manner for the total field by using the equations of electrodynamics. Consequently, we shall always take "mechanical excitons" to mean excitations obtained in the absence of or neglecting not only the long-wave transverse electromagnetic field, but also the potential macroscopic (long-wave) electric field (see, for example, <sup>[8]</sup>). From the point of view of solving the mechanical problem, this means that the potential macroscopic field  $\mathbf{E}_{||}$  (if it does not vanish) is discarded from the equations of motion and we are thus taking an approximate rather than complete account of the Coulomb interaction.<sup>†</sup> Actually, of course, in specific calculations this is the natural procedure, a fact clearly reflected in Sec. 44 of the book<sup>[12]</sup>. As regards terminology, the article<sup>[14]</sup> defines as "mechanical excitons" all the exact (and only exact) solutions of the Coulomb problem, and real excitons are called "optical excitons"; previously in article<sup>[14a]</sup>, the same real excitons were called "polaritons." As already mentioned, we are using a different terminology. A

<sup>\*</sup>We assume that there are no "free charges" and no absorption, and therefore  $\text{div } \mathbf{D} = \text{div } (\mathbf{E} + 4\pi\mathbf{P}) = 0$  (see Sec. 1a, where the quantity  $\mathbf{D}'$  is used with  $\mathbf{D} = \mathbf{D}'$  for a non-absorbing non-magnetic medium; in the present introduction we use for simplicity the vectors  $\mathbf{D}$  and  $\mathbf{P}$  rather than  $\mathbf{D}'$  and  $\mathbf{P}'$ ). In state (3) the polarization vector is

$$\mathbf{P} = \int \Psi_{\mathbf{k}l}^* \left( \frac{1}{V} \sum \mathbf{e} \mathbf{r}_i - \mathbf{P}_u \right) \Psi_{\mathbf{k}l} d\mathbf{r}_1 \dots d\mathbf{r}_{NV},$$

where  $\mathbf{P}_u$  is the ion polarization,  $N$  the electron concentration, and  $V$  the volume of the crystal.

<sup>†</sup>As is well known, the macroscopic field  $\mathbf{E}$  can be separated in a consistent manner (see, for example,<sup>[12]</sup> Secs. 30 and 44). Thus, neglect of the influence of the field  $\mathbf{E}$  on the eigenfunctions and natural frequencies can be carried out in a fully defined manner. We note also that by macroscopic field (in particular, the field  $\mathbf{E}$ ) we mean here any field whose Fourier expansion contains only long waves ( $\lambda \gg a$ ; for more details see Sec. 1a).

factor of importance to us is the fact that the "mechanical exciton" is the name used for an excitation considered without account of the long-wave field. As to the account of the short-wave (microscopic) field, the restriction to the Coulomb interaction only, as is implied in the use of (3), is of no significance in principle. Furthermore, it is more correct to assume that in the calculation of the energies and wave functions of the "mechanical excitons" account is taken of the entire short-wave interaction existing under given specific conditions (in addition to the Coulomb interaction with account of exchange, we obviously may also deal with magnetic interaction). Since in practice the discussion is nevertheless usually restricted to the Coulomb interaction, we shall likewise mention only this interaction; however, this is done solely for the sake of simplifying the discussion.

To explain the foregoing and for further exposition, we consider qualitatively a simple model. Namely, we discuss a system of NV anisotropic harmonic oscillators, located in the sites of a rhombic lattice (the two-fold axes coincide with  $x, y, z$ ). Each isolated oscillator has three different natural frequencies  $\omega_l = \omega_{x,y,z}$ , and the normal modes correspond to vibrations (variation of the electric dipole moment  $\mathbf{p}$ ) of the oscillator along the lattice axis  $x, y$ , or  $z$ . In the limit of sufficiently large lattice constants  $a_x, a_y$ , and  $a_z$  or in the case of small "oscillator strengths" (i.e., sufficiently weak interaction between oscillators), the normal frequencies of the system can be regarded as NV-fold degenerate and also equal to  $\omega_{x,y,z}$ . As the oscillators come closer together, the frequencies split and the normal oscillations have the form  $\mathbf{p}_{l,i} = \mathbf{p}_{l,i,0} \exp i[\mathbf{k} \cdot \mathbf{r}_i - \omega_l(\mathbf{k})t]$  where  $\mathbf{p}_{l,i}$  is the dipole moment of the  $i$ -th oscillator located at the point  $\mathbf{r}_i$ . If we consider only Coulomb interaction (specifically, dipole-dipole interaction) between oscillators, then the long-wave normal oscillations separate quite distinctly into transverse polarization oscillations ( $\mathbf{p}_{l,i} \cdot \mathbf{k} = 0$ ), longitudinal oscillations ( $\mathbf{p}_{l,i} \times \mathbf{k} = 0$ ), and all others. In the case of transverse polarization oscillations, there is no longitudinal electric field  $\mathbf{E} = \mathbf{E}_{||}$ , since  $\text{div } \mathbf{P} = \text{div} \left( \frac{1}{V} \sum \mathbf{p}_i \right) = 0$ ,  $\text{div } \mathbf{D} = \text{div} (\mathbf{E}_{||} + 4\pi\mathbf{P}) = 0$ ,  $\text{curl } \mathbf{E}_{||} = 0$ , and thus  $(\mathbf{k} \cdot \mathbf{E}_{||}) = 0$  and  $\mathbf{k} \times \mathbf{E}_{||} = 0$ . From symmetry considerations it is clear that transverse normal oscillations are possible if the moments  $\mathbf{p}_{l,i}$  and consequently the electric polarization  $\mathbf{P}$  are directed along one of the axes  $x, y$ , or  $z$ , whereas the wave vector  $\mathbf{k}$  lies in the corresponding coordinates plane (for example,  $P_x \neq 0, P_y = P_z = 0, k_x = 0$ ). In a rhombic lattice there are obviously three "zones" of such waves with frequencies  $\omega_x (k_x = 0, k_y, k_z)$ ,  $\omega_y (k_x, k_y = 0, k_z)$  and  $\omega_z (k_x, k_y, k_z = 0)$ . In section 1b these waves ("polarization waves") will be analyzed macroscopically. Since the field in "polarization waves" is  $\mathbf{E}_{||} = 0$ , these waves

should be classified as "mechanical excitons" and are obtained at the same time in the exact solution of the Coulomb problem.

Waves traveling along one of the axes and polarized along the same axis (for example,  $P_x \neq 0, P_y = P_z = 0, k_y = k_z = 0$ ), are longitudinal waves in which  $\mathbf{E} = \mathbf{E}_{||} \neq 0$  and  $\mathbf{D} = 0$ . Finally, waves can exist in which  $\mathbf{D} \neq 0$  and  $\mathbf{E} = \mathbf{E}_{||} \neq 0$ . In these waves, as in all others, the vector  $\mathbf{D} = \mathbf{E} + 4\pi\mathbf{P}$  is transverse ( $\mathbf{k} \cdot \mathbf{D} = 0$  by virtue of  $\text{div } \mathbf{D} = 0$ ). But the vector  $\mathbf{E} = \mathbf{E}_{||}$  must be longitudinal (a strictly Coulomb problem is considered). Therefore the vector  $\mathbf{P}$  in these waves ("fictitious" longitudinal waves; see Sec. 1b) is neither longitudinal nor transverse. Only such normal waves can propagate in this model in an arbitrary direction ( $k_x \neq 0, k_y \neq 0, k_z \neq 0$ ).<sup>\*</sup> The longitudinal and "fictitious" longitudinal waves, regarded as exact solutions of the Coulomb problem, are not "mechanical excitons" in our terminology. To each of these waves, however, there corresponds a "mechanical exciton" which is a normal oscillation with characteristics that are obtained when the effect of the field  $\mathbf{E}_{||}$  is neglected. Using as an example a longitudinal wave propagating along the  $x$  axis, we shall explain this statement. The equation of motion for the oscillator can then be written  $\ddot{x}_i + \omega_{xi}^2 x_i g_{xi} = e f^{1/2} E_{||} / m$ , where  $p_i = f^{1/2} x_i$  ( $f$ —"oscillator strength") and  $g_i$ —force exerted by all other oscillators, after subtracting the long-range part (the latter is precisely included in the field  $\mathbf{E}_{||}$ ).<sup>†</sup> In a longitudinal wave we have

$$\mathbf{D} = \mathbf{E}_{||} + 4\pi\mathbf{P} = 0, \quad P = \frac{e f^{1/2} \sum x_i}{V} \approx e f^{1/2} N x_i$$

(we consider long waves) and  $\mathbf{E}_{||} = -4\pi e f^{1/2} N x_i$ . Thus, an account of the field  $\mathbf{E}_{||}$  changes the square of the natural frequency by  $\omega_0^2 = 4\pi e^2 f N / m$ , and this change can be quite large and exceed the splitting due to the force  $g_i$ .<sup>‡</sup> In our example, obviously, the "mechanical

<sup>\*</sup>The foregoing statements follow without further detailed analysis from the macroscopic consideration (see Sec. 1b), and also become clear if one bears in mind the limiting case of uncoupled (non-interacting) oscillators (when the coupling is weak, these properties of the model remain patently unchanged).

<sup>†</sup>The force  $g_i$  contains also a long-range term (the "polarization correction") connected with the difference between the macroscopic field  $\mathbf{E}$  and the "effective field"  $\mathbf{F}$ . For a cubic lattice made up of point-like dipoles, as is well known,  $\mathbf{F} - \mathbf{E} = 4\pi\mathbf{P}/3$  (as  $\mathbf{k} \rightarrow 0$ ). Consequently in a cubic lattice polarized along the  $x$  axis, the force  $g_i$  contains a term  $(4\pi e f^{1/2}/3)\mathbf{P} \approx (4\pi e^2 f N/3m)x_i$ . This term can be accounted for by suitably changing the frequency  $\omega_x$ ; the same pertains to crystals with lower symmetry, if we disregard the replacement of the numerical coefficient  $4\pi/3$  by some other coefficient. In connection with the foregoing and with the qualitative character of this discussion, we shall not include explicitly the difference between the effective field and the macroscopic field, assuming the frequencies  $\omega_{x,y,z}$  to be suitably modified, and we shall regard  $g_i$  as accounting only for the short-range interaction (the correct classical calculation for the cubic lattice is found in<sup>[15]</sup>; see also Sec. 4b).

<sup>‡</sup>See the foregoing footnote.

ical exciton" is a longitudinal wave with frequency  $\omega_x + \delta\omega_x$ , where  $\delta\omega_x$  is the frequency change due to the forces  $g_{xi}$ . The frequency of the real longitudinal exciton, on the other hand, is  $\omega_{||} = \omega_x + \delta\omega_x + \omega_0$ .

The foregoing remarks and the exposition that follows should leave no doubt of the advisability of distinguishing in crystal optics between real excitons and certain approximate solutions (images), called "mechanical excitons," the difference lying precisely in whether the effect of the long-wave electromagnetic field is accounted for or not.

What is the connection between real and "mechanical" excitons? If we bear in mind only the principal aspect of the problem (and not a specific quantitative comparison), the answer to this question is perfectly clear from both general considerations and from an examination of any simple model. The latter path is more illustrative and shorter, and consequently we return to the discussed model of anisotropic oscillators.

In the case of longitudinal waves the real exciton (i.e., the exact solution with account of the entire electromagnetic interaction) differs from the corresponding mechanical exciton in the inclusion of the long-wave field  $E_{||}$ . This leads, as was already explained, to a change in the natural frequencies  $\omega_{||}(k)$ . For transverse mechanical excitons ("polarization waves")  $P_{\perp} \neq 0$  and  $E = E_{||} = 0$ . The presence of alternating polarization  $P_{\perp}$  gives rise to a transverse electromagnetic field, which leads to a generally significant difference between the real transverse and mechanical excitons. Indeed, the equation of motion for the  $i$ -th oscillator, which makes its contribution, say, to the transverse normal oscillation along the  $y$  axis, can be written in the form  $\ddot{y}_i + \omega_y^2 y_i + g_{yi} = e f^{1/2} E_y / m$ . To clarify the qualitative picture in the long-wave region, we can put  $g_{yi} = 0$  (we assume the "polarization correction" to be accounted for by modifying the frequency  $\omega_y$ ) and  $P_y = e f^{1/2} N y_i$ . Then in the field  $E_y = E_{y0} \exp[i(k \cdot r - \omega t)]$  we have

$$P_y = \frac{e^2 f N E_y}{m(\omega_y^2 - \omega^2)} = \frac{\epsilon'_y - 1}{4\pi} E_y, \quad \epsilon_y = \epsilon'_y = 1 + \frac{4\pi e^2 f N}{m(\omega_y^2 - \omega^2)}, \quad \omega_y = \omega_y(k_x, k_y = 0, k_z). \quad (4)$$

For transverse waves the field equations lead in this case to the relation  $\epsilon'_y E_y = \hat{n}^2 E_y$ , i.e.,

$$\hat{n} = n + i\alpha = \sqrt{\epsilon_y} = \sqrt{1 + \frac{4\pi e^2 f N}{m(\omega_y^2 - \omega^2)}}. \quad (4')$$

Naturally, if the frequency  $\omega_y$  is real, then there is no absorption (as was indeed assumed). In the region  $\epsilon_y = \hat{n}^2 = -\kappa^2 < 0$  and neglecting spatial dispersion, the average energy flux in the wave is zero (when a wave strikes such a medium from the outside, total reflection occurs; see, for example, [2] and Sec. 1c below). In the transparency region ( $\kappa = 0$  and  $\hat{n}^2 = n^2 > 0$ ) we have

$$k = \frac{\omega}{c} n(\omega, s) = \frac{\omega}{c} \sqrt{1 + \frac{4\pi e^2 f N}{m(\omega_y^2 - \omega^2)}} \quad (5)$$

In the limit corresponding to classical crystal optics,  $\omega_y$  is independent of  $k$  and it is easy to express  $\omega$  explicitly in terms of  $k$ .

Thus, for a real exciton the dispersion equation  $\omega = \omega(k)$  has in this case the form (5), whereas for the mechanical exciton  $\omega = \omega_y(k)$ . In the simplest case, when the  $k$  dependence of  $\omega_y$  can be neglected (the primitive calculation\* above is sufficiently convincing without further analysis only under these conditions), the foregoing analysis is of course equivalent to the well-known elementary dispersion theory. We wanted to emphasize here, however, the close connection between exciton theory and problems well known long ago. The use of a different terminology or of unaccustomed images frequently makes this connection insufficiently obvious as evidenced by the contents of certain articles.

In classical crystal optics ( $\omega_{x,y,z} = \text{const}$ ) plots of the functions  $n = [\epsilon_{x,y,z}(\omega)]^{1/2}$  are usually employed. Such a plot for the case of (5) is shown in Fig. 1a. In exciton theory the relation  $\omega = \omega(k)$  is more frequently used, and the curve for our example is shown in Fig. 1b.

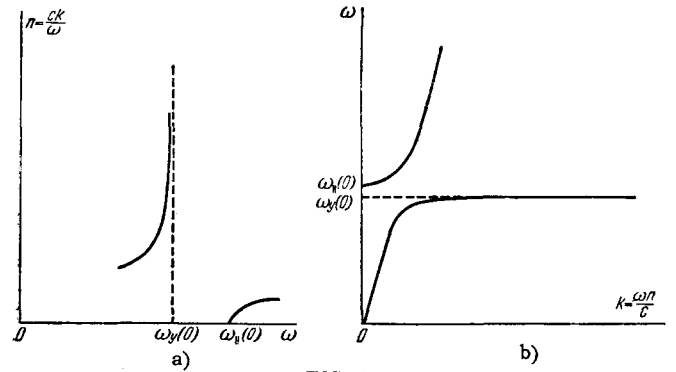


FIG. 1

We note that the frequency  $\omega_{||} = [\omega_y^2 + 4\pi e^2 f N / m]^{1/2}$  corresponds to the condition  $\epsilon_y(\omega) = 0$ ; this frequency  $\omega_{||}$  is precisely equal to the frequency of the longitudinal wave propagating along the  $y$  axis (see Sec. 1b). Near the frequency  $\omega = \omega_{||}$  we obviously have

$$\epsilon_y(\omega) = \left( \frac{d\epsilon_y}{d\omega} \right)_{\omega_{||}} (\omega - \omega_{||}) = n^2 = \frac{c^2 k^2}{\omega_{||}^2},$$

or

$$\omega = \omega_{||} + \frac{c^2}{\omega_{||}^2 (d\epsilon_y/d\omega)_{\omega_{||}}} k^2. \quad (6)$$

If we compare (6) with the relation  $W = \hbar\omega = \hbar\omega_0 + \hbar^2 k^2 / 2m_{\text{eff}}$ , we can speak<sup>[14]</sup> of an effective exciton mass  $m_{\text{eff}} = \hbar^2 \omega_{||}^2 (d\epsilon_y/d\omega)_{\omega_{||}} / 2c^2$  near the frequency  $\omega_0$ . The use of this term naturally adds nothing to the understanding of classical dispersion theory and will not be applied to real excitons.

If  $\omega_y = \omega_y(k)$  for the mechanical exciton, then  $\epsilon_y$  depends on  $k$ ; this corresponds to allowance for spa-

\*See Sec. 4b for rigorous calculations.

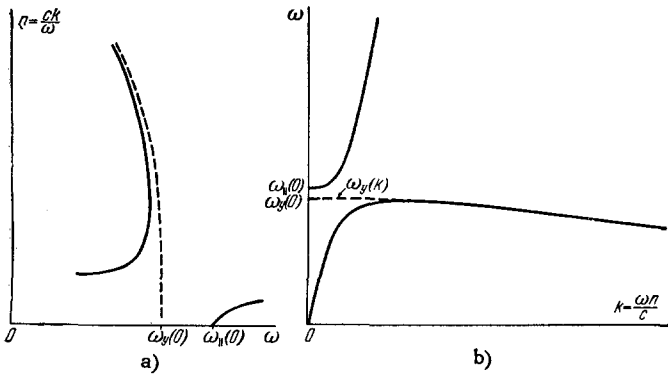


FIG. 2.

tial dispersion. The character of the curves  $n(\omega)$  and  $\omega(k)$  under similar conditions with  $k_x = k_y = 0$ ,  $k_z = k \neq 0$  (or with  $k_y = k_z = 0$ ,  $k_x = k \neq 0$ ) and with a certain definite dependence of  $\omega_y$  on  $k$  is clear from Fig. 2. We note that for optical vibrations of an ionic lattice the function  $\omega_y(k)$  corresponds in form in the simplest case precisely to the dashed curve of Fig. 2. A transition from mechanical vibrations (mechanical excitons) to real electromagnetic waves (excitons) for this case can also be easily traced, as was recently done.<sup>[12,16,17]</sup> The result is qualitatively the same as for the oscillator model, and is illustrated in Fig. 2. This is understandable, for dispersion theory is quite general in character and only an account of absorption can change the curves of Figs. 1 and 2 appreciably (provided, naturally, we speak of only a single resonant frequency and a single function  $\epsilon_{x,y,z}$ ; see Sec. 3b).

We now consider a "mechanical exciton" of a third type (i.e., neither longitudinal nor transverse relative to the polarization  $\mathbf{P}$ ). In our model such excitons include, first, all mechanical normal waves with  $k_x \neq 0$ ,  $k_y \neq 0$ ,  $k_z \neq 0$ . The vector  $\mathbf{P}$  has in this case a transverse component, inducing a transverse electromagnetic field. The corresponding real exciton is therefore neither longitudinal nor transverse (we refer to the field  $\mathbf{E}$ ), and its dispersion law  $\omega = \omega(\mathbf{k})$  differs from that of the mechanical exciton, generally speaking, even more than in the case of the transverse excitons. In the limit as  $k \rightarrow 0$  the picture can be readily explained using the formulas of classical crystal optics or by employing the same oscillator model. We shall return to this question later (see Sec. 1b).

