Two-stage energy relaxation of a photoexcited electron-hole plasma in a CdS crystal

R. Baltrameyunas, A. Zhukauskas, V. Latinis, and S. Yurshenas
V. Kapsukas Vil'nyus State University

(Submitted 11 May 1987)
Pis'ma Zh. Eksp. Teor. Fiz. **46**, No. 2, 67–69 (25 July 1987)

Two stages have been found in the cooling of a nonequilibrium electron-hole plasma in the polar semiconductor CdS (the energy relaxation times are $\tau_1 \leqslant 15$ ps and $\tau_2 = 400$ ps). These stages reflect the initial stages of the relaxation to thermodynamic equilibrium in the phonon subsystem of the highly excited crystal.

Several studies (those cited in Ref. 1 and also Refs. 2–4) have established that the energy relaxation of the dense electron-hole plasma, which is excited in a semiconductor by intense picosecond light pulses, occurs considerably more slowly than would be expected on the basis of the elementary models of the electron-phonon interaction. Attempts to explain the nature of the processes which determine the rate of this relaxation in polar crystals (GaAs and CdSe) have run into several difficulties. For example, an attempt to explain the experimental data in terms of the amplification of longitudinal optical (LO) lattice vibrations[1,2] has required adopting relaxation times for the nonequilibrium population of LO phonons (25–60 ps) which are longer than the known values (<10 ps; Ref. 5). Another approach, based on the screening of the electron-phonon interaction by nonequilibrium charge carriers,[6] leads to a satisfactory description of the experimental results of Refs. 4 and 7 only in a model which uses a simplified energy-loss spectrum of a hot electron gas.[8] Finally, appealing to a model of the heating of the crystal itself[3] presupposes the instantaneous establishment of a thermodynamic equilibrium among all of the lattice vibration modes.

We have carried out a detailed analysis of the spectra and temporal characteristics of the emission from a highly excited CdS single crystal. As a result of this analysis, it has become possible, for the first time, to clearly distinguish two stages in the cooling of a nonequilibrium electron-hole plasma. These two stages reflect the sequential onset of several mechanisms that retard the energy relaxation.

The experimental apparatus consists of an ND:YAG laser with passive mode locking (the pulse length is 30 ps), a microcomputer, and a CAMAC-standards computer-controlled luminescence spectrometer. The time resolution of the apparatus is provided by a CS_2 Kerr optical shutter. The luminescence spectra are recorded photoelectrically through a buildup of the signal at various points with amplitude discrimination of the exciting pulses.

Figure 1 shows several characteristic luminescence spectra of the electron-hole plasma in a CdS single crystal excited by the third harmonic of the laser light ($h\nu_0 = 3.50$ eV, power density of 500 MW/cm^2) recorded at various times after the pump pulse. The shape of the short-wave part of the spectra indicates that the effective temperature of the electron-hole plasma, T_e, is significantly higher than the initial

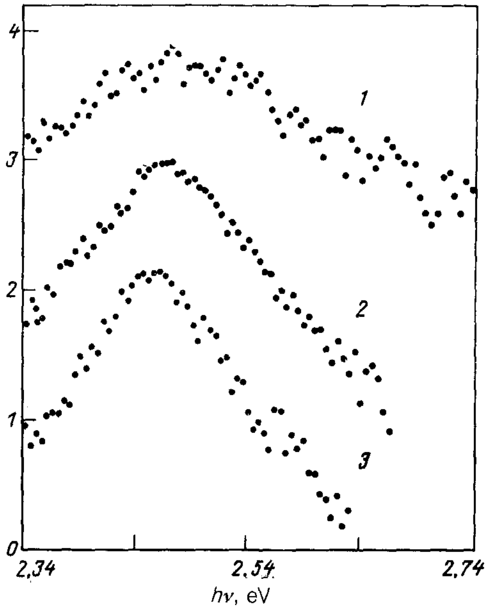

FIG. 1. Luminescence spectra of a CdS single crystal ($T = 295$ K) at various times after the excitation pulse. 1—20 ps; 2—80 ps; 3—280 ps. The intensity scales for the different spectra have been shifted by arbitrary distances.

