Drag of excitons by heat-generated phonon pulses in silicon

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A study was made of the influence of heat-generated phonon pulses on the exciton luminescence emitted by silicon crystals. The drag of excitons by a directional acoustic phonon flux was observed for the first time in silicon. Strong reflection of phonons was observed for an atomically clean freshly cleaved surface of Si in contact with liquid helium. The ballistic component in the propagation of phonons of $\hbar\Omega \geqslant 3.8$ meV frequency was observed for the first time in silicon.

Silicon occupies a special place among materials included in studies of the properties of nonequilibrium hf acoustic phonons. The availability of pure and "perfect" Si single crystals in which the low-temperature mean free path of subterahertz phonons is sufficiently long has made it possible to carry out a great variety of experiments on ballistic phonon beams in silicon. These were the experiments on the focusing of phonons, visualization of phonon beams, reflection of phonons from a surface, interaction of phonons with impurity atoms, etc.

The present paper describes the first experimental investigation[1)] of the interaction of nonequilibrium acoustic phonons with exciton states in silicon. These phonons were injected into samples kept at helium temperature by a surface heater (heat-pulse method). The interaction of phonons with excitons was deduced from the effects observed in the exciton luminescence spectra of silicon.

The drag of an exciton gas by a directional flux of acoustic phonons was observed for the first time for silicon. We also observed and studied important features of the behavior of both types of intrinsic excitations of Si in the form of excitons (surface recombination of excitons) and hf acoustic phonons (interaction with the surface for various conditions on the surface, dynamics of phonons in a "hot" part of a crystal near a heater, etc.).

§1. EXPERIMENTAL METHOD AND RESULTS

We used oriented single-crystal rectangular plates of ~ 1 cm dimensions prepared from dislocation-free pure silicon (resistivity $\sim 10^4\ \Omega\cdot$cm, difference between acceptor and donor concentrations $N_a - N_d \sim 10^{12}$ cm^{-3}). These plates were immersed in pumped liquid helium ($T = 1.7$ K) inside a cryostat.

One of the faces was illuminated (inset in Fig. 1) by an obliquely incident beam of light from a cw Ar laser ($\lambda = 5145$ Å, $P \lesssim 100$ mW, focused beam diameter 0.5–1 nm), which excited a sample to a depth of $\sim 1\,\mu$m (Ref. 2). Free excitons (*FE*) were generated in the excited layer and these excitons diffused into the sample creating an exciton cloud near the surface. The exciton luminescence was observed along the normal to the surface. Special measurements carried out under pulsed excitation conditions showed that the *FE* luminescence exhibited nonexponential decay kinetics. A steep initial fall of the intensity was due to fast surface recombination of free excitons and the exponential tail was determined by the lifetime of free excitons in the bulk. The measured *FE* lifetime was $\tau_0 = 1.0 \pm 0.1\,\mu$s in the bulk, in good agreement with the results reported in Ref. 3 for Si samples of similar quality.

The observed steady-state luminescence spectrum contained, in agreement with Ref. 4, lines at $E_1 = 1.097$ eV, $E_2 = 1.099$ eV, and $E_3 = 1.144$ eV, associated with the radiative annihilation of free excitons accompanied by the creation of optical ($\omega_{TO} = 57.5$ meV, $\omega_{LO} = 55.3$ meV) and acoustic ($\omega_{TA} = 18.8$ meV) phonons in the lattice. The spectrum recorded at $T = 1.7$ K showed also clearly lines in the region of $E = 1.093$ eV, which were due to radiative annihilation (assisted by optical phonons) of excitons bound to impurities (*BE*). Nonequilibrium phonons were injected into the sample by pulses (2×10^{-7} s) generated by electric-current heating of a constantan film h of area 1×1 mm and of thickness 100 Å, evaporated on one of the faces of a sample. The energy density of an electrical pulse W was within the range $W = 0.3$–$30\,\mu$J/mm^2, which corresponded to a heater temperature $T_h = 10$–30 K. When the heater was switched on, a continuous spectrum of phonons was injected into a crystal and it traveled into the interior of a sample. We

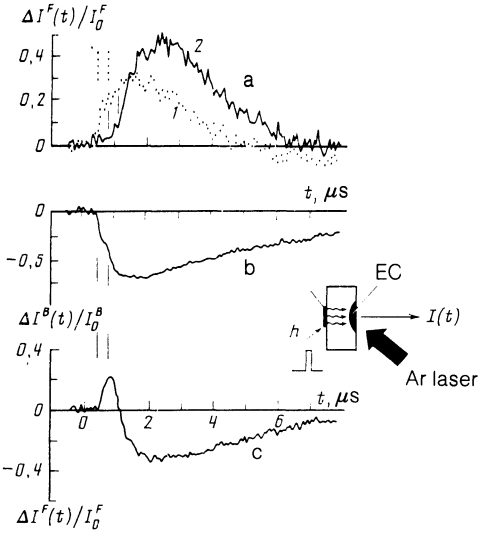

FIG. 1. Free-exciton $\Delta I^F(t)/I_0^F$ (a, c) and bound-exciton $\Delta I^B(t)/I_0^B$ (b) luminescence pulses emitted from an exciton cloud (EC) in samples with different states of the surface: a), b) (111) surface freshly cleaved in helium; c) oxidized cleaved surface. Propagation of phonons along $\mathbf{q} \| \langle 111 \rangle$, $R = 3.8$ mm (*a*, curve 1; b, c) and along $\mathbf{q} \| \langle 001 \rangle$, $R = 6.6$ mm (*a*, curve 2). Heating energy density $W = 10\,\mu$J/mm^2. The inset shows the experimental symmetry. The vertical segments give the calculated values of the time of arrival of ballistic *LA* and *TA* phonons.

