SUPPLEMENTARY LIGHT WAVES IN CRYSTALS AND EXCITON ABSORPTION*

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Usp. Fiz. Nauk 77, 309-319 (June, 1962)

1. INTRODUCTION

In crystal optics, the medium is characterized by a dielectric tensor $\epsilon_{ij}(\omega, \mathbf{k})$ (ω —frequency, $\mathbf{k}$ —electromagnetic wave vector). Spatial dispersion, i.e., the dependence of ϵ_{ij} on $\mathbf{k}$, was introduced long ago,[1] but until recently it was assumed to be significant only in considerations of the rotation of the plane of polarization of light in gyrotropic crystals, and to introduce in all other respects only negligible corrections in crystal optics, on the order of a/λ (a —lattice constant, λ —wavelength of light). Even in discussions of spatial dispersion in crystals it has been tacitly assumed until 1957[2] that ordinary birefringence is maintained, i.e., for a given propagation direction $\mathbf{s}$ there are two waves with induction vectors $\mathbf{D}_1$ and $\mathbf{D}_2$, which are perpendicular to each other and to $\mathbf{s}$.

This estimate of the smallness of the effects connected with spatial dispersion is in most cases correct. However, as we have shown as long ago as 1957,[3] if the frequency of the light approaches some exciton-absorption band, the foregoing estimates are utterly incorrect, and an account of spatial dispersion modifies ϵ_{ij} significantly even when $\lambda \gg a$. As a result, the frequency dependence of the refractive indices becomes appreciably changed, arbitrarily large deviations from the known relation between the integral absorption of the band and the oscillator strength of the elementary crystal cell become possible, etc. The most interesting result of [3] was the predicted existence of supplementary light waves in the vicinity of the exciton absorption bands, in addition to the universally known birefringence waves.

It was shown, for example, that two light waves having the same polarization and propagation direction but different velocities can exist in the crystal.

Following the publication of [3], the theory of supplementary waves advanced rapidly and during the elapsed four years many papers were published on this subject.[4-34] Supplementary light waves were observed experimentally in anthracene[23,24] and in cuprous oxide.[26] Attempts to observe these waves experimentally in a few other crystals also yielded favorable preliminary results.

Investigations show that supplementary waves occur only near the exciton absorption bands. By exciton is meant here a crystal excitation with a single continuous quantum number—the quasi momentum $\mathbf{k}$ (all other quantum numbers are discrete).[3] It is also required that the aforementioned exciton represent with sufficient accuracy a stationary state of the crystal, i.e., that it have a sufficiently long lifetime with respect to thermal transitions (scattering by phonons, transition to the "multi-phonon wind" etc.). Since the lifetime (path time) of the exciton increases with decreasing temperature,[28,29] the additional waves and the associated phenomena should manifest themselves more distinctly at low temperatures.

*Paper delivered at the plenary session of the Eighth Congress on Low-Temperature Physics in Kiev, October 13, 1961.

2. SUPPLEMENTARY LIGHT WAVES IN THE ACCOUNT OF ABSORPTION

If the frequency of light ω is close to $\omega_0 \equiv \mathscr{E}(0)/\hbar$ where $\mathscr{E}(\mathbf{k})$ is an isolated exciton energy band, then the specific dipole moment of crystal polarization $\mathbf{P}(\mathbf{r}, t)$ caused by the passage of the light wave is[3,9]

$$\mathbf{P} = \mathbf{P}_0 e^{i(\mathbf{kr}-\omega t)} + \mathbf{P}_0^* e^{-i(\mathbf{kr}-\omega t)}, \quad \mathbf{P}_0 = \beta(\omega, \mathbf{k}) E_0^{\text{ext}}, \tag{1}$$

where β is a tensor of the second rank,

$$\beta_{xy} = \beta'_{xy} + \frac{Q_{xy} + \alpha_{xy}}{\mathscr{E}(k) + H^a - \hbar\omega}, \tag{2}$$

$$Q_{xy} = V(\mathbf{P}_{0\mathbf{k}})_x (\mathbf{P}^*_{0\mathbf{k}})_y, \quad \mathbf{P}_{0\mathbf{k}} = \langle \Psi^{0*} \hat{\mathbf{P}}(0) \Psi_{\mathbf{k}} \rangle. \tag{3}$$

Here β'_{xy} —constants, V —volume of the unit cell of the crystal, Ψ^0 and Ψ_k —the ground and exciton states of the crystal, $\mathbf{k}$ —quasi momentum of the exciton, which coincides with the wave vector of light, and $\hat{\mathbf{P}}(0)$ —specific dipole moment operator at the macropoint $\mathbf{r} = 0$. The energy is measured in such a way that in the state Ψ the energy is zero. The scalar H^a and the tensor α are complex; they vanish if the exciton lifetime is infinite[3] and are proportional to the probability of scattering or thermal decay of the exciton,[9] which is assumed small.

