

# Accumulation of the decay products of nonequilibrium optical phonons in a GaAs-Al<sub>x</sub>Ga<sub>1-x</sub>As superlattice

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An increment in the electron temperature in a photoexcited GaAs-Al<sub>x</sub>Ga<sub>1-x</sub>As superlattice stems from a long-lived nonequilibrium population of vibrational modes with a frequency half that of a longitudinal optical (*LO*) phonon.

Our understanding of the relaxation of the excess energy of photoexcited electrons in semiconductors has gone through several stages of evolution since the advent of laser research methods. The original model of the interaction of hot photoelectrons with an unexcited lattice<sup>1</sup> had to be supplemented with an account of nonequilibrium optical phonons.<sup>2</sup> Experimental data obtained in Ref. 3 showed that in the late stage of the cooling of the nonequilibrium electron-hole plasma the energy relaxation is slowed down by nonequilibrium phonons of a second generation. A concentration of these phonons near the subharmonic frequency  $\omega_0/2$  ( $\omega_0$  is the frequency of an optical phonon) has been observed during coherent excitation of optical vibrations in a crystal.<sup>4</sup> The present letter reports a new result: The accumulation of long-lived products of the decay of *LO* phonons is capable of serving as a governing mechanism for retarding the energy relaxation of charge carriers.

It has been pointed out that there are distinctive features in the growth of the electron temperature in a photoexcited semiconductor superlattice as the pump intensity and the excess energy of the nonequilibrium electron-hole pairs increase. The structure studied in the present experiments included alternating layers of GaAs and Al<sub>0.41</sub>Ga<sub>0.59</sub>As, 100 Å of each, and each  $\approx 50$  Å thick. A sample in helium vapor (the

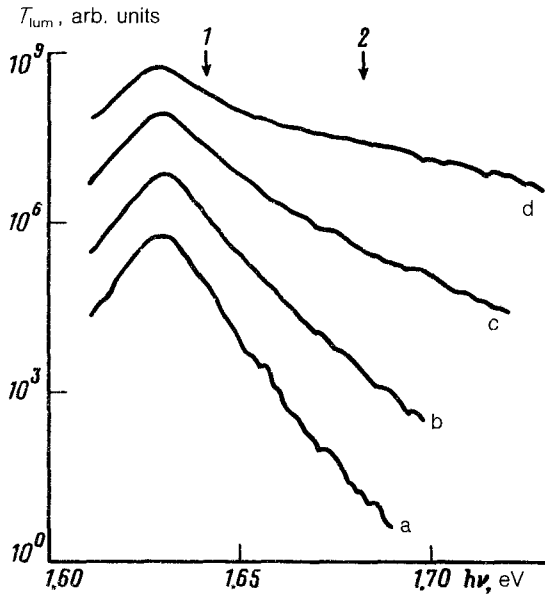


FIG. 1. Luminescence spectra of the GaAs-Al<sub>0.41</sub>Ga<sub>0.59</sub>As superlattice for a pump photon energy of 1.77 eV and for various flux densities of the photons which are absorbed,  $G$  (in units of  $10^{22}$  cm<sup>-2</sup>·s<sup>-1</sup>),  $T_c$  (K): a—7.5, 63; b—21.6, 92; c—61.7, 174; d—177, 312. The intensity scales for the various spectra have been shifted by arbitrary amounts. Arrow 1) position of the band gap,  $E_g = 1.641$  eV; arrow 2) energy of the photon corresponding to the threshold for the emission of an LO phonon by an electron,  $E_g + \hbar\omega_0(m_e/m_h + 1)$ .

bath temperature was 6 K) was excited by the beam from an oxazine dye laser (pulse length of 10 ns) directed along the axis of the superlattice. The energy of the pump photon,  $h\nu_0$ , did not exceed 1.77 eV, so the excess energy of the holes was not sufficient for a transition of the holes into the AlGaAs layer. The electrons were excited only within the first subband. The resulting luminescence spectra (Fig. 1) are governed in the region  $h\nu > E_g$  ( $E_g$  is the band gap) by an interband recombination of nonequilibrium charge carriers. The effective temperature of these carriers can be found from  $T_c = -[k_B \partial(\ln I_{lum}) / \partial(h\nu)]^{-1}$ . The effective temperature of the electron system in this structure differed from that in a bulk GaAs crystal<sup>1</sup> in that it was more sensitive to the deposited power level. In the first place, the rate of influx of the excess energy  $G(h\nu_0 - E_g)$  ( $G$  is the flux density of photons which are absorbed) required to reach a given  $T_c$  (e.g., 100 K) in a single crystal is about an order of magnitude higher than that in the present case. Second, the temperature  $T_c$  increases twice as fast, approximately in accordance with  $G(h\nu_0 - E_g) \sim \exp(-\delta E/k_B T_c)$ , where  $\delta E$  is equal to half the energy of an LO phonon, rather than its total energy, as in the case of a bulk crystal. This behavior persists as the pump level is varied at a fixed  $h\nu_0$  and also as the energy of the pump photon is varied at a fixed  $G$  (Fig. 2). The features observed in the heating of the photoelectrons could hardly be attributed to a specific feature of the electric-phonon interaction in a quasi-two-dimensional structure, since the probability for the emission of an intralayer LO phonon or an interlayer LO phonon could change by at most a factor of two, up or down, respectively, in the case of 50-Å layers.<sup>5</sup> On the other hand, over a broad range of the pump intensity we do not observe a depletion in the distribution function of the hot photoelectrons near the energy  $E = \hbar\omega_0$  (arrow 2 in Fig. 2 shows the corresponding energy of the pump photon) which would be caused by a substantial difference in the temperatures of the