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carried out experiments of two types differing in respect of the relative positions of the heater (h) on the faces of a sample and in respect of the region of soft optical excitation of the surface, i.e., the exciton cloud region.

1. In the main series of experiments the heater h and the exciton cloud were located on the opposite faces of a sample (inset in Fig. 1). The phonons which traveled from h to the exciton cloud on the opposite face altered the intensities of the luminescence lines due to free (FE) and bound (BE) excitons. We observed phonon-induced luminescence pulses $I(t)$ in the case of the FE and BE lines; we determined the differential signal $\Delta I(t) = I(t) - I_0$, where I_0 is the steady-state luminescence intensity at $T = 1.7$ K in the absence of heat pulses. In the case for the FE luminescence, on which our attention was concentrated, we determined the integrated (over the spectrum) intensities of the lines E_1 and E_2 measurements in the spectral intervals within the individual lines gave similar dependences $\Delta I^F(t)$].

We established that the FE luminescence pulses $\Delta I^F(t)$ had different profiles and depended strongly on the number of experimental conditions. First of all, we observed a very strong dependence of $\Delta I^F(t)$ on the state of the surface of a sample near which the exciton cloud is generated. Moreover, the nature of $\Delta I^F(t)$ depended on the energy density W released by the heater on the distance R traveled by phonons from h to the exciton cloud, and on the direction of the straight line joining h and the exciton cloud relative to the crystallographic axes of the sample.

We shall now consider the role of the state of the surface. We used silicon samples of thickness amounting to several millimeters and the working faces (those with h and the exciton cloud) of a sample were oriented along the (111) plane and subjected to heating characterized by moderate energy densities ($W \sim 10\,\mu J/mm^2$). Figure 1a shows the luminescence pulses $\Delta I^F(t)$ determined for a sample when an electron cloud was created near a freshly cleaved (111) surface formed and stored in liquid helium. Curve 1 was retained when the direction between h and the exciton cloud was normal to the cleaved face (inset in Fig. 1), i.e., when the direction of phonon propagation (vector $\mathbf{q}$) between h and the exciton cloud was the $\langle 111\rangle$ axis. Curve 2 was obtained when phonons propagated from h to the exciton cloud along $\mathbf{q} \| \langle 001\rangle$ at an angle to the (111) cleaved surface [in this case the heater h and the exciton cloud on the opposite (111) faces of a sample were shifted suitably and were not along the same normal to the faces].

Clearly, in both cases the luminescence pulses had a positive sign. The beginning of the leading edge of $\Delta I^F(t)$ corresponded to the shortest arrival times of phonons traveling from h to the exciton cloud. This was evidence of ballistic propagation along the line from h to the exciton cloud in the case of the phonons which were responsible for the increase in the FE luminescence intensity (the calculated times for the transit from h to the exciton cloud in the case of ballistic LA and TA phonons are identified by vertical segments in Fig. 1). It is clear from Fig. 1a that in the $\mathbf{q} \| \langle 001\rangle$ case the amplitude of an $\Delta I^F(t)$ pulse exceeded greatly the amplitude of a pulse in the case when $\mathbf{q} \| \langle 111\rangle$, although the latter was obtained for a much shorter distance between the exciton cloud and the photon source (the distances between h and the exciton cloud are given in the caption of Fig. 1). This difference between the amplitudes was evidence of sharp focusing of TA phonons propagating in Si along $\langle 001\rangle$ (relatively weak focusing of LA phonons occurred along the 111 axis).[5] This manifestation of focusing confirmed additionally the ballistic regime of phonon propagation, which induced a positive pulse of the FE luminescence (Fig. 1a).

Figure 1b shows the phonon-induced BE luminescence pulses $\Delta I^B(t)$. The sign of these pulses was negative, corresponding to a reduction in the BE luminescence under the influence of nonequilibrium phonons. We also investigated how the $\Delta I^F(t)$ pulses were affected by oxidation of the (111) surface which was cleaved in silicon and near which an exciton cloud was induced. A sample was taken out of the helium and kept for 20 h in air at room temperature. This was followed by reimmersion of the sample in liquid helium at $T = 1.7$ K and measurements of the phonon-induced luminescence pulses (Fig. 1c). Clearly, the $\Delta I^F(t)$ pulses had a complex alternating-sign profile. At times corresponding to the ballistic transit of phonons from h to the exciton cloud there was a short positive peak, which changed to a more extended negative signal corresponding to a reduction in the intensity of the FE luminescence.

