Phonon-induced transitions between exciton subbands in silicon

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A resonant absorption of nonequilibrium acoustic phonons on transitions between exciton subbands has been observed. The threshold kinetic energy of the excitons has been found to be $E_{\vec{k}} = 0.1$ meV.

A splitting ΔE of exciton subbands which results from a lifting of the degeneracy of the energy states of heavy and light holes at $\vec{k} = 0$ by the field of electrons in side valleys[1] may give rise to numerous optical and transport phenomena in multivalley semiconductors with a degenerate exciton band (Ge, Si, etc.).[2] In the present study we have observed a threshold in the absorption of nonequilibrium acoustic phonons which stems from intersubband exciton transitions in Si.

In the experiments, the phonon-induced transitions between exciton subbands were detected by measuring the integral intensity of the differential luminescence signal induced by nonresonant acoustic phonons ($\delta\Sigma$), the intensity ratio of the luminescence of excitons involving the emission of longitudinal (I_{EX}^{LO}) and transverse (I_{EX}^{TO}) optical phonons ($\eta = I_{EX}^{LO}/I_{EX}^{TO}$), and the shape of the exciton luminescence spectra (the *LO–TO* doublet) (Fig. 1). The single-crystal Si:B samples, with boron densities $N_B < 10^{13}$ cm^{-3} and dimensions $\sim 2 \times 5 \times 10$ mm, were exposed to a steady-state excitation beam from an Ar* laser (see the inset in Fig. 1). The beam intensity was below the threshold corresponding to condensation into droplets of an electron–hole liquid. A lateral surface of the sample was excited optically (in a spot 5×5 mm in size, with an excitation G uniform within $\delta G/G < 0.1$) in order to prevent effects stemming from a spatial variation of the exciton distribution (phonon drag, etc.). A strip ~ 200 μm $\times 5$ mm in size was "cut out" at the center of this spot, and the luminescence from this strip was detected with a DFS-24 double monochromator and a digital-sampling photon-counting system. A constantan heater with dimensions of 0.1×1 mm was deposited on an end of the Si sample. This heater generated nonequilibrium acoustic

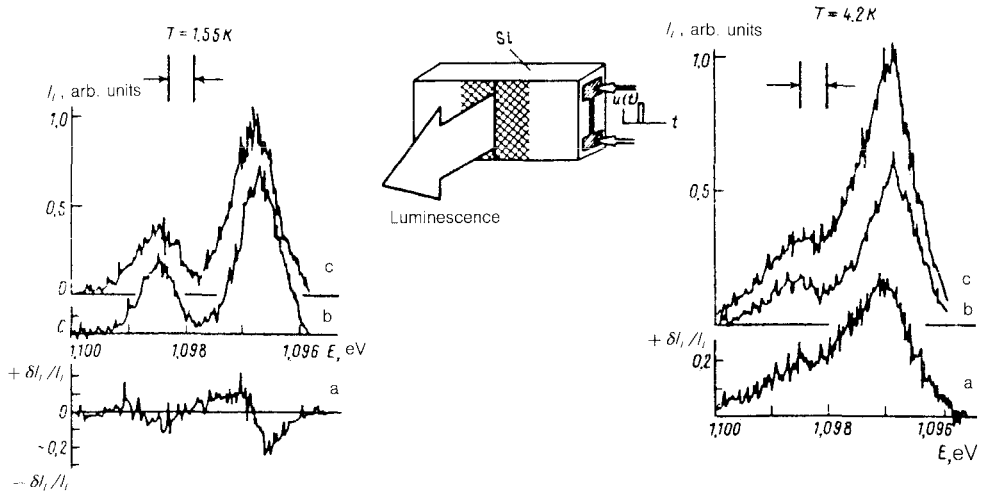

FIG. 1. Luminescence spectra of free excitons (*LO–TO* replicas). a—Differential spectrum; b—luminescence spectrum in the absence of nonequilibrium phonons; c—luminescence spectrum during the injection of nonequilibrium phonons. The inset shows the experimental geometry; the hatched region on the sample is the excitation spot.

