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Abstract

Full Text

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COMPLEX BIREFRACTION IN SINGLE CRYSTALS OF CUPROUS OXIDE

(Presented by Academician I. V. Obreimov, May 18, 1961)

Until recently it was believed that, in the absence of deformations, complex refraction of light can occur only in anisotropic crystals. In crystals with cubic lattice symmetry, according to the concepts of classical crystal optics, complex refraction should be absent, and the structure of the intrinsic absorption spectra should not reveal any dependence on the direction of propagation or on the polarization of the light waves.

The theory ([1,2]), developed with allowance for spatial dispersion, predicts that in cubic crystals as well, in the region of exciton absorption bands, complex refraction may occur. It has been found experimentally that in single crystals of cuprous oxide the longest-wavelength line of the discrete absorption spectrum is anisotropic with respect to the direction of propagation of light and its polarization ([3]).

Below we present experimental data that reveal a nonmonotonic dependence of the absorption on the thickness of a Cu_2O crystal. Similar experiments were previously carried out on single crystals of anthracene ([4]), which do not belong to the cubic crystal system. The authors regard the data obtained as experimental proof of the existence in an anthracene crystal, in the region of exciton absorption, of additional waves predicted by the theory of spatial dispersion.

Fig. 1. Scheme of the passage of light through a Cu_2O crystal

To measure the absorption curve of the line assigned to the yellow exciton series of Cu_2O, to which the number $n = 1$ is assigned in the hydrogen-like spectrum ([5]), a Fabry-Perot interferometer coupled to an ISP-51 prism spectrograph was used. Details of the operation of the apparatus have been described earlier ([6]). The measurements were performed in polarized light on single-crystal plates that were cooled to a temperature of -180° in a cryostat.

The scheme of the passage of light through the crystal is shown in Fig. 1. Here

Fig. 2. Oscillatory dependence of absorption for an exciton line of cuprous oxide on the thickness of a single-crystal plate

Figure 2: Fig. 2. Oscillatory dependence of absorption for an exciton line of cuprous oxide on the thickness of a single-crystal plate

abcd is a Cu_2O plate cut so that its large faces are parallel to the (110) plane of the cube; the direction of propagation of the light coincides with ML. If the light wave is polarized so that the oscillations of the electric vector $E_{0\perp}$ coincide with the direction of the second-order axis C_2, then the line under investigation is not observed in the absorption spectrum. In this case the attenu—

the intensity of the beam in the spectral region corresponding to the line under study is the same as in the adjacent regions of the step (^{6}), within which the absorption coefficient shows no dependence on the character of the polarization of the light or on the thickness of the plate. The amplitude of the electric vector of the wave that has passed under these conditions through the crystal is denoted by $E_\perp$.

In the case of oscillations of the electric vector $E_{0\|}$, parallel to the fourth-order axis C_4, a sharp absorption line is observed in the absorption spectrum against the background of the step. Thus, when light propagates along the direction of the second-order axis of a Cu_2O crystal, the longest-wavelength exciton absorption line is completely polarized.

Fig. 2. Oscillatory dependence of absorption for an exciton line of cuprous oxide on the thickness of a single-crystal plate

Measurements of the absorption curves corresponding to the line under study as a function of the plate thickness were carried out for the polarization $E_{0\|}$. The absorption coefficients were calculated from the results of microphotometry by means of the formula

$$k_\nu = \frac{1}{d} \lg \frac{I_0(\nu)}{I(\nu)}, \tag{1}$$

where d is the thickness of the crystalline plate; $I_0(\nu)$ is the intensity of the light that has passed through the crystal next to the line; $I(\nu)$ is the intensity of the light that has passed through the crystal next to the line; $I(\nu)$ is the intensity of the light within the line under study.

The results of the measurements are shown in Fig. 2, where the ordinate axis gives the values of the absorption coefficient in decimal reciprocal centimeters, and along the abscissa axis is the thickness of the crystalline plate. Each peak-shaped curve represents the contour of the line under study for the crystal thickness corresponding to the abscissa of the peak maximum. The figure also gives a wavelength scale over the range in which the line contour was measured.

The intensity of light absorption within the line under study, just as its half-width, displays a nonmonotonic dependence on the crystal thickness, which indicates that the Lambert-Bouguer law, expressed by relation (1) and used for calculating the absorption coefficients, is not applicable here. The appearance of oscillations is probably connected with interference of waves which, upon propagation in the crystal, acquire a path difference depending on the thickness. The observed period of the oscillations cannot be explained on the basis of the assumption of interference of multiply reflected waves in the pre-

within the crystalline layer, since the refractive index of Cu_2O in the region studied has a value of about 3, which should make the period of the oscillations three orders of magnitude smaller than that observed. Evidently, the oscillations are connected with the interference of waves propagating in the crystal with different velocities.

If two waves with different complex wave vectors $k_1 = k_1^0 + ik_1'$ and $k_2 = k_2^0 + ik_2'$ propagate in the medium, then, according to (4), the intensity of the light emerging from the plate should be determined by the relation

$$I \sim |a|^2 e^{-2k_1' d} + |b|^2 e^{-2k_1' d} + 2|(ab^*)| e^{-(k_1'+k_2')d} \cos\left[(k_1^0 - k_2^0)d + \alpha_0\right], \quad (2)$$

where a and b are the initial amplitudes of the two waves; d is the thickness of the crystalline plate; α_0 is the initial phase difference. In the case of ordinary birefringence, the oscillations of the electric vector of the ordinary and extraordinary waves occur in mutually perpendicular planes. Therefore, if the plane of polarization of the light wave is parallel or perpendicular to the principal plane of the crystal, then $|(ab^*)| = 0$, and the intensity of the light passing through the crystal should decrease monotonically as the thickness increases. A nonmonotonic dependence of absorption on the thickness of the crystal should be observed when $|(ab^*)| \neq 0$.

To explain the experimental results obtained, it is necessary to assume that in a Cu_2O crystal, in a direction parallel to the second-order symmetry axis C_2, at least two light waves propagate with different velocities, and that they have the same directions of oscillation of the electric vector, oriented parallel to the fourth-order axis C_4.

The appearance of oscillations in this case must be connected with the interference of waves which, on leaving the crystal, have electric-vector amplitudes $E_{\parallel}^{+}$ and $E_{\parallel}^{-}$ (Fig. 1).

On the basis of the foregoing, the following may be regarded as established: first, complex refraction of light can also occur in the case of cubic crystals; second, in a Cu_2O crystal, when light propagates along the second-order axis, two waves with different propagation velocities are observed, but with identical polarizations, for which the oscillations of the electric vectors occur in a direction perpendicular to the direction of the C_2 axis.

Both results find no explanation within the framework of ordinary crystal optics and agree with a theory that takes account of the effects of spatial dispersion (7). The exciton absorption line investigated in Cu_2O is a very convenient object for such observations, since the optical transition corresponding to it is forbidden in the dipole approximation, and it is observed owing to the effects of spatial dispersion.

In conclusion, the authors consider it a pleasant duty to express their gratitude to Yu. I. Gritsenko for kindly providing single crystals of cuprous oxide, and also to Prof. A. A. Shishlovsky and Prof. S. I. Pekar for their interest in this work.

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