The experiments described above for one sample (compare Figs. 1a and 1c) demonstrated that the profiles and even the signs of the FE luminescence pulses induced by nonequilibrium phonons depended decisively on the state of the Si surface near which excitons were excited. In the case of an atomically clean cleaved surface we observed a positive $\Delta I^F(t)$ pulse, whereas after oxidation of a cleaved surface $\Delta I^F(t)$ the luminescence pulse was mainly negative, but it had a short positive peak at the leading edge.

Similar "bipolar" signals were observed not only after oxidation of a freshly cleaved face, but generally in the case of "ordinary" Si surfaces formed in air (for example by polishing or etching in CP-4 solution) when an oxide layer should form on the Si surface (Fig. 2). The relative amplitude of the $\Delta I^F(t)/I_0^F$ signal was independent of the state of the ordinary surface of silicon, but this state affected strongly the steady-state intensity I_0^F (because of surface recombination of excitons). This conclusion was drawn from a comparison of the curves in Fig. 2 obtained for a ground surface before and after etching, which increased the intensity of the steady-state luminescence I_0^F by a factor of 5.

Figure 3, which shows the luminescence pulses determined for a sample with an ordinary (etched) surface, demonstrates the dependence of the $\Delta I^F(t)$ pulses on the density of the energy released in the heater. Clearly (compare curves a, b, c) an increase in W increased the amplitude of the negative signal and extended greatly the duration ($t > 10\,\mu s$ for curve c) of the negative pulse tail. At the maximum energy

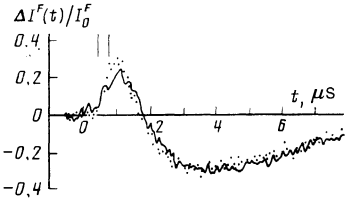

FIG. 2. Free-exciton $\Delta I^F(t)/I_0^F$ luminescence pulses obtained for a chemically etched (continuous curve) and ground (dots) (111) surface of Si, $\mathbf{q} \| \langle 001\rangle$, $W = 10\,\mu J/mm^2$.

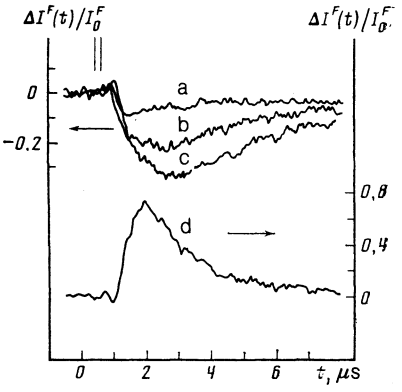

FIG. 3. Free-exciton $\Delta I^F(t)/I_0^F$ luminescence pulses for an ordinary surface of Si obtained for different values of W (μJ/mm^2): *a*) 0.6; *b*) 10; *c*) 20; *d*) 30; $\mathbf{q} \| \langle 001 \rangle$.

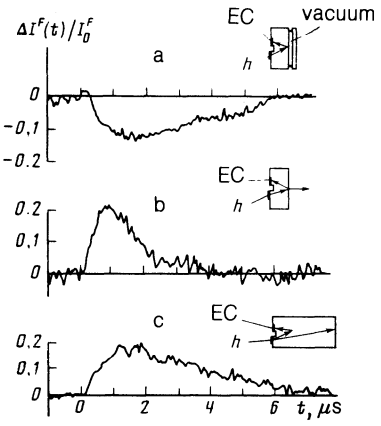

FIG. 5. Free-exciton $\Delta I^F(t)/I_0^F$ luminescence pulses obtained in the experimental geometries shown as insets. The face opposite to the surface with h and the exciton cloud (EC) was in vacuum (a) or in liquid helium (b, c). The distance to the opposite face was L = 1.2 mm (a, b) or 10 mm (c).

density of $W = 30\,\mu$J/mm^2, when liquid helium started boiling near the surface where the exciton cloud was created, a positive luminescence pulse was observed (Fig. 3d). It should also be added that positive $\Delta I^F(t)$ pulses were observed for an exciton cloud near an ordinary surface every time (irrespective of the energy density released by the heater) when this surface was in vacuum.

Figure 4 shows the *FE* luminescence pulses obtained for samples of two thicknesses, i.e., for two values of the distance R from h to the exciton cloud near the etched surface of silicon. One of the samples was extremely thin ($R = 0.3$ mm) and a very low power was delivered to the heater h ($W = 0.7\,\mu$J/mm^2). As is usual for an ordinary surface, a negative pulse was observed (Fig. 4a). However, an increase in the distance from h to the exciton cloud up to $R = 3.8$ mm, while retaining a low energy density ($W = 0.7\,\mu$J/mm^2), reversed the sign of the pulse so that $\Delta I^F(t) > 0$ (Fig. 4b).

We considered above the phonon-induced *FE* luminescence pulses. The influence of phonons on the *BE* luminescence in silicon was also investigated and in all cases it was found that "negative" $\Delta I^B(t)$ pulses were observed under the influence of heat pulses: this indicated that the heat pulses reduced the *BE* luminescence intensity (Fig. 1b).