From (5) or from Figs. 1 and 2 it is clear that at large values of  $k$  (or  $n$ ) but at frequencies  $\omega \sim \omega_{\parallel}(0) \sim \omega_y(0)$  we have approximately

$$\omega(\mathbf{k}) \approx \omega_y(\mathbf{k}), \quad k \rightarrow \infty, \quad \omega \lesssim \omega_{\parallel}(0). \quad (7)$$

In other words, with increasing  $k$  the properties of real transverse excitons approach those of the transverse mechanical excitons. This result is quite general and has a clear-cut physical meaning [Eq. (7) is the resonance condition in (5)]. As regards the frequencies of non-transverse (with respect to  $\mathbf{P}$ ) me-

chanical excitons, they coincide as  $k \rightarrow 0$  with the frequencies of transverse excitons having the same direction of oscillation, for when  $k = 0$  the polarization direction cannot play any role if the long-range long-wave field  $\mathbf{E}_{\parallel}$  is not included. Since in the region of interest to us the spatial dispersion is weak (the vector  $\mathbf{k}$  is small compared with  $1/a_{x,y,z}$ ), the difference between the frequencies of the mechanical excitons with given polarization  $\mathbf{P}$  but different  $\mathbf{k}$  will also be small. This is, however, no longer the case for the exact solutions of the Coulomb problem with  $\mathbf{E}_{\parallel} \neq 0$  (i.e., for the longitudinal and "fictitious" longitudinal waves); for example, the frequency of the longitudinal wave differs from the frequencies of the transverse waves by an amount on the order of  $\omega_0 = [4\pi e^2 fN/m]^{1/2}$ , and the "plasma" frequency  $\omega_0$  in a condensed medium is usually quite high.

Whereas the properties of real and mechanical excitons are the same only in the limiting cases [see, for example, (7)], the connection between them, of course, always exists. This is evident from the same expressions (4) and (5), which show that the dielectric constant and consequently all the properties of real excitons are determined by the frequencies of the mechanical excitons. On the other hand, if the general dispersion formula is used, it is clear that the energy (frequencies) and eigenfunctions of the mechanical excitons completely determine under certain conditions the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$ . We shall return to the last question in Sec. 4. We must emphasize at the same time that it is not the purpose of the present article to present either a detailed and complete exposition of the quantum mechanical theory of mechanical excitons or of specific calculations of  $\epsilon_{ij}(\omega, \mathbf{k})$ . The corresponding remarks made above and those that follow in Secs. 3 and 4, are essentially prompted by methodological considerations. For the same reason, the subject is developed in the article in most cases with sufficient detail, even when dealing with elementary problems. It seems to us that such an exposition is justified, for the tremendous literature on exciton theory shows much confusion and misunderstanding. We note, finally, that by developing in relatively detailed fashion certain aspects, the authors do not pretend at all to provide a complete explanation of the problems touched upon in exciton theory (the same pertains also to the list of cited references).

## 1. THE COMPLEX DIELECTRIC TENSOR $\epsilon_{ij}(\omega, \mathbf{k})$ AND NORMAL WAVES IN A MEDIUM

a) The tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  and its properties. The equations of the electromagnetic field will be written in the form\*

$$\begin{aligned} \text{rot } \mathbf{B} &= \frac{1}{c} \frac{\partial \mathbf{D}'}{\partial t}, \quad \text{div } \mathbf{D}' = 0, \\ \text{rot } \mathbf{E} &= -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}, \quad \text{div } \mathbf{B} = 0. \end{aligned} \quad (1.1)$$

\*rot = curl.

Here  $\mathbf{E}$  is the intensity of the electric field and  $\mathbf{B}$  the magnetic induction, occurring in the expression for the force  $\mathbf{F} = e(\mathbf{E} + \mathbf{v} \times \mathbf{B}/c)$  acting on a particle with charge  $e$  and velocity  $\mathbf{v}$ . The charge density  $\rho_0$  and the current density  $\mathbf{j}_0$  corresponding to the external source are assumed to vanish in (1.1) (otherwise  $\text{curl } \mathbf{B} = \frac{1}{c} \frac{\partial \mathbf{D}'}{\partial t} + \frac{4\pi}{c} \mathbf{j}_0$  and  $\text{div } \mathbf{D}' = 4\pi\rho_0$ ). The electric induction  $\mathbf{D}'$  used in (1.1) is defined by the relation

$$\frac{\partial \mathbf{D}'}{\partial t} = \frac{\partial \mathbf{E}}{\partial t} + 4\pi \mathbf{j}, \quad (1.2)$$

where  $\mathbf{j}$  is the density of the current induced in the medium.\*

To avoid misunderstanding, we emphasize that we shall not refer at all to averaging of fields over small regions. Such a procedure is unnecessary, nor is it feasible in a theory of material electrodynamics in which spatial dispersion is consistently taken into account.

In linear electrodynamics, the connection between  $\mathbf{D}'$  and  $\mathbf{E}$  can be written in the following general form

$$D'_i(\mathbf{r}, t) = \int_{-\infty}^t dt' \int d\mathbf{r}' \hat{\epsilon}_{ij}(t-t', \mathbf{r}, \mathbf{r}') E_j(\mathbf{r}', t'); \quad (1.3)$$

summation over repeated indices is implied henceforth.

The dependence of the kernel of this integral equation on the difference  $t-t'$  is due to the assumed homogeneity of the properties of the medium in time (in other words, the properties of the medium are assumed constant in time). On the other hand, if some factor (for example, an alternating external pressure) causes the properties of the medium to vary in time, then  $\hat{\epsilon}_{ij} = \epsilon_{ij}(t, t', \mathbf{r}, \mathbf{r}')$ . Further, the integration with respect to  $t'$  in (1.3) is only in the interval from  $-\infty$  to  $t$ , as dictated by the causality principle, viz: the induction  $\mathbf{D}'(\mathbf{r}, t)$  is determined only by past and present values of the field  $\mathbf{E}$ , i.e., for values  $t' \leq t$ . If the medium is also spatially homogeneous (all its points are equivalent), then

$$D'_i(\mathbf{r}, t) = \int_{-\infty}^t dt' \int d\mathbf{r}' \hat{\epsilon}_{ij}(t-t', \mathbf{r}-\mathbf{r}') E_j(\mathbf{r}', t'). \quad (1.4)$$

Let us take the Fourier transform of this formula, putting

$$E_j(\mathbf{r}, t) = \int E_j(\mathbf{k}, \omega) e^{i(\mathbf{k}\mathbf{r}-\omega t)} d\omega d\mathbf{k}$$

[we use the same symbols  $E_j$  for the transforms

\*By  $\mathbf{j}$  we denote here the current density in a system which is in a definite state, or after averaging with the aid of a statistical matrix (by the same token, fluctuation phenomena are disregarded; fluctuations are discussed in<sup>[10]</sup>). A traditional notation in field theory uses in place of  $\mathbf{D}'$  the expression  $\mathbf{j} + \partial\mathbf{P}/\partial t + \mathbf{c} \times \text{curl } \mathbf{M}$ , where  $\mathbf{j}$  is the conduction current density,  $\mathbf{P} = (\mathbf{D} - \mathbf{E})/4\pi$  is the electric polarization and  $\mathbf{M}$  is the magnetization (in other words, the quantity  $\mathbf{D}'$  which we use and the customarily employed induction  $\mathbf{D}$  coincide only for a non-absorbing non-magnetic medium).

$E_j(\mathbf{k}, \omega)$  and for the originals  $E_j(\mathbf{r}, t)$ , but this will not lead to confusion, since the arguments are indicated; similar notation is used for other quantities]. Incidentally, to obtain  $D_j(\mathbf{k}, \omega)$  there is not even need for the Fourier transformation, and it is sufficient to put in (1.4)  $\mathbf{E}(\mathbf{r}', t') = \mathbf{E}(\mathbf{k}, \omega) \exp[i(\mathbf{k} \cdot \mathbf{r}' - \omega t')]$ . As a result we get

$$D'_i(\mathbf{k}, \omega) = \epsilon_{ij}(\omega, \mathbf{k}) E_j(\mathbf{k}, \omega),$$

$$\epsilon_{ij}(\omega, \mathbf{k}) = \int_0^\infty d\tau \int d\mathbf{R} e^{-i(\mathbf{k}\mathbf{R}-\omega\tau)} \hat{\epsilon}_{ij}(\tau, \mathbf{R}), \quad (1.5)$$

and the component  $D_i(\mathbf{k}, \omega)$  is connected only with the components  $E_j(\mathbf{k}, \omega)$  having the same values of  $\omega$  and  $\mathbf{k}$ ; this holds by virtue of the temporal and spatial homogeneity of the medium, i.e., by virtue of the dependence of  $\epsilon_{ij}$  only on the differences  $\tau = t-t'$  and  $\mathbf{R} = \mathbf{r}-\mathbf{r}'$ .

$\epsilon_{ij}(\omega, \mathbf{k})$  is called the complex dielectric tensor.

The frequency dependence of  $\epsilon_{ij}(\omega, \mathbf{k})$  corresponds to frequency dispersion, while the dependence on the wave vector corresponds to spatial dispersion. The region in which the kernel  $\hat{\epsilon}_{ij}(\tau, \mathbf{R})$  is of any significance whatever is determined by the characteristic frequencies of the medium  $\omega_l$  (and also by the reciprocals of the relaxation times) and by the characteristic dimensions  $a_l$ .

The frequencies  $\omega_l$  usually lie within a rather broad range. The dimensions  $a_l$  ("molecular action radius" etc), to the contrary can in many cases be regarded as small. In liquids and solids, the  $a_l$  are the dimensions of the molecules, the distances between atoms, or the lattice constants; all these quantities are usually of the same order of magnitude and are very small compared with the wavelengths, which lie in the optical range. It is therefore understandable why in optics spatial dispersion plays generally a less important role than frequency dispersion.

If spatial dispersion is neglected, then  $\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}(\omega, 0) \equiv \epsilon_{ij}(\omega)$ , and in a homogeneous isotropic medium, of course,  $\epsilon_{ij}(\omega) = \epsilon(\omega)\delta_{ij}$  (an example of a homogeneous but anisotropic medium is a liquid crystal). If spatial dispersion is taken into account in an isotropic non-gyrotropic medium, then

$$\epsilon_{ij}(\omega, \mathbf{k}) = (\delta_{ij} - k_i k_j / k^2) \epsilon^{\text{tr}}(\omega, k) + (k_i k_j / k^2) \epsilon^{\text{l}}(\omega, k),$$

with

$$\epsilon^{\text{tr}}(\omega, 0) = \epsilon^{\text{l}}(\omega, 0) = \epsilon(\omega)$$

(for details, see below). We note that the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$ , as can be seen from (1.1) and (1.5), describes also the magnetic properties of the medium [if  $\mathbf{D}'$  depends also on  $\mathbf{B}$ , relation (1.3) remains in force, since  $\mathbf{B}$  can be expressed in terms of  $\mathbf{E}$  with the aid of the equation  $\text{curl } \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}$ ]. The permeability  $\mu_{ij}(\omega)$  need be introduced in addition only on making the transition to the tensor  $\epsilon_{ij}(\omega)$ . The corresponding limiting transition is quite unusual;<sup>[10]</sup>

we shall not discuss this problem here, since  $\mu_{ij}(\omega) = \delta_{ij}$  for optical frequencies (see also [1], Sec. 60). This circumstance is already implied in the assumption  $\epsilon_{ij}(\omega, 0) = \epsilon_{ij}(\omega)$ . We note also that formula (1.5) or its analytical continuation determine, generally speaking, the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  not only for real but also for complex values of  $\omega$  and  $\mathbf{k}$ . Also the tensor  $\epsilon_{ij}$  does not lose its physical meaning, since it relates  $\mathbf{D}'$  with the field  $\mathbf{E}$ , the amplitude of which builds up or attenuates in time and in space. To be sure, such amplitudes increase without limit at infinity in corresponding regions of the complex plane. However, a field in the form  $\mathbf{E} = \mathbf{E}_0 \exp[i(\mathbf{k} \cdot \mathbf{r} - \omega t)]$  with  $\omega$  and  $\mathbf{k}$  real likewise does not correspond to reality, since such a wave fills all of space-time. For any specific physical formulation of the problem it becomes necessary to deal with an integral with respect to the frequencies and the wave vectors (i.e., with wave packets; this includes also problems with boundary and initial conditions) and strictly speaking only finite values of  $t$  and  $r$  are significant. Therefore, in the region where the functions  $\epsilon_{ij}(\omega, \mathbf{k})$  are defined, they can be used without danger. Generally speaking, the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  has an inverse  $\epsilon_{ij}^{-1}(\omega, \mathbf{k})$ , so that

$$E_i(\mathbf{k}, \omega) = \epsilon_{ij}^{-1}(\omega, \mathbf{k}) D_j(\mathbf{k}, \omega). \quad (1.6)$$

A crystal is not a spatially-homogeneous medium, but for long waves ( $\lambda_0 \gg a$ ) the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  can be used in this case, too. The interaction of  $\epsilon_{ij}(\omega, \mathbf{k})$  in crystals will be analyzed in Sec. 2a; here we confine ourselves to any medium for which (1.5) and (1.6) hold.

The energy relations will be discussed in Sec. 1c. We merely note now that when  $\omega$  and  $\mathbf{k}$  are real the heat released in a monochromatic wave is proportional to the difference  $\epsilon_{ij}(\omega, \mathbf{k}) - \epsilon_{ji}^*(\omega, \mathbf{k})$ , where the asterisk denotes the complex conjugate (see also Sec. 1c). Thus, under the indicated conditions there is no energy absorption if the tensor  $\epsilon_{ij}$  is Hermitian

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ji}^*(\omega, \mathbf{k}). \quad (1.7)$$

The tensor  $\epsilon_{ij}$  is generally speaking complex even for real  $\omega$  and  $\mathbf{k}$ . In both this case and the general case it is convenient to resolve  $\epsilon_{ij}$  into two Hermitian parts  $\epsilon'_{ij}$  and  $\epsilon''_{ij}$ :

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon'_{ij}(\omega, \mathbf{k}) + i\epsilon''_{ij}(\omega, \mathbf{k}) \equiv \delta_{ij} + i \frac{4\pi}{\omega} \sigma_{ij}(\omega, \mathbf{k}), \quad (1.8)$$

$$\epsilon'_{ij} = \epsilon'_{ji}, \quad \epsilon''_{ij} = \epsilon''_{ji},$$

where we introduce also the complex conductivity tensor

$$\sigma_{ij}(\omega, \mathbf{k}) = \sigma'_{ij}(\omega, \mathbf{k}) + i\sigma''_{ij}(\omega, \mathbf{k})$$

which is occasionally employed (we assume that  $\omega \neq 0$  and the tensors  $\sigma'_{ij}$  and  $\sigma''_{ij}$  are Hermitian).\*

\*The mixed notation<sup>[2]</sup>  $\epsilon'_{ij} = \epsilon_{ij} \pm i4\pi\sigma_{ij}/\omega$  is also used, where the total tensor is denoted by the prime, and its Hermitian part is written without the prime [here, unlike in (1.8),  $\sigma_{ij}$  is a Hermitian tensor]. The sign of  $\sigma_{ij}$  corresponds to the sign in the expression for the wave  $\exp[\pm i(\mathbf{k} \cdot \mathbf{r} - \omega t)]$ ; the plus sign is chosen in the present article.

It is obvious from (1.7) that  $\epsilon''_{ij} = 0$  in the absence of absorption and when  $\omega$  and  $\mathbf{k}$  are real.

A real field  $\mathbf{E}$  should produce real induction  $\mathbf{D}'$ , by virtue of which [see (1.4)–(1.5) and Sec. 1c]

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}^*(-\omega, -\mathbf{k}), \quad (1.9)$$

with  $\omega$  and  $\mathbf{k}$  assumed real here. On the other hand, if  $\omega$  is real and the vector  $\mathbf{k}$  complex (a situation which we shall deal with), then

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}^*(-\omega, -\mathbf{k}^*). \quad (1.9a)$$

By virtue of the symmetry of the kinetic coefficients we have [for a proof see, for example [10]]; it is a direct generalization of the corresponding proof for the tensor  $\epsilon_{ij}(\omega)$

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ji}(\omega, -\mathbf{k}). \quad (1.10)$$

It is assumed here and generally in what follows that the induction of the permanent magnetic field is  $\mathbf{B}_{\text{ext}} = 0$ ; otherwise

$$\epsilon_{ij}(\omega, \mathbf{k}, \mathbf{B}_{\text{ext}}) = \epsilon_{ji}(\omega, -\mathbf{k}, -\mathbf{B}_{\text{ext}}).$$

If the medium has a center of symmetry (i.e., the gas molecules or the unit cells of the crystal have a center of symmetry), then the directions  $\mathbf{k}$  and  $-\mathbf{k}$  are equivalent and

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ji}(\omega, \mathbf{k}). \quad (1.11)$$

Generalizing in natural fashion the customary definitions, we shall call a medium in which condition (1.11) is satisfied non-gyrotropic, while a medium not satisfying this condition is called gyrotropic or naturally-active [it must be borne in mind here that relation (1.11) may be satisfied also in the absence of a center of symmetry; in other words, the absence of a center of symmetry is necessary but not sufficient for the occurrence of gyrotropy; see Sec. 2b].

For a non-gyrotropic medium and for  $\mathbf{B}_{\text{ext}} = 0$ , when relation (1.11) holds true, the tensors  $\epsilon'_{ij}(\omega, \mathbf{k})$  and  $\epsilon''_{ij}(\omega, \mathbf{k})$  are real, since a symmetrical Hermitian tensor is always real [see (1.8) and (1.11)]. This takes place, in particular, in the absence of spatial dispersion and when  $\mathbf{B}_{\text{ext}} = 0$ . (The induction  $\mathbf{B}_{\text{ext}} \neq 0$  when there is an external field present or in ferromagnets.) In this connection,  $\epsilon'_{ij}$  and  $\epsilon''_{ij}$  usually denote the real and imaginary parts of  $\epsilon_{ij}$ . We shall do the same whenever there is no danger of misunderstanding.

By virtue of the causality principle which we already used in (1.3) and (1.4), the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  has definite analytical properties. The use of these properties enables us to relate in integral fashion the real and imaginary parts of  $\epsilon_{ij}$  for real  $\omega$  and  $\mathbf{k}$ . The entire analysis is similar to that used for an isotropic medium without spatial dispersion (see, for example, [1]). As a result (see also [10])

$$\text{Re } \epsilon_{ij}(\omega, \mathbf{k}) - \delta_{ij} = \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\text{Im } \epsilon_{ij}(\omega', \mathbf{k})}{\omega' - \omega} d\omega',$$

$$\text{Im } \epsilon_{ij}(\omega, \mathbf{k}) = -\frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\text{Re } \epsilon_{ij}(\omega', \mathbf{k}) - \delta_{ij}}{\omega' - \omega} d\omega'. \quad (1.12)$$



The integrals are taken here in the sense of the principal values and for simplicity we put

$$\lim_{\omega \rightarrow 0} \omega_{ij} \varepsilon(\omega, \mathbf{k}) \rightarrow 0.$$

The Hermitian tensors  $\epsilon'_{ij}$  and  $\epsilon''_{ij}$  can be represented in the form

$$\begin{aligned} \epsilon'_{ij} &= \epsilon'_{ij,c} + i\epsilon_{ij,a}, & \epsilon''_{ij} &= \epsilon''_{ij,c} + i\epsilon''_{ij,a}, & \text{Re } \epsilon_{ij} &= \epsilon'_{ij,c} - \epsilon''_{ij,a}, \\ \epsilon'_{ij,c} &= \epsilon'_{ji,c}, & \epsilon''_{ij,c} &= \epsilon''_{ji,c}, & \epsilon'_{ij,a} &= -\epsilon'_{ji,a}, & \epsilon''_{ij,a} &= -\epsilon''_{ji,a}, \\ \text{Im } \epsilon_{ij} &= \epsilon''_{ij,c} + \epsilon''_{ij,a}, \end{aligned} \quad (1.8a)$$

where all the tensors with subscripts *c* and *a* are real. If  $\mathbf{B}_{\text{ext}} = 0$  and condition (1.11) is satisfied, then  $\epsilon'_{ij,a} = \epsilon''_{ij,a} = 0$  and we can simply put in (1.12)  $\text{Re } \epsilon_{ij} = \epsilon'_{ij}$  and  $\text{Im } \epsilon_{ij} = \epsilon''_{ij}$ . In the general case, on the other hand, we obtain from (1.12) and (1.8a)

$$\begin{aligned} \epsilon'_{ij,c}(\omega, \mathbf{k}) - \delta_{ij} &= \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\epsilon''_{ij,c}(\omega', \mathbf{k}) d\omega'}{\omega' - \omega}, & \epsilon''_{ij,c}(\omega, \mathbf{k}) \\ &= -\frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\{\epsilon_{ij,c}(\omega', \mathbf{k}) - \delta_{ij}\} d\omega'}{\omega' - \omega}, \\ \epsilon'_{ij,a} &= \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\epsilon''_{ij,a}(\omega', \mathbf{k}) d\omega'}{\omega' - \omega}, & \epsilon''_{ij,a}(\omega, \mathbf{k}) &= -\frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\epsilon'_{ij,a}(\omega', \mathbf{k}) d\omega'}{\omega' - \omega}. \end{aligned} \quad (1.12a)$$

We note that in the references known to us<sup>[10,18]</sup> the formulas in (1.12) are derived with both  $\omega$  and  $\mathbf{k}$  assumed real. Yet both quantities can be regarded real only in the limiting case of a transparent medium. Since (1.12) and (1.12a) involve integration with respect to the frequency, the assumed transparency over the entire interval is patently inadmissible. Relations (1.12) and (1.12a) can therefore be used in the presence of spatial dispersion for an analysis of wave propagation in a medium only if they are generalized to include complex values of  $\mathbf{k}$ . Relation (1.10) is obtained (see<sup>[10]</sup>) using (1.12) and is consequently proved in<sup>[10]</sup> only for real  $\mathbf{k}$ . Since  $\mathbf{k}$  enters (1.12) as a parameter, it seems to us that these expressions and formula (1.10) should hold true also for complex  $\mathbf{k}$ . It is assumed here that the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  is uniquely defined for the complex values of  $\mathbf{k}$  under consideration. The absence of singularities of  $\epsilon_{ij}(\omega, \mathbf{k})$  in the corresponding range of variables (analyticity) is probably of importance, too. In the case of weak spatial dispersion of interest to us (see Sec. 2b), the tensor  $\epsilon_{ij}$  or the other tensors employed have indeed no inadmissible singularities near the point  $\mathbf{k} = 0$ , at which the formulas (1.12) are known to be valid. Allowance for the fact that the velocity of the signal does not exceed *c* leads<sup>[18,10]</sup> to dispersion relations that are more general than (1.12).

