

Soviet-era science, translated into English

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1961

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Abstract

Full Text

PHYSICS

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QUADRUPOLE ABSORPTION COEFFICIENT AND OPTICAL LIFETIME OF THE GROUND STATE OF EXCITONS IN A Cu_2O CRYSTAL

In our works ^(1,2) the phenomenon of optical anisotropy of cubic crystals, caused by the effect of spatial dispersion, was discovered. This phenomenon was observed in the exciton absorption spectrum of Cu_2O when studying its longest-wavelength line $\lambda 6125 \text{ \AA}$ ($T = 77^\circ \text{ K}$), usually associated with excitation of the ground state of excitons (the member $n = 1$ of the yellow hydrogen-like series ⁽³⁾). We established that absorption in this line depends on the direction of propagation of light in the cubic lattice of Cu_2O and is anisotropic with respect to intensity and polarization state. The anisotropy of absorption of the line $\lambda 6125 \text{ \AA}$ was interpreted by us as a consequence of the quadrupole character of the optical transition upon excitation of the ground state of excitons. Study of the spatial distribution of absorption showed that this distribution corresponds to absorption of light by three incoherent "plane" quadrupoles oriented normal to the three cubic axes of the crystal ^(1,2). In work ⁽⁴⁾ it was shown that such a form of quadrupole absorption, observed in Cu_2O , corresponds to a quadrupole transition theoretically possible in crystals of the group O_h from the ground state of the crystal (Γ_1) to a triply degenerate exciton level whose wave functions form the basis of the irreducible representation Γ'_{25} . Theoretically, the intensity of quadrupole absorption for the transition $\Gamma_1 \rightarrow \Gamma'_{25}$ as a function of the direction of propagation of light in the crystal lattice (in a spherical coordinate system associated with the triad of cubic axes of the crystal, according to Fig. 2 of work ⁽²⁾) is given by the expressions

$$K_{\parallel}(\theta, \varphi) = K_0 (1 - \sin^2 2\varphi \sin^2 \theta),$$

$$K_{\perp}(\theta, \varphi) = \frac{1}{4} K_0 (\sin^2 2\varphi \sin^2 2\theta + 4 \cos^2 2\theta), \quad (1)$$

where K_{\parallel} and K_{\perp} are the coefficients of light absorption in polarization states with oscillations of the electric vector respectively in the meridional plane and perpendicular to it, and K_0 is a constant coefficient.

Fig. 1. Exciton absorption line $\lambda 6125 \text{ \AA}$

Figure 1: Fig. 1. Exciton absorption line $\lambda 6125 \text{ \AA}$

The results presented in ⁽²⁾ of a qualitative investigation of polarized absorption in the line $\lambda 6125 \text{ \AA}$ for the most characteristic directions L of light passage in the Cu_2O lattice (along symmetry axes of different order $L \parallel C_4$, $L \parallel C_3$, and $L \parallel C_2$, and when varying the direction L in the crystallographic planes of the cube and the rhombododecahedron), generally speaking, convincingly show that the observed spatial distribution of absorption can be described by formulas (1). The assignment of the line $\lambda 6125 \text{ \AA}$ to the quadrupole transition $\Gamma_1 \rightarrow \Gamma'_{25}$ is also confirmed by subsequent experiments on the study of splitting of the line under deformation of crystals ⁽⁵⁾ and by a magnetic field ⁽⁶⁾. However, in view of the fundamental significance of the optical anisotropy of cubic crystals—so far experimentally established only in a single case, Cu_2O —

Of great interest is a **quantitative** investigation of the spatial distribution of the absorption coefficient of the line $\lambda 6125 \text{ \AA}$. Such an investigation is important both for comparison with the theoretical formulas (1) and for obtaining the value of the absorption coefficient in single crystals, since the existing measurements of the absorption magnitude of the anisotropic line $\lambda 6125 \text{ \AA}$ refer to polycrystals ^{(7)*}. Measurement of the absorption coefficient in the line is of interest for determining the oscillator strength of the quadrupole transition and the lifetime of the excited state.