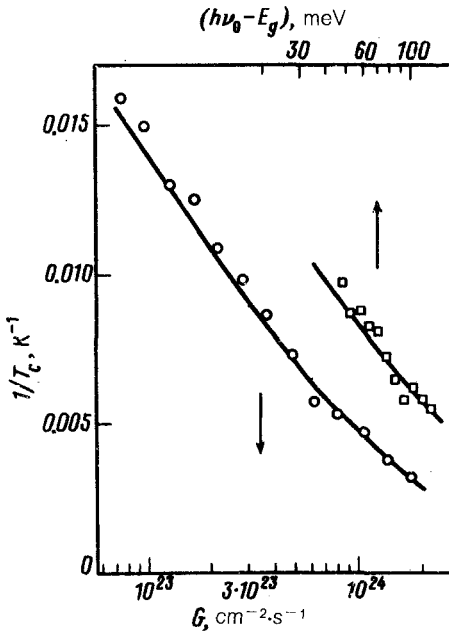


FIG. 2. Reciprocal electron temperature versus (O) the excitation level  $G$  ( $h\nu_0 = 1.77$  eV) and versus (□) the excess energy of the equilibrium electron-hole pair,  $h\nu_0 - E_g$  ( $G = 6.17 \times 10^{23}$  cm $^{-2}$ ·s $^{-1}$ ). The solid lines show  $N_2(T_e) \sim G(h\nu_0 - E_g)$ .

charge carriers and the optical phonons.<sup>6</sup> The effective electron temperature here evidently differs only slightly from the “temperature” of the nonequilibrium  $LO$  phonons,  $T_1$ , which are concentrated in the long-wavelength region of  $\mathbf{k}$  space<sup>2</sup>: ( $T_e - T_1 \ll T_1$ ). However, the average occupation number of these phonons,  $N_1 = [\exp(\hbar\omega_0/k_B T_1) - 1]^{-1} \approx \exp(-\hbar\omega_0/k_B T_1)$ , increases quadratically with increasing power of their excitation, according to the results shown in Fig. 2. This conclusion can be explained by assuming that the “temperature” of the nonequilibrium phonons of the first generation ( $LO$  phonons) is in turn sustained by the coalescence of the products of their decay which have accumulated and which are characterized by a “temperature”  $T_2$  satisfying  $T_1 - T_2 \ll T_2$ . In GaAs, the nonequilibrium phonons of the second generation are short-wavelength longitudinal acoustic ( $LA$ ) phonons, whose wave numbers lie in an interval of width  $q_0$  (the wave number of decaying  $LO$  phonons) which is centered at  $Q \approx 6 \times 10^7$  cm $^{-1}$ . The average “temperature” of the phonons in this interval, with  $q_0 \ll Q$ , and under the assumption that the occupation of the lower-frequency modes is insignificant, is given by the balance equation

$$N_2 \equiv [\exp(\hbar\omega_0/k_B T_2) - 1]^{-1} = \frac{2G(h\nu_0 - E_g)\tau_2}{\hbar\omega_0 d} [(2\pi^2)^{-1} Q^2 q_0]^{-1},$$

where  $d$  is the thickness of the region which is pumped, and  $\tau_2$  is the relaxation time of the nonequilibrium population. It should be noted that these acoustic phonons, with a wavelength equal to two lattice constants, must undergo a strong scattering by the interfaces between layers, even in a high-quality structure. The superlattice seems to

constitute a trap for such phonons, in which the time  $\tau_2$  is determined more by anharmonic than by transport processes, in contrast with the situation in a single-photon-excited bulk crystal. Assuming  $T_2 = T_c$ , we find an upper limit on  $\tau_2$  from the equation given above. It turns out to be 2 (5) ns for  $q_0 = q_{e(h)} = (2m_{e(h)}\omega_0/\hbar)^{1/2}$  (in the second case, it is assumed that the hole-phonon energy-transfer mechanism is predominant).

In summary, a multistep process in which the excess energy of photoexcited charge carriers is converted into vibrations of the crystal lattice may in general be retarded by various generations of nonequilibrium phonons. In microscopic semiconductor structures (including superlattices), where it is difficult for short-wavelength oscillations to propagate, the bottleneck in the energy transfer may turn out to be an ensemble of vibrational modes near a subharmonic of the optical phonons. The temperature of the phonons in this bottleneck is capable of determining the energy spectrum of the electron system, which is in fact a pseudohot system under such conditions.

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