Certain inequalities for the derivatives of  $\epsilon_{ij}$  with respect to  $\omega$  follow from (1.12) and from the principle of increasing entropy (see<sup>[19]</sup> and Sec. 1c). We note finally that all properties (in particular the symmetry property) of the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  extend to the inverse tensor  $\epsilon_{ij}^{-1}(\omega, \mathbf{k})$ . The use of the tensor  $\epsilon_{ij}$  or  $\epsilon_{ij}^{-1}$ , i.e., of relations (1.5) or (1.6), is equivalent over a wide range and is dictated by convenience.

b) Normal electromagnetic waves in a medium. Transverse and longitudinal waves. "Fictitious" longitudinal waves and "polarization waves." We shall seek the solution of the field equations (1.1) in the form of plane waves

$$\mathbf{E}_l = \mathbf{E}_{0l} e^{i(\mathbf{k}\mathbf{r} - \omega t)}, \quad \mathbf{B}_l = \mathbf{B}_{0l} e^{i(\mathbf{k}\mathbf{r} - \omega t)}, \quad \mathbf{D}'_l = \mathbf{D}'_{0l} e^{i(\mathbf{k}\mathbf{r} - \omega t)}, \quad (1.13)$$

where  $\mathbf{E}_{0l}$ ,  $\mathbf{B}_{0l}$ , and  $\mathbf{D}'_{0l}$  are constants.

We then have\*

$$\begin{aligned} \mathbf{D}' &= -\frac{c}{\omega} [\mathbf{k}\mathbf{B}], & \mathbf{k}\mathbf{D}' &= 0, \\ \mathbf{B} &= \frac{c}{\omega} [\mathbf{k}\mathbf{E}], & \mathbf{k}\mathbf{B} &= 0. \end{aligned} \quad (1.14)$$

From this, eliminating  $\mathbf{B}$ , we obtain the "wave equation"

$$\mathbf{D}' = \frac{c^2}{\omega^2} \{k^2 \mathbf{E} - \mathbf{k}(\mathbf{k}\mathbf{E})\}. \quad (1.15)$$

The waves (1.13) can obviously be solutions of the field equations<sup>†</sup> only if the relation between  $\mathbf{D}'$  and  $\mathbf{E}$  is such that only waves with the same  $\omega$  and  $\mathbf{k}$  are coupled. This is precisely the situation under conditions when the tensors  $\epsilon_{ij}(\omega, \mathbf{k})$  or  $\epsilon_{ij}^{-1}(\omega, \mathbf{k})$  can be introduced.

Substituting (1.5) in (1.15) we obtain a system of linear equations

$$\frac{\omega^2}{c^2} \epsilon_{ij} E_j - k^2 E_i + k_i k_j E_j = 0. \quad (1.16)$$

The condition for the existence of a nontrivial solution for this system leads to a dispersion equation relating  $\omega$  with  $\mathbf{k}$  (the symbols  $\Delta$  or  $|\dots|$  denote determinants):

$$\Delta_1(\omega, \mathbf{k}) = \left| \frac{\omega^2}{c^2} \epsilon_{ij}(\mathbf{k}, \omega) - k^2 \delta_{ij} + k_i k_j \right| = 0. \quad (1.17)$$

If the tensor  $\epsilon_{ij}$  is replaced by  $\epsilon_{ij}^{-1}$ , we obtain

$$\Delta_2(\omega, \mathbf{k}) = \left| \frac{\omega^2}{c^2} \delta_{ij} - k^2 \epsilon_{ij}^{-1}(\omega, \mathbf{k}) + k_i k_j \epsilon_{ij}^{-1}(\omega, \mathbf{k}) \right| = 0. \quad (1.18)$$

When both tensors  $\epsilon_{ij}$  and  $\epsilon_{ij}^{-1}$  exist simultaneously, equations (1.17) and (1.18) are of course equivalent. Unless otherwise stipulated, this situation is assumed throughout.

The roots of (1.17), which are the frequencies of the normal waves, will be designated by the index *l*, i.e., denoted by  $\omega_l = \omega_l(\mathbf{k})$ , where  $\omega_l$  and  $\mathbf{k}$  are, generally speaking, complex. Whether one of these quantities,  $\omega_l$  or  $\mathbf{k}$ , can be chosen as the real variable depends on the formulation of the physical problem (see, for example,<sup>[2]</sup>). In crystal optics the frequency  $\omega$  is usually real (this is the frequency of the light from the external source). But even when the frequency  $\omega$  is real in general  $\mathbf{k} = \mathbf{k}' + i\mathbf{k}''$  and only in the transparency region do we have approximately  $\mathbf{k} = \mathbf{k}'$  ( $\mathbf{k}''$

\* $[\mathbf{k}\mathbf{B}] = \mathbf{k} \times \mathbf{B}$ .

†An analysis of waves of the type (1.13) may prove insufficient for values of  $\omega$  and  $\mathbf{k}$  corresponding to multiple roots of the dispersion equation. This question, which is not very significant, will be discussed in Sec. 3a.

and  $\mathbf{k}''$  are real). We shall consider below as a rule only homogeneous plane waves, for which

$$\mathbf{k} = \frac{\omega}{c} \hat{n}\mathbf{s}, \quad \hat{n}(\omega, \mathbf{s}) = n + i\kappa, \quad (1.19)$$

where  $\mathbf{s} = \mathbf{k}/k$  is a unit real vector and the frequency  $\omega$  is real. When the vector  $\mathbf{s}$  can occur, it is implied without further explanation that the transition to homogeneous waves has been made. Under conditions (1.19), obviously, the statement that  $\mathbf{k}$  is complex with  $\omega$  real denotes that  $\kappa \neq 0$ .

Equation (1.15) now has the form

$$\mathbf{D}' = \hat{n}^2 \{\mathbf{E} - \mathbf{s}(\mathbf{s}\mathbf{E})\}. \quad (1.20)$$

The dispersion equation (1.17) has in this case the form

$$\begin{aligned} \epsilon_{ij} \left( \omega, \frac{\omega}{c} \hat{n}\mathbf{s} \right) - \hat{n}^2 (\delta_{ij} - s_i s_j) \Big| \\ = \epsilon_{ij} s_i s_j \hat{n}^4 - [(\epsilon_{ij} s_i s_j) \epsilon_{il} - s_i \epsilon_{il} \epsilon_{lj} s_j] \hat{n}^2 + |\epsilon_{ij}| = 0. \end{aligned} \quad (1.21)$$

The induction  $\mathbf{D}'$  is always transverse (i.e.,  $\mathbf{s} \cdot \mathbf{D}' = 0$ ) and the terms "transverse" and "longitudinal" waves will be employed henceforth, as usual, only with respect to the vector  $\mathbf{E}$ . In the general case normal waves in an anisotropic medium are neither transverse nor longitudinal. Under certain conditions, however, the waves can be longitudinal or transverse, and these cases will be pointed out. For transverse waves, in accordance with (1.20),

$$\mathbf{D}' = \hat{n}^2 \mathbf{E}, \quad \mathbf{s}\mathbf{E} = 0, \quad \mathbf{s}\mathbf{D}' = 0. \quad (1.22)$$

By way of illustration we shall indicate in the present section the character of different normal waves for a rhombic crystal in the limit as  $\mathbf{k} \rightarrow 0$ , i.e., assuming  $\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}(\omega)$  (classical crystal optics). For such a crystal the principal axes of the tensor  $\epsilon_{ij}$  coincide for all  $\omega$  with the two-fold axes  $x, y, z$  (we refer to crystal classes  $D_2$  and  $D_{2h}$ ; in class  $C_{2v}$  the  $z$  axis is parallel to the two-fold axis and  $x$  and  $y$  are perpendicular to the symmetry planes). In this coordinate system the diagonal elements of  $\epsilon_{ij}$ , with values  $\epsilon_x, \epsilon_y$ , and  $\epsilon_z$ , are all different (it is assumed that there is no accidental degeneracy). In transverse waves the vector  $\mathbf{E}$  is parallel to any one of the axes  $x, y$ , or  $z$ , and the vector  $\mathbf{k}$  lies in one of the coordinate planes. In the frame of the principal axes  $x, y$ , and  $z$  we have for transverse waves

$$n_{1,2,3}^2 = \epsilon_{x,y,z}(\omega). \quad (1.23)$$

In an arbitrary frame we have for transverse waves [see (1.16), (1.19), and (1.23)]

$$|\epsilon_{ij}(\omega, \mathbf{k}) - \hat{n}^2 \delta_{ij}| = 0. \quad (1.22a)$$

This equation also holds true in the presence of spatial dispersion.

We note that transverse waves correspond only to those solutions of (1.23) that are compatible with the transversality condition  $\mathbf{s} \cdot \mathbf{E} = 0$ . It is therefore obvi-

ously inconvenient and senseless to use (1.22a). For longitudinal waves, as is clear from (1.20)

$$\mathbf{D}' = 0, \quad \mathbf{E} = E\mathbf{s}. \quad (1.24)$$

When  $\mathbf{D}'_i = \epsilon_{ij}$  and  $E_j = 0$ , the vector  $\mathbf{E}$  can be non-vanishing only if

$$|\epsilon_{ij}(\omega, \mathbf{k})| = 0. \quad (1.25)$$

In addition, one must be certain that the vector is longitudinal for the solution under consideration, i.e., that  $\mathbf{E} = E\mathbf{s}$ . Longitudinal waves are considered quite frequently in plasma applications and have been sufficiently well investigated (see, for example, [2]). Nonetheless we make several remarks concerning these waves. It follows from the field equations (1.14) that in the longitudinal waves  $\mathbf{B} = 0$  and they can thus be regarded with allowance for the Coulomb field only.\*

In the absence of spatial dispersion, the condition (1.25) determines the possible longitudinal-wave frequencies  $\omega_{||}$ , which are independent of  $\mathbf{k}$ . Therefore the group velocity  $\mathbf{u} = d\omega/d\mathbf{k} = 0$  and consequently only an account of the spatial dispersion can lead to the propagation of longitudinal waves (in the sense that  $\mathbf{u} \neq 0$ ).

In an isotropic non-gyrotropic medium, as already indicated in Sec. 1a, we have

$$\epsilon_{ij}(\omega, \mathbf{k}) = (\delta_{ij} - s_i s_j) \epsilon^{\text{tr}}(\omega, k) + s_i s_j \epsilon^{\text{l}}(\omega, k). \quad (1.26)$$

We arrive at this expression by recognizing that a second-rank tensor can be formed in an isotropic medium only by using the tensor  $\delta_{ij}$  ( $\delta_{ij} = 0$  when  $i \neq j$ ,  $\delta_{ij} = 1$  when  $i = j$ ) and the tensor  $s_i s_j$ . The introduction of the symbols  $\epsilon^{\text{tr}}$  and  $\epsilon^{\text{l}}$  is connected with the fact that in a transverse field  $\mathbf{E}$ , when  $\mathbf{s} \cdot \mathbf{E} = 0$ , we have

$$\mathbf{D}' = \epsilon^{\text{tr}}(\omega, k) \mathbf{E} \quad (\text{for } \mathbf{s}\mathbf{E} = 0). \quad (1.27)$$

In a longitudinal field, when  $\mathbf{E} = E\mathbf{s}$ ,

$$\mathbf{D}' = \epsilon^{\text{l}}(\omega, k) \mathbf{E} \quad (\text{for } \mathbf{E} = E\mathbf{s}). \quad (1.28)$$

It is easily seen that waves in an isotropic medium can be either longitudinal or transverse. For transverse waves we have in this case [see (1.22) and (1.27)]

$$\hat{n}_{\perp}^2 = \epsilon^{\text{tr}} \left( \omega, \frac{\omega}{c} \hat{n}_{\perp} \right); \quad (1.29)$$

and for longitudinal waves, according to (1.24) and (1.28),

$$\epsilon^{\text{l}} \left( \omega, \frac{\omega}{c} \hat{n}_{\parallel} \right) = 0. \quad (1.30)$$

Of course, (1.30) can also be obtained by substituting (1.26) in the general relation (1.25). When spatial dispersion is neglected  $\epsilon^{\text{tr}} = \epsilon^{\text{l}} = \epsilon(\omega)$  and the frequen-

\*The equality  $\mathbf{B} = 0$  in longitudinal waves pertains, naturally only to the field of the wave, as is assumed in (1.14). In the presence of an external magnetic field  $\mathbf{B}_{\text{ext}} = \text{const}$ , the longitudinal wave can propagate only in the direction of  $\mathbf{B}_{\text{ext}}$  (see [2]).

cies of the longitudinal waves are determined by the equation  $\epsilon(\omega_{\parallel}) = 0$ .

For rhombic crystals and  $k = 0$  Eq. (1.25) for longitudinal waves has in the frame of the principal axes the form

$$\epsilon_x(\omega) \epsilon_y(\omega) \epsilon_z(\omega) = 0.$$

In the absence of accidental degeneracy these longitudinal waves are directed along one of the principal axes  $x$ ,  $y$ ,  $z$  and have a frequency  $\omega$  satisfying the corresponding equation  $\epsilon_x(\omega) = 0$ ,  $\epsilon_y(\omega) = 0$ , or  $\epsilon_z(\omega) = 0$ . Only if the vector  $\mathbf{k}$  is parallel to one of the axes  $x$ ,  $y$ , or  $z$  are all normal waves in a rhombic crystal transverse or longitudinal, as is clear even from symmetry considerations. When the vector  $\mathbf{k}$  lies in the coordinate plane (but not parallel to an axis), one of the normal waves is transverse and the other is neither longitudinal nor transverse.\* The latter pertains to all normal waves in the case of a vector  $\mathbf{k}$  lying outside the coordinate planes. We have in mind here the optical region or, formally, finite values of  $\mathbf{k}$ , and at the same time only solutions that satisfy all the field equations. If we consider large values of  $k$  (short waves), then waves with finite values of  $\mathbf{D}'$  and with non-zero field  $\mathbf{E}$  come quite close to longitudinal. This is clear directly from the wave equation (1.20): in the transparency region we have  $\hat{n} = n$  and for a finite frequency  $\omega$  the limit  $k = \omega n/c \rightarrow \infty$  corresponds to the limit  $n \rightarrow \infty$ ; however when  $\hat{n} = n \rightarrow \infty$  the induction  $\mathbf{D}'$ , in accordance with (1.20), is finite only when  $\mathbf{E} = \mathbf{s}(\mathbf{s} \cdot \mathbf{E})$ , i.e., only for longitudinal waves if  $\mathbf{E} \neq 0$ . On going over to absolute values, the same pertains to an arbitrary  $\hat{n}$ , but actually if absorption is taken into account the modulus  $|\hat{n}|$  is finite and the transition to the limit  $|\hat{n}| \rightarrow \infty$  cannot be realized. If spatial dispersion is taken into account,  $|\hat{n}|$  is generally speaking finite even if absorption is neglected (see Sec. 3b). These factors, however, are not significant here since the limiting transition  $k \rightarrow \infty$  has a formal significance, and physically we are dealing only with the fact that waves with  $\mathbf{E} \neq 0$  are nearly longitudinal when  $k = \omega |\hat{n}|/c$  is sufficiently large.

To determine the conditions under which  $n$  increases, we first use the well-known equation which determines  $n$  in classical crystal optics for a uniaxial non-absorbing crystal:

$$n_{\perp}^2 = \epsilon'_{\perp} = \epsilon'_x = \epsilon'_y, \quad \frac{1}{n_z^2} = \frac{s_x^2 + s_y^2}{\epsilon'_z(\omega)} + \frac{s_z^2}{\epsilon'_{\perp}(\omega)} \quad (1.31)$$

(the  $z$  axis coincides with an optical axis, say the four-fold axis in the case of a tetragonal crystal).

The refractive index  $n_z$  for the extraordinary wave becomes infinite if

$$\epsilon'_{\perp}(\omega) (s_x^2 + s_y^2) + \epsilon'_z(\omega) s_z^2 = 0. \quad (1.32)$$

\*In such waves the direction of the energy flux  $\mathbf{S}$  (or of the group-velocity vector  $\mathbf{u} = d\omega/d\mathbf{k}$ ) does not coincide with  $\mathbf{k}$  (see also Sec. 1c).

In order to satisfy this condition, the indices  $\epsilon'_x$  and  $\epsilon'_z$  should obviously have different signs, but can have no singularities (poles) whatever.

Within the limits of classical crystal optics, if the principal axes of the tensors  $\epsilon'_{ij}(\omega)$  and  $\epsilon''_{ij}(\omega)$  coincide, it is convenient to transform to the system of these axes. In this system the dispersion equation (1.21) has the form

$$\begin{aligned} & (\epsilon_x s_x^2 + \epsilon_y s_y^2 + \epsilon_z s_z^2) \hat{n}^4 \\ & - [\epsilon_x (\epsilon_y + \epsilon_z) s_x^2 + \epsilon_y (\epsilon_x + \epsilon_z) s_y^2 + \epsilon_z (\epsilon_x + \epsilon_y) s_z^2] \hat{n}^2 \\ & + \epsilon_x \epsilon_y \epsilon_z = 0. \end{aligned} \quad (1.33)$$

The root of this equation can become infinite for finite values of  $\epsilon_x$ ,  $\epsilon_y$ , and  $\epsilon_z$  only under a condition that is a direct generalization of (1.32):

$$\epsilon_x^2 s_x^2 + \epsilon_y^2 s_y^2 + \epsilon_z^2 s_z^2 = 0. \quad (1.34)$$

On the other hand, if we do not change to the principal-axes frame, then we can readily see that (1.34) is written in the form

$$\epsilon_{ij}(\omega) s_i s_j = 0. \quad (1.35)$$

The frequencies  $\omega'_{\parallel}$ , satisfying (1.35) or the more general expression (1.39) given below, will be called the frequencies of the "fictitious" longitudinal waves; in these waves  $\mathbf{D}' \neq 0$  and  $\mathbf{E} = \mathbf{E}\mathbf{s}$ .