Fig. 1. Exciton absorption line $\lambda 6125 \text{ \AA}$ for propagation of light in the crystal lattice of Cu_2O along C_4 (a) and C_3 (b). Thickness of the single crystals $d = 1.27 \text{ mm}$ (a) and $d = 1.35 \text{ mm}$ (b).

In the present note we give the results of a quantitative investigation of the light-absorption coefficients of the line $\lambda 6125 \text{ \AA}$ when light passes along the symmetry axes of the 4th and 3rd orders ($L \parallel C_4$, $L \parallel C_3$), when the absorption in the line is not polarized. Determination of these “isotropic” ($K_{\parallel} = K_{\perp}$) coefficients $K(C_4)$ and $K(C_3)$ is important for comparison with theory, since, according to formulas (1), the ratio of the coefficients for $L \parallel C_4$ ($\theta = 0$) and $L \parallel C_3$ ($\varphi = \pi/4$, $\theta = \arccos 1/\sqrt{3}$) should be equal to $K(C_4)/K(C_3) = 3$. The absence of absorption polarization for $L \parallel C_4$ and $L \parallel C_3$ also simplifies the experiment and reduces the errors in measuring the absorption coefficients.

The measurements were carried out on plates cut from a single-crystal block of Cu_2O , oriented in such a way that their polished planes coincided with the crystallographic planes of the cube and the octahedron. For the case $L \parallel C_4$, two plates of thickness $d = 0.69 \text{ mm}$ and $d = 1.27 \text{ mm}$ were used; for $L \parallel C_3$, plates of thickness $d = 1.05 \text{ mm}$ and $d = 1.35 \text{ mm}$ were used.** Unpolarized light from a stabilized incandescent lamp fell normally on the plates, which were at $T = 77^\circ \text{ K}$, and, consequently, propagated in the crystals along the C_4 or C_3 axes.

* In note (7) the contour of the line $\lambda 6125 \text{ \AA}$ is given, measured in polarized light on a Cu_2O single crystal for one direction of light propagation, which is not specified exactly. We note that the measurements, made in (8), of almost completely polarized linear absorption of $\lambda 6125 \text{ \AA}$ for many different positions of the polarization plane of the incident light are unnecessary, capable of giving, without adding anything new, only a trivial result.

** The authors express their gratitude to V. T. Agekyan for assistance in preparing the cuprous-oxide samples.

Measurements of the transmission spectra of the samples were carried out with a DFS-12 spectrometer, used as a single monochromator (dispersion at the output $\sim 9.5 \text{ \AA/mm}$, slit width 0.01–0.02 mm). The spectral transmission curve of Cu_2O in the region of the line $\lambda 6125 \text{ \AA}$ was automatically recorded on the tape of the spectrometer's electronic potentiometer. Fig. 1 gives the original recording curves of the spectral absorption in the line $\lambda 6125 \text{ \AA}$ for two single crystals of different orientation ($L \parallel C_4$ and $L \parallel C_3$), having approximately the same thickness. From the curves it was possible to calculate the values (in inverse centimeters) of the absorption coefficient K in the line (relative to the surrounding background line of continuous absorption)*. In addition to the absorption coefficient at the line maximum K_{max} , the integral absorption coefficient $S = \int K(\nu) d\nu$, summed over the entire line contour, was also measured. The results of determining these quantities for the cases of light propagation along the crystal axes C_4 and C_3 are given in Table 1. The table also gives the half-width of the line $\delta\nu$, defined as $\delta\nu = S/K_{\text{max}}$.

