Population inversion in sublevels of $n = 1$ excitons of the Cu_2O yellow series in a magnetic field

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(Submitted 29 April 1985)
Pis'ma Zh. Eksp. Teor. Fiz. **41**, No. 12, 514–516 (25 June 1985)

The effect of the excitation conditions on the luminescence spectrum of excitons of a cuprous oxide crystal in a magnetic field has been studied. During resonant excitation of the upper ($M = +1$) component of the Γ_5^+ level, a population inversion among exciton sublevels is observed.

Cuprous oxide is a semiconductor with a direct forbidden transition. The $1S$ exciton ground state of the yellow series, formed by electrons from the Γ_6^+ band and holes from the Γ_7^+ band, is split by an exchange interaction into the triply degenerate Γ_5^+ (an orthoexciton) and the nondegenerate Γ_2^+ (a paraexciton).[1] A transition to the orthoexciton level is allowed in the quadrupole approximation, while a transition to the paraexciton level is forbidden in the dipole and quadrupole approximations. A zero-phonon line of the paraexciton is observed in crystals subjected to a uniaxial deformation[2] or a magnetic field.[3] The Γ_2^+ level lies 96 cm^{-1} below the Γ_5^+ level, and it is obvious that the luminescence of the orthoexciton at liquid-helium temperatures can be observed only as a result of a deviation from a thermodynamic equilibrium between the levels of the orthoexciton and the paraexciton.[4]

In this letter we report a study of the exciton luminescence of a Cu_2O crystal in a magnetic field during resonant excitation. The results show that in the steady state a thermodynamic equilibrium is not established among the sublevels of the Γ_5^+ excitons during resonant excitation.

The Cu_2O crystals are grown by a hydrothermal method. The luminescence is excited by a tunable dye laser whose output line has a half-width ≈ 0.5 cm^{-1}.

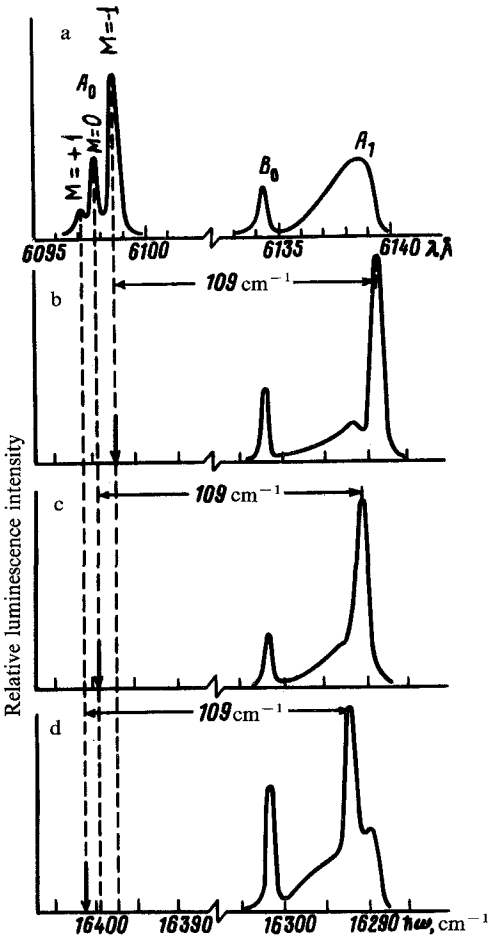

FIG. 1. Effect of the wavelength of the exciting light on the exciton luminescence spectrum in the Cu_2O crystal. Nonresonant excitation: a—$\lambda_e = 6040$ Å, resonant excitation of Zeeman components; b—$M = -1$; c—$M = 0$; d—$M = +1$. Here A_0 and B_0 are zero-phonon transitions of orthoexcitons and paraexcitons, respectively, and A_1 is a repetition of the orthoexciton with a phonon with $\hbar\omega = 109$ cm^{-1}. The arrows show the frequencies of the exciting light. $T = 2$ K, $H = 50$ kOe.

In a magnetic field, the Γ_5^+ level splits into three sublevels with[1] $M = \pm 1.0$. Figure 1a is a part of the luminescence spectrum found, including zero-phonon transitions of the orthoexciton (A_0) and of the paraexciton (B_0) and repetitions of the orthoexciton with a Γ_3^- phonon ($\hbar\omega = 109$ cm$^-$) (A_1), recorded during nonresonant excitation ($\lambda_e = 6040$ Å). We see from Fig. 1a that in a field $H = 50$ kOe at $T = 2$ K, with nonresonant excitation, the intensity of transitions from the lowest Γ_5^+ sublevel ($M = -1$) and, correspondingly, the population of this sublevel are about an order of magnitude higher than those for the highest sublevel, Γ_5^+ ($M = +1$). During resonant excitation, the picture changes radically. In addition to a contradiction of the phonon repetitions,[5] there is a redistribution of their intensities (Fig. 1, b, c, d). The relative

intensities of the transitions are determined in this case not by the level energies but by the excitation conditions. For example, during excitation of the highest sublevel the intensity of the corresponding phonon satellite is twice that corresponding to the lowest level (Fig. 1b). A similar picture is observed if the resonance condition is satisfied for the central component of the Γ_5^+ triplet ($M = 0$; Fig. 1c). A similar redistribution of intensities occurs in the spectrum of phonon repetitions of Γ_4^- ($\hbar\omega = 150$ cm^{-1}).

The Cu_2O crystals grown by the hydrothermal method have a bright exciton luminescence, which is much more intense than the Raman scattering. To verify that resonant Raman scattering was indeed negligible, we carried out some control measurements at the same time as the measurements of the orthoexciton intensity I_0: We measured the intensity (I_p) of the indirect-emission band of the paraexciton, for which the condition for resonant excitation did not hold. We found that the relative intensity I_0/I_p is essentially independent of the excitation conditions (resonant or nonresonant). Consequently, during resonant excitation the change in the band intensity results not from an increase in the Raman intensity but from a redistribution of the luminescence intensity in the spectrum of phonon repetitions of the orthoexciton. A population inversion is accordingly observed during resonant excitation of the upper sublevels (Fig. 1, c and d).

During steady-state excitation of the upper level (with $\Delta E_{12} > kT$), we have $N_2\gamma_{12} = N_1\gamma$, where N_2 and N_1 are the populations of the upper and lower sublevels, respectively, γ_{12} is the probability for a transition between them, and γ is the total probability for the recombination of excitons. We thus find $\gamma_{12} = \gamma N_1/N_2$ or, since $N_1/N_2 = I_1/I_2$, $\gamma_{12} = \gamma I_1/I_2$, where I_1 and I_2 are the luminescence intensities of the corresponding phonon satellites. In a field $H = 50$ kOe at $T = 2$ K, as we have already mentioned, we have $I_1 \approx 0.5 I_2$; we thus have $\gamma_{12} \approx 0.5\gamma$. Substituting the results of the direct measurements of the orthoexciton lifetime in the hydrothermal Cu_2O crystals at liquid-helium temperatures,[6] we find $\gamma_{12} \approx 1.5 \times 10^8$ s^{-1}.

We wish to thank S. V. Pogarev for considerable assistance in this study.

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Translated by Dave Parsons