As already mentioned, the "fictitious" longitudinal waves satisfy the field equations only as  $\mathbf{k} \rightarrow \infty$ . However, if we consider the equation (1.35) independently of the field equations, a procedure that has a certain meaning (see below), then the frequencies  $\omega'_{\parallel}$  are found to depend on  $\mathbf{s}$  even when  $k = 0$  [the absolute value of  $k$  does not enter in (1.35)]. This means that the frequencies  $\omega'_{\parallel}$  are analytic functions of  $\mathbf{k}$  as  $k \rightarrow 0$ .

The frequencies  $\omega'_{\parallel}$  correspond to resonance—infinite values for the curves of  $n(\omega)$ —only for non-transverse waves. The latter limitation is connected with the fact that strictly transverse waves [see (1.22) and (1.23)] cannot approach longitudinal waves in any manner. At the same time, the index  $n(\omega)$  can tend to infinity for these waves too.

It is clear from (1.20) that for non-longitudinal waves, and particularly for transverse waves [see (1.21)] we have

$$\mathbf{E} = 0 \text{ (for } |\hat{n}| \rightarrow \infty \text{ and finite } \mathbf{D}'). \quad (1.36)$$

In the simple case (1.23), the pole  $|\hat{n}| \rightarrow \infty$  corresponds to a pole of one of the quantities  $\epsilon_{x,y,z}(\omega)$ .

If we do not consider the field equations, then the induction  $\mathbf{D}'$  can differ from zero for a zero field  $\mathbf{E}_i = \epsilon_{ij}^{-1}(\omega, \mathbf{k}) D'_j = 0$  only when

$$|\epsilon_{ij}^{-1}(\omega, \mathbf{k})| = 0. \quad (1.37)$$

Waves with frequencies  $\omega_{\mathbf{P}}(\mathbf{k})$ , satisfying simultaneously this equation and the condition  $\mathbf{k} \cdot \mathbf{D}' = 0$ , will be called "polarization waves" [in such waves the generalized polarization  $\mathbf{P} = (\mathbf{D}' - \mathbf{E})/4\pi = \mathbf{D}'/4\pi$  does not

vanish when  $\mathbf{E} = 0$ ]. As follows from (1.36) and the statements above, the "polarization waves" satisfy the complete system of equations only as  $k \rightarrow \infty$ .

The "fictitious" longitudinal waves and the "polarization waves," satisfying the field equations only as  $|\hat{n}| \rightarrow \infty$ , cannot exist in reality, and their analysis is only ancillary. The latter is connected predominantly with the fact that these waves are obtained in the solution of the purely Coulomb problem  $\text{div } \mathbf{D}' = 0$ ,  $\text{curl } \mathbf{E} = 0$  for arbitrary  $\mathbf{k}$  [the equations of the Coulomb problem are obtained formally from the general field equations (1.1) with  $c \rightarrow \infty$ ]. For homogeneous plane waves the last two equations mean that

$$s\mathbf{D}' = 0, \quad \mathbf{E} = E\mathbf{s}. \quad (1.38)$$

"Polarization waves" satisfy these equations of the Coulomb problem if the induction in these waves, in addition to meeting condition (1.37), is also transverse ( $\mathbf{s} \cdot \mathbf{D}' = 0$ ). "Fictitious" longitudinal waves ( $\mathbf{D}' \neq 0$ ,  $\mathbf{E} = E\mathbf{s} \neq 0$ ) satisfy equations (1.38) if

$$s_i D'_i = s_i \varepsilon_{ij}(\omega, \mathbf{k}) E_j = s_i s_j \varepsilon_{ij}(\omega, \mathbf{k}) E = 0,$$

i.e., under the condition

$$\varepsilon_{ij}(\omega, \mathbf{k}) s_i s_j = 0. \quad (1.39)$$

This equation generalizes directly the condition (1.35) and becomes identical with it in the limit of classical crystal optics.

The fact that the solutions of the Coulomb problem ("fictitious" longitudinal waves and "polarization waves") satisfy for  $k \rightarrow \infty$  the complete system of field equations is of course connected with the possibility of neglecting the transverse field\* (or, as is sometimes stated, neglecting the retardation) as  $k \rightarrow \infty$ . It is indeed clear directly from the field equations (1.14) that as  $k \rightarrow \infty$  and for finite  $\omega \neq 0$ ,  $\mathbf{E}$ , and  $\mathbf{D}'$  we have  $\mathbf{B} \rightarrow 0$  and  $\mathbf{k} \times \mathbf{E} = 0$ . The same is clear from the dispersion equation (1.21), since this equation is satisfied if (1.39) holds and  $\hat{n}^2 \rightarrow \infty$ .

The longitudinal waves (1.24), of course, satisfy the equations of the Coulomb problem. If we disregard these waves, then the solution of the Coulomb problem in crystal optics is of importance only from the point of view of calculating the tensor  $\varepsilon_{ij}(\omega, \mathbf{k})$  for some particular model. An important role is then assumed, as already mentioned in the introduction and as will be discussed further in Sec. 4, by the "polarization waves" and by the approximate solutions obtained when  $\mathbf{E} = 0$  and going over into the "fictitious" longitudinal waves when the influence of the long-wave

\*If we are dealing with the microscopic field, it is necessary to take into account in the field equations (1.1) the appearance of the charge tensor  $\rho_0$  and current density  $\mathbf{j}_0$ , which are connected with the particles. Consequently the neglect of the transverse field does not apply to magnetic fields, which are important in the analysis of the magnetic interaction between particles in the crystal.

**Table I.** Normal plane and homogeneous waves in an anisotropic medium ( $\mathbf{E}_l = E_{0l} e^{i(\mathbf{k}\mathbf{r} - \omega t)}$ ,  $\mathbf{k} = \frac{\omega}{c} \hat{n}(\omega) \mathbf{s}$ ,  $s^2 = 1$ )

<p>a) Complete system of field equations (finite <math>k</math>):</p> <p>General Case:</p> $\mathbf{D}' = \hat{n}^2 \{\mathbf{E} - \mathbf{s}(\mathbf{s}\mathbf{E})\}, \quad s\mathbf{D}' = 0, \quad  \varepsilon_{ij}(\omega, \mathbf{k}) - \hat{n}^2(\delta_{ij} - s_i s_j)  = 0;$ <p>Transverse Waves:</p> $\mathbf{D}' = \hat{n}^2 \mathbf{E}, \quad s\mathbf{D}' = 0, \quad \mathbf{s}\mathbf{E} = 0;$ <p>Longitudinal Waves:</p> $\mathbf{D}' = 0, \quad \mathbf{E} = E\mathbf{s} \neq 0, \quad  \varepsilon_{ij}(\omega, \mathbf{k})  = 0.$ <p>b) Solutions of the Coulomb problem (<math>\mathbf{s} \cdot \mathbf{D}' = 0</math>, <math>\mathbf{E} = E\mathbf{s}</math>):</p> <p>Longitudinal Waves:</p> $\mathbf{D}' = 0, \quad \mathbf{E} = E\mathbf{s} \neq 0, \quad  \varepsilon_{ij}(\omega, \mathbf{k})  = 0;$ <p>"Fictitious" Longitudinal Waves:</p> $\mathbf{D}' \neq 0, \quad s\mathbf{D}' = 0, \quad \mathbf{E} = E\mathbf{s} \neq 0, \quad \varepsilon_{ij}(\omega, \mathbf{k}) s_i s_j = 0;$ <p>"Polarization Waves:"</p> $\mathbf{D}' \neq 0, \quad s\mathbf{D}' = 0, \quad \mathbf{E} = 0, \quad  \varepsilon_{ij}^{-1}(\omega, \mathbf{k})  = 0.$ <p>Formally, as <math>k \rightarrow \infty</math>, the "fictitious" longitudinal waves and the "polarization waves" satisfy also the complete system of field equations.</p>
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field  $\mathbf{E}$  on the frequencies and wave polarization is taken into account.

In conclusion we present Table I, which lists the relations for normal waves in an anisotropic medium.

Formally, as  $k \rightarrow \infty$ , the "fictitious" longitudinal waves and the "polarization waves" satisfy also the complete system of field equations.

c) Energy and some other relations for waves in an anisotropic medium. In the analysis of quantities that are quadratic in the field (for example, the energy density), the fields must be regarded as real. We therefore put

$$\mathbf{E}(\mathbf{r}, t) = \frac{1}{2} (\mathbf{E}_0 e^{-i(\omega t - \mathbf{k}\mathbf{r})} + \mathbf{E}_0^* e^{i(\omega t - \mathbf{k}^*\mathbf{r})}), \quad (1.40)$$

where  $\mathbf{E}_0$  is constant for a monochromatic wave and varies slowly for a quasi-monochromatic field (this means that  $\mathbf{E}_0$  changes little within a time  $\sim 1/\omega$  and over a distance  $\sim 1/k$ ). The frequency  $\omega$  in (1.40) is assumed real in accordance with the character of the problems of interest to us. Expressions analogous to (1.40) are used for  $\mathbf{D}'$  and  $\mathbf{B}$ .

If we neglect the derivatives of  $\mathbf{E}_0$ ,  $\mathbf{B}_0$ , and  $\mathbf{D}'_0$  we can rewrite (1.14) in the form

$$\omega \mathbf{D}'_0 = -c [\mathbf{k}\mathbf{B}_0], \quad \omega \mathbf{B}_0 = c [\mathbf{k}\mathbf{E}_0]; \quad (1.41a)$$

$$\omega \mathbf{D}'_0^* = -c [\mathbf{k}^*\mathbf{B}_0^*], \quad \omega \mathbf{B}_0^* = c [\mathbf{k}^*\mathbf{E}_0^*]. \quad (1.41b)$$

Multiplying (1.41a) by  $\mathbf{E}_0$  and  $\mathbf{B}_0$  respectively, we get

$$\mathbf{D}'_0 \mathbf{E}_0 = \mathbf{B}_0 \mathbf{B}_0 = c\mathbf{k} [\mathbf{E}_0 \mathbf{B}_0]. \quad (1.42)$$

In a transparent medium  $\mathbf{k} = \mathbf{k}^*$  and, as is clear from (1.41), we also have

$$\mathbf{D}'_0 \mathbf{E}_0^* = \mathbf{B}_0 \mathbf{B}_0^*. \quad (1.43)$$

Further, we obtain from (1.41a) after multiplying by  $\mathbf{E}^*$  and  $\mathbf{B}_0^*$  respectively

$$\omega (\mathbf{D}'_0 \mathbf{E}_0^* \pm \mathbf{B}_0 \mathbf{B}_0^*) = ck \{ [\mathbf{E}_0 \mathbf{B}_0^*] \pm [\mathbf{E}_0^* \mathbf{B}_0] \}. \quad (1.44)$$

In addition, of course,

$$k \mathbf{D}'_0 = 0, \quad k \mathbf{B}_0 = 0, \quad \mathbf{D}'_0 \mathbf{B}_0 = 0, \quad \mathbf{B}_0 \mathbf{E}_0 = 0. \quad (1.45)$$

Differentiating (1.44) with respect to  $k_l$  and taking (1.5) into account, we obtain expressions that become highly simplified for a transparent medium if the upper sign is chosen in (1.44) (for the general case see [19], where the earlier literature is also cited). For a transparent medium we thus arrive at the relation

$$\begin{aligned} \overline{W} \mathbf{u} &= \overline{S}, \quad \mathbf{u} = \frac{\partial \omega}{\partial \mathbf{k}}, \quad \epsilon_{ij} = \epsilon'_{ij}, \quad \overline{S} = \overline{S}_0 + \overline{S}_1, \quad \overline{S}_0 = \frac{c}{16\pi} \{ [\mathbf{E}_0 \mathbf{B}_0^*] \\ &+ [\mathbf{E}_0^* \mathbf{B}_0] \}, \\ S_{1l} &= -\frac{\omega}{16\pi} \frac{\partial \epsilon'_{ij}(\omega, \mathbf{k})}{\partial k_l} E_{0j} E_{0i}^*, \\ \overline{W} &= \frac{1}{16\pi} \left\{ \frac{\partial \omega \epsilon'_{ij}(\omega, \mathbf{k})}{\partial \omega} E_{0j} E_{0i}^* + \mathbf{B}_0 \mathbf{B}_0^* \right\} \end{aligned} \quad (1.46)$$

Here  $\mathbf{u}$  —group velocity,  $W$  —time-averaged energy density,  $\mathbf{S}_0$  —Poynting vector, and  $S_1$  —energy flux arising only in the presence of spatial dispersion (the superior bar denotes averaging with respect to time or, more accurately, with respect to the high frequency; for more details see [1,2,10,19,20]). In the absence of spatial and frequency dispersion

$$\overline{W} = \overline{W}_0 = \frac{\mathbf{D}'_0 \mathbf{E}_0^* + \mathbf{B}_0 \mathbf{B}_0^*}{16\pi}, \quad \overline{S} = \overline{S}_0 \quad (1.47)$$

and choosing the upper sign we obtain from (1.46) and (1.44)

$$\overline{W}_0 \mathbf{u} = \overline{S}_0, \quad \overline{W}_0 = \frac{\mathbf{k}}{\omega} \overline{S}_0, \quad \frac{\mathbf{u} \mathbf{k}}{\omega} = 1, \quad (1.48)$$

i.e.,  $u_k = v_{ph}$ , where  $u_k = \mathbf{u} \cdot \mathbf{k} / k$  and  $v_{ph} = \omega / k$  — phase velocity in the direction of  $\mathbf{k}$ .

Relations other than (1.41) and (1.46) but of the same type can also be derived. [19]

In the presence of absorption or in the case of a non-stationary process, an important role in the analysis of the energy relations is played by the Poynting equation

$$\frac{1}{4\pi} \mathbf{E} \frac{\partial \mathbf{D}'}{\partial t} + \frac{\partial}{\partial t} \frac{B^2}{4\pi} = -\frac{c}{4\pi} \text{div} [\mathbf{E} \mathbf{B}], \quad (1.49)$$

which follows from the field equations (1.1).

For the field (1.40) relation (1.49) averaged over the high frequency becomes [19a]

$$\begin{aligned} \frac{\partial \overline{W}}{\partial t} + \frac{1}{2} \sigma_{ij} E_{0j} E_{0i}^* - \frac{i}{4} \left\{ \frac{\partial \sigma_{ij}^*}{\partial \omega} \frac{\partial E_{0i}}{\partial t} E_{0j}^* - \frac{\partial \sigma_{ij}}{\partial \omega} \frac{\partial E_{0i}^*}{\partial t} E_{0j} \right\} \\ + \frac{i}{4} \left\{ \frac{\partial \sigma_{ij}^*}{\partial k_l} \frac{\partial E_{0j}}{\partial x_l} E_{0i}^* - \frac{\partial \sigma_{ij}}{\partial k_l} \frac{\partial E_{0i}^*}{\partial x_l} E_{0j} \right\} = -\text{div} \overline{S}, \end{aligned} \quad (1.50)$$

where the expressions for  $\overline{W}$  and  $\overline{S}$  are indicated in (1.46) and the quantity  $\sigma_{ij} = \omega \epsilon''_{ij} / 4\pi$  (the conductivity) is introduced with  $\epsilon_{ij} = \epsilon'_{ij} + i\epsilon''_{ij}$  [the Hermitian tensors  $\epsilon_{ij}$ ,  $\sigma_{ij}$ , and their derivatives are taken in (1.50)

for real values of the frequency  $\omega$  and the vector  $\mathbf{k}$ ; a more general expression, suitable for complex  $\mathbf{k}$ , is also derived in [19a]]. It is clear from (1.50) (see [19a,20] for details) that the physical interpretation of the various terms in this equation is in general difficult. On the other hand, if we are dealing for example with a monochromatic wave, (i.e., when  $\partial \mathbf{E}_{0i} / \partial t = 0$ ) in the absence of spatial dispersion [20] or in an almost transparent medium (real vector  $\mathbf{k}$ ; see [10]) then the quantity  $\frac{1}{2} \sigma_{ij} E_{0j} E_{0i}^*$  is the heat dissipated from unit volume per unit time.\*

Starting from the dispersion relations (1.12) we can obtain in the case of an equilibrium medium [19] several inequalities for the derivatives  $\partial \epsilon'_{ij} / \partial \omega$  and their combinations. For example,

$$\frac{\partial \epsilon'_{ii}(\omega, \mathbf{k})}{\partial \omega} > 0, \quad \frac{\partial \epsilon'_{ii}}{\partial \omega} > \frac{2(3 - \epsilon'_{ii})}{\omega}. \quad (1.51)$$

Recognizing that the group velocity  $\mathbf{u}$  is the signal velocity in a transparent medium, it becomes quite obvious that in such a medium

$$u = \left| \frac{d\omega}{d\mathbf{k}} \right| < c, \quad (1.52)$$

to include the vacuum we must naturally write this inequality in the form  $u \leq c$ . In the general case  $\omega = \omega(k, \alpha, \beta)$ , where  $\alpha = k_x / k$  and  $\beta = k_y / k$  ( $\gamma = k_z / k$ ,  $\alpha^2 + \beta^2 + \gamma^2 = 1$ ). Hence, as can be readily seen,

$$\frac{\partial \omega}{\partial k_x} = \frac{\partial \omega}{\partial k} \alpha + \frac{\partial \omega}{\partial \alpha} \frac{1 - \alpha^2}{k} - \frac{\partial \omega}{\partial \beta} \frac{\alpha \beta}{k}$$

etc.

We therefore have directly

$$u_k = \frac{\mathbf{k}}{k} \frac{d\omega}{d\mathbf{k}} = \frac{\partial \omega}{\partial k} = \frac{c}{\frac{d(\omega n)}{d\omega}} = u \cos \varphi, \quad (1.53)$$

where  $\varphi$  —angle between  $\mathbf{u}$  and  $\mathbf{k}$ . According to (1.52) and (1.53)

$$\left| \frac{d(\omega n)}{d\omega} \right| > 1. \quad (1.54)$$

In the absence of spatial dispersion we can show that the angle  $\varphi < \pi/2$  (i.e., the vectors  $\mathbf{u}$  and  $\mathbf{k}$  always make an acute angle) and consequently

$$\frac{d(\omega n)}{d\omega} > 1. \quad (1.54a)$$

In addition, in the absence of spatial dispersion in a non-absorbing medium the quantity  $\hat{n}^2 = (n + i\kappa)^2$  is always real, and in the presence of absorption the product  $n\kappa$  is positive. [19] In the presence of spatial dispersion the angle between  $\overline{S}_0$  and  $\mathbf{k}$  remains acute,

\*In the present article we are treating an absorbing medium which is in particular a medium in thermal equilibrium. We note that a non-equilibrium medium (crystal) may be not absorbing, but radiating, as is the case in lasers. For an isotropic medium without spatial dispersion this means that  $\epsilon'' = 4\pi\sigma/\omega < 0$  and  $n\kappa = 2\pi\sigma/\omega < 0$ , as is clear from the relation  $\epsilon = \epsilon' + i\epsilon'' = (n + i\kappa)^2$ .

but the angle between  $\mathbf{u}$  and  $\mathbf{k}$  can be arbitrary. The quantity  $\hat{n}^2$  can be complex in this case also for a non-absorbing medium, and the sign of  $n\kappa$  can be arbitrary (see Sec. 3). In the case of complex  $\hat{n}^2$ , the field decreases on penetrating the medium not monotonically (as in the case when  $\hat{n} = -\kappa^2$  is real), but in oscillatory fashion; therefore the time-averaged Poynting vector is  $\bar{\mathbf{S}}_0 \neq 0$ . On the other hand, if total reflection takes place (no absorption, but the wave strikes an extensive non-transparent medium with  $\kappa = 0$ ), then the time-averaged total energy flux within the medium,  $\bar{\mathbf{S}} = \bar{\mathbf{S}}_0 + \bar{\mathbf{S}}_1$ , of course vanishes. Since  $\mathbf{S}_1 = 0$  in the absence of spatial dispersion, the equality  $\bar{\mathbf{S}}_0 = 0$  must be satisfied in this case (normal incidence on the medium) for total reflection. It is therefore clear why in the absence of spatial dispersion the field, under total reflection conditions, cannot decrease with depth in a non-absorbing medium in oscillatory fashion (i.e., with index  $n \neq 0$ ).

Let us discuss in conclusion the effect of spatial dispersion on the scalar product of the inductions  $\mathbf{D}'$  for different homogeneous normal waves propagating in a given direction  $\mathbf{s}$ .

