Investigation of the role of damping in absorption of light by excitons

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It is observed that integral absorption of light in the region of quadrupole exciton transition in Cu_2O tends to zero with decreasing temperature. This leads to violation of the classical relation between the area under the absorption curve and the oscillator strength of the transition. The mechanism of the observed phenomenon is discussed.

One of the basic problems in exciton physics is that of the mechanism whereby light is absorbed by crystals near frequencies that resonate with the excitation energy of the exciton. According to present-day concepts[1,2] there should be no absorption if there are no exciton-scattering processes. On the other hand, at sufficiently large damping, the classical relation between the area under the absorption curve and the exciton-transition oscillator strength should be satisfied. There should therefore exist a range of damping parameters in which the decrease of the parameter is accompanied by a decrease of the integral absorption. The relation indicated is violated in this region, which can be called the region of "nonclassical" absorption. This result was recently obtained theoretically in[3].

It appears that the present study provides the first direct confirmation of the existence of such a "nonclassical" absorption.

Experimental procedure and results. We investigated the temperature dependence of the integral absorption of the half-width of the $n=1$ quadrupole line in a Cu_2O crystal. Inasmuch as the manifestation of the "nonclassical" absorption can be expected only at low values of the damping, the quality of the samples played a decisive role. The selected samples having a narrow $n=1$ line, and by the same token reduced greatly the contribution of the defects and impurities to the damping. From the methodological point of view, we investigated also crystals in which the $n=1$ line was broadened. It turned out that at $T=4.2$ °K the $n=1$ line of good samples is very narrow. Therefore investigations at low temperatures were carried out with the aid of a Fabry-Perot etalon, which was crossed with a DFS-12 spectrometer. Figure 1a shows a plot of a section of the continuous spectrum, while Fig. 1b shows a typical plot of the $n=1$ absorption line at $T=4.2$ °K. Figure 1c shows for comparison the He-Ne laser emission line. At $T>35$ °K the line contour was investigated with the aid of the DFS-12 spectrometer.

Figure 2 shows experimental plots of the temperature dependence of the integral absorption and of the half-width of the $n=1$ line. Curves a and b pertain to a good crystal (No. 1) with a narrow absorption line. We investigated only crystals of this type and established that the half-width and the integral absorption behave quite identically in these crystals. Curves e and f represent the approximate course of the integral absorption in crystals Nos. 2 and 3, in which the half-width of the $n=1$ line was 0.8 and 0.4 cm^{-1}, respectively, at $T=4.2$ °K.

Discussion of results. As seen from Figs. 2a, the integral absorption in the $n=1$ line of crystal No. 1 is constant in the temperature interval 120—60 °K and decreases by a factor 4.5 as the temperature is further lowered to 1.8 °K. Thus, at temperatures $T<60$ °K the usual relations between the area under the absorption curve and the oscillator strength should not hold.

A qualitative explanation of the difference in the behavior of the integral absorption in the region of low ($T<60$ °K) and high ($T>60$ °K) temperatures can be obtained by starting from the polariton nature of elementary excitations in the region of exciton resonance. According to Hopfield,[1] the polariton can be represented as a photon that is periodically transformed into an exciton of frequency f:

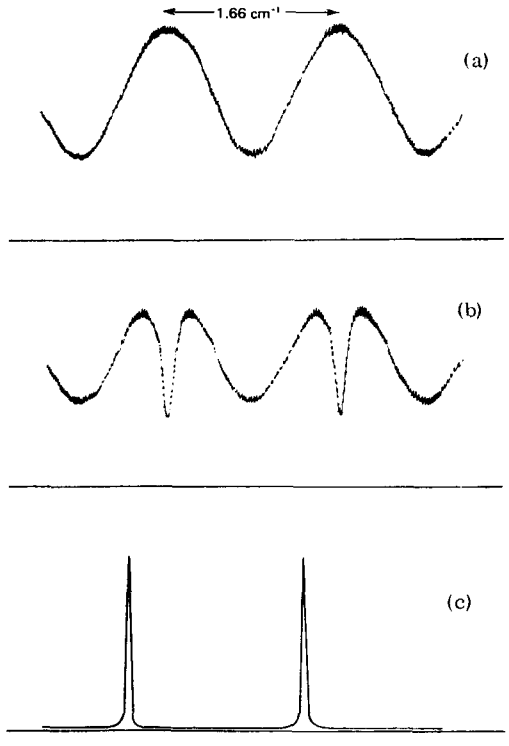

FIG. 1. Spectra recorded with the aid of a Fabry-Perot etalon: a) continuous spectrum, b) $n=1$ absorption line of Cu_2O crystal at $T=4.2$ °K, c) He-Ne laser emission line.