The external electric field perturbing the crystal is

$$\mathbf{E}^{\text{ext}} = \mathbf{E}_0^{\text{ext}} e^{i(\mathbf{kr}-\omega t)} + (\mathbf{E}_0^{\text{ext}})^* e^{-i(\mathbf{kr}-\omega t)}, \tag{4}$$

where

$$\mathbf{E}_0^{\text{ext}} \equiv \mathbf{E}_0 - \mathbf{E}'_0, \quad \mathbf{E}'_0 = -4\pi(\mathbf{P}_0\mathbf{s})\mathbf{s}, \quad \mathbf{s} \equiv \frac{\mathbf{k}}{|\mathbf{k}|}.$$

Here $\mathbf{E}_0$ —amplitude to the total macrofield and $\mathbf{E}'_0$ —amplitude of the vortex-free field, generated without retardation by the fictitious dielectric-polarization charges that arise in the crystal upon passage of the wave. If no extraneous charges are introduced into the crystal, then $\mathbf{E}_0^{\text{ext}}$ coincides with the vortical (transverse) part of $\mathbf{E}_0$:

$$\mathbf{E}_0^{\text{ext}} = \eta \mathbf{E}_0, \quad \eta_{xy} = \delta_{xy} - s_x s_y. \tag{5}$$

If, as is customary, we assume that the electric field, the induction, and the magnetic field are proportional to $\exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$, where $k = \omega n \mathbf{s}/c$ and n is the refractive index of the wave, then Maxwell's equations reduce to

$$\mathbf{D}_0 = n^2 \eta \mathbf{E}_0; \ \mathbf{D}_0 = \gamma \mathbf{E}_0, \text{ where } \gamma_{xy}(\omega, \mathbf{k}) = \delta_{xy} + 4\pi [\beta(\omega, \mathbf{k})\eta]_{xy}. \tag{6}$$

Equation (6) has a solution if

$$(\mathbf{s}, \gamma\mathbf{s})\, n^4 + [(\mathbf{s}, \gamma^2\mathbf{s}) - (\mathbf{s}, \gamma\mathbf{s})\,\mathrm{sp}\,\gamma]\, n^2 + \begin{vmatrix} \gamma_{xx} & \gamma_{xy} & \gamma_{xz} \\ \gamma_{yx} & \gamma_{yy} & \gamma_{yz} \\ \gamma_{zx} & \gamma_{zy} & \gamma_{zz} \end{vmatrix} = 0. \tag{7}$$

This equation determines the values of n. If we neglect the dependence of γ on $\mathbf{k}$, then (7) becomes quadratic in n^2 and the two ordinary birefringence waves are obtained. On the other hand, if the dependence of γ on $\mathbf{k}$, i.e., on n, is taken into account, then (7) is of higher degree in n, and defines not two but more values of n, with as many plane-wave solutions of Maxwell's equations. This gives rise to the supplementary waves. In general, $\mathscr{E}(\mathbf{k})$ cannot be expanded in powers of k_x, k_y, and k_z,[7,9] nor can it be expanded in powers of $|\mathbf{k}|$ for a given direction $\mathbf{s}$. Let us examine the case of a crystal with a symmetry center, when this expansion contains only even powers:

$$\mathscr{E}(\mathbf{k}) = \mathscr{E}_s + \frac{\hbar^2 |\mathbf{k}|^2}{2M_s} + \ldots, \tag{8}$$

$\mathscr{E}_S$ and M_S are independent of $|\mathbf{k}|$, but do depend on the direction $\mathbf{s}$.

If we introduce the notation $\gamma' = I + 4\pi\beta'\eta$, $I_{xy} = \delta_{xy}$:

$$\left.\begin{aligned} \mu_s &= \frac{2M_s c^2}{\hbar\omega}\left(1 - \frac{\mathscr{E}_s + H^a}{\hbar\omega}\right) \cong \frac{2M_s c^2}{\hbar\omega_0^2}\left(\omega - \omega_0 - \frac{H^a}{\hbar}\right), \\ B_s &= \frac{8\pi M_s c^2}{\hbar^2\omega^2}(Q + \alpha) \cong \frac{8\pi M_s c^2}{\hbar\omega_0^2}(Q + \alpha), \end{aligned}\right\} \tag{9}$$

then substitution of (2) and (8) in (6) yields

$$\gamma = \gamma' + \frac{B_s \eta}{n^2 - \mu_s}. \tag{10}$$

In this case the degree of Eq. (7) with respect to n^2 will depend on the symmetry of the crystal in the propagation direction $\mathbf{s}$. This equation can be solved for specific cases.