From (1.20), recognizing also that  $\mathbf{s} \cdot \mathbf{D} = 0$ , we obtain for any two normal waves with given  $\mathbf{s}$  (waves  $\mathbf{D}'_1, \mathbf{E}_1, \hat{n}_1$ , and  $\mathbf{D}'_2, \mathbf{E}_2, \hat{n}_2$ )

$$\mathbf{D}'_1 \mathbf{D}'_2 = \hat{n}_1^2 \mathbf{E}_1 \mathbf{D}'_2 = \hat{n}_2^2 \mathbf{E}_2 \mathbf{D}'_1. \quad (1.55)$$

Consequently for different  $\hat{n}_1$  and  $\hat{n}_2$ , taking the symmetry property (1.10) into account, we have

$$\mathbf{D}'_1 \mathbf{D}'_2 \left( \frac{1}{\hat{n}_1^2} - \frac{1}{\hat{n}_2^2} \right) = \left\{ \varepsilon_{ij} \left( \omega_2, \frac{\omega_2}{c} \hat{n}_2 \mathbf{s} \right) - \varepsilon_{ij} \left( \omega_1, -\frac{\omega_1}{c} \hat{n}_1 \mathbf{s} \right) \right\} E_{1i} E_{2j}. \quad (1.56)$$

In the absence of spatial dispersion and when  $\omega_1 = \omega_2$ , we conclude from (1.56) that the solutions 1 and 2 are orthogonal (i.e., under these conditions  $\mathbf{D}'_1 \cdot \mathbf{D}'_2 = 0$ ). In the presence of spatial dispersion this orthogonality property does not obtain, generally speaking, even for identical  $\omega_1$  and  $\omega_2$ , since  $\hat{n}_1^2 \neq \hat{n}_2^2$ . In an isotropic medium or in propagation along an optical axis, when  $\hat{n}_1 = \hat{n}_2$ , degeneracy sets in, and the vectors  $\mathbf{D}'_1$  and  $\mathbf{D}'_2$  in the normal waves can be chosen orthogonal. In addition, in the case of gyrotropic media [see (2.26) below], it can be readily seen that  $\mathbf{D}'_1$  and  $\mathbf{D}'_2$  remain orthogonal when  $\hat{n}_1 \neq \hat{n}_2$ , in spite of the presence of gyrotropy—spatial dispersion of first order with respect to  $\mathbf{k}$ .

In analogy with the derivation of (1.56), we readily obtain the equation

$$\mathbf{D}'_1 \mathbf{D}'_2^* \left( \frac{1}{\hat{n}_1^2} - \frac{1}{(\hat{n}_2^2)^*} \right) = \left[ \varepsilon_{ij}^* \left( \omega_2, \frac{\omega_2}{c} \hat{n}_2 \mathbf{s} \right) - \varepsilon_{ji} \left( \omega_1, \frac{\omega_1}{c} \hat{n}_1 \mathbf{s} \right) \right] E_{1i} E_{2j}^*. \quad (1.56a)$$

When  $\omega_1 = \omega_2$ , in the absence of spatial dispersion, and when absorption is neglected the tensor  $\varepsilon_{ij}$  is Hermitian, and  $\hat{n}^2$  is real. Under these conditions and when  $\hat{n}_1^2 \neq \hat{n}_2^2$ , obviously  $\mathbf{D}'_1 \mathbf{D}'_2^* = 0$ . If we disregard spatial

dispersion, then even for a transparent medium (index  $\hat{n}$  real, tensor  $\varepsilon_{ij}$  Hermitian) the condition  $\mathbf{D}'_1 \mathbf{D}'_2^* = 0$  is violated even when  $\omega_1 = \omega_2$  but  $\hat{n}_1^2 \neq \hat{n}_2^2$ . We note, finally, that in the analysis of wave propagation in a medium use is made occasionally of the tensor

$$\varepsilon_{\perp,ij}^{-1} = \eta_{il} \varepsilon_{lm}^{-1} \eta_{mj}, \quad (1.57)$$

where  $\eta_{ij}$  is the projection tensor

$$\eta_{ij} = \delta_{ij} - s_i s_j. \quad (1.58)$$

By virtue of (1.20) and  $\mathbf{s} \cdot \mathbf{D}' = 0$ , obviously,

$$\frac{1}{\hat{n}_2^2} \mathbf{D}'_2 = \eta_{il} E_l = \eta_{il} \varepsilon_{ij}^{-1} \mathbf{D}'_j = \eta_{il} \varepsilon_{lm}^{-1} \eta_{mj} \mathbf{D}'_j = \varepsilon_{\perp,ij}^{-1} \mathbf{D}'_j. \quad (1.59)$$

The symmetry and Hermitian properties of the tensor  $\varepsilon_{ij}^{-1}$  extend also to the tensor  $\varepsilon_{\perp,ij}^{-1}$ , since the tensor  $\eta_{ij}$  is real and symmetrical. By way of an example of the use of the tensors  $\varepsilon_{\perp,ij}^{-1}$  and  $\eta_{ij}$  let us write the condition (1.39) for the existence of "fictitious" longitudinal waves in a different form. Namely, the two relations  $\mathbf{E}_1 = \varepsilon_{ij}^{-1} \mathbf{D}'_j$  and  $s_i \mathbf{D}'_i = 0$  can be combined in

one equation  $\mathbf{E}_1 = \varepsilon_{ij}^{-1} \eta_{lj} \mathbf{D}'_j$ , since  $s_i \eta_{ij} \mathbf{D}'_j = 0$  automatically. It is clear, therefore, that when  $\mathbf{D}' \neq 0$  the vector  $\mathbf{E}$  is non-vanishing and longitudinal (i.e.,  $\mathbf{E} = \mathbf{E} \mathbf{s}$  or  $\eta_{ij} \mathbf{E}_j = 0$ ) only if the determinant

$$|\eta_{il} \varepsilon_{lm}^{-1} \eta_{mj}| = |\varepsilon_{\perp,ij}^{-1}| \quad (1.60)$$

vanishes. The tensor  $\varepsilon_{\perp,ij}$ , which is the inverse of  $\varepsilon_{\perp,ij}^{-1}$ , is also sometimes introduced.<sup>[25]</sup> Since  $\mathbf{E}_l$

$= \varepsilon_{lj}^{-1} \mathbf{D}'_j = \varepsilon_{lm}^{-1} \eta_{mj} \mathbf{D}'_j$ , we obviously have

$$\eta_{il} E_l = \varepsilon_{\perp,ij}^{-1} \mathbf{D}'_j$$

and

$$\begin{aligned} D_i &= \varepsilon_{\perp,ij} \eta_{jl} E_l = \varepsilon_{\perp,ij} E_{\perp,lj}, \\ \varepsilon_{\perp,im} \varepsilon_{\perp,mj}^{-1} &= \delta_{ij}, \quad E_{\perp,li} = \eta_{lj} E_j. \end{aligned} \quad (1.61)$$

Thus, the tensor  $\varepsilon_{\perp,ij}$  enables us to express the induction  $\mathbf{D}'$  (if  $\mathbf{s} \cdot \mathbf{D}' = 0$ ) in terms of the transverse electric field  $\mathbf{E}_{\perp}$ . The tensor  $\varepsilon_{\perp,ij}$  does not exist if (1.60) holds true. This is readily understandable, for if (1.60) holds then  $\mathbf{D}' \neq 0$  for longitudinal  $\mathbf{E}$ , i.e., when  $\mathbf{E}_{\perp} = 0$ .

Generally speaking, the tensor  $\varepsilon_{\perp,ij}$  is not an analytic function of  $\mathbf{k}$  as  $\mathbf{k} \rightarrow 0$ , even if the tensor  $\varepsilon_{ij}$  is analytic (the latter does hold true over a wide range; see Sec. 2b). In addition, knowing  $\varepsilon_{\perp,ij}$ , it is not always possible to determine  $\varepsilon_{ij}$ . Even for these reasons alone it is already clear that the tensors  $\varepsilon_{\perp,ij}$  and  $\varepsilon_{\perp,ij}^{-1}$  can at best only be of ancillary value, although they can prove to be convenient for writing down certain expressions. Indeed, in the case of non-longitudinal waves the expression for  $\hat{n}^2$  is expressed quite simply in terms of  $\varepsilon_{\perp,ij}^{-1}$  and  $\varepsilon_{\perp,ij}$ . Thus, according to (1.59), the dispersion equation has the form

$$\left| \frac{\delta_{ij}}{\hat{n}^2} - \varepsilon_{\perp,ij}^{-1} \right| = 0. \quad (1.62)$$

Further, from (1.20) and (1.61) it is clear that

$$\epsilon_{\perp,ij} E_{\perp,j} = \hat{n}^2 E_{\perp,i}, \quad (1.63)$$

and the dispersion equation is written in the form

$$|\epsilon_{\perp,\alpha\beta} - \hat{n}^2 \delta_{\alpha\beta}| = \hat{n}^4 - \epsilon_{\perp,\alpha\alpha} \hat{n}^2 + |\epsilon_{\perp,\alpha\beta}| = 0. \quad (1.64)$$

Here  $\alpha, \beta = 1, 2$  and the  $z$  axis is in the direction of  $\mathbf{k}$ ; in this system

$$\eta_{ij} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

$$\epsilon_{\perp,\alpha\alpha} = \epsilon_{\perp,11} + \epsilon_{\perp,22}; |\epsilon_{\perp,\alpha\beta}| = \epsilon_{\perp,11} \epsilon_{\perp,22} - \epsilon_{\perp,12} \epsilon_{\perp,21}$$

and in the absence of spatial dispersion

$$n_{1,2}^2 = \frac{\epsilon_{\perp,11} + \epsilon_{\perp,22}}{2} \pm \frac{1}{2} \sqrt{(\epsilon_{\perp,11} - \epsilon_{\perp,22})^2 + 4\epsilon_{\perp,12}\epsilon_{\perp,21}}. \quad (1.65)$$

In place of the tensor  $\epsilon_{\perp,ij}$  we can equally well use the tensor

$$\epsilon_{ij} = \epsilon_{\perp,ij} + s_i s_j,$$

since

$$D'_i = \epsilon_{\perp,ij} E_{\perp,j} = \epsilon_{ij} E_{\perp,j}.$$

We can show that

$$\epsilon_{\perp,ij} = 2(\delta_{ij} - s_i s_j) - \epsilon_{ij} + \frac{\epsilon_{il} \epsilon_{mj} s_l s_m}{\epsilon_{rl} s_r s_l}. \quad (1.66)$$

In the microscopic theory, if only the transverse field is used as the perturbation, we obtain none other than the tensor  $\epsilon_{\perp,ij}$  (see Sec. 4b).

## 2. THE TENSOR $\epsilon_{ij}(\omega, \mathbf{k})$ IN CRYSTALS

a) Introduction of the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  for the case of crystals. Crystals are not spatially homogeneous, since, for example, the lattice sites are not equivalent to other points. Therefore the use of the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$ , introduced for a homogeneous medium, has certainly only limited application when it comes to crystals. Before we proceed to the discussion of crystals, we rewrite in different form the equation (1.3), which pertains to an arbitrary medium (the properties of the medium being independent of the time). We put

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_j(\mathbf{r}, \omega) e^{-i\omega t},$$

or

$$\mathbf{E}(\mathbf{r}, t) = \int \mathbf{E}_j(\mathbf{k}', \omega) e^{i(\mathbf{k}'\mathbf{r} - \omega t)} d\mathbf{k}',$$

and use an analogous notation for  $\mathbf{D}'$ . Substituting these expressions in (1.3), we obtain

$$\left. \begin{aligned} D'_i(\mathbf{r}, \omega) &= \int \epsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}') E_j(\mathbf{r}', \omega) d\mathbf{r}', \\ \epsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}') &= \int \hat{\epsilon}_{ij}(\tau, \mathbf{r}, \mathbf{r}') e^{i\omega\tau} d\tau, \\ D'_i(\mathbf{k}, \omega) &= \frac{1}{(2\pi)^3} \int D'_i(\mathbf{r}, \omega) e^{-i\mathbf{k}\mathbf{r}} d\mathbf{r} \\ &= \int \epsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}') E_j(\mathbf{k}', \omega) d\mathbf{k}', \\ \epsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}') &= \frac{1}{(2\pi)^3} \int \epsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}') e^{i(-\mathbf{k}\mathbf{r} + \mathbf{k}'\mathbf{r}')} d\mathbf{r} d\mathbf{r}'. \end{aligned} \right\} \quad (2.1)$$

For an unbounded homogeneous medium

$$\hat{\epsilon}'_{ij}(\tau, \mathbf{r}, \mathbf{r}') = \epsilon_{ij}(\tau, \mathbf{r} - \mathbf{r}')$$

and since

$$\frac{1}{(2\pi)^3} \int e^{i(\mathbf{k} - \mathbf{k}')\mathbf{r}'} d\mathbf{r}' = \delta(\mathbf{k} - \mathbf{k}'),$$

we see that

$$\epsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}') = \epsilon_{ij}(\omega, \mathbf{k}) \delta(\mathbf{k} - \mathbf{k}').$$

We thus arrive at Eq. (1.5) for a homogeneous medium, as we should.

The crystal is an inhomogeneous medium whose properties remain unchanged under a displacement equal to any lattice vector  $\mathbf{a}$  (translational symmetry). In the crystal, therefore,

$$\epsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}') = \epsilon_{ij}(\omega, \mathbf{r} + \mathbf{a}, \mathbf{r}' + \mathbf{a}). \quad (2.2)$$

A function possessing the property (2.2) can be written in the form

$$\epsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}') = \sum_{\mathbf{b}} g_{\mathbf{b}}(\omega, \mathbf{r} - \mathbf{r}') e^{-i2\pi\mathbf{b}\mathbf{r}'},$$

where

$$\mathbf{b} = n_1 \mathbf{b}_1 + n_2 \mathbf{b}_2 + n_3 \mathbf{b}_3$$

is an arbitrary reciprocal-lattice vector ( $n_j$  are integers and  $\mathbf{b}_j$  the three fundamental reciprocal-lattice vectors, i.e.,  $\exp(i2\pi\mathbf{a} \cdot \mathbf{b}) = 1$ ). Substituting such an expression for  $\epsilon_{ij}(\omega, \mathbf{r}, \mathbf{r}')$  in (2.1), we obtain

$$\epsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}') = \sum_{\mathbf{b}} \epsilon_{ij}^{\mathbf{b}}(\omega, \mathbf{k}) \delta(\mathbf{k}' - \mathbf{k} - 2\pi\mathbf{b}), \quad (2.3)$$

where

$$\epsilon_{ij}^{\mathbf{b}}(\omega, \mathbf{k}) = \int g_{\mathbf{b}}(\omega, \mathbf{R}) e^{-i\mathbf{k}\mathbf{R}} d\mathbf{R}$$

and obviously, if we include only the term with  $\mathbf{b} = 0$ , we get

$$\epsilon_{ij}(\omega, \mathbf{k}, \mathbf{k}') = \epsilon_{ij}^{\mathbf{b}=0}(\omega, \mathbf{k}) \delta(\mathbf{k}' - \mathbf{k})$$

and

$$D'_i(\mathbf{k}, \omega) = \epsilon_{ij}^{\mathbf{b}=0}(\omega, \mathbf{k}) E_j(\mathbf{k}, \omega).$$

Thus, the tensor  $\epsilon_{ij}^{\mathbf{b}=0}(\omega, \mathbf{k})$  corresponds to the dielectric tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  and the latter can be used in the electrodynamics of crystals whenever it is sufficient to retain one term with  $\mathbf{b} = 0$  in the right half of (2.3), or when all the remaining terms can be expressed by means of this first term.

According to (2.1) and (2.3), the electric field  $\mathbf{E}$  and the induction  $\mathbf{D}'$  are related by the equation

$$D'_i(\mathbf{k}, \omega) = \sum_{\mathbf{b}} \epsilon_{ij}^{\mathbf{b}}(\omega, \mathbf{k}) E_j(\mathbf{k} + 2\pi\mathbf{b}, \omega), \quad (2.4)$$

which enables us to write the wave equation (1.15) in the form

$$\frac{\omega^2}{c^2} \sum_{\mathbf{b}} \epsilon_{ij}^{\mathbf{b}}(\omega, \mathbf{k}) E_j(\mathbf{k} + 2\pi\mathbf{b}, \omega) - k^2 E_i(\mathbf{k}, \omega) + k_i k_j E_j(\mathbf{k}, \omega) = 0. \quad (2.5)$$

The determinant  $\Delta(\omega, \mathbf{k})$  of the system (2.5) determines the dispersion equation

$$\Delta(\omega, \mathbf{k}) = 0, \quad (2.6)$$

the roots of which (generally speaking, complex) are

$$\omega_l = \omega_l(\mathbf{k}), \quad l = 1, 2, 3, \dots \quad (2.7)$$

and correspond to different branches (modes) and zones of the "normal" frequencies of the electromagnetic field in the crystal.

In optics the interest lies in the region of relatively long waves ( $k \ll b \sim 1/a$ ) and relatively low frequencies ( $\omega \ll cb \sim c/a$ ). Let us assume that in this frequency region some Fourier component of the electric field  $E_j(\mathbf{k}, \omega)$ , corresponding to small  $k$ , is considerably greater than all the other Fourier components in (2.4) and (2.5) corresponding to  $\mathbf{b} \neq 0$ .

From (2.5) we can readily establish by successive approximations that such an assumption is generally speaking confirmed, and the field components with  $\mathbf{b} \neq 0$  turn out to be smaller than  $E_j(\mathbf{k}, \omega)$  by a factor  $(\omega/2cb)^2 \epsilon^{-b}(\omega, 2\pi\mathbf{b}) \lesssim (a/\lambda_0)^2$  (one can think that usually  $\epsilon^{-b}(\omega, 2\pi\mathbf{b}) \lesssim 1$ ).<sup>\*</sup> Therefore, if we eliminate

<sup>\*</sup>We denote the running wave vector by  $\mathbf{k}'$  [the symbol  $\mathbf{k}$  was used in (2.5)] and put  $\mathbf{k}' = 2\pi\mathbf{b} + \mathbf{k}$ ,  $|\mathbf{k}| \ll |2\pi\mathbf{b}|$ . We then have in first approximation

$$\left(\frac{\omega}{2\pi c}\right)^2 \epsilon_{ij}^{-b}(\omega, 2\pi\mathbf{b}) E_j(\mathbf{k}, \omega) - b^2 E_i(2\pi\mathbf{b}, \omega) + b_i b_j E_j(2\pi\mathbf{b}, \omega) = 0.$$

From this we get precisely  $E(2\pi\mathbf{b}, \omega) \sim \left(\frac{\omega}{2\pi cb}\right)^2 \epsilon_{ij}^{-b}(\omega, 2\pi\mathbf{b}) E(\mathbf{k}, \omega)$ ,

if we disregard the possibility of occurrence of a small coefficient in the denominator, owing to the smallness of the determinant  $|\delta_{ij} - b_i b_j/b^2|$ . Even if the crystal is under the influence of a homogeneous field  $E(0, \omega)$ , the induced current and  $\mathbf{D}'$  will in general be deeply modulated, in view of the influence of the lattice sites with periods  $\sim a$ , which corresponds to wave vectors  $\sim 1/a \sim b$ . This is why we assume  $\epsilon_{ij}^{-b}(\omega, 2\pi\mathbf{b}) \lesssim 1$ . It must be noted in addition that both the last assumption and specially the use of the estimate  $|\delta_{ij} - b_i b_j/b^2| \sim 1$  are worthy of a more detailed analysis. For this purpose it would be useful to carry out the corresponding calculations for some simple model.

Under the assumptions made above we have for small  $k \ll 2\pi b$   $E(\mathbf{k} + 2\pi\mathbf{b}, \omega) \approx E(2\pi\mathbf{b}, \omega) \sim \left(\frac{a}{\lambda_0}\right)^2 E(\mathbf{k}, \omega)$  and the first term in (2.5) has the form  $\frac{\omega^2}{c^2} \left\{ \epsilon_{ij}^{b=0}(\omega, \mathbf{k}) \times E_j(\mathbf{k}, \omega) + \text{terms of the order of } \epsilon^{b \neq 0}(\omega, \mathbf{k}) \left(\frac{a}{\lambda_0}\right)^2 E(\mathbf{k}, \omega) \right\}$ .