2. In contrast to the experiments described above in which the heater h and the exciton cloud were on the opposite faces of a sample, we also carried out experiments of a different type in which h and the exciton cloud were on the same face of a sample of Si (insets in Figs. 5*a*, 5*b*, and 5*c*). A cut 0.65 mm wide and 0.53 mm deep was made between h and the exciton cloud. This cut prevented "ballistic" arrival of phonons in the exciton cloud from the region of the heater h. The luminescence signal emitted by the exciton cloud could only be induced by the phonons generated by h and reaching the exciton cloud along a curved path that bypassed the cut, i.e., it could only be due to phonons reflected from the opposite face of the sample or scattered in the bulk.

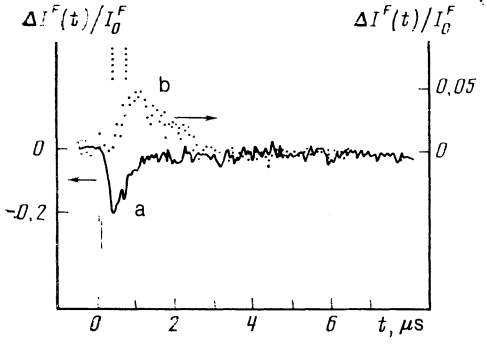

FIG. 4. Free-exciton $\Delta I^F(t)/I_0^F$ luminescence pulses obtained for an ordinary surface of Si. Distance from h to the exciton cloud R (mm): *a*) 0.3; *b*) 3.8; $W = 0.7\,\mu$J/mm^2, $\mathbf{q} \| \langle 111 \rangle$.

Our experiments were carried out on samples for which both surfaces (that with h and the exciton cloud and the opposite one) were ordinary (subjected to oxidation in air after etching). The face with h and the exciton cloud was always in contact with liquid helium ($T = 1.7$ K), but the conditions on the opposite face could be altered: it could be either in vacuum or it could be in contact with superfluid helium. Figures 5a and 5b showed the phonon-induced $\Delta I^F(t)$ luminescence pulses obtained for a thin sample with the distance between the faces amounting to $L = 1.2$ mm. When the face opposite to h and the exciton cloud was in vacuum, a negative $\Delta I^F(t)$ pulse was observed (Fig. 5a). However, when vacuum at this face was replaced with superfluid helium, there was a radical change in the pulse profile, which became positive (Fig. 5b). Figure 5c shows the $\Delta I^F(t)$ signal determined using a "thick" sample ($L = 10$ mm). This signal was positive and its form was independent of the conditions on the face opposite to the surface carrying h and the exciton cloud.

§2. DISCUSSION OF RESULTS

a. Mechanisms of interaction between nonequilibrium phonons and an exciton cloud

It is clear from §1 that the profiles of the phonon-induced free-exciton luminescence pulses $\Delta I^F(t)$ can vary greatly, depending on the actual experimental conditions. Before explaining the experimental results, we consider first possible microscopic mechanisms whereby nonequilibrium acoustic phonons injected into a sample at $T = 1.7$ K act on the exciton gas and its luminescence.

Firstly, dissociation of bound excitons accompanied by the absorption of acoustic phonons is possible and it in-

creases the concentration of free excitons. The phonons active in this process have frequencies exceeding the threshold value $\hbar\Omega > \Delta E$, where $\Delta E = 3.8$ meV is the lowest binding energy of bound excitons.[6] The existence of $BE \rightarrow FE$ exciton processes accompanied by phonon absorption is well known from steady-state experiments[7] demonstrating enhancement of the *FE* luminescence (and weakening of the *BE* luminescence) on increase in the temperature of silicon. In our case the process of $BE \rightarrow FE$ dissociation under the influence of nonequilibrium phonon pulses should obviously result in enhancement of the *FE* luminescence and weakening of the *BE* luminescence.

Secondly, nonequilibrium acoustic phonons may be scattered by free excitons and the latter may acquire the energy and momentum of phonons (heating of the exciton gas). It follows from the laws of conservation of energy and momentum in the case of a parabolic exciton band that phonons participating in elementary scattering processes may have frequencies not exceeding

$$\hbar\omega' = 2m_{ex}s^2 + 2(2m_{ex}s^2E)^{1/2}, \tag{1}$$

where s is the velocity of sound; E is the kinetic energy of an exciton; m_{ex} is the translational mass of an exciton. Accurate determination of the maximum frequency in the case of Si is difficult because of the complex nature of the exciton spectrum of Si (Ref. 8) due to the strong anisotropy of the electron masses in ⟨001⟩ valleys and a quadruple degeneracy of the valence band extremum at the point Γ. However, ignoring the 0.3 meV "crystal" splitting of the bottom of the exciton band of silicon,[9] we can estimate the maximum value of ω' by substituting in Eq. (1) the largest values of the following parameters: $m_{ex} = m_{\parallel}^{e} + m^{h}$ (where m_h is the mass of a heavy hole in Si and $m_{\parallel}^{e} = 0.92m_0$ is the "heavy" longitudinal mass of an electron) and the velocity $s_{LA} = 8.43 \times 10^5$ cm/s. It is found that the scattering by free excitons at $T = 1.7$ K is dominated by low-frequency phonons with $\hbar\omega < 1.8$ meV. Phonons of these frequencies travel in a clear ballistic manner in pure silicon.[5]