We consider below excitons whose photoexcitation is allowed in the dipole approximation $\mathbf{P}_{0\mathbf{k}} \cong \lim_{|\mathbf{k}|\to 0} \mathbf{P}_{0\mathbf{k}}$. Let us examine, for simplicity, the case of a crystal with symmetry not lower than rhombic, in which the principal axes of the tensor ϵ_{ij} coincide with the twofold or fourfold crystal axes. We choose the latter as the Cartesian coordinates. If the beam direction $\mathbf{s}$ coincides with one of these coordinate axes, then the latter remain the principal axes of ϵ_{ij} also if the dependence of ϵ_{ij} on $\mathbf{k}$ is allowed for. In this case $\mathbf{P}_{0\mathbf{k}}(|\mathbf{k}| \to 0)$ can be directed only along one of the aforementioned principal axes. Let us assume that $s_x = s_y = 0$ and $s_z = 1$ for the light beam and for the exciton interacting with it. The equation (7) can then be solved in elementary fashion. The polarization of the exciton and of the light wave can in this case be either longitudinal [3,9,17]

$$\mathbf{P}_{0\mathbf{k}} \| \mathbf{E}_0 = \mathbf{E}_0' = -4\pi \mathbf{P}_0 \| \mathbf{s}, \ \mathbf{D}_0 = 0, \quad n^2 = \mu_{\|s}(\omega), \tag{11}$$

or transverse (to be specific, parallel to Ox):

$$\mathbf{P}_{0\mathbf{k}} \| 0x, \ E_{0y} = E_{0z} = 0,$$

$$E_{0x} = \begin{cases} E_+ \ n_+^2 = \frac{1}{2}(\mu_s + \gamma'_{xx}) + \sqrt{\frac{1}{4}(\mu_s - \gamma'_{xx})^2 + B_{sxx}}, \\ E_- \ n_-^2 = \frac{1}{2}(\mu_s + \gamma'_{xx}) - \sqrt{\frac{1}{4}(\mu_s - \gamma'_{xx})^2 + B_{sxx}}. \end{cases} \tag{12}$$

We see that there exist two waves of the same polarization and with different refractive indices. For the exciton polarization considered above ($\mathbf{P}_{0\mathbf{k}} \equiv \mathbf{P}_{0\mathbf{k}}^{(x)} \| Ox$) it is possible to have, in addition to the waves (12), still another ordinary transverse light wave, which does not interact at all with the given exciton:

$$E_{0x} = E_{0z} = 0, \quad E_{0y} \neq 0, \quad n = \sqrt{\gamma'_{yy}}. \tag{13}$$

If the principal axes x and y of the tensor ϵ_{ij} are degenerate, i.e., if z is the optical axis of the crystal (rhombohedral, tetragonal, hexagonal, and cubic systems), then the exciton is also doubly degenerate, with $\mathbf{P}_{0\mathbf{k}}^{(x)}$ and $\mathbf{P}_{0\mathbf{k}}^{(y)}$ equal to each other and simultaneously different from zero. In this case only transverse waves of type (12) exist in the vicinity of the exciton band (four waves, assuming the waves with different polarizations to be different), and their amplitudes $\mathbf{E}_\pm$ are arbitrarily oriented in the x, y plane. There are no waves of type (13).

If the lifetime (scattering time) of the exciton is infinite, then the quantities H^a and α in (2) vanish,[9] and consequently the light is not absorbed at all in the crystal, no matter how strong the phototransition oscillator may be. The dispersion curves corresponding to formulas (12) for this case are the continuous lines of Figs. 1 and 2. The plots correspond to the following parameters:

$$\mathscr{E}_s = 2 \text{ eV}, \quad a = 10 a_0 \quad \left(a_0 = \frac{\hbar^2}{me^2} = 0.529 \text{ Å}\right), \quad \gamma'_{xx} = 2.$$

The oscillator strength, referred to the crystal elementary cell, is f = 0.1.