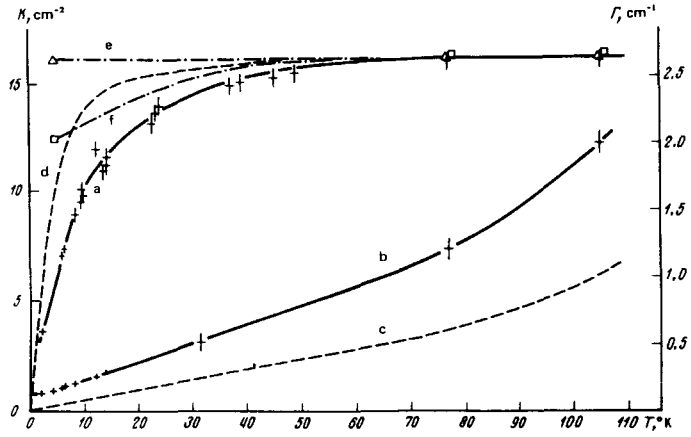

FIG. 2. Temperature dependences of the integral absorption K and of the half-width Γ of the $n=1$ line in Cu_2O: a, b) $K(T)$ and $\Gamma(T)$ for crystal No. 1; c, d) theoretical plots of $\gamma(T)$ and $K(T)$; e, f) $K(T)$ for crystals Nos. 2 and 3.

$$f = \omega_p (\pi F/\epsilon)^{1/2}, \tag{1}$$

where ω_p is the plasma frequency, F is the exciton-transition oscillator strength, and ϵ is the dielectric constant. Polariton scattering will occur only at those instants when the polariton is in the exciton states, so that the criterion for violation of the classical ratio for the integral absorption is the inequality $f \geq \gamma$, where γ is the frequency of the exciton-scattering acts. The estimates that follow are in satisfactory agreement with the experimental results. Indeed, assuming $F = 3\times10^{-10}$,[4] $\epsilon = 7.5$, and $\omega_p = 6.5\times10^{15}$ sec^{-1} we obtain $f = 7\times10^{10}$ sec^{-1} (0.35 cm^{-1}). From the experimental plot in Fig. 2b we see that this value of the damping is realized already at 20 °K. The "nonclassical" character of the absorption is observed also in this temperature region.

It is also of interest to compare our results with the conclusions of[3], where a quantum-statistical method was used to obtain an approximate expression for the coefficient $K(\omega)$ of light absorption by excitons:

$$K(\omega) = \frac{\gamma f^2/2c}{(\omega-\omega_0)^2 + \gamma^2 + f^2/4}. \tag{2}$$

The line half-width is in this case

$$\Gamma = 2\sqrt{\gamma^2 + f^2/4}, \tag{3}$$

and the integral absorption is

$$K = \int K(\omega)\, d\omega = \frac{\pi f^2 \gamma/2c}{\sqrt{\gamma^2 + f^2/4}}. \tag{4}$$

Expression (4) describes both the limiting cases $f \ll \gamma$, when $K = \pi f^2/2c$, and $f \gg \gamma$, when $K = \pi f\gamma/c$, as well as the transition region where $f \sim \gamma$.

Assuming that the experimental temperature dependence of Γ of the $n=1$ line satisfies relation (3), and obtaining $f = 2.4\times10^{10}$ sec^{-1} (0.2 cm^{-1}) and $\gamma(T)$ (Fig. 2c) in this manner, we have plotted, using (4), the theoretical temperature dependence of the integral absorption (Fig. 2d).[1)] In spite of the good agreement between experiment and theory, further research is necessary to draw final conclusions.

It must be emphasized once more that the dependence of the integral absorption on the temperature is clearly pronounced only in good crystals. This is illustrated by the curves e and f, which reflect the behavior of the integral absorption in "poor" crystals Nos. 2 and 3. We note that the observed change in the value of the integral absorption of the exciton-transition line is not connected with spatial-dispersion effects, and is only the consequence of quantum oscillations of the excitation energy between the photon and exciton states. A quite similar problem was posed and solved in papers devoted to a theoretical investigation of nonradiative transitions in crystals with impurities.[6]

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[1)] To obtain agreement between the integral absorptions at high temperatures, it was necessary to introduce the empirical factor 35 in the numerator of (4). The appearance of this factor may be due to the fact that formulas (2)—(4), which were obtained for the dipole case, cannot be used directly for a quadrupole transition.[5]

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