Table 1

	$K_{\text{max}}, \text{cm}^{-1}$	$S, \text{cm}^{-1} \cdot \text{cm}^{-1}$	$\delta\nu, \text{cm}^{-1}$
$L \parallel C_4$	5.0 ± 0.2	17.6 ± 1.0	3.5
$L \parallel C_3$	1.7 ± 0.1	5.6 ± 0.4	3.3

From the found values of the absorption coefficients for $L \parallel C_4$ and $L \parallel C_3$, their ratio was calculated. The ratio of the absorption coefficients at the line maximum gives $K_{\text{max}}(C_4)/K_{\text{max}}(C_3) = 2.9 \pm 0.3$. The ratio of the integral absorption coefficients is $S(C_4)/S(C_3) = 3.1 \pm 0.4$. These experimental values are close to the theoretical value of the ratio, equal to 3, which occurs in the case of the quadrupole transition $\Gamma_1 \rightarrow \Gamma'_{25}$. Thus, the results of the quantitative study of the absorption of the line $\lambda 6125 \text{ \AA}$ convincingly confirm the conclusion that the line $n = 1$ is associated with a quadrupole-type transition $\Gamma_1 \rightarrow \Gamma'_{25}$, for which the spatial distribution of absorption is given by formulas (1).

With the aid of formulas (1), one can calculate that “average” absorption coefficient \bar{K} which should be observed, for example, in spectra of polycrystalline Cu_2O samples under complete averaging of the anisotropic absorption due to the different orientation of the individual single crystallites relative to the light beam. Obviously,

$$\bar{K} = \frac{1}{8\pi} \int_0^\pi \int_0^{2\pi} [K_{\parallel}(\theta, \varphi) + K_{\perp}(\theta, \varphi)] \sin \theta d\theta d\varphi.$$

Substitution of the expressions for K_{\parallel} and K_{\perp} from (1) and integration over the sphere gives $\bar{K} = 3/5 K_0 = 3/5 K(C_4)$. Similarly, the average integral absorption coefficient is $\bar{S} = 3/5 S(C_4)$. Substituting here the experimentally found values $K(C_4)$ and $S(C_4)$, we obtain $\bar{K} = 3.0 \text{ cm}^{-1}$ and $\bar{S} = 10.6 \text{ cm}^{-1} \cdot \text{cm}^{-1}$. These quantities agree satisfactorily with the values measured in (7) on polycrystalline Cu_2O samples: $K = 2.53 \text{ cm}^{-1}$ ($T = 96^\circ \text{ K}$) and $S = 8.3 \text{ cm}^{-1} \cdot \text{cm}^{-1}$ ($T = 77^\circ \text{ K}$).

Using the value of the average integral absorption coefficient found by us,

$$\int \bar{K} d\nu = 10.6 \text{ cm}^{-1} \cdot \text{cm}^{-1},$$

one can, from formula (9)

$$\int K d\nu \simeq 10^{-13} \frac{(n^2 + 2)^2}{n} Nf,$$

determine the oscillator strength of the optical transition. Calculated per one unit cell of the Cu_2O crystal lattice, with $1/N = (4.25 \text{ \AA})^3$, the oscillator strength is $f = 3 \cdot 10^{-10}$. As can be seen, in order of

* The calculation of K was carried out by the formula $K = \frac{1}{d} \ln \frac{I_\phi}{I}$, where I_ϕ is the background intensity on the transmission curves, measured from zero transmission, and I is the intensity in the line. This determination made it possible not to take into account the reflection of light at the crystal surfaces, which is weak in the region of the line and nonselective.

in magnitude it is close to the usual values for quadrupole transitions. Knowing f , one can calculate the lifetime τ of the corresponding excited state with respect to spontaneous emission

$$\left(\frac{1}{\tau} = \frac{8\pi^2 e^2 \nu^2}{mc^3} f \right).$$

It is equal to $\tau \approx 20 \text{ sec}$. Thus, the “optical” lifetime of the ground state of an exciton in the Cu_2O lattice turns out to be extremely large. This explains the observed absence of exciton luminescence in Cu_2O : the excitons annihilate through more probable nonradiative mechanisms, which apparently determine the actual lifetime of the exciton excitation in the Cu_2O lattice. Taking into account the insufficiency of the theoretical investigation of the question, the

values obtained for f and τ should apparently be regarded as estimates of the orders of magnitude of these quantities.

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Received
17 II 1961

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