Figure 1 corresponds to the case $M_S = m$, and in this case $B_{SXX} = 58400$; Figure 2 corresponds to the same values of M_S and B_{SXX}, but with negative sign. This new type of frequency dependence of the refractive indices (12) was first derived in our paper [3] (see also [4]).

In the case of a finite but large exciton lifetime with respect to thermal transitions, when H^a and α are small, we can neglect α and the real part of H^a in (2), but the imaginary part of H^a must be retained, since H^a depends on ω and $\mathbf{k}$ in the considered re-

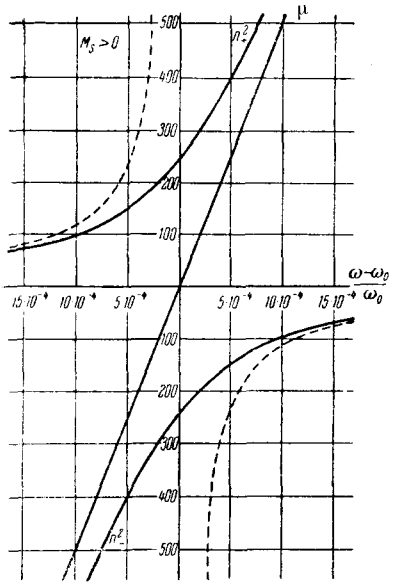

FIG. 1. Dependence of the refractive indices of the transverse light waves on the frequency in the absence of absorption.

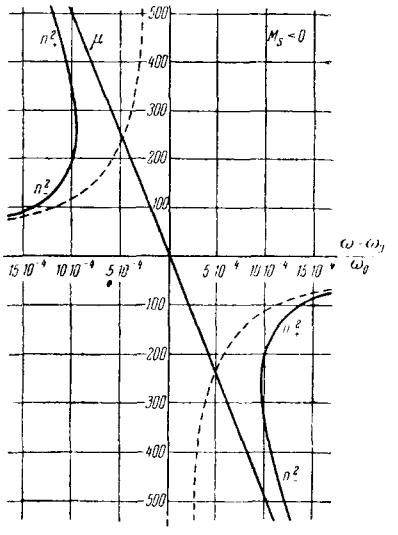

FIG. 2. Dependence of the refractive indices of transverse light waves on the frequency in the absence of absorption.

gion of $\mathcal{E}(\mathbf{k}) - \hbar\omega \approx 0$.[9] But since $\mathbf{k}$ is small, it is possible to replace $H^a(\omega, \mathbf{k})$ by $H^a(\omega, 0)$. The frequency dependence of H^a has not yet been investigated. If we assume that it is sufficiently smooth and recognize that the term H^a is in general significant only in the small frequency range $\omega \approx \omega_0$, we can assume approximately that H^a is a constant imaginary quantity. Then μ_S in (9) becomes complex, $\mu_S = \mu'_S + i\mu''_S$, where[9]

$$\mu''_s = \frac{2M_S c^2}{\hbar^2\omega_0^2} iH^a, \quad iH^a > 0. \tag{14}$$

As a result, if the refractive indices are represented in the form $n = n' + in''$, their frequency dependence, which is determined by formula (12), has the form shown in Figs. 3 and 4. We assumed here the same values of the parameters as in Figs. 1 and 2, but introduce an imaginary addition (14) to μ_S, namely $\mu''_S = 10$, which corresponds to $iH^a/\hbar = 6.1 \times 10^{10}$ sec^{-1}. Figures 3 and 4 correspond to the cases $M_S = m$ and

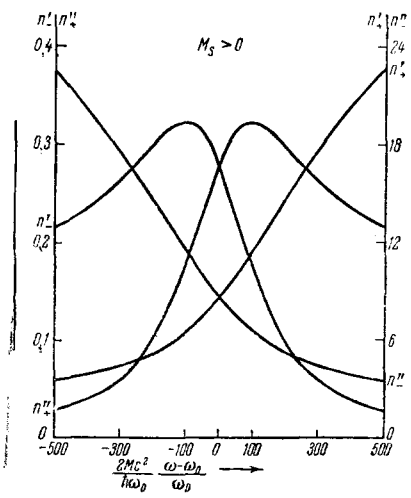

FIG. 3. Dependence of the real (n′) and imaginary (n″) parts of the refractive indices of the transverse light waves on the frequency when absorption is taken into account.