The processes of the scattering of nonequilibrium phonons on free excitons (accompanied by moderate heating of the exciton gas) should not by themselves influence the integral *FE* luminescence intensity, because these processes do not alter the total number of free excitons in an exciton cloud in a crystal. However, we should bear in mind that injection of nonequilibrium phonons into a sample of Si from a surface heater creates a directional flux of the injected phonons and this flux is characterized by an anisotropic distribution of the phonon momenta. If such a distribution is retained in the exciton cloud region, then excitons may be dragged by the phonon flux. The phonon drag of excitons (for a review see Ref. 10) has been observed earlier in experiments carried out on a small number of materials such as Ge (Ref. 11) and CdS (Ref. 12). The drag of excitons by nonequilibrium phonons may influence the *FE* luminescence intensity if excitons are transferred by the phonon flux to the part of a crystal with different *FE* recombination conditions (times), which affect the number of free excitons in an exciton cloud. In particular, in both experimental schemes (Figs. 1 and 5), when an exciton cloud is formed near the face of an Si sample and h is outside the exciton cloud, the phonon drag of free excitons along the normal to the surface should quench the luminescence intensity because of reduction in the number of free excitons due to their accelerated surface recombination.

It therefore follows that in an analysis of the influence of thermal phonon pulses on the luminescence emitted from a surface exciton cloud in Si we have to allow for two opposite factors. One of them enhances the *FE* luminescence because of the $BE \rightarrow FE$ dissociation of bound excitons under the influence of "high-frequency" phonons (characterized by $\hbar\Omega > 3.8$ meV). The second is associated with the drag of free excitons toward the surface under the influence of a flux of "low-frequency" phonons (characterized by $\hbar\omega < 1.8$ meV) and it quenches the *FE* luminescence.

b. Calculation of the influence of nonequilibrium phonons on the exciton-cloud luminescence (steady-state case)

We shall consider a one-dimensional model problem which is based on the main experimental geometry in which a surface exciton cloud and a source of nonequilibrium phonons h are located on the opposite faces of a sample along a line x perpendicular to the surface boundary (inset in Fig. 1). We shall study the behavior of free and bound excitons in the region of the cloud in the presence of nonequilibrium phonons with occupation numbers that vary with time (t). The exciton gas densities $N_F(x, t)$ for free excitons and $N_B(x, t)$ for bound excitons are described by the equation of continuity allowing for the drag of free excitons and the equation representing the balance between the free and bound exciton states. In the case of one bound state of excitons the system of equations becomes[12]

$$\frac{\partial N_F(x,t)}{\partial t} - D\frac{\partial^2 N_F(x,t)}{\partial x^2} - v(t)\frac{\partial N_F(x,t)}{\partial x} + w_{FB}N_F(x,t) - w_{BF}(t)N_B(x,t) = 0, \tag{2a}$$

$$\frac{\partial N_B(x,t)}{\partial t} + [w_0 + w_{BF}(t)]N_B(x,t) - w_{FB}N_F(x,t) = 0 \tag{2b}$$

subject to the boundary conditions (at $x = 0$) allowing for surface recombination (at a velocity w) and for a constant surface optical pumping of free excitons (g):

$$D\frac{\partial N_F(x,t)}{\partial x}\Big|_{x=0} + v(t)N_F(x,t)|_{x=0} - wN_F(x,t)|_{x=0} + g = 0. \tag{3}$$

The quantity D in Eqs. (2a) and (3) is the diffusion coefficient of free excitons in an exciton region, which in the case of pure Si is governed by the processes of the scattering, discussed above in §2a, of free excitons on low-frequency ($\hbar\omega < 1.8$ meV) phonons. We shall assume that the value $D = 10^2$ cm^2/s (Ref. 13) corresponding to the scattering by thermal phonons (at $T = 1.7$ K) is independent (because of the weak temperature dependence $D \propto T^{-1/2}$) of the number of nonequilibrium phonons, i.e., it is independent of time. Here, $v(t)$ denotes the drift velocity of free excitons under the influence of a flux of "low-frequency" ($\hbar\omega < 1.8$ meV) nonequilibrium phonons directed toward the surface. It follows from Ref. 12 that

$$v = s\Delta\bar{n}_\omega/\bar{n}_\omega, \tag{4}$$

where s is the velocity of sound, $\bar{n}_\omega$ are the total occupation numbers of low-frequency phonons (including thermal phonons), and $\Delta\bar{n}_\omega = (\bar{n}_\omega^{+} - \bar{n}_\omega^{-})$ is the anisotropy (describing the phonon flux) of the occupation numbers of phonons with the x projections of the momentum directed