lattice temperature of the crystal ($T = 295$ K). Furthermore, analysis of the spectral position of the maxima of the emission bands leads to the conclusion that the trivial heating of the crystal lattice, which results in a narrowing of the band gap, is not manifested in this case. Figure 2 shows the time evolution of the heating of the electron-hole plasma, $\Delta T_e = T_e - T$. We see that in the initial stage there is a rapid cooling of the plasma, with a time constant equal to or slightly less than 15 ps (this value was found by a deconvolution analysis). This cooling is followed by an abrupt increase in the energy relaxation time of the electron-hole plasma, to 400 ps. The change in rates in the cooling kinetics of the plasma occurs at a certain delay after the

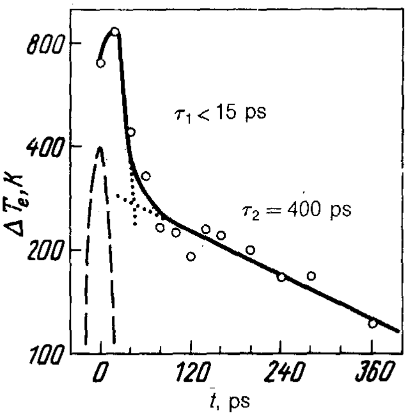

FIG. 2. Kinetics of the heating of the electron-hole plasma in highly excited CdS. Dashed line—shape of the pump pulse in logarithmic scale.

time at which the density of nonequilibrium charge carriers reaches its maximum (during the decay of the pump pulse). The mechanism for the retardation of the energy relaxation of the electron-hole plasma thus cannot be identified as a screening of the electron-phonon interaction. The energy relaxation rate may decrease over time because of a nonequilibrium multistep filling of the lattice vibration modes of the crystal, beginning at the highest-frequency modes, in the following way: During the pump pulse, a hot electron-hole plasma forms in the crystal. It causes a deviation from equilibrium of the phonons which are coupled most strongly with the electrons and holes: the LO vibrations in the long-wavelength region of the Brillouin zone. At the end of the excitation pulse, the injection of fast electrons into the crystal ceases, and the kinetics of the temperature T_e reflects the relaxation of the nonequilibrium population of the LO phonons that have been accumulated, because of the absorption of these phonons by the plasma. The depopulation of nonequilibrium LO phonons due to the anharmonic decay proceeds with a characteristic time constant τ_1 until, in a certain interval of wave numbers, lower-frequency, long-lived lattice-vibration modes become heated. These modes are the final states of this decay. As a result, the probability for the formation of a long-wavelength LO phonon approaches the probability for the decay of the phonon, and in the next step the cooling rate of the electron-hole plasma is determined by the slow relaxation of nonequilibrium low-frequency phonons with a characteristic time constant τ_2. If it is assumed that an LO phonon decays into two phonons of identical energy, then the ratio τ_2/τ_1 should be about[9] 2^5, in agreement with the experimental data reported here.

In summary, the multistep cooling of a nonequilibrium electron-hole plasma which is seen in these experiments apparently reflects the successive buildup of nonequilibrium phonons in bounded regions of **k** space, as a result of constraints imposed by the quasimomentum selection rules on energy transfer processes in the excited crystal.

[1] R. Luzzi and A. R. Vasconcellos, in Semiconductors Probed by Ultrafast Laser Spectroscopy, Vol. 1 (ed. R. R. Alfano), Academic, Orlando, 1984, pp. 135–169.
[2] W. Pötz and P. Kocevar, Phys. Rev. B **28**, 7040 (1983).
[3] T. Amand and J. Collet, J. Phys. Chem. Sol. **46**, 1053 (1985).
[4] R. Baltrameyunas, A. Zhukauskas, V. Latinis, V. Styapankyavichyus, and S. Yurshenas, Fiz. Tekh. Poluprovodn. **21**, 932 (1987) [Sov. Phys. Semicond. **21**, 568 (1987)].
[5] D. von der Linde, J. Kuhl, and H. Klingenberg, Phys. Rev. Lett. **44**, 1505 (1980).
[6] M. Pugnet, J. Collet, and A. Cornet, Solid State Commun. **38**, 531 (1981).
[7] R. J. Seymour, M. R. Junnarkar, and R. R. Alfano, Solid State Commun. **41**, 657 (1982).
[8] E. J. Yoffa, Phys. Rev. B **23**, 1909 (1981).
[9] P. G. Klemens, Phys. Rev. **148**, 845 (1966).

Translated by Dave Parsons