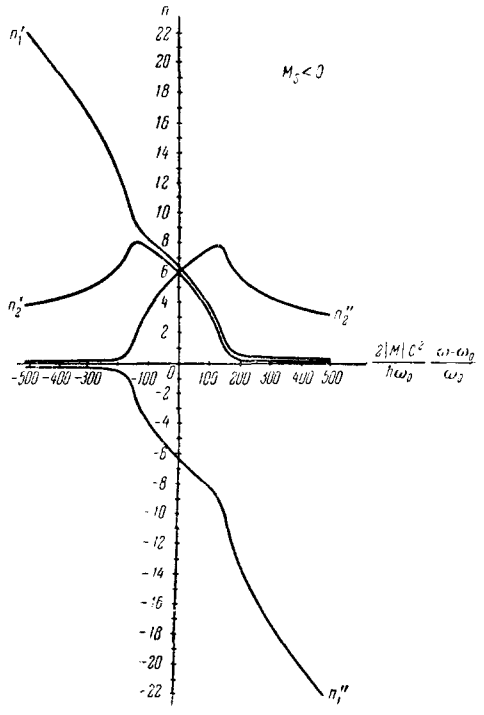

FIG. 4. Dependence of the real (n′) and imaginary (n″) parts of the refractive indices of transverse light waves on the frequency when absorption is taken into account.

$M_S = -m$, respectively. In Fig. 4 we have $n_1 = n_-$, $n_2 = n_+$ when $-\mu'_S > \gamma'_{XX}$ and $n_1 = n_+$, $n_2 = n_-$ when $-\mu'_S < \gamma'_{XX}$.

Attention is called to the almost symmetrical form of the curves in Figs. 3 and 4. The symmetry should actually occur in the region where $|n^2_\pm| \gg \gamma'_{XX}$. We can then discard by way of an approximation the term γ'_{XX} in (12) and obtain as a result

$$\begin{aligned} n'_-(\omega - \omega_0) &= \pm n''_+(\omega_0 - \omega); \\ n''_-(\omega - \omega_0) &= \pm n'_+(\omega_0 - \omega). \end{aligned} \tag{15}$$

As can be seen from Fig. 3, the exponential damping coefficient of the wave is much larger in the n″

space for the "minus" wave than for the "plus" wave. The dependence of n''_+ on ω goes through a maximum, while the ω-dependence of n''_- is monotonic. If the illuminated crystal slab is sufficiently thick, only the "plus" wave, which is less attenuated, will pass through it. The intensity of the light transmitted through the slab will depend monotonically on the thickness l, namely $J \sim \exp(-2\omega n''_+ l/c)$. Consequently, when $M_S > 0$ the Lambert-Bouguer law should hold in the case of thick plates, and we can introduce the concept of an absorption coefficient $2\omega n''_+/c$ as a specific characteristic of the matter. The form of the absorption band is given by the $n''_+(\omega)$ curve of Fig. 3. Its maximum lies on the violet side of the "dispersion frequency" ω_0.

On the other hand, if the slab is thin both waves (12) emerging from the slab have comparable amplitudes. Since these waves are coherent and have the same polarization, they will interfere with each other after emerging from the slab. The phase difference of these waves, $\omega(n'_+ + n'_-)l/c$, depends on the thickness l of the slab. As a result, the intensity of the light transmitted through the slab will be not a monotonic but an oscillating function of l.[23] (This will be discussed in greater detail in the next section). It is therefore impossible to retain the concept of absorption coefficient as a specific characteristic of the matter.

In the case when $M_S < 0$, as can be seen from Fig. 4, we have $n''_1 > n''_2$ only on the violet side of the maximum of the absorption band. Therefore in the case of a thick slab only in this frequency range will the "2" wave alone be transmitted, and $2\omega n''_2/c$ assumes the sense of a specific light absorption coefficient. On the red side of the maximum of the band we can have $n''_1 \approx n''_2$ and both waves will pass equally well through a slab of arbitrary thickness. We cannot therefore retain the concept of specific coefficient of light absorption, in view of the mentioned unavoidable interference of the two waves on emerging from the slab.

If the experimenter ignores this interference and determines the frequency dependence of the coefficient of absorption in traditional fashion, assuming that the intensity of the transmitted light decreases monotonically and exponentially with increasing l, then the band shapes obtained experimentally for different values of l will not be reproducible, as was pointed out in [35].

According to Fig. 4, n'_1 and n''_1 have opposite signs. If light is incident from vacuum on a semi-infinite crystal, the sign preceding the complex value $n_1 = n'_1 + in''_1$ should be chosen such[1] as to make the wave $\exp[i(\omega n_1 z/c - \omega t)]$ attenuate as it penetrates in the crystal. Let z increase with increasing depth in the crystal; we should then have $n''_1 > 0$ and $n'_1 < 0$. It is interesting that in this case the phase velocity of the wave is directed outward from the crystal towards its surface, i.e., opposite the incoming wave. On the other hand, the group velocity is directed inside the crystal.