toward the surface ($\bar{n}_\omega^+$) and away from it ($\bar{n}_\omega^-$). The time dependence $v(t)$ reflects the kinetics of the flux $\Delta\bar{n}_\omega/n_\omega$ of low-frequency phonons. The fourth term in Fig. 2a describes the capture of free excitons by bound states with the probability $w_{FB} \sim 10^6\,\text{s}^{-1}$ (Ref. 3), which is the main channel for the loss of free excitons and governs the experimentally determined (at $T = 1.7$ K) lifetime of free excitons in the bulk amounting to $\tau_0 = w_{FB}^{-1} = 1\,\mu$s. Under the conditions of our experiments the quantity w_{FB} can be regarded as constant. The fifth term in Eq. (2a) describes the dissociation of bound excitons resulting in the creation of free excitons under the influence of nonequilibrium high-frequency phonons characterized by $\hbar\Omega > 3.8$ meV. The dissociation probability $w_{BF} \propto \bar{n}_\Omega(t)$ reflects the kinetics of the occupation numbers $\bar{n}_\Omega$ of these phonons. The quantity w_0 in Eq. (2b) is the probability of annihilation of a bound exciton as a result of nonradiative Auger recombination of bound excitons ($w_0^{-1} = 0.6$–$1\,\mu$s—see Ref. 3 and the references cited there). In Eqs. (2)–(4) the occupation numbers of nonequilibrium phonons $\Delta\bar{n}_\omega/\bar{n}_\omega$ and $\bar{n}_\Omega$, which influence the drag of excitons (drift velocity v) and the dissociation of bound excitons (probability w_{BF}), are assumed to be independent only of time but not of the coordinate. This is justified by the small thickness of the exciton cloud ($\sim 100\,\mu$m, see below) compared with the spatial scale of changes in the nonequilibrium phonon concentration in a sample when heat pulses are injected, as found experimentally in measurements of the integrated intensity of the luminescence emitted by the whole exciton cloud.

In the first stage of investigation of the system of equations (2) we shall consider its steady-state solution by assuming that the parameters of low- and high-frequency phonons injected into crystals are constant (then the velocity of drift of free excitons v and the probability w_{BF} of $BE \rightarrow FE$ dissociation are independent of time). The solution of the steady-state problem makes it possible to analyze important experimental features of the response of the luminescence to nonequilibrium phonons, particularly the sign of the momentum $\Delta I^F(t)$.

The steady-state solution of Eq. (2) subject to the boundary conditions of Eq. (3) is of the form

$$N_F(x, t) = g(D/r + w - v)^{-1}\exp(-x/r) \tag{5}$$

where r is the "thickness" of a surface exciton cloud which decays exponentially with depth in a sample:

$$r = (D\tau)^{1/2}(\beta + (\beta^2+1)^{1/2})^{-1}. \tag{6}$$

The time

$$\tau = \frac{w_0 + w_{BF}}{w_0 w_{FB}} = \tau_0 + \frac{w_{BF}}{w_0 w_{FB}} \tag{7}$$

which occurs in Eq. (6) is the real lifetime of free excitons in the bulk of a crystal. It is longer than the lifetime of free excitons τ_0 in the absence of nonequilibrium high-frequency phonons because of the $BE \rightarrow FE$ dissociation processes that cause return of excitons to the free state. The quantity

$$\beta = {}^1/_2 v(\tau/D)^{1/2} \tag{8}$$

is a parameter representing the relative role of the phonon drag process which governs v and the $BE \rightarrow FE$ dissociation process influencing the time τ.

It is worth noting that in this discussion the thickness of the exciton cloud of Eq. (6) is independent of the surface exciton recombination rate w. In the absence of nonequilibrium phonons (when $\tau = \tau_0$ and $\beta = 0$) the thickness of the exciton cloud of Eq. (6) given by $r_0 = (D\tau_0)^{1/2}$ is $r_0 \sim 100$ μm for the value $D \sim 10^2$ cm^2/s known from Ref. 13 for pure Si. It is clear from Eqs. (6) and (8) that in the presence of nonequilibrium phonons and when the drag is weak ($\beta \ll 1$) the thickness of the exciton cloud $r = (D\tau)^{1/2}$, increases compared with the original value $r_0 = (D\tau_0)^{1/2}$ because of an increase in the exciton lifetime τ in the bulk; in the case of a strong drag ($\beta \gg 1$) the thickness of the exciton cloud is $r = D/v$ and it is independent of the free exciton lifetime τ in the bulk.

The luminescence intensity I^F is proportional to the total number of free excitons in the exciton cloud. Integration of Eq. (5) with respect to x gives

$$I^F \sim g\tau[1 + w(\tau/D)^{1/2}(\beta + (\beta^2+1)^{1/2})]^{-1}. \tag{9}$$

In the absence of nonequilibrium phonons ($v = 0$, $\tau = \tau_0$, $\beta = 0$) the luminescence intensity is

$$I_0^F \sim g\tau_0[1 + w(\tau_0/D)^{1/2}]^{-1}. \tag{10}$$

The relative intensity of the luminescence signal in the presence of nonequilibrium phonons is

$$\frac{I^F}{I_0^F} = \frac{\tau}{\tau_0}\frac{1 + w(\tau_0/D)^{1/2}}{1 + w(\tau/D)^{1/2}(\beta + (\beta^2+1)^{1/2})}. \tag{11}$$