phonons in the energy band $\sim$0–5 meV with occupation numbers well above the equilibrium values [the applied power was <25 W; the length of the electrical pulse was $U(t) = 0.1$–$1\ \mu$s] (Fig. 1). The following properties were recorded simultaneously (Fig. 1): (a) the differential luminescence spectrum, (b) the luminescence spectrum in the absence of phonons, and (c) the luminescence spectrum at the time of arrival of the ballistic pulse. The experiments were carried out in He^4 over the temperature interval $T = 1.5$–4.2 K.

Figure 1 shows two families of luminescence spectra from a series of spectra measured in He^4 at two temperatures, $T = 1.5$ K and $T = 4.2$ K. In each case we see a heating of the exciton gas by the nonequilibrium phonons (a shift of the peak of the luminescence line and an increase in the intensity on the high-energy wing of the line at a fixed position of the red boundary of the line). However, while the total integral of the differential spectrum at $T = 1.5$ K is $\delta\Sigma^{LO+TO} \sim 0$ (this result is evidence that an exciton energy redistribution in the lower band due to an interaction with nonequilibrium acoustic phonons is playing a leading role), at $T = 4.2$ K the injection of the nonequilibrium phonons leads to a significant increase in the relative luminescence intensity of the *TO* component (Fig. 2): The value of η is half that in the absence of phonons. The heating of the excitons is significantly more effective in this case than at $T = 1.5$ K. A qualitative change in the nature of the interaction of the excitons with the acoustic phonons first appears at $T \sim 2.3$ K (Fig. 2): a sharp increase in the integral intensity $\delta\Sigma^{TO}/\Sigma^{TO}$ at $T \simeq 1.9$–2 K and a relatively slow decrease from $T > 2.7$ K up to 4.2 K.

Let us briefly discuss the results of these experiments. Figure 3 shows $E_{\pm}(\vec{k})$ dispersion curves for excitons in Si, plotted on the basis of Ref. 3, and for acoustic

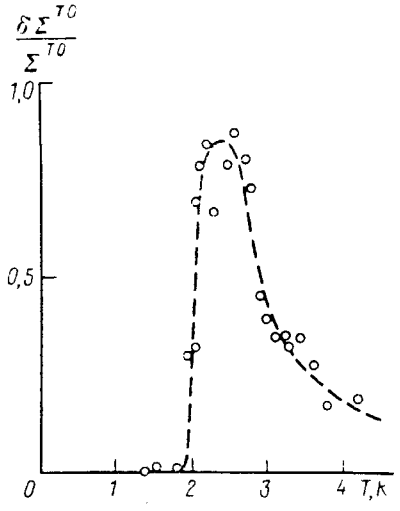

FIG. 2. Points: Relative integral luminescence of the *TO* replica of a free exciton versus the He^4 temperature (the temperature was varied by pumping off the He^4 vapor). Here $\delta\Sigma^{TO}$ is the integral intensity of the differential signal at the *TO* line induced by nonequilibrium phonons, and Σ^{TO} is the integral intensity of the *TO* line in the absence of nonequilibrium phonons. Dashed line: Average experimental behavior.

phonons. In a phonon-absorption event an exciton undergoes an oblique transition within a subband; beginning at certain energies of the initial state, it also undergoes an intersubband transition. The phonons which interact with the excitons satisfy the conservation laws (Fig. 2)

$$\hbar\Omega_{\vec{q}} = E_+(\vec{k}_f) - E_-(\vec{k}_i), \tag{1}$$

where the + and − specify the corresponding exciton subband,[3] $\vec{k}_i$ and $\vec{k}_f$ are the wave vectors of the initial and final states of the exciton, and q is the phonon wave vector. By varying the temperature of the He^4 bath, we can shift the peak ($E_{\max} = kT/2$) of the Boltzmann distribution of excitons in the band and thereby alter the initial energy of the bulk of the excitons participating in the absorption process. We can reach a region on the $E_-(\vec{k})$ dispersion curve from which transitions

