Cooling of interacting-exciton gas

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(Submitted 10 June 1985)
Pis'ma Zh. Eksp. Teor. Fiz. **42**, No. 2, 63–65 (25 July 1985)

A gas of strongly interacting excitons in CdSe crystals is brought for the first time into a thermodynamic quasiequilibrium with the lattice ($T = 80$ K) by means of selective photoexcitation.

The excitation of a dense exciton gas is usually accompanied by its heating.[1–3] Although the causes of this phenomenon are not yet fully understood, it is clear that superheating of the exciton system prevents the formation of excitonic molecules, Bose-Einstein condensate, and electron-hole liquid. The importance of understanding the mechanism responsible for the heating of excitons and for the control of their effective temperature is therefore clear. In this letter we report the preliminary results on the control of the energy spectrum of a dense exciton system in a semiconductor of the group $A^{\rm II}B^{\rm VI}$ (CdSe) by the method of selective photoexcitation.

A freshly cleaved CdSe single crystal (the lattice temperature is $T = 80$ K, the ground-state energy of the exciton A is $E_0^{1s} = 1.816$ eV, the width of the energy gap is $E_g = 1.832$ eV) is pumped by a beam from an organic-dye nanosecond laser continuously tunable over the range of photon energies $h\nu_0 = 1.79$–2.19 eV (the line width is ~ 0.3 meV). Figure 1 shows several luminescence spectra measured by means of a double monochromator upon excitation by a polarized light ($\mathbf{E}\perp\mathbf{c}$) with different exciting photon energies $h\nu_0$. At higher pump levels, the main spontaneous emission band

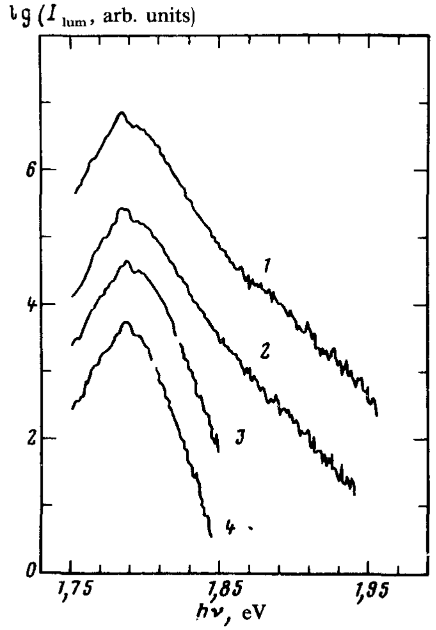

FIG. 1. Luminescence spectra of a CdSe crystal excited by light with various photon energies: $h\nu_0 = 2.1883$ eV (1), 2.0731 eV (2), 1.8223 eV (3), 1.8056 eV (4). The intensity scale of each spectrum is shifted arbitrarily.

with a peak at 1.798 eV results from exciton-exciton interaction (see Ref. 4).[1)] In the energy range $h\nu > E_0^{1s}$, the short-wavelength wing of this band decays exponentially, appropriately representing the effective temperature, T_x, of the excitons.[3] At a constant flux density of the incident photons (6×10^{24} cm$^{-2}\cdot$s^{-1}) the intensity of the given luminescence band remains essentially the same at exciting photon energies $h\nu_0 > 1.804$ eV. We can assert, therefore, that the density of exciton gas is maintained at a constant level. At $h\nu_0 > 2.07$ eV, we can single out in the far short-wave region of the spectra a second exponential wing (spectra 1 and 2 in Fig. 1), resulting from the emission of a hot electron-hole plasma which is trapped at the absorption depth of the exciting light.[5] The effective temperature of the hot surface electron-hole plasma, T_e, decreases rapidly with decreasing energy of the exciting photon ($T_e = 244$ K and 176 K at $h\nu_0 = 2.1883$ eV and 2.0731 eV, respectively), and at $h\nu_0 < 2.07$ eV the second exponential wing is virtually impossible to identify.

Figure 2 is a plot of the effective temperature of the constant-density exciton gas versus the energy of the exciting photon. This plot can be divided into three characteristic regions.

1. The region $h\nu_0 > 1.852$ eV $= E_g + 3kT$ corresponds to the generation of hot electron-hole pairs and possibly excitons whose kinetic energy is much higher than their binding energy. The effective temperature T_x in this case is much higher (by ~ 50 K) than the lattice temperature, although it is virtually independent of the energy of the exciting photon and of T_e which is closely associated with this energy. This behavior is evidently caused by the localization of the rapid initial relaxation of the initial excess energy $h\nu_0 - (E_g + 3kT)$ of the nonequilibrium charge carriers at the depth to which the exciting light penetrates the crystal.[5] Consequently, there is no flow of energy from the "hot" surface of the crystal to the excitons that are trapped at a much greater diffusion depth. However, the heating of excitons which are clearly captured by the cold electron-hole plasma seems to be caused by the dominance of a capture process in which the total energy of an electron-hole pair is completely converted to the total energy of the exciton (i.e., an exciton with a kinetic energy on the order of the binding energy is produced).

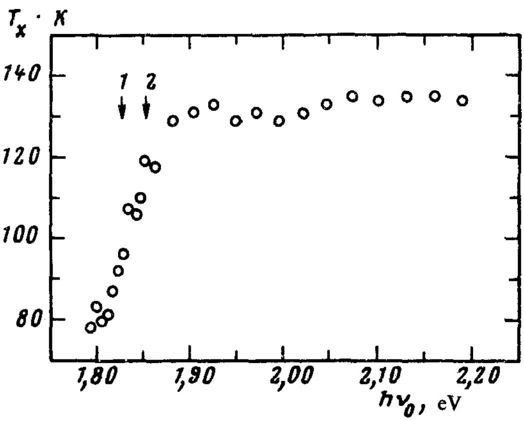

FIG. 2. The effective temperature of excitons versus the energy of the exciting photon at an excitation level 6×10^{24} cm^{-2} s^{-1}. The arrows indicate the energies $E_0^{1s} + \frac{3}{2}kT$ (1) and $E_g + 3kT$ (2).

2. The second region is conventionally bounded by the exciting-photon energies which correspond to the adiabatic excitation of the free electron-hole pairs ($E_g + 3kT = 1.852$ eV) and the excitons ($E_0^{1s} + \frac{3}{2}kT = 1.826$ eV), respectively. Here we have a rapid but incomplete cooling of the exciton system (from 120 K to 92 K), indicating that one of the heating sources in this case is the initial kinetic energy of the exciton. A finite superheating of the excitons due to adiabatic excitation allows us to assume, however, that the radiative exciton-exciton collisions, which are essentially exothermal processes,[3] are the second cause of heating.

3. A total cooling of the exciton gas to the lattice temperature can be achieved by further reducing the exciting-photon energy from $E_0^{1s} + \frac{3}{2}kT$ to an energy slightly lower than E_0^{1s}. The responsibility for such a compensation of the collision mechanism of heating may be borne by the excitation of excitons with a kinetic energy lower than the average energy and also by the process which is the inverse of the radiative exciton-exciton collision, in which a light quantum with an energy $h\nu_0 \lesssim E_0^{1s}$ is absorbed because of a concomitant energy loss by another exciton.

In summary, we were able to determine for the first time by the method of selective photoexcitation the contribution of various processes to the superheating of excitons and we were able to cool a dense exciton system.

[1)]The second emission peak at 1.789 eV, whose intensity strongly depends on the pump level, appears to be superluminescent.

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Translated by S. J. Amoretty