At the same time, in accord with (2.4), we have accurate to small terms

$$D'_i(2\pi\mathbf{b}, \omega) = \epsilon_{ij}^{-b}(\omega, 2\pi\mathbf{b}) E_j(\mathbf{k}, \omega) \quad \text{and} \quad D'_i(\mathbf{k}, \omega) = \epsilon_{ij}^{b=0}(\omega, \mathbf{k}) E_j(\mathbf{k}, \omega).$$

From this it is clear that in order of magnitude

$$\frac{D'(2\pi\mathbf{b}, \omega)}{D'(\mathbf{k}, \omega)} \sim \frac{\epsilon^{-b}(\omega, 2\pi\mathbf{b})}{\epsilon^{b=0}(\omega, \mathbf{k})}$$

and thus this relation is far from small when  $\epsilon^{-b} \sim 1$  and  $\epsilon^{b=0} \lesssim 10$ . In other words, if the spatial modulation is deep, by virtue of which  $\epsilon^{-b}(\omega, 2\pi\mathbf{b}) \sim 1$ , the smallness of the amplitudes  $E(2\pi\mathbf{b}, \omega)$  is not at all simultaneously connected with smallness of the amplitudes  $D'(2\pi\mathbf{b}, \omega)$ . For the latter reason the wave equation for  $\mathbf{D}'$  can, generally speaking, not be used [similar to Eq. (2.5) for  $\mathbf{E}$ ] to obtain equations of the type (2.8) by successive approximations.

in the corresponding approximation all terms with  $\mathbf{b} \neq 0$  from (2.5), we obtain for  $E(\mathbf{k}, \omega)$  an equation that coincides with (1.16):

$$\frac{\omega^2}{c^2} \epsilon_{ij}(\omega, \mathbf{k}) E_j - k^2 E_i + k_i k_j E_j = 0. \quad (2.8)$$

Here  $\epsilon_{ij}(\omega, \mathbf{k})$  differs from  $\epsilon_{ij}^{b=0}(\omega, \mathbf{k})$  by terms of order  $(a/\lambda_0)^2$ . These additional terms, in addition, depend weakly on  $\mathbf{k}$ , provided  $k/2\pi b \sim ak = 2\pi an/\lambda_0 \ll 1$ ; consequently in first approximation  $\epsilon_{ij}^b(\omega, \mathbf{k}) \approx \epsilon_{ij}^b(\omega, 0)$ . In other words, in the long-wave region it is necessary to include the additional terms if frequency dispersion is considered with accuracy to  $\sim (a/\lambda_0)^2$ . What is of interest, on the other hand, are effects of order  $an/\lambda_0$  (natural optical activity), effects of order  $(an/\lambda_0)^2 = (a/\lambda)^2$  when  $n \gg 1$  (longitudinal waves, waves near the absorption lines), and terms proportional to  $(a/\lambda_0)^2$ , but dependent on the direction (the optical anisotropy of cubic crystals with  $n \sim 1$ ). As a result we can, generally speaking, put  $\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}^{b=0}(\omega, \mathbf{k})$  in (2.8) for the long-wave region of interest to us.

Equation (2.8) leads to the dispersion equation (1.17), with limitation to rather low frequencies ( $\omega \ll c/a \sim 10^{18} \text{ sec}^{-1}$ ) and long waves ( $\lambda_0 \gg a \sim 3 \times 10^{-8} \text{ cm}$ ), which does correspond to the optical region; the harmonics of the electric field, corresponding to short waves, are small and the principal part of the field is given by (2.8) with a certain tensor  $\epsilon_{ij}(\omega, \mathbf{k})$ . By the same token, it is possible to employ this tensor in crystal optics in the same way as in a homogeneous medium.<sup>[8]</sup> This was indeed the past procedure, but a more precise statement of the conditions and of the meaning of the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  as applied to crystals is essential. It is sufficient to point out that when the tensor  $\epsilon_{ij}(\omega, \mathbf{k} \neq 0)$  is used, the ratio  $a/\lambda$  is assumed non-vanishing, while on the other hand, the medium (crystal) is considered spatially-homogeneous. The validity of such a procedure is not obvious a priori.

The situation involved in the formalism employed was described above. Physically, everything reduces to having the lattice constant  $a$  play a double role. On the one hand, the parameter  $a$  characterizes the spatial inhomogeneity of the medium, an inhomogeneity which is neglected on going over to  $\epsilon_{ij}(\omega, \mathbf{k})$ . On the other hand, the same parameter  $a$  describes the "molecular action radius"  $l$  — the interval of values of  $R$ , for which the kernel  $\epsilon_{ij}(\omega, \mathbf{r}, \mathbf{r} + \mathbf{R})$  in (2.1) differs appreciably from zero. Letting the radius  $l$  approach zero corresponds to neglecting spatial dispersion, as was already noted above. If we introduce from the very outset two formally independent parameters  $a$  (the lattice constant) and  $l$  (the "molecular action radius"), the picture may become clearer. Actually, however, usually  $a \sim l$  in the crystal, and for complicated crystals with anomalously large lattice parameter,  $a \gg l$ . The inequality  $a \ll l$  (or, formally, the tendency of  $a$  to zero with  $l \neq 0$ ) is apparently impossible for a real crystal.



b) Case of weak spatial dispersion ( $a/\lambda \ll 1$ ). The introduction of the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  and its use in the wave equation (2.8) for a crystal lattice is, generally speaking, possible if

$$\left(\frac{a}{\lambda_0}\right)^2 \ll 1 \quad (2.9)$$

(we disregard for the time being some additional aspects pointed out in Sec. 2a).

In optics the condition (2.9) can be assumed to be satisfied at all times, at the same time, however, it still does not indicate that the spatial dispersion is small. The point is, as was already emphasized, that spatial dispersion is characterized by a parameter  $a/\lambda = an/\lambda_0$ . It is obvious that at large values of the refractive index  $n$  this parameter  $an/\lambda_0$ , and consequently also the spatial dispersion, can be appreciable even if the inequality (2.9) holds true. Under such conditions  $\epsilon_{ij}(\omega, \mathbf{k})$  can turn out to be a rather complicated function of  $\mathbf{k}$ , and if we imagine  $\epsilon_{ij}(\omega, \mathbf{k})$  to be expanded in powers of  $\mathbf{k}$ , this series will contain many terms (the expansion parameter is precisely the quantity  $an/\lambda_0$ ). The wave equation (2.8) can in this case have many solutions, i.e., the dispersion equation (1.17) will have many roots  $\omega_l(\mathbf{k})$ . On the other hand, if spatial dispersion is neglected, the dispersion equation has only two roots corresponding to the ordinary and extraordinary waves, and under certain conditions an additional root  $\omega_{||} = \text{const}$  for the longitudinal wave.

In connection with the foregoing, a very important fact is that in crystal optics the spatial dispersion is weak, i.e., the inequality mentioned in the introduction applies

$$a/\lambda = an/\lambda_0 \ll 1. \quad (2.10)$$

To satisfy this inequality we can put in practice  $\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}^{p=0}(\omega, \mathbf{k})$ . This is an important factor from the point of view of calculating  $\epsilon_{ij}(\omega, \mathbf{k})$  from the microscopic theory. Since such calculations have only limited significance, an incomparably more important factor is the fact that condition (2.10) allows us to expand the tensors  $\epsilon_{ij}(\omega, \mathbf{k})$  or  $\epsilon_{ij}^{-1}(\omega, \mathbf{k})$  in powers of  $\mathbf{k}$  of which only the first two or three terms need be retained.<sup>[5]</sup> We thus use the expansion

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}(\omega) + i\gamma_{ijl}(\omega)k_l + \alpha_{ijlm}(\omega)k_l k_m, \quad (2.11)$$

or, for homogeneous plane waves, when  $\mathbf{k} = \omega \hat{\mathbf{n}}/c$ :

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}(\omega) + i\gamma_{ijl}(\omega) \frac{\omega}{c} \hat{n}_l + \alpha_{ijlm}(\omega) \left(\frac{\omega}{c}\right)^2 \hat{n}_l \hat{n}_m. \quad (2.12)$$

Analogously, for the inverse tensor

$$\epsilon_{ij}^{-1}(\omega, \mathbf{k}) = \epsilon_{ij}^{-1}(\omega) + i\delta_{ijl}(\omega)k_l + \beta_{ijlm}(\omega)k_l k_m. \quad (2.13)$$

The use of the tensor  $\epsilon_{ij}$  and  $\epsilon_{ij}^{-1}$ , whether in general or in the form (2.11)–(2.13), is equivalent over a wide range and is dictated by considerations of convenience.\*

\*In spite of the fact that the tensor  $\epsilon_{ij}$  is used more frequently, the advantages gained by using the tensor  $\epsilon_{ij}^{-1}$  in certain cases have been noticed long ago (see, for example,<sup>[21,22]</sup>).

Exceptions are cases in which several components of the tensors  $\epsilon_{ij}(\omega)$  or  $\epsilon_{ij}^{-1}(\omega)$  tend to infinity (increase strongly). For example, if some component  $\epsilon_{ij}(\omega)$  tends to infinity, then the expansions (2.11)–(2.12) are insufficient for the corresponding component of  $\epsilon_{ij}(\omega, \mathbf{k})$ , since all the terms are vanishingly small compared with  $\epsilon_{ij}(\omega)$ . At the same time, an increase in  $\epsilon_{ij}$  usually brings about an increase in  $\hat{\mathbf{n}}$ , i.e., an increase in the role of the spatial dispersion. In such a case we can use expansion (2.13), which is particularly effective when  $\epsilon_{ij}^{-1}(\omega)$  decreases. Analogously, in the region where  $\epsilon_{ij}^{-1}(\omega)$  increases strongly, we must use the expansion (2.11)–(2.12), and not (2.13). We shall henceforth make use of the expansions (2.11)–(2.13).

Let us now discuss the conditions under which they can be used, apart from the initial requirement (2.10). Expressions (2.11)–(2.13) are meaningful only if the functions  $\epsilon_{ij}(\omega, \mathbf{k})$  and  $\epsilon_{ij}^{-1}(\omega, \mathbf{k})$  can in general be expanded near the point  $\mathbf{k} = 0$ . In this connection we recall that as  $\mathbf{k} \rightarrow 0$  the functions  $\epsilon_{ij}(\omega, \mathbf{k})$  and  $\epsilon_{ij}^{-1}(\omega, \mathbf{k})$  tend respectively to  $\epsilon_{ij}(\omega)$  and  $\epsilon_{ij}^{-1}(\omega)$  and are independent of both the modulus and the direction of  $\mathbf{k}$ . Further,  $\epsilon_{ij}(\omega, \mathbf{k})$  and  $\epsilon_{ij}^{-1}(\omega, \mathbf{k})$  are integral quantities, obtained by spectral summation (see Sec. 4). Therefore, even if the integrand has a few sufficiently weak singularities (for example, at the natural frequencies of the unperturbed problem), the components of  $\epsilon_{ij}$  will have no singularities. In addition, as already mentioned in the introduction and as will be pointed out in Sec. 4, in the cases known to us the natural frequencies of the correctly chosen unperturbed problem have no singularities near  $\mathbf{k} = 0$ . Thus, there are no grounds whatever for doubting that the functions  $\epsilon_{ij}$  and  $\epsilon_{ij}^{-1}$  have no essential singularities when  $\mathbf{k} \rightarrow 0$ .

Expansions of the type (2.11)–(2.13) using only a few terms may not be sufficient in the following unique situation. Let, for example,  $\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon(\omega, \mathbf{k}) \delta_{ij}$ , with

$$\epsilon(\omega, \mathbf{k}) = \epsilon(\omega) + \frac{\nu k^2}{\frac{\omega - \omega_l}{\omega_l} - \mu k^2}; \quad (2.14)$$

Under certain conditions this expression describes the variation of  $\epsilon(\omega, \mathbf{k})$  near a quadrupole absorption line. So long as the term  $\mu k^2$  is insignificant, we deal here with an expansion such as (2.11). In the opposite case, however,

$$[\epsilon(\omega, \mathbf{k}) - \epsilon(\omega)]^{-1} = \frac{\frac{\omega - \omega_l}{\omega_l} - \mu k^2}{\nu k^2},$$

which corresponds to neither (2.11) nor (2.13). It is easy to write an expression that generalizes formula (2.14) to include any crystal in the sense of the phenomenological expansion (2.11)

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}(\omega) + i\gamma_{ijl}(\omega)k_l + \alpha_{ijlm}(\omega, \mathbf{k})k_l k_m,$$

$$\epsilon_{ij}^{-1}(\omega, \mathbf{k}) = \xi_{ijlm}(\omega) + i\eta_{ijl}(\omega)k_l + \zeta_{ijkl}(\omega)k_l k_m. \quad (2.15)$$

Similarly we can replace  $\gamma_{ijl}(\omega)$  in (2.11) by  $\gamma_{ijl}(\omega, \mathbf{k})$ , with

$$\gamma_{ijl}^{-1}(\omega, \mathbf{k}) = \gamma_{ijl}^{-1}(\omega) + i\mu_{ijlm}(\omega)k_m + \nu_{ijlmn}(\omega)k_l k_n.$$

For a non-gyrotropic cubic crystal formula (2.15) is equivalent to (2.14), with

$$\alpha_{ijlm}k_l k_m = \delta_{ij} \frac{\nu(\omega)k^2}{\omega - \omega_l - \mu k^2}.$$

In a more complicated case the use of the combined expansion (2.15) leads to cumbersome expressions, which can hardly be of practical interest. It is more important, however, that formulas such as (2.14) and (2.15) need be used only in exceptional cases. In fact, the expansions (2.11)–(2.15) are in the parameter  $a/\lambda$ , i.e., the coefficients  $\gamma$  and  $\delta$  are of order  $a$  while the coefficients  $\alpha$ ,  $\beta$ ,  $\nu$ , and  $\mu$  are of order  $a^2$ . Therefore, as is clear from (2.14), the term  $\mu k^2$  in the denominator need be taken into account only when  $|\omega - \omega_l|/\omega_l \sim \mu k^2 \sim (2\pi a/\lambda)^2$ , while at the same time

$$\frac{\nu k^2}{\omega - \omega_l} \sim \mu k^2 \sim \varepsilon(\omega) \sim 1.$$

The latter means that the quadrupole line makes a contribution to  $\varepsilon$ , comparable with the contribution of the dipole lines. The corresponding value of the ratio  $(\omega - \omega_l)/\omega_l$  is on the order of  $10^{-5}$ – $10^{-6}$ , which is equivalent to approaching the center of the line within  $\Delta\lambda \sim 10^{-2} \text{ \AA}$  in the case of weak absorption. Even when such conditions are realized, the term  $\mu k^2$  in (2.14) or the dependence of  $\alpha_{ijlm}$  on  $\mathbf{k}$  in (2.15) still need to be taken into account in a very narrow region near the center of the line. Outside these regions, expansions (2.11)–(2.13) are more convenient, and the entire "quadrupole effect" finds its reflection in the terms  $\alpha_{ijlm}k_l k_m$  or  $\beta_{ijlm}k_l k_m$ , which thus play the principal role. Consequently, a changeover to expressions of the type of (2.14) and (2.15) corresponds in a certain sense to an examination of higher-order effects. The corresponding limitation on the range of validity of the expressions (2.11) and (2.13) is therefore perfectly natural. It is interesting that a similar higher-order effect is encountered in the experiments (see Sec. 3f). Summarizing, we can state that the use of formulas (2.11)–(2.13) in crystal optics with spatial dispersion is a fully consistent method,\* although it does occasionally need some further generalization, such as the use of expressions similar to (2.15).

\*When series such as (2.11)–(2.13) are substituted in the dispersion equations (1.17) and (1.18), as was done in<sup>[5]</sup>, one obtains for the refractive index  $\hat{n}$  algebraic equations whose order increases with the number of terms retained in the series. The new roots  $\hat{n}$ , however, correspond to ever increasing values of  $k$ . Consequently, in the determination of only those first few roots  $\hat{n}$  which can be considered in connection with condition (2.9) and by virtue of the influence of absorption (see<sup>[5]</sup> and Sec. 3b, below), the retention of only the first terms of the series is again fully consistent and justified. Thus, remarks to the contrary, made in<sup>[23]</sup>, appear to us to be incorrect.

We note that some references<sup>[14,24,25]</sup> use expressions for  $\varepsilon_{ij}(\omega, \mathbf{k})$  in crystals that are in some respect more general than (2.11)–(2.13). Thus, the expression used in<sup>[14]</sup> is

$$\varepsilon_{ij}(\omega, \mathbf{k}) = \varepsilon_{ij}^0 + \frac{g_{ij}}{\omega - \omega_l - F(\mathbf{k})}, \quad (2.16)$$

where  $\varepsilon_{ij}^0$ ,  $\omega_l$ , and  $g_{ij}$  are constants. In fact, however, we must put in (2.16)  $F(\mathbf{k}) = f + g_l k_l + h_{lm} k_l k_m$ , since inclusion of terms of higher order in  $\mathbf{k}$  is in general beyond the limits of the initial approximation. Equation (2.16) then reduces to (2.13) and is suitable for use only near the center of the line.\* Formulas suitable for a wide range of frequencies are given in<sup>[24]</sup>, but these extend beyond the limits where expansions (2.11) and (2.13) are valid, and have essentially an extrapolative character. It seems to us that the most correct procedure is not to resort to extrapolation formulas for  $\varepsilon_{ij}(\omega, \mathbf{k})$ , thereby taking explicit account of the weakness of the spatial dispersion. For  $\varepsilon_{ij}(\omega)$ , to the contrary, it is convenient to use extrapolation formulas near the absorption line, for example by putting in the principal-axis frame

$$\varepsilon_j(\omega) = \varepsilon_j' + i\varepsilon_j'' = \varepsilon_{0j} - \frac{\omega_{0j}^2}{\omega^2 - \omega_{0j}^2 - i\nu_j \omega}. \quad (2.17)$$

The tensors contained in (2.11)–(2.13) satisfy certain relations that follow from (1.7) and (1.9)–(1.11). Thus, by virtue of (1.10),

$$\begin{aligned} \varepsilon_{ij}(\omega) &= \varepsilon_{ji}(\omega), \quad \varepsilon_{ij}^{-1}(\omega) = \varepsilon_{ji}^{-1}(\omega), \quad \alpha_{ijlm}(\omega) = \alpha_{jilm}(\omega), \\ \gamma_{ijl}(\omega) &= -\gamma_{jil}(\omega), \quad \delta_{ijl}(\omega) = -\delta_{jil}(\omega), \quad \beta_{ijlm}(\omega) = \beta_{jilm}(\omega). \end{aligned} \quad (2.18)$$

In addition, the tensors  $\alpha_{iklm}$  and  $\beta_{iklm}$  can always be chosen such as to make  $\alpha_{ijlm} = \alpha_{ijml}$  and  $\beta_{ijlm} = \beta_{ijml}$  (we assume here that this choice was indeed made). We note also that the magnetic induction of the external field  $\mathbf{B}_{\text{ext}}$  is assumed equal to zero everywhere except in Sec. 3d. In the presence of a center of symmetry and for a nongyrotropic medium in general, it follows from (1.11) that

$$\gamma_{ijl} = 0, \quad \delta_{ijl} = 0. \quad (2.19)$$

In the absence of absorption, the tensors  $\varepsilon_{ij}(\omega)$ ,  $\varepsilon_{ij}^{-1}(\omega)$ ,  $\gamma_{ijl}$ ,  $\alpha_{ijlm}$ ,  $\delta_{ijl}$ , and  $\beta_{ijlm}$  are all real [see (1.7) and (2.18)].

All tensors simplify considerably for crystals with high symmetry, let alone for an isotropic medium.