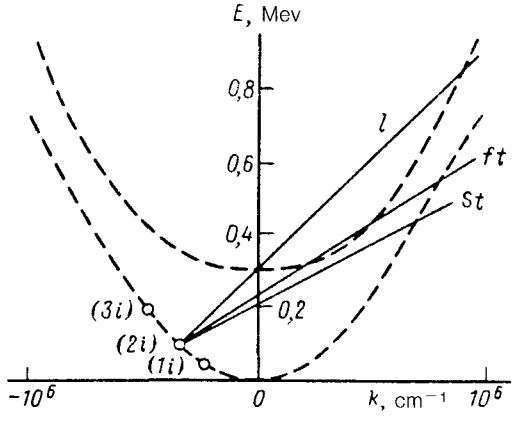

FIG. 3. Dashed lines—$E_\pm(\vec{k})$ dispersion curves calculated in accordance with Ref. 3; solid lines—dispersion curves for acoustic phonons ($c_l = 9.19\times10^5$ cm/s, $c_{ft} = 5.87\times10^5$ cm/s, $c_{lt} = 4.89\times10^5$ cm/s).[4] [$\hbar\Omega_{\vec{q}} = c_s qh$; $s = l, ft, st$; $1i$ ($T = 1$ K), $2i$ ($T = 2.3$ K), and $3i$ ($T = 4.2$ K) are the initial states of the excitons for the absorption of acoustic phonons].

can occur to the upper subband (Fig. 3). These are the conditions—during the injection of nonequilibrium phonons with occupation numbers exceeding the equilibrium values into the sample—under which we observe the threshold change in the quantities $\delta\Sigma^{TO}/\Sigma^{TO}$ and η as a result of a resonant absorption of acoustic phonons during intersubband transitions[1] (Figs. 1 and 2). The population of the upper exciton subband, which determines the luminescence intensity and the factors $\delta\Sigma^{TO}/\Sigma^{TO}$ and η, depends on the occupation numbers of the acoustic phonons participating in the absorption in this case. The maximum of $\delta\Sigma^{TO}/\Sigma^{TO}=f(T)$ near the "threshold" energy $E=E_{\max}=kT/2\simeq 0.1$ meV (Fig. 2) is linked with a nonparabolic nature of the $E_{\pm}(\vec{k})$ subbands and thus a corresponding increase in the density of states in this region of exciton energies. The threshold exciton energy determined from an analysis of the luminescence characteristics under these conditions, $E_{\max}\simeq 0.1$ meV, along with the acoustic-vibration quantum $\hbar\Omega_{\vec{q}}=0.2$ meV, estimated in accordance with (1), makes up the quantity $E_{\text{thresh}}=E_{\max}+\hbar\Omega_{\vec{q}}=0.3$ meV, which is equal to the energy splitting ΔE of the exciton band.[2] Above the threshold temperature, the exciton luminescence spectrum is governed by a superposition of the contributions from the two subbands. This circumstance is responsible for the high effectiveness of the heating of excitons by nonequilibrium phonons which can be inferred from the shape of the luminescence line under these conditions.

In summary, the results of these studies show convincingly that a threshold absorption of nonequilibrium acoustic phonons by excitons has been observed. The existence of this threshold is attributed to a "crystal" splitting of the valence band at $\vec{k}=0$. It has been established experimentally that the exciton gas is heated by nonequilibrium acoustic phonons. Studies of this sort, by the technique of frequency-resolved phonon spectroscopy, can provide valuable information about the dispersion of the exciton subbands near $\vec{k}=0$ (about the exact value of the splitting ΔE, the curvature of the subbands near $\vec{k}=0$, etc.).

[1)] The modulation of the lines of exciton-impurity complexes caused by a radiationless decay induced by nonequilibrium phonons does not change significantly anywhere in the temperature interval $T=1.4$–4.2 K under these experimental conditions.

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