If terms of higher power in $|\mathbf{k}|$ are retained in the expansion (8), higher powers of n will appear in the denominator of (10). The degree of Eq. (7) in n will then increase and additional roots will appear. Does this mean further appearance of supplementary waves, on top of those considered above? An investigation shows that the new roots n do not have any physical meaning, since they correspond to values of $|\mathbf{k}|$ so large that the expansion (8) diverges (we get $\lambda < a$).

It must not be thought that introduction of spatial dispersion into the dielectric tensor should lead in general to additional electromagnetic waves. The point is that the supplementary roots n do not arise in the overwhelming majority of cases, or else have no physical meaning. The exciton is apparently still the only exception to this rule. For example, inclusion of spatial dispersion does not lead to additional waves in a crystal near non-exciton absorption bands. Another example is a plasma (ionosphere), in which an account of the spatial dispersion of the electric tensor greatly distorts the frequency dependence of the refractive indices (the discrete frequency spectrum of the longitudinal waves changes into a band spectrum, and dispersion curves such as shown in Figs. 1 and 2 appear in a magnetic plasma), but no additional waves arise—three waves exist in the plasma whether spatial dispersion is included or not. There are likewise three waves in a magnetic plasma.*

3. EXPERIMENTAL OBSERVATION OF SUPPLEMENTARY WAVES

We have previously proposed[4,23] several experiments with which to check the foregoing theory and, in particular, to detect supplementary waves. Some of these experiments have already been performed and have actually confirmed the existence of the supplementary light waves predicted by the theory.[3] We report here briefly the results of these experiments

Assume that a monochromatic light wave is normally incident from vacuum on a plane-parallel crystal slab and gives rise to two waves in the slab with complex wave vectors $k_+ = k'_+ = ik''_+$ and $k_- = k'_- + ik''_-$. On leaving the slab, the two waves interfere with each other and as a result the intensity of the transmitted light J should be an oscillating function of the slab thickness l:[23]

$$J \sim |E|^2 = |\mathbf{a}_+|^2 e^{-2k''_+ l} + |\mathbf{a}_-|^2 e^{-2k''_- l} + 2|(\mathbf{a}_+, \mathbf{a}^*_-)| \cos[(k'_+ - k'_-)l + \alpha_0]. \quad (16)$$

Here $\mathbf{a}_+$ and $\mathbf{a}_-$ —amplitudes of the electric field of the waves after emergence from the slab, with allow-

*This may explain why the theoreticians, who investigated for years electromagnetic waves in media with spatial dispersion but did not consider specially the case of the exciton, did not raise the question of the supplementary waves until 1957.

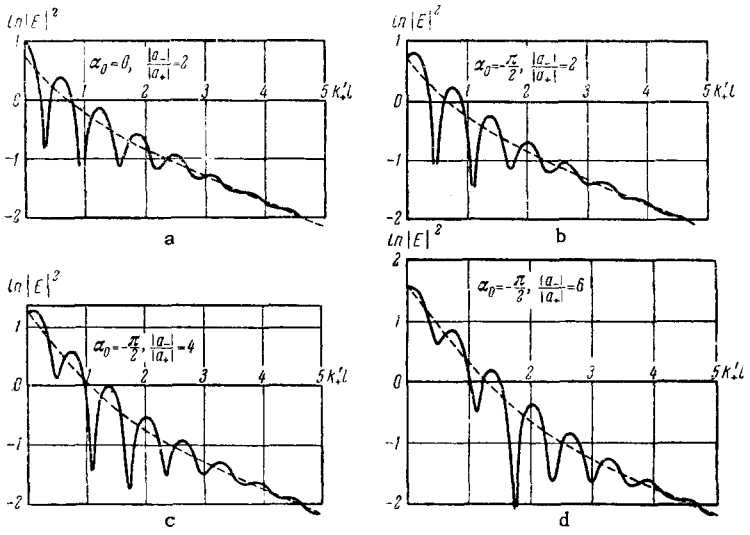

FIG. 5. Theoretical dependence of the square of the modulus of the amplitude of the electric field of the transmitted wave on the thickness of a crystal slab. $(k'_+ - k'_-)/k''_+ = 10$; $k''_-/k''_+ = 3$.

ance for the reflection of light from both surfaces of the slab, but without account of attenuation in the slab (we neglect multiply reflected waves); α_0 is determined by the equation $(\mathbf{a}_+ \cdot \mathbf{a}_-^*) = |(\mathbf{a}_+ \cdot \mathbf{a}_-^*)| e^{i\alpha_0}$. Equation (16) is plotted in Fig. 5.