In an isotropic nongyrotropic medium the tensor  $\varepsilon_{ij}(\omega, \mathbf{k})$  has the form (1.26). If the spatial dispersion is weak, then [see (1.26)–(1.30) and (2.12)]

\*In<sup>[25]</sup> and in some other articles, the approach used in<sup>[5]</sup> and above in this article, connected with the expansion of  $\varepsilon_{ij}(\omega, \mathbf{k})$  in powers of  $\mathbf{k}$ , is compared to some extent with different calculations that include various elements of microscopic theory. It seems to us that one cannot agree with these comparisons and with many other ideas advanced in the literature. Since the corresponding critical remarks have already been mentioned in<sup>[6]</sup>, we do not deem it necessary to discuss them in the present article.

$$\epsilon_{ij}(\omega, \mathbf{k}) = \epsilon(\omega) \delta_{ij} - \alpha_{\perp}(\omega) \hat{n}^2 (\delta_{ij} - s_i s_j) - \alpha_{\parallel}(\omega) \hat{n}^2 s_i s_j,$$

$$n_{\perp}^2 = \frac{\epsilon(\omega)}{1 + \alpha_{\perp}(\omega)}, \quad n_{\parallel}^2 = \frac{\epsilon(\omega)}{\alpha_{\parallel}(\omega)} \quad (2.20)$$

[the factor  $(\omega/c)^2$ , contained in (2.12), is included in  $\alpha_{\parallel}$  and  $\alpha_{\perp}$ ]. Near the pole  $\epsilon(\omega)$  an expansion of the type (2.13) must be used, and for an isotropic medium

**Table II.** Properties of the tensor  $\epsilon_{ij}(\omega)$  and the pseudo-tensor  $f_{ij}(\omega)$  ( $\epsilon_{ij} = \epsilon_{ji}$ ,  $f_{ij} = f_{ji}$ ,  $\gamma_{ijl} = \epsilon_{ijm} f_{ml}$ )

System	$\epsilon_{ij}$	Principal axes of the tensor	$f_{ij}$
Triclinic	$\begin{pmatrix} \epsilon_{11} & \epsilon_{12} & \epsilon_{13} \\ \epsilon_{12} & \epsilon_{22} & \epsilon_{23} \\ \epsilon_{13} & \epsilon_{23} & \epsilon_{33} \end{pmatrix}$	Not fixed	$f_{ij}=0$ in class $C_i$ $f_{ij} = \begin{pmatrix} f_{11} & f_{12} & f_{13} \\ f_{12} & f_{22} & f_{23} \\ f_{13} & f_{23} & f_{33} \end{pmatrix}$ in class $C_1$
Monoclinic	$\begin{pmatrix} \epsilon_{11} & 0 & \epsilon_{13} \\ 0 & \epsilon_{22} & 0 \\ \epsilon_{13} & 0 & \epsilon_{33} \end{pmatrix}$	y axis directed along 2-fold axis or perpendicular to the symmetry plane.	$f_{ij}=0$ in class $C_{2h}$ $f_{ij} = \begin{pmatrix} f_{11} & 0 & f_{13} \\ 0 & f_{22} & 0 \\ f_{13} & 0 & f_{33} \end{pmatrix}$ in class $C_2$ $f_{ij} = \begin{pmatrix} 0 & f_{12} & 0 \\ f_{12} & 0 & f_{23} \\ 0 & f_{23} & 0 \end{pmatrix}$ in class $C_s$
Rhombic	$\begin{pmatrix} \epsilon_{11} & 0 & 0 \\ 0 & \epsilon_{22} & 0 \\ 0 & 0 & \epsilon_{33} \end{pmatrix}$	Axes x, y, and z directed along 2-fold axes, and in class $C_{2v}$ the x and y axes are perpendicular to the symmetry planes.	$f_{ij}=0$ in class $C_{2h}$ $f_{ij} = \begin{pmatrix} f_{11} & 0 & 0 \\ 0 & f_{22} & 0 \\ 0 & 0 & f_{33} \end{pmatrix}$ in class $D_2$ $f_{ij} = \begin{pmatrix} 0 & f_{12} & 0 \\ f_{12} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$ in class $C_{2v}$
Tetragonal	$\begin{pmatrix} \epsilon_{\perp} & 0 & 0 \\ 0 & \epsilon_{\perp} & 0 \\ 0 & 0 & \epsilon_{\parallel} \end{pmatrix}$	In classes $C_4$ , $S_4$ , and $C_{4h}$ only the z axis (4-fold axis) is fixed. In classes $D_4$ , $C_{4v}$ , $D_{2d}$ , and $D_{4h}$ all axes are fixed.	$f_{ij}=0$ in classes $C_{4h}$ , $C_{4v}$ , and $D_{4h}$ $f_{ij} = \begin{pmatrix} f_{\perp} & 0 & 0 \\ 0 & f_{\perp} & 0 \\ 0 & 0 & f_{\parallel} \end{pmatrix}$ in classes $C_4$ and $D_4$ $f_{ij} = \begin{pmatrix} f_{11} & f_{12} & 0 \\ f_{12} & f_{11} & 0 \\ 0 & 0 & 0 \end{pmatrix}$ in class $S_4$ $f_{ij} = \begin{pmatrix} 0 & f_{12} & 0 \\ f_{12} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$ in class $D_{2d}$
Trigonal	$\begin{pmatrix} \epsilon_{\perp} & 0 & 0 \\ 0 & \epsilon_{\perp} & 0 \\ 0 & 0 & \epsilon_{\parallel} \end{pmatrix}$	In classes $C_3$ and $C_{3i}$ only the z axis is fixed (3-fold axis). In classes $D_3$ , $C_{3v}$ , and $D_{3d}$ all axes are fixed.	$f_{ij}=0$ in classes $C_{3i}$ , $C_{3v}$ , and $D_{3d}$ $f_{ij} = \begin{pmatrix} f_{\perp} & 0 & 0 \\ 0 & f_{\perp} & 0 \\ 0 & 0 & f_{\parallel} \end{pmatrix}$ in classes $C_3$ and $D_3$
Hexagonal	$\begin{pmatrix} \epsilon_{\perp} & 0 & 0 \\ 0 & \epsilon_{\perp} & 0 \\ 0 & 0 & \epsilon_{\parallel} \end{pmatrix}$	In classes $C_6$ , $C_{3h}$ , and $C_{6h}$ only the z axis (6-fold axis) is fixed.	$f_{ij}=0$ in classes $C_{3h}$ , $C_{6h}$ , $C_{6v}$ , $D_{3h}$ , $D_{6h}$ $f_{ij} = \begin{pmatrix} f_{\perp} & 0 & 0 \\ 0 & f_{\perp} & 0 \\ 0 & 0 & f_{\parallel} \end{pmatrix}$ in classes $C_6$ and $D_6$
Cubic	$\begin{pmatrix} \epsilon & 0 & 0 \\ 0 & \epsilon & 0 \\ 0 & 0 & \epsilon \end{pmatrix}$	All axes fixed (x, y and z are 2-fold axes in classes T and $T_h$ and 4-fold axes in classes O, $T_d$ and $O_h$ ).	$f_{ij}=0$ in classes $T_h$ , $T_d$ and $O_h$ $f_{ij} = \begin{pmatrix} f & 0 & 0 \\ 0 & f & 0 \\ 0 & 0 & f \end{pmatrix}$ in classes T and O
Isotropic medium	$\begin{pmatrix} \epsilon & 0 & 0 \\ 0 & \epsilon & 0 \\ 0 & 0 & \epsilon \end{pmatrix}$	Choice of axes arbitrary	$f_{ij}=0$ in the presence of a center of symmetry $f_{ij} = \begin{pmatrix} f & 0 & 0 \\ 0 & f & 0 \\ 0 & 0 & f \end{pmatrix}$ in the absence of a center of symmetry

$$\varepsilon_{ij}^{-1} = \varepsilon^{-1}(\omega) \delta_{ij} + \beta_{\perp}(\omega) \hat{n}^2 (\delta_{ij} - s_i s_j) + \beta_{\parallel}(\omega) \hat{n}^2 s_i s_j. \quad (2.21)$$

By virtue of the condition  $\mathbf{s} \cdot \mathbf{D}' = 0$ , we have obviously  $\mathbf{E} = [1/\varepsilon + \beta_{\perp}(\omega) \hat{n}_{\perp}^2] \mathbf{D}$ , and from (1.20) we obtain

$$\beta_{\perp} n_{\perp}^4 + \frac{\hat{n}_{\perp}^2}{\varepsilon} - 1 = 0, \quad n_{\perp}^2 = -\frac{1}{2\beta_{\perp} \varepsilon} \pm \sqrt{\left(\frac{1}{2\beta_{\perp} \varepsilon}\right)^2 + \frac{1}{\beta_{\perp}}}. \quad (2.22)$$

We shall have more to say about this solution in Sec. 3b. For the longitudinal wave we obtain from (2.21) the condition  $\varepsilon(\omega_{\parallel}) = 0$ . The spatial dispersion for the longitudinal wave is disregarded when the expansion (2.21) is used.

We note that formulas (2.20) and (2.21) correspond to the following relations

$$\begin{aligned} \mathbf{D} &= \varepsilon \mathbf{E} - \alpha_{\perp}(\omega) \frac{c^2}{\omega^2} \text{rot rot } \mathbf{E} + \alpha_{\parallel}(\omega) \frac{c^2}{\omega^2} \text{grad div } \mathbf{E}, \\ \mathbf{E} &= \frac{\mathbf{D}}{\varepsilon} + \beta_{\perp}(\omega) \frac{c^2}{\omega^2} \text{rot rot } \mathbf{D} - \beta_{\parallel}(\omega) \frac{c^2}{\omega^2} \text{grad div } \mathbf{D}. \end{aligned} \quad (2.23)$$

The expressions (2.23) are the most general relations between the two vectors in an isotropic nongyrotropic medium with neglect of all derivatives of order higher than the second.

The conditions connected with the symmetry of the crystals, with respect to the tensors  $\varepsilon_{ij}$  and  $\gamma_{ijkl}$ , are well known (see, for example, [1,22,26]). Nonetheless for the sake of convenience we recall the corresponding results (the symmetry properties for the inverse tensors are obviously the same as for the original tensors and their components will not be written out below).

A symmetrical tensor of the second rank, specifically  $\varepsilon_{ij}(\omega)$ , has at most six independent components. For the corresponding second-order characteristic surface  $\varepsilon_{ij} x_i x_j = 1$  this corresponds to the lengths of three axes and to three parameters (angles) determining the orientations of these axes. The symmetry of the tensor  $\varepsilon_{ij}$  is the same for all crystal classes of a given crystal system. This can be readily verified by determining  $\varepsilon_{ij}$  for the least symmetrical class of each system. In doing this, it is useful to bear in mind the following fact, which is obvious from the properties of second-order surfaces: in a plane perpendicular to the 3-fold and higher-order axes, the section through the surface degenerates into a circle. Therefore, for example, even for the least symmetrical crystal class T in the cubic system the characteristic surface degenerates into a sphere, i.e.,  $\varepsilon_{ij} = \varepsilon \delta_{ij}$  (class-T crystals have four 3-fold axes, corresponding to the space diagonals of a cube).

In the tetragonal, trigonal (rhombohedral) and hexagonal systems we can, by aligning the z axis with the 4-fold, 3-fold, or 6-fold axis respectively, reduce

the tensor  $\varepsilon_{ij}$  to the form  $\begin{pmatrix} \varepsilon_1 & 0 & 0 \\ 0 & \varepsilon_2 & 0 \\ 0 & 0 & \varepsilon_3 \end{pmatrix}$  etc (Table II).\*

\*The tensor simplification resulting from symmetry is discussed in particular detail in [26]. We note that we use for the crystal classes the Schoenflies notation, which is most widely used in the physics literature (the correspondence between this notation and the international notation is given in [26]).

By virtue of (2.18), the tensor  $\gamma_{ijkl}$  (and  $\delta_{ijkl}$ ) has the following properties:

$$\begin{aligned} \gamma_{xx, l} = \gamma_{yy, l} = \gamma_{zz, l} = 0, \quad \gamma_{xy, l} = -\gamma_{yx, l}, \quad \gamma_{yz, l} = -\gamma_{zy, l}, \\ \gamma_{zx, l} = -\gamma_{xz, l} \quad (l = 1, 2, 3 \equiv x, y, z); \end{aligned}$$

The tensors  $\gamma_{ijkl}$  and  $\delta_{ijkl}$  have thus in general nine independent components and can be written in the form

$$\gamma_{ijkl} = e_{ijm} g_{ml}, \quad \delta_{ijkl} = e_{ijm} f_{ml}, \quad (2.24)$$

where  $e_{ijm}$  is a unit pseudotensor of third rank ( $e_{123} = 1$ ,  $e_{213} = -1$ ,  $e_{112} = 0$  etc;  $e_{ijm}$  is unchanged by mirror reflection) and  $g_{ml}$  and  $f_{ml}$  are pseudotensors of the second rank. Further, one can write

$$\gamma_{ijkl} k_l = e_{ijm} g_{ml} k_l = e_{ijm} g'_m, \quad \delta_{ijkl} k_l = e_{ijm} f_{ml} k_l = e_{ijm} f'_m, \quad (2.25)$$

where the pseudovectors (i.e., axial vectors)  $g'$  and  $f'$  are introduced.

Neglecting in (2.11)–(2.13) terms that are quadratic in  $\mathbf{k}$ , we have

$$\begin{aligned} D'_i &= \varepsilon_{ij}(\omega, \mathbf{k}) E_j = \varepsilon_{ij}(\omega) E_j - i [g'_E]_i, \\ E_i &= \varepsilon_{ij}^{-1}(\omega, \mathbf{k}) D'_j = \varepsilon_{ij}^{-1}(\omega) D'_j - i [f'_D]_i. \end{aligned} \quad (2.26)$$

Substituting these relations into the wave equation (1.15), we can verify (see Sec. 3a and, for example, [1], Secs. 82, 83) that only the scalar product  $f'_{ij} s_i s_j$  or  $f'_i s_i$  plays a role in this equation. Consequently the refractive indices and the ratio of the components of the vector  $\mathbf{D}'$  are obviously independent of the antisymmetrical part of  $f'_{ij}$ , i.e., this tensor can be chosen in symmetric form.\* For an isotropic medium and cubic

\*The antisymmetrical parts of the pseudotensors  $g_{ij, a}$  and  $f_{ij, a}$  lead to pseudovectors  $g'_a$  and  $f'_a$ , which have the form  $\text{const} \cdot [\mathbf{h} \times \mathbf{k}]$ ; here  $\mathbf{h}$  is a certain unit vector connected with the crystal. Obviously, such a vector can exist only for crystals in which one direction is preserved (without change of sign) under all symmetry transformations. In other words, the vector  $\mathbf{h}$  exists only for pyroelectric crystals  $C_1, C_s, C_2, C_{2v}, C_4, C_{4v}, C_3, C_{3v}, C_6, C_{6v}$ ; see, for example [1], Sec. 13). From this and from Table III it is clear that for crystals of classes  $C_{3v}, C_{4v}$ , and  $C_{6v}$  the entire tensor  $f_{ij}$  is antisymmetrical, but these crystals are usually not regarded as gyrotropic (only the symmetrical part of the tensor  $f_{ij}$  is indicated in Table II). Actually, it follows from (2.26) that the vector  $f'_a$  leads to the appearance of a longitudinal component of  $\mathbf{E}$  only (we assume that  $\mathbf{k} = \omega \mathbf{n} s / s$ ). Therefore the tensor  $f_{ij, a}$  makes no contribution whatever to the refractive indices and to the ratio between the components of the vector  $\mathbf{D}$  in the solution of the wave equation for the isotropic crystal (see, for example, Sec. 3a). At the same time, the vector  $\mathbf{E}$ , generally speaking, has elliptic polarization even when  $f_{ij} = f_{ij, a}$ . In this case, however, the degree of ellipticity is proportional to  $\mathbf{k}$ , i.e., we are speaking of an effect of order  $(a/\lambda)$ . Thus, in examining only circular or near circular polarization, crystals of classes  $C_{3v}, C_{4v}$ , and  $C_{6v}$  are nongyrotropic, and for all other crystals the tensor  $f_{ij}$  can be regarded as symmetrical.

We shall proceed in this fashion henceforth (this pertains to Table II). In addition, in examining effects of second order in  $\mathbf{k}$  we shall henceforth assume for simplicity that (2.11)–(2.13) contain no terms linear in  $\mathbf{k}$ , although these terms may also lead to second-order effects.

“Weak gyrotropy” (the degree of ellipticity  $\sim a/\lambda$ ) which should be observed in pure form in crystals of classes  $C_{3v}, C_{4v}$ , and  $C_{6v}$  (this was pointed out earlier in [27]) seems to us, at the same time, fully deserving of study.

**Table III.** Properties of the tensor  $\alpha_{ijlm}$  ( $\alpha_{ijlm} = \alpha_{jilm}$ ;  
 $\alpha_{ijlm} = \alpha_{ijml}$ ; the properties of the tensor  $\beta_{ijlm}$   
 are analogous to those of  $\alpha_{ijlm}$ )

System	Components $\alpha_{ijlm}$ (in addition to those indicated, the components obtained by virtue of the conditions $\alpha_{ijlm} = \alpha_{jilm} = \alpha_{ijml}$ also differ from zero)
Triclinic	All 36 components $\alpha_{ijlm}$ differ from zero (possibility of fixing the axes is not taken into account in this table; see the text and table II)
Monoclinic	20 components differ from zero; the following components vanish: $\alpha_{xyyy}, \alpha_{zyyy}, \alpha_{yyyx}, \alpha_{yyyz}, \alpha_{xxxy}, \alpha_{yxxx}, \alpha_{zzzy}, \alpha_{yzzz}, \alpha_{xzxy}, \alpha_{xyxz}, \alpha_{xxyz}, \alpha_{yzxx}, \alpha_{zxzy}, \alpha_{zyzx}, \alpha_{zzxy}, \alpha_{xyzz}$
Rhombic	12 independent components differ from zero: $\alpha_{xxxx}, \alpha_{yyyy}, \alpha_{zzzz}, \alpha_{xxyy}, \alpha_{xxzz}, \alpha_{yyxx}, \alpha_{zzxx}, \alpha_{xyxy}, \alpha_{xzxx}, \alpha_{yyzz}, \alpha_{zzyy}, \alpha_{yzzy}$
Tetragonal	In classes $D_4, C_{4v}, D_{2d}$ , and $D_{4h}$ seven independent components differ from zero: $\alpha_{xxxx} = \alpha_{yyyy}, \alpha_{zzzz}, \alpha_{xxyy} = \alpha_{yyxx}, \alpha_{xxzz} = \alpha_{yyzz}, \alpha_{zzxx} = \alpha_{zzyy}, \alpha_{xzzx} = \alpha_{yzzy}$ and $\alpha_{xyxy}$ In classes $C_4, S_4$ , and $C_{4h}$ three additional independent components differ from zero: $\alpha_{xyxx} = -\alpha_{xyyy}, \alpha_{xxxy} = -\alpha_{yyxy}, \alpha_{xzyz} = -\alpha_{yzxz}$
Trigonal	In classes $C_3$ and $C_{3i}$ there are 12 independent components: $\alpha_{xxxx} = \alpha_{yyyy}, \alpha_{xxyy} = \alpha_{yyxx}, \alpha_{xxzz} = \alpha_{yyzz}, \alpha_{xxyx} = \alpha_{yyxz} = -\alpha_{yyyz}, \alpha_{xxzx} = -\alpha_{yyzx} = -\alpha_{xyyz}, \alpha_{xxxy} = \alpha_{xyyy} = -\alpha_{yyxy} = -\alpha_{xyxx}, \alpha_{zzxx} = \alpha_{zzyy}, \alpha_{zzzz}, \alpha_{yzxx} = -\alpha_{yzzy} = \alpha_{xzxy}, \alpha_{xzzx} = -\alpha_{xzzy} = -\alpha_{yzxy}, \alpha_{yzzy} = \alpha_{xzzz}, \alpha_{yzxz} = -\alpha_{xzzy}, \alpha_{xyxy} = +\alpha_{xxxx} - \alpha_{xxyy}$ . In classes $D_3, C_{3v}$ and $D_{3d}$ we have in addition $\alpha_{xxzx} = \alpha_{xxxy} = \alpha_{yzxz} = \alpha_{xzzx} = 0$
Hexagonal	In classes $C_6, C_{3h}$ and $C_{6h}$ there are 8 independent components (the components not included are equal to zero): $\alpha_{xxxx} = \alpha_{yyyy}, \alpha_{xxyy} = \alpha_{yyxx}, \alpha_{xxzz} = \alpha_{yyzz}, \alpha_{zzxx} = \alpha_{zzyy}$ and $\alpha_{zzzz}, \alpha_{xyyy} = \alpha_{xxxy} = -\alpha_{yyxy} = -\alpha_{xyxx}, \alpha_{yzzy} = \alpha_{xzzz}, \alpha_{yzxz} = -\alpha_{xzzy}, \alpha_{xyxy} = \alpha_{xxxx} - \alpha_{xxyy}$ . In classes $D_6, C_{6v}, D_{3h}$ , and $D_{6h}$ , we have in addition $\alpha_{xxxy} = \alpha_{yzxz} = 0$ (other components connected with the foregoing are also equal to zero in view of the relations given above).
Cubic	In classes $T$ and $T_h$ , four independent components differ from zero: $\alpha_1 = \alpha_{xxxx} = \alpha_{yyyy} = \alpha_{zzzz}, \alpha_2 = \alpha_{xxzz} = \alpha_{yyxx} = \alpha_{zzyy}, \alpha_3 = \alpha_{xyxy} = \alpha_{yzzy} = \alpha_{zxxz}, \alpha_4 = \alpha_{zzxx} = \alpha_{xxyy} = \alpha_{yyzz}$ For classes $O, T_d$ , and $O_h$ , we have in addition $\alpha_2 = \alpha_4$
Isotropic medium	The same components as in the cubic system differ from zero, but in addition to $\alpha_2 = \alpha_4$ we also have $\alpha_3 = \alpha_1 - \alpha_2$ (thus, we have a total of two independent components).

crystals (of course, without center of symmetry, for otherwise  $g_{ij} = f_{ij} = 0$ ; in addition, it must be borne in mind that the tensor  $g_{ij}$  may vanish also in the absence of a center of symmetry, as occurs, for example, for cubic crystals of class  $T_d$ ):

$$\left. \begin{aligned} g_{ij} &= g\delta_{ij}, f_{ij} = f\delta_{ij}, g' = gk, f' = fk, \\ D' &= \epsilon E - ig[kE], \\ E &= \frac{D'}{\epsilon} - if[kD']. \end{aligned} \right\} \quad (2.27)$$

In this case not only  $D' \cdot k = 0$ , but also  $E \cdot k = 0$  when  $D' \neq 0$ . On the other hand, if  $D' = 0$  and  $E = E_s$  (longitudinal waves), then the gyrotropy becomes immaterial. The form of the symmetrical tensor  $f_{ij}$  for different crystal classes is indicated in Table II (for more details see [1], Sec. 33, and [22, 26, 28]).

