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PHYSICS

1965

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Abstract

Full Text

UDC 548.0:535+535.343

PHYSICS

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CONTOUR OF EXCITON ABSORPTION LINES IN A CUPROUS OXIDE CRYSTAL

(Presented by Academician B. P. Konstantinov, 12 IV 1965)

In solid-state spectroscopy the cuprous oxide crystal occupies a place as important as the hydrogen atom in atomic spectroscopy. Many works have been devoted to a comprehensive study of the optical properties of Cu_2O (see ⁽¹⁾). However, such fundamental quantities as the absorption coefficient, half-width, oscillator strength, and also the line contour have been studied insufficiently. At the same time, it is precisely for Cu_2O that the theory of the contour ^(2,3) and the absorption magnitudes ^(4,5) of exciton lines have been developed in greatest detail.

Fig. 1. Contour of exciton lines of the “yellow” series of cuprous oxide: a– $T = 160^\circ \text{ K}$; b– $T = 4.2^\circ \text{ K}$

In the long-wavelength edge region of the fundamental absorption of the Cu_2O crystal, two groups of hydrogen-like ($n \geq 2$, n is the quantum number) absorption lines are observed, which by their position in the spectrum are usually called the “yellow” and “green” series and are located in the regions 6150–5700 and 5500–5400 Å, respectively.

Using a recording spectral setup with dispersion $D = 3.2 \text{ \AA/mm}$ at temperatures T equal to 160 and 4.2° K , we investigated the spectral distribution of the absorption coefficient of mono- and polycrystals of Cu_2O (crystal thickness $d = 5\text{--}300 \mu$) ^(6–9). The exciton lines rise above the background of continuous absorption of indirect band-to-band transitions; the absorption in the background is approximately the same as in the lines. The monotonic course of the continuous background outside the lines was extrapolated by us into the region of the lines by a smooth curve under the assumption that the lines do not overlap one another. The extrapolation of the continuous background is reliable in the region

Fig. 2 and Fig. 3

Figure 2: Fig. 2 and Fig. 3

of the “yellow” series and less reliable (because of the large half-width of the lines) in the region of the “green” series.

In the spectral distribution of absorption of very perfect single crystals of Cu_2O we have found lines of the “yellow” series with $n = 1, 2, 3, 4, 5, 6$ at $T = 160^\circ \text{ K}$ (Fig. 1a) and with $n = 1, 2, 3, 4, 5, 6, 7$ at $T = 4.2^\circ \text{ K}$

Fig. 2. Contour of the exciton lines of the “green” series of cuprous oxide: $a-T = 160^\circ \text{ K}$; $b-T = 4.2^\circ \text{ K}$

Fig. 3. Dependence of the logarithm of the area of the contour of the lines of the “yellow” series on the square of the quantum number: $a-T = 160^\circ \text{ K}$; $b-T = 4.2^\circ \text{ K}$

(Fig. 1b), and also lines of the “green” series with $n = 2, 3$ at $T = 160^\circ \text{ K}$ (Fig. 2a) and with $n = 2, 3, 4$ at $T = 4.2^\circ \text{ K}$ (Fig. 2b) (see Tables 1 and 2; K_{\max} —absorption at the maximum; $\Delta\nu$ —half-width; $\delta = (2\nu_{\max} - \nu_- + \nu_+) : (\nu_+ - \nu_-)$ —asymmetry of the bands, where ν_{\max} and ν_- , ν_+ are the frequency values at the band maximum and at the half-width level, respectively; f —oscillator strength).