Oscillating curves of this type were obtained experimentally for anthracene at 20°K near the exciton absorption band, the maximum of which is at 25,200 cm^{-1}. Preliminary results of the experiment were published in [23] and more accurate results of a more carefully performed repeat experiment, using an improved procedure, were published in [24]. Figure 6 shows one of the curves obtained in [24]. It corresponds to a light frequency 25,108 cm^{-1}. The experimental points are marked with the probable errors in both directions. The light was transmitted through two parallel polarizers, one (Glan prism) ahead of the crystal and the other (spar) behind the crystal. The monoclinic crystal axis of anthracene, which coincides with the principal axis of the tensor ϵ_{ij}, was oriented strictly parallel to the electric field vector of the polarized light wave. Consequently only one of the two ordinary birefringence waves was produced in the crystal. The interference shown in Fig. 6 denotes the appearance of a second wave of the same polarization in the crystal, i.e., of a supplementary wave.

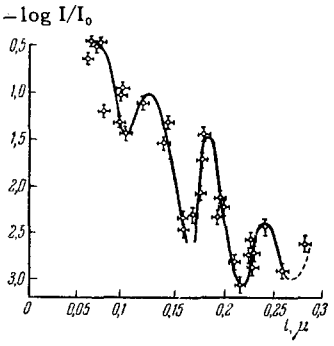

FIG. 6. Experimentally obtained dependence of the intensity of the transmitted light on the thickness of the slab. Anthracene, 20°K (thickness in microns).

It must be emphasized that under the conditions of the described experiment the interference of the waves multiply reflected from the surfaces of the slab was completely eliminated, since the intensity of the beam after passing three times through the plate is two or three orders of magnitude lower than the intensity of the beam passing once through the slab, whereas in Fig. 6, the value of J oscillated some 30 times. In addition, in order to explain the observed period of the oscillations of J with increasing l, it would be necessary to ascribe to anthracene a refractive index of 3.45, whereas the actual value exceeds 5 at this frequency and temperature. [36]

Figure 6 shows quite clearly the periodicity of the maxima and minima: the distance between the maxima is $\Delta l = 0.056$, 0.060, and 0.057 μ. The distances between the minima are $\Delta l = 0.063$ and 0.055 μ. Such a periodicity would not be natural for a simple straggle of the points.

It must be emphasized that each of the 30 experimentally measured points on Fig. 6 is obtained for its own independently grown crystal. Different points represent different independent experiments (sometimes separated by a long time interval). Consequently the near-lying points found in many places on the curves illustrate the reproducibility of the experimental results.

The value of $k'_+ - k'_-$ can be calculated with the aid of (16) from the average period of the curve which is equal to 0.058 μ. A value of 6.9 is obtained for $n'_+ - n'_-$. The experimental curve of Fig. 6 is similar to the theoretical curves of Fig. 5. Other than the theory of supplementary light waves, we see no possible explanation for the observed two waves of like polarization.

Another crystal in which it was possible to observe experimentally the supplementary light waves predicted by the theory is cuprous oxide. The first (with longest wavelength) absorption band of the yellow ex-

citon series was investigated. This band is forbidden in the dipole approximation, and consequently formula (12) does not apply to it. The theory of supplementary waves for the case of a dipole-forbidden line is developed in detail for cubic crystals in [20,21]. It is shown there, in particular, that if the light propagates, along the (1, 1, 0) direction, it breaks up into waves that can have only two polarizations: 1) with electric field parallel to the (0, 0, 1) direction, and 2) with electric field parallel to the (1, −1, 0) direction. At one of these polarizations the light wave ignores completely the considered exciton—it does not interact with the exciton, it is not absorbed, and no peak is produced on the dispersion curve in the corresponding frequency region. Nor do supplementary waves arise. If the Cartesian y axis is oriented along this polarization, we obtain relations of the type (13) for such a wave:

$$E_{0x} = E_{0z} = 0, \quad E_{0y} \neq 0, \quad n = \sqrt{\gamma'}. \tag{17}$$

In a cubic crystal γ' is a constant.