In the limiting case where there is no drag ($v = 0$, $\beta = 0$) and only the $BE \rightarrow FE$ dissociation by $\hbar\Omega > 3.8$ meV phonons is allowed for, we find that $\tau > \tau_0$ and it follows from Eq. (11) that $I^F/I_0^F > 1$, i.e., the sign of the differential signal $\Delta I^F = I^F - I_0^F$ is positive ($\Delta I^F > 0$). In the other limiting case when the dissociation of bound excitons can be ignored and only the drag of free excitons need be allowed for (when $\tau = \tau_0$ and $\beta = {}^1/_2 v(\tau_0/D)^{1/2}$), it follows from Eq. (11) that $I^F/I_0^F < 1$, i.e., the differential signal is now negative ($\Delta I^F < 0$). In general, the signal I^F/I_0^F is a nonlinear function of the exciton lifetime τ and of the velocity of their drift v. Consequently, it depends in a complex manner on the occupation numbers of high-frequency phonons characterized by $\hbar\Omega > 3.8$ meV which govern the value of τ given by Eq. (7) and on the anisotropy of the momentum distribution $\Delta\bar{n}_\omega/\bar{n}_\omega$ of low-frequency phonons with $\hbar\omega < 1.8$ meV; this anisotropy determines the quantity v in Eq. (4). When the experimental conditions change, $\bar{n}_\Omega$ and $\Delta\bar{n}_\omega/\bar{n}_\omega$ vary differently and this gives rise to the experimentally observed dependences of the sign (and profile) of the signal on such experimental conditions as the state of the surface of a sample, the power of a heater, the distance from the heater to the exciton cloud, etc. (see §1).

c. Comparison of experiments with calculations. Sign of the phonon-induced exciton luminescence pulses

A qualitative interpretation of the results obtained in the main experimental geometry in the case of "transmission" of phonons from h to an exciton cloud (Fig. 1) will begin with an explanation of the dependence of the sign of a pulse $\Delta I^F(t)$ on the state of the surface of Si near which the exciton cloud is excited. On the whole, this dependence can be attributed to the known[5,14] influence of the state of the

surface on the conditions of emission into superfluid helium of low-frequency ($\hbar\omega < 1.8$ meV) phonons responsible for the drag of excitons. In fact, low-frequency phonons reaching ballistically the surface region of the exciton cloud from the source h are partly reflected at the boundary between Si and helium and are returned back into the sample and partly emerge into the helium, and the nature of the reflection (specular or diffuse) and the fraction of the reflected phonons depend on the conditions on the surface.[5,14]

The reflection of phonons from a freshly cleaved surface in He, when this surface is nearly ideal, has not yet been investigated in the case of silicon. However, by analogy with the results obtained for cleaved alkali fluoride crystals,[14] we can expect a practically 100% (specular) reflection of phonons from the boundary between a freshly cleaved Si surface and helium. Consequently, in a thin surface part of the exciton cloud, where the forward and reverse reflected ballistic phonon fluxes are superimposed, the anisotropy of the momentum distribution of nonequilibrium phonons is largely cancelled out: $\Delta\bar{n}_\omega \approx 0$. It follows from Eq. (4) that the drag of free excitons is absent (drift velocity $v = 0$) and the signal ΔI^F should be positive because of the $BE \rightarrow FE$ dissociation (see §2b), which is indeed observed exprrimentally (Fig. 1a). The existence of the $BE \rightarrow FE$ dissociation processes is confirmed directly by the quenching of the BE luminescence under the influence of heat pulses ($\Delta I^B < 0$, see Fig. 1b).

In the case of an "ordinary" silicon surface with an oxidized layer it is found, as established in a number of experiments,[5] that phonons of frequencies $\lesssim 1$ meV are scattered diffusely by the surface and effectively emerge in liquid helium (giving rise to an anomalously low Kapitza resistance at the interface between a solid and liquid helium). Therefore, a ballistic flux of phonons from a heater is uncompensated in the exciton cloud region ($\Delta\bar{n}_\omega \neq 0$). This flux drags free excitons toward the surface of Si, where they effectively recombine. This should quench the luminescence ($\Delta I^F < 0$), which is indeed observed experimentally for ordinary surfaces (Fig. 1c). Obviously, under these experimental conditions the drag of free excitons manifested in the luminescence predominates over the effect of the $BE \rightarrow FE$ dissociation (we shall consider later the origin of a short "positive" peak preceding negative pulses—see Fig. 1c).

The role of the boundary conditions is manifested also in the experiments on the dependence $\Delta I^F(t)$ on the power of the heat pulses (Fig. 3). We can see from Fig. 3 that when the heating power h is sufficiently high, so that helium boils on the opposite face of the sample where an exciton cloud is formed, the negative sign of ΔI^F typical of the ordinary surface changes to positive. This is due to the fact that when an He gas pillow forms at the surface of a crystal, low-frequency phonons can no longer emerge from the sample. They are reflected into the sample, compensating the anisotropy $\Delta\bar{n}_\omega$ of the direct flux from a heater, which is essential for the drag responsible for the sign of $\Delta I^F < 0$. We can similarly explain also the observed sign of $\Delta I^F > 0$ for an ordinary surface in vacuum, when again no phonons emerge from a sample.