Let us proceed to examine the properties of the tensor  $\alpha_{ijlm}$  (and  $\beta_{ijlm}$ ). By virtue of the symmetry of the tensor  $\alpha_{ijlm}$  with respect to the indices  $ij$ , and separately with respect to the indices  $lm$  [see (2.18)], this tensor has in general 36 components (in place of the 81 components of an arbitrary 4th-rank tensor).

Further decrease in the number of independent components is connected with the specific symmetry of the crystals. The general principle reduces, as is known,

to the requirement that the tensor components (physical quantities) remain unchanged under the transformations allowed by the symmetry of the crystal (for more details see [26, 28]).

Triclinic crystals either have no symmetry elements (class  $C_1$ ), or else have a center of symmetry (class  $C_i$ ). However, the presence or absence of a center of symmetry does not impose any conditions on a 4th-rank tensor. Consequently, 36 independent coefficients  $\alpha_{ijlm}$  remain in the triclinic system. To be sure, the choice of axes, which in this case is arbitrary, enables us to fix three coefficients. It seems more rational, however, to calculate the number of independent components without account of the possibility of free choice of axes.

It is sufficient here to recall as an example the tensor  $\epsilon_{ij}(\omega)$  (or any other second-rank tensor). The number of independent components of  $\epsilon_{ij}$  in the triclinic, monoclinic, and rhombic systems is respectively 6, 4, and 3 (see Table II). At the same time in all these cases in the system of principal axes the tensor  $\epsilon_{ij}$  has three independent components. However, the difference between the crystals of these systems is very large, inasmuch as in a rhombic crystal the

principal axes are fixed, but in a triclinic crystal they must be determined, this being equivalent to three additional parameters.

We shall indicate below the possibility of choice of the principal axes (i.e., we note the cases when this choice is not fixed by considerations connected with the symmetry of the crystal), but in calculating the number of independent components this circumstance is disregarded.\*

In monoclinic crystals of classes  $C_2$  and  $C_{2h}$  there is a two-fold axis, usually chosen to be the  $y$  axis. In rotation about such an axis, the coordinates are transformed as follows:  $x \rightarrow -x$  and  $z \rightarrow -z$ . The components of tensors with odd sums of the number of  $x$  and  $z$  indices reverse sign under this transformation, while physically both systems of coordinates are perfectly equivalent, and in this lies the meaning of the assertion that a two-fold axis is present. Consequently, the corresponding 16 components of the tensor  $\alpha_{ijlm}$  vanish, namely the components

$$\alpha_{xyyy}, \alpha_{zyyy}, \alpha_{yypx}, \alpha_{yyyz}, \alpha_{xxxy}, \alpha_{yxxx}, \alpha_{zzzy}, \alpha_{yzzz}, \alpha_{zxxy}, \alpha_{xyxz}, \alpha_{xxyz}, \alpha_{yzxx}, \alpha_{zzzy}, \alpha_{zyyx}, \alpha_{zzxy}, \alpha_{xyzz}.$$

The class  $C_s$  has only a mirror plane perpendicular to the  $y$  axis, and by virtue of this symmetry the components  $\alpha_{ijlm}$  with odd number of  $y$  indices should vanish. This requirement leads to the same result as for classes  $C_2$  and  $C_{2h}$ . Thus, in monoclinic crystals the tensor  $\alpha_{ijlm}$  has 20 independent components. In crystals of the monoclinic system the symmetry properties fix only one crystal axis (the  $y$  axis) and by choosing the other axes we can reduce the number of components of the tensor  $\alpha_{ijlm}$  by one.

Classes  $D_2$  and  $D_{2h}$  of the rhombic system have three two-fold axes. We see readily that in this case the following 12 components with even number of each of the indices  $x, y,$  and  $z$  (the axes  $x, y,$  and  $z$  are two-fold axes) differ from zero:

$$\alpha_{xxxx}, \alpha_{xyyy}, \alpha_{xxzz}, \alpha_{yyxx}, \alpha_{zzxx}, \alpha_{xyxy}, \alpha_{xzzz}, \alpha_{yyyy}, \alpha_{yyzz}, \alpha_{zzyy}, \alpha_{yzzz}, \alpha_{zzzz} \quad (2.28)$$

(in addition, of course,  $\alpha_{xyxy} = \alpha_{yxxy} = \alpha_{xyyx}$  etc.).

The third class of the rhombic system, class  $C_{2v}$ , has one two-fold axis (the  $z$  axis) and two mutually perpendicular mirror planes passing through this axis. Reflection in these planes, i.e., the transformations  $x \rightarrow -x$  or  $y \rightarrow -y$ , leave only the components of the tensor with even number of indices  $x$ , and simultaneously, with even number of indices  $y$  unchanged. But from this it follows for the fourth-rank tensor that the number of indices  $z$  must be even, i.e., we again arrive at the scheme (2.28). Thus, crystals of the rhombic system have 12 independent components  $\alpha_{ijlm}$ .

\*The possibility of the choice of axes is taken into consideration in<sup>[29]</sup> in an evaluation of the number of independent components of the elastic-modulus tensor. This explains the difference in the number of independent components between<sup>[29]</sup> and<sup>[26]</sup>.

For classes  $D_4, C_{4v}, D_{2d},$  and  $D_{4h}$  of the tetragonal system, in addition to symmetry elements corresponding to one of the classes of the rhombic system, the  $z$  axis is a 4-fold rotation or inversion axis. Rotation through an angle  $\pi/2$  about this axis should leave the components of the tensor (physical properties) unchanged, and at the same time the transformation  $x \rightarrow -y, y \rightarrow -x$  occurs. It follows therefore that some of the coefficients of (2.28) are equal to each other, and only seven among them are independent (the remaining  $\alpha_{ijlm}$ , in addition to those obtained by virtue of the conditions  $\alpha_{ijlm} = \alpha_{ji\bar{l}\bar{m}} = \alpha_{ijm\bar{l}}$  are equal to zero in the chosen frame):

$$\alpha_{xxxx} = \alpha_{yyyy}, \alpha_{zzzz}, \alpha_{xxyy} = \alpha_{yyxx}, \alpha_{xxzz} = \alpha_{zzxx}, \alpha_{zzxx} = \alpha_{zzyy}, \alpha_{zzzz} = \alpha_{yzyz}, \alpha_{xyxy} \quad (2.29)$$

For classes  $C_4, S_4,$  and  $C_{4h}$  of the tetragonal system, having only a 4-fold rotation or inversion axis (and also a mirror plane perpendicular to it in the case of class  $C_{4h}$ ), we cannot start from (2.28). As was already illustrated above, we arrive at the conclusion that the other non-vanishing coefficients, in addition to those in (2.29), for these classes are

$$\alpha_{xyxx} = -\alpha_{xyyy}, \alpha_{xxxz} = -\alpha_{yxyy}, \alpha_{xzyz} = -\alpha_{yzzz} \quad (2.30)$$

Thus, for classes  $C_4, S_4,$  and  $C_{4h}$  there are 10 independent components. At the same time, for these classes the symmetry of the crystal singles out only the  $z$  axis, and thus there is one degree of freedom in the choice of the coordinate frame.

Crystal classes of the cubic system have no non-vanishing components other than those for rhombic crystals [see (2.28)].\* The number of independent components, however, decreases greatly. Thus, even the four 3-fold axes, which are present in all classes of the cubic system (body diagonals of the cube), lead to the equivalence of all components of the tensor under the substitution  $xyz \rightarrow yzx \rightarrow zxy$ . We are, therefore, left with only four independent components

$$\alpha_1 = \alpha_{xxxx} = \alpha_{yyyy} = \alpha_{zzzz}, \alpha_2 = \alpha_{xxzz} = \alpha_{yyxx} = \alpha_{zzyy}, \alpha_3 = \alpha_{xyxy} = \alpha_{yzyz} = \alpha_{zzxx}, \alpha_4 = \alpha_{zzxx} = \alpha_{xyxy} = \alpha_{yzyz} \quad (2.31)$$

For classes  $T$  and  $T_h$ , no further simplifications arise. In classes  $T_d, O,$  and  $O_h$  we have, in addition

$$\alpha_2 = \alpha_4 \quad (\text{classes } T_d, O, O_h) \quad (2.32)$$

(2.32) follows in obvious fashion from (2.31), if we recognize that the axes of the cube,  $x, y,$  and  $z,$  are 4-fold

\*This statement is connected with the fact that all classes of the cubic system share symmetry elements with at least one of the classes of the rhombic system (in addition to having other symmetry elements) (see, for example, the illustrative table 21 in<sup>[26]</sup>). We recall also that the axes  $x, y,$  and  $z$  are fixed for all classes of the cubic system (two-fold axes for classes  $T, T_h,$  and  $T_d$ ; 4-fold axes for classes  $O$  and  $O_h$ ).

inversion axes (class  $T_d$ ) or rotation axes (classes  $O$  and  $O_h$ ).\*

In an isotropic medium, as is clear from (2.20), there are only two independent components  $\alpha_{ijlm}$ . In tensor notation for an arbitrary Cartesian frame

$$\left. \begin{aligned} \alpha_{xxxx} = \alpha_{yyyy} = \alpha_{zzzz} = \alpha_1 = -\frac{c^2}{\omega^2} \alpha_{||}, \quad \alpha_{xxyy} = \alpha_{xxzz} \\ = \alpha_{yyxx} = \alpha_{yyzz} = \alpha_{zzxx} = \alpha_{zzyy} = \alpha_2 = \alpha_4 = -\frac{c^2}{\omega^2} \alpha_{\perp}, \\ \alpha_{xyxy} = \alpha_{xzzz} = \alpha_{yzyz} = \alpha_3 = \alpha_1 - \alpha_2 = \frac{c^2}{\omega^2} (\alpha_{\perp} - \alpha_{||}). \end{aligned} \right\} (2.33)$$

The difference between the cubic crystals of classes  $T_d$ ,  $O$ , and  $O_h$  lies obviously only in the existence of the relation  $\alpha_3 = \alpha_1 - \alpha_2$ .

In crystals of the triclinic and hexagonal systems, an account of the symmetry properties calls for simple analytic transformations, which we shall not carry out here (see, for example [26,29] for some of these transformations applied to some other tensors). There is no further need for it, particularly since the symmetry properties of the tensor  $\alpha_{ijlm}$  (including those given above) can in fact be assumed known from the literature. The point is that the symmetry of the tensor  $\alpha_{ijlm}$  is the same as in the long-employed tensor of piezo-optical coefficients  $\pi_{ijlm}$ , which relates the variation of  $\epsilon_{ij}^{-1}$  with the stress tensor  $\sigma_{lm}$  (thus,  $\delta\epsilon_{ij}^{-1} = \pi_{ijlm}\sigma_{lm}$ ; see [26]). Precisely the same symmetry is of course possessed by the tensor of elastooptical coefficients  $p_{ijlm}$  (here  $\delta\epsilon_{ij}^{-1} = p_{ijlm}u_{lm}$ , where  $u_{lm}$  is the strain tensor). Therefore the symmetry properties of the tensor  $\alpha_{ijlm}$  for the trigonal and hexagonal systems will be taken from [26] (see Table 15 of [26]). This was not done for other systems in view of the simplicity of the derivation and the desire to emphasize certain aspects. A summary of all the values of  $\alpha_{ijlm}$  is given in Table III.

We shall not consider simultaneously terms of first and second order in  $\mathbf{k}$ , in view of the remark already made concerning the smallness of the spatial dispersion. We shall therefore be interested not in the general expressions (2.11)–(2.13), but in expression (2.26) for the gyrotropic medium and the following expression for a nongyrotropic medium

$$\left. \begin{aligned} \epsilon_{ij}(\omega, \mathbf{k}) = \epsilon_{ij}(\omega) + \left(\frac{\omega}{c}\hat{n}\right)^2 \alpha_{ijlm}(\omega) \hat{n}^2 s_l s_m, \\ \epsilon_{ij}^{-1}(\omega, \mathbf{k}) = \epsilon_{ij}^{-1}(\omega) + \left(\frac{\omega}{c}\hat{n}\right)^2 \beta_{ijlm}(\omega) \hat{n}^2 s_l s_m. \end{aligned} \right\} (2.33')$$

The tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  can, of course, always be diagonalized by choosing the corresponding (principal) axes.† The direction of these axes for arbitrary  $\mathbf{s}$

\*We point out a certain inaccuracy in [5]: the statements made with respect to the values of  $\alpha_{ijlm}$  in tetragonal and cubic crystals are valid only for the more symmetrical classes of these systems [the schemes (2.29) and (2.31)–(2.32)].

†If the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  is not Hermitian, then we must consider  $\epsilon_{ij}^i$  and  $\epsilon_{ij}^j$  separately, and the principal axes of these tensors (more accurately, the eigenvectors, which in general are complex) do not coincide in the general case. Unless otherwise stipulated, we refer in the text, for the sake of brevity, only to the tensor  $\epsilon_{ij}^i$ , which is considered real.

does not coincide with either  $\mathbf{s}$  or with the axes of the tensor  $\epsilon_{ij}(\omega)$ ; if the axes of the tensor  $\epsilon_{ij}(\omega)$  are fixed (i.e., they are free of the degeneracies present in cubic and uniaxial crystals), the axes of the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  are close to the axes of  $\epsilon_{ij}(\omega)$ , in view of the smallness of the  $\mathbf{s}$ -independent terms in (2.33).

In crystal optics with spatial dispersion, naturally, a great interest is attached to those principal axes  $\epsilon_{ij}(\omega, \mathbf{k})$ , the direction of which coincides with  $\mathbf{s}$ . For rhombic crystals such axes are  $x, y$ , and  $z$  [see (2.28) and Table III]. If, for example, the vector  $\mathbf{s}$  is directed along the  $x$  axis, then the principal values of the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  are

$$\begin{aligned} \epsilon_1 = \epsilon_{xx}(\omega, \mathbf{k}) = \epsilon_{xx}(\omega) + \left(\frac{\omega}{c}\hat{n}\right)^2 \alpha_{xxxx}, \quad \epsilon_2 = \epsilon_{yy}(\omega, \mathbf{k}) = \epsilon_{yy}(\omega) \\ + \left(\frac{\omega}{c}\hat{n}\right)^2 \alpha_{yyxx}, \quad \epsilon_3 = \epsilon_{zz}(\omega, \mathbf{k}) = \epsilon_{zz}(\omega) + \left(\frac{\omega}{c}\hat{n}\right)^2 \alpha_{zzxx}. \end{aligned}$$

In tetragonal crystals of classes  $D_4, C_{4v}, D_{2d}$ , and  $D_{4h}$ , for a vector  $\mathbf{s}$  directed along the axes  $x$  and  $y$ , the tensor  $\epsilon_{ij}(\omega, \mathbf{k})$  turns out to be reduced to the principal axes, and the principal values are different. On the other hand if the vector is directed along the  $z$  axis (along the 4-fold axis), then

$$\epsilon_1 = \epsilon_2 = \epsilon_{\perp}(\omega) + \left(\frac{\omega}{c}\hat{n}\right)^2 \alpha_{xxzz}, \quad \epsilon_3 = \epsilon_{||} + \left(\frac{\omega}{c}\hat{n}\right)^2 \alpha_{zzzz}.$$

Without dwelling on crystals of other systems, let us proceed to cubic crystals. In this case (see Table III)

$$\left. \begin{aligned} \epsilon_{xx} = \epsilon + \left(\frac{\omega}{c}\hat{n}\right)^2 \{\alpha_1 s_x^2 + \alpha_4 s_y^2 + \alpha_2 s_z^2\}, \quad \epsilon_{xy} = 2 \left(\frac{\omega}{c}\hat{n}\right)^2 \alpha_3 s_x s_y, \\ \epsilon_{yy} = \epsilon + \left(\frac{\omega}{c}\hat{n}\right)^2 \{\alpha_2 s_x^2 + \alpha_1 s_y^2 + \alpha_4 s_z^2\}, \quad \epsilon_{xz} = 2 \left(\frac{\omega}{c}\hat{n}\right)^2 \alpha_3 s_x s_z, \\ \epsilon_{zz} = \epsilon + \left(\frac{\omega}{c}\hat{n}\right)^2 \{\alpha_4 s_x^2 + \alpha_2 s_y^2 + \alpha_1 s_z^2\}, \quad \epsilon_{yz} = 2 \left(\frac{\omega}{c}\hat{n}\right)^2 \alpha_3 s_y s_z. \end{aligned} \right\} (2.34)$$

[for classes  $O, T_d$  and  $O_h$ , in addition,  $\alpha_2 = \alpha_4$ ; the factor two in the expressions for  $\epsilon_{xy}, \epsilon_{xz}$ , and  $\epsilon_{yz}$  appears in connection with the summation in (2.33) of terms proportional to  $s_x s_y$  and  $s_y s_x$ ]. It is, therefore, obvious that the axes of the cube  $x, y$ , and  $z$  are the principal axes of the tensor, if the vector  $\mathbf{s}$  is directed along any of the axes  $x, y, z$ . In this case the corresponding second-degree surface for  $\alpha_2 = \alpha_4$  degenerates into a surface of revolution (ellipsoid or hyperboloid). If the vector  $\mathbf{s}$  is directed along the body diagonals of the cube ( $|s_x| = |s_y| = |s_z| = 1/\sqrt{3}$ ), then

$$\begin{aligned} \epsilon_{xx} = \epsilon_{yy} = \epsilon_{zz} = \epsilon + \frac{1}{3} \left(\frac{\omega}{c}\hat{n}\right)^2 (\alpha_1 + \alpha_2 + \alpha_4), \\ |\epsilon_{xy}| = |\epsilon_{xz}| = |\epsilon_{yz}| = 2 \left(\frac{\omega}{c}\hat{n}\right)^2 \frac{\alpha_3}{3}. \end{aligned} \quad (2.35)$$

[Part II of this article (Secs. 3 and 4) will be published in one of the future issues of UFN.]

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