In the case of the second of the polarizations, the light wave interacts with the exciton, becomes absorbed, albeit weakly (dipole-forbidden band), and a weak peak should appear on the dispersion curve. In lieu of (12) we obtain

$$E_{0y} = E_{0z} = 0, \quad E_{0x} = E_{\pm} \neq 0,$$
$$n_{\pm}^2 = \frac{1}{2}(\gamma' + b_s + \mu_s) \pm \sqrt{\frac{1}{4}(\gamma' + b_s - \mu_s)^2 + b_s\mu_s}. \tag{18}$$

Consequently, two waves with this polarization should exist—the supplementary wave appears; b_S is a constant considerably smaller than B_S (which is contained in formula (12) for the dipole-forbidden bands). As before, μ_S is determined by (11).

In a cubic crystal there can also exist exciton bands for which waves of either type (17) alone or of type (18) alone exist in both polarizations. But these do not include the already mentioned investigated excited band in cuprous oxide.

The experiments on the longest-wave band of the yellow exciton series in cuprous oxide have fully confirmed the theoretical predictions. Thus, independently of the development of the theory, it was observed first in [22] and then in [25] that a wave with polarization 1) is absorbed, whereas a wave with polarization 2) is not absorbed at all. This fact, which agrees completely with our theory, allows us to conclude that the exciton band belongs to the F_2, B′, or E′ symmetry in the group-theoretical classification.[20,27] In all three cases theory predicts that two waves of type (18) should be observed in the case of polarization 1), while one wave of type (17) should appear for polarization 2). To check this, a special experiment was set up,[26] analogous to that with anthracene. The existence of two waves of like polarization was proved by the oscillations present in the dependence on the intensity of the transmitted light and on the thickness of the cuprous-oxide slab. The results of the experiments are shown in Fig. 7. The ordinates represent the absorption coefficient in the line, K_l, after subtracting the background absorption coefficient; $K_l \equiv (1/l) \ln [J_0/J(\lambda)]$ cm^{-1} (J_0 —intensity of the transmitted light at a frequency alongside the line, and J the intensity of the transmitted light with frequency falling inside the line).

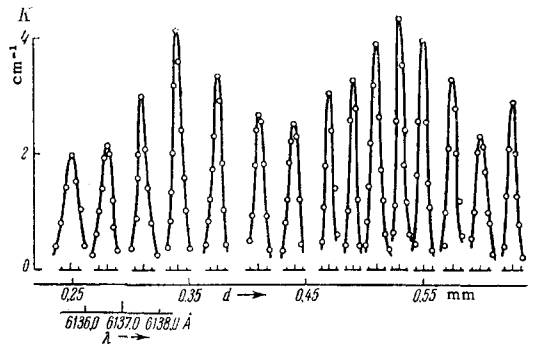

FIG. 7. Experimentally observed dependence $K = (1/l) \times \ln [J_0/J(\lambda)]$ on thickness of the slab. Cuprous oxide, 93° K.

The abscissa axis represents the thickness of the crystal slab. Each peaked curve represents the line shape $K(\lambda)$ at a thickness corresponding to the abscissa of the maximum of the peak. Figure 7 contains also a scale for the wavelengths at which the contour of the band was measured. The figure shows the oscillations of J as a function of the thickness of the slab, demonstrating the presence of two waves with polarization 1). No such oscillations with polarization 2) were observed, indicating that only one wave exists, in accord with the theory.

The observed period of oscillations, ≈ 0.2 mm, cannot be attributed to interference of the waves multiply reflected from the surfaces of the slab. The latter would result in a period on the order of 0.6×10^{-3} mm.

We note that the observed total polarization of the absorption band, and also the presence of an additional wave in polarization 1) and its absence in polarization 2), contradict that version of the theory[6] in which the spatial dispersion is introduced arbitrarily by representing $\epsilon_{ij}^{-1}(\omega, \mathbf{k})$ in the form of a polynomial in k_x, k_y, and k_z. The reason for this contradiction lies in the fact that in most cases neither ϵ_{ij} nor ϵ_{ij}^{-1} are analytic functions of k_x, k_y, and k_z (for more details see [37]) and cannot be represented by polynomials.

An account of spatial dispersion, in addition to leading to supplementary waves, leads also to ordinary birefringence in cubic crystals. The latter was considered theoretically in [1,38-40,6,18] and in other works. This birefringence should be more clearly pronounced near the exciton absorption bands. Apparently, however, it will be easier to observe experimentally in dipole-allowed bands. The possibilities and conditions

favorable for an experimental detection of birefringence of cubic crystals were considered in [18].

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Translated by J. G. Adashko