Table 1

Values of K_{\max} (cm^{-1}), $\Delta\nu$ (cm^{-1}), δ , and $f \cdot 10^6$ for the lines of the “yellow” and “green” series

n	$\lambda, \text{ \AA}$	$\nu, \text{ cm}^{-1}$	K_{\max}	$\Delta\nu, \text{ cm}^{-1}$	δ	f
“Yellow” series, $T = 4.2^\circ \text{ K}$						
2	5770.8	17 328.6	172	26.3	0.266	1.77
3	5735.0	17 436.8	238	8	0.162	0.98
4	5722.8	17 473.9	248	4.1	0.11	0.38
5	5716.8	17 492.3	141	2.9	0.07	0.125
6	5713.1	17 503.6	65	2.3	0	0.042
7	5711.3	17 509	38	1.15	0	0.013
“Yellow” series, $T = 160^\circ \text{ K}$						
1	6125.3	16 325.7	3	4	0	$5 \cdot 10^{-5}$

n	$\lambda, \text{Å}$	ν, cm^{-1}	K_{max}	$\Delta\nu, \text{cm}^{-1}$	δ	f
2	5792.7	17 263.2	220	33	0.364	2.48
3	5756.6	17 371.2	210	13	0.154	0.87
4	5743.8	17 410.1	120	10.4	0.05	0.32
5	5738.1	17 427.5	38	6.7	0	0.067
6	5734.1	17 439.4	7	5.5	0	0.018
“Green” series,						
$T = 4.2^\circ \text{K}$						
2	5469	18 285	1400	110	0.255	65.6
3	5419	18 454	510	50	0.20	8.4
4	5402	18 512	110	?	?	1.6
“Green” series,						
$T = 160^\circ \text{K}$						
2	5496	18 195	800	235	0.575	60.6
3	5444	18 369	143	77	?	3.7

The main features of the line contours are as follows: 1) all lines of the “green” series and many lines of the “yellow” series are strongly asymmetric, with the asymmetry of the lines decreasing sharply with increasing n ; according to the theory [2], this indicates that the character of the interaction of excitons with phonons changes with increasing n from weak to strong, while the exciton energy bands have a maximum at $K = (0, 0, 0)$; 2) lowering the temperature from 160 to 4.2°K leads, for the “yellow” series, to a strong, complex, and different change in the absorption intensity for different lines; the regularity empirically found by us, $\lg f \sim -n^2$, differs sharply from the theoretically predicted $f \sim (n^2 - 1) : n^5$ [4, 5] (Fig. 3 presents the dependences of the logarithms of the areas of the line contours S_n on n^2 ; $S_n \sim f$); 3) the values of K_{max} upon lowering the tem-

peratures increase strongly for $n > 3$ and change little for $n = 2$ and 3; the small value of K_{max} characterizes the optical transitions of the “yellow” and “green” exciton series, respectively, as forbidden and weakly allowed, in agreement with the theory ⁽⁴⁾.

In ^(10–12), at T equal to 77.3, 20, and 4.2°K , the line contour of the “yellow” series with $n = 2, 3, 4$ and of the “green” series with $n = 2, 3$ of polycrystalline Cu_2O was studied (see also ⁽¹³⁾). A careful comparison of our results with the data of ^(10–12) shows the following: 1) the values of K_{max} coincide in the different works at $T \geq 77.3^\circ \text{K}$, but at $T = 4.2^\circ \text{K}$ and $n > 3$ they differ considerably;

2) the values of $\Delta\nu$ according to ^(11,12) are considerably smaller than our $\Delta\nu$; asymmetry was found in ^(11,12) only on the lines with $n = 2$ and 3 ; 3) the values of f in ^(11,12) agree with the theory ⁽⁴⁾ owing to the failure in ^(11,12) to detect the asymmetry of many lines. It is necessary to emphasize the great importance, for studies of line contours, of the dispersion of the spectrometer and the quality of the samples*.

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Received
8 III 1965

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* The dispersion of Gorban' s spectrometers ⁽¹⁰⁾ is smaller than ours by approximately a factor of 10, and that of Nikitine, Grun, and Siskind ^(11,12) by a factor of 5. It is possible that many discrepancies between the results of works ⁽¹⁰⁻¹²⁾ and our data arise because of insufficient dispersion and the polycrystalline nature of the Cu₂O samples in works ⁽¹⁰⁻¹²⁾.

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