Screening of excitons in cuprous oxide

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The absorption spectrum of Cu_2O at a high density of free carriers produced by two-photon light excitation, is investigated. A successive dissociation of excitons to $n = 3$ and a contraction of the forbidden band as a result of small absolute displacement of exciton lines was observed.

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Several mechanisms are capable of influencing the absorption spectrum of a crystal in which a high carrier density is produced. An exact evaluation of all possible contributions is unrealistic, and for this reason it is not a simple matter to predict the experimental result by summing these contributions. It must be kept in mind that the absolute energy of an exciton resonance is determined by the Coulomb interaction and by the width of the forbidden band, which are functions of the free-carrier density.

Serious basic drawbacks are inherent to many experimental studies of screening produced as a result of excitation of the crystals by light (for example, Refs. 1–5). These studies were performed on the luminescence and reflection spectra of crystals with a direct allowed transition due to excitation in the band $h\nu > \epsilon_g^0$. Under these conditions a layer with a thickness of only 10^{-5}–10^{-6} cm is exicted and the surface properties rather than volume properties are analyzed under excitation conditions that are highly nonuniform with depth. If, however, the absorption spectra of these crystals are examined, then the samples with a thickness of $\leqslant 10^{-4}$–10^{-5} cm must be used for a one-photon excitation that is uniform with depth, and in this case the influence of surface regions becomes important again.

Excluding the surface effects, we can give the following reasons for the change in the absorption spectrum at large n_{eh}: 1) Coulomb-interaction screening—a decrease of the exciton binding energy and oscillator strength; 2) enhancement of the exchange interaction of free carriers; 3) the Burstein-Moss effect; 4) screening of the interaction of carriers with impurities; 5) attenuation of the polaron effect.

To these normally investigated effects we should add the weakening of the momentum selection rules at large n_{eh} and the absorption of light quanta with an energy lower than ϵ_g^0, as is the case, for example, in the interaction of photons with phonons or in the tunneling in an electric field: a large density of hot carriers allows absorption of a quantum with an energy lower than ϵ_g^0 with a simultaneous cooling of one of the hot free carriers.

Cuprous oxide is a convenient material for studying the absorption spectrum produced as a result of strong excitation. The multiple yellow series contains states with radii and binding energies of different orders of magnitude. The small absorption coefficient makes it possible to record the spectrum of thick samples in which the

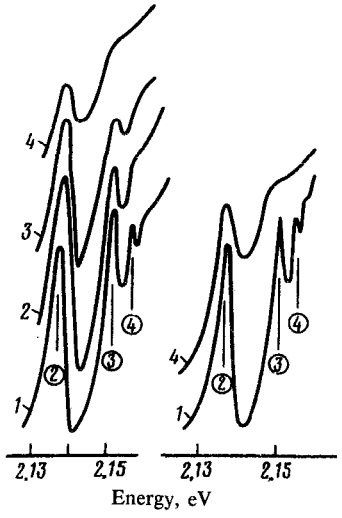

FIG. 1. Screening of the exciton lines as the excitation density increases: 1, $I_0 = 0$; 2, $I_0 = 20$; 3, $I_0 = 40$; 4, $I_0 = 60$ MW/cm^2 (the numbers in circles are the principal quantum numbers n). Two curves for one position of the zero are shown at the right. $T = 4$ K.

surface effects are negligible. Finally, the luminescence and the tails of the impurity absorption are very weak near the absorption edge of the test samples.

The absorption spectrum of Cu_2O ($\epsilon_g^0 = 2.17$ eV) at $T = 4$ and 77 K in our experiment was recorded by pulse synchronous detection. The carriers were generated by using the two-photon method—the first harmonic of a YAG-Nd^{3+} laser, $h\nu = 1.17$ eV; there was almost no absorption in our samples at this frequency. The two-photon absorption coefficient is $K = 0.31_0$ cm^{-1}, where I_0 is the excitation intensity at the surface. The K value is sufficiently large in order to obtain a high density n_{eh}, but at the same time a sample with a thickness of 40 μm can be assumed to be uniformly excited in the bulk at pumping levels less than 60 MW/cm^2.

As I_0 increases, the absorption edge shifts toward longer wavelengths (contraction of the forbidden band) and the successive dissociation of the exciton lines can be observed (Fig. 1). The lifetime of free carriers and excitons, which is estimated from the data on the cyclotron resonance and luminescence kinetics, is close to 10^{-9} sec. Measurements of the incident radiation power density and of the K values make it possible to estimate the n_{eh} values, at which the exciton lines with a certain quantum number n disappear (see Table I).

TABLE I

n	5	4	3
n_{eh} exp	9×10^{4}	8×10^{16}	7×10^{17} cm^{-3}
n_{eh} theor	3×10^{16}	6×10^{16}	1×10^{17} cm^{-3}

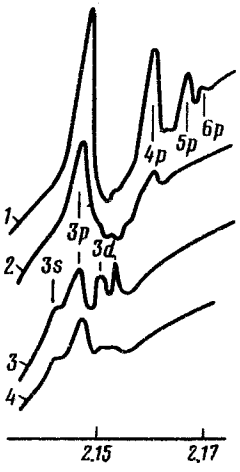

FIG. 2. Exciton screening at external electric field $F = 0$ (curves 1 and 2) and $F = 5$ kV/cm (curves 3 and 4). Excitation density is $I_0 = 0$ (curves 1 and 3) and $I_0 = 20$ MW/cm^2 (curves 2 and 4). $T = 4$ K.

A critical examination of the theoretical studies of the spectrum under screening conditions showed that in our case we should use the results of the work of Egri[6] and Gay.[7] We can see in Table I that for $n = 3$ and 4 the experimental values exceed the theoretical values by several factors. The discrepancy is not surprising if we bear in mind the simple premises of the theory and the errors in the determination of the excitation power and the moment of dissociation. At $n = 5$, however, the theoretical estimate is larger. The reason for this must be the influence of the electric fields of lattice imperfections and the temperature—the boundary of the continuous spectrum in shifted relative to ϵ_g^0 which is calculated from the exciton series, and this, in turn, affects the states with a small binding energy. The influence of the external electric field F on screening is demonstrated in Fig. 2: at $F = 5$ kV/cm the light excitation strongly screens the 3d line, but at $F = 0$ the influence of the same excitation on the line with $n = 3$ is negligible.

Some previous studies showed the presence of a strong shift in the reflection, photo-conductivity, and luminescence exciton maxima with no significant change in the shape of the lines and the oscillator strength. At first glance, this is consistent with the Debye-Hückel screening theory, but the agreement breaks down if the narrowing of the forbidden band is taken into account. The true cause of the shift observed in Refs. 1 and 2 is the change in the depth dependence[8] of the exciton-resonance energy due to illumination of the crystal.

An important result of our experiment is the absence of noticeable shifts of the exciton lines—narrowing of the forbidden band and decrease of the exciton binding energy until it dissociates compensate each other to some extent.

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