The role of the conditions facilitating the emission of low-frequency phonons from a sample accounts also for the sign of the $\Delta I^F(t)$ pulses observed in the experimental geometry of Figs. 5a and 5b (inset) when the phonon flux enters the exciton cloud not directly from the heater, but after reflection of phonons from the opposite face of a thin ($L = 1.2$ mm) sample. If this face is in vacuum, then low-frequency phonons characterized by $\hbar\omega < 1.8$ meV are reflected totally by this face and then reach the exciton cloud, increasing strongly the drag of free excitons; this explains the signal sign $\Delta I^F < 0$ typical of this case (Fig. 5a). If the same face is in liquid helium, a considerable fraction of low-frequency phonons escapes into helium, so that the flux of phonons reflected from the face is much weaker and does not ensure in the exciton cloud the strong free-exciton drag necessary for luminescence quenching. The sign of ΔI^F then becomes positive (Fig. 5b) and it is due to the $BE \rightarrow FE$ processes of dissociation by high-frequency $\hbar\Omega > 3.8$ meV phonons which arrive in the exciton cloud as a result of scattering in the bulk of a sample. This conclusion follows from the preservation of the sign of the signal ΔI^F when a thin sample is replaced with a thick one ($L = 10$ mm) and the reflecting face is very far from the exciton cloud (Fig. 5c).

Therefore, the experimentally observed influence of the state of the silicon surface on the phonon-induced $\Delta I^F(t)$ luminescence signals is associated with the influence of this state on the reflection of phonons from the boundary with helium and, therefore, on the magnitude of the directional phonon flux that drags free excitons toward the surface. It follows from Fig. 2 that the phonon-induced relative change in the luminescence intensity $\Delta I^F(t)/I_0^F$ is independent of the state of an ordinary surface, i.e., it is independent of the velocity surface recombination of free excitons on which the intensity of the luminescence itself depends strongly. It follows from Eq. (11) that I^F/I_0^F ceases to depend on w if

$$w(\tau_0/D)^{1/2} \gg 1 \tag{12}$$

and then

$$\frac{I^F}{I_0^F} = \frac{(\tau/\tau_0)^{1/2}}{\beta + (\beta^2+1)^{1/2}}. \tag{13}$$

The inequality (12) allows us to estimate a lower limit of the velocity of surface recombination of excitons $\omega \gg (D/\tau_0)^{1/2} \sim 10^4$ cm/s (if $D = 10^2$ cm^2/s and $\tau_0 = 1\,\mu$s). A comparison of Eqs. (11) and (6) shows that when the condition (12) is satisfied, we have $I^F/I^F_0 = r/r_0$, i.e., the intensity of the FE luminescence reflects directly the thickness r of the exciton cloud.

Several factors may be responsible for a phenomenon observed in experiments characterized by a low heater power, viz., reversal of the "usual" negative sign of the signal $\Delta I^F(t)$ for an ordinary surface when the distance between h and the exciton cloud is increased tenfold and the phonons propagate along the ⟨111⟩ axis (Fig. 4). The most important is the geometric factor (finite size of the heater h, phonon focusing, mode and angular dependences of the exciton-phonon interaction), an allowance for which can in principle result in predominance of the $BE \rightarrow FE$ dissociation over the drag of free excitons at high values of R.

d. Profile of the phonon-induced luminescence pulses

In §2 we discussed the problem of the sign and magnitude of the observed luminescence signals ΔI^F using the steady-state solution of the system of equations (2) in the presence of nonequilibrium phonons with time-independent occupation numbers. The observed profile of the luminescence pulses $\Delta I^F(t)$ depends both on the phonon kinetics in

the exciton cloud region (i.e., on the kinetics of the occupation numbers $\bar{n}_\Omega$ of high-frequency phonons and on the flux of low-frequency phonons ($\Delta\bar{n}_\omega/\bar{n}_\omega$) and on the "inertia" of the exciton cloud acting as a phonon detector based on the emitted luminescence. The inertia of the exciton cloud can be represented by a response time $\tilde{t}$ which corresponds to the time of a transition of an exciton cloud from an initial state in the absence of nonequilibrium phonons to a new steady state with the fixed value of $\bar{n}_\Omega$ and $\Delta\bar{n}_\omega/\bar{n}_\omega$. Obviously, the kinetics of changes in $\Delta I^F(t)$ observed experimentally on a time scale much greater than $\tilde{t}$, reflects the phonon kinetics, whereas at times shorter than $\tilde{t}$ it should reflect additionally the inertia of the exciton cloud.

A rigorous determination of the inertia of this cloud requires solution of transient equations (2a) and (2b), but an approximate value of t can be estimated for two extreme cases. In one of them the drag of free excitons is ignored and only the $BE \rightarrow FE$ dissociation process under the influence of high-frequency phonons is considered. In this case we have $\tilde{t} \sim \tau \gtrsim 1\,\mu$s, where τ is the lifetime of free excitons in the bulk given by Eq. (7). Clearly, this type of inertia is largely responsible for the broadening of the leading edge of positive $\Delta I^F(t)$ pulses, induced by ballistic high-frequency phonons (Fig. 1a). In the other extreme case an allowance is made only for the strong phonon drag of free excitons, which occurs in the time $\tilde{t} \sim r_0/v = (D\tau_0)^{1/2}/v \sim 10^{-7}$ s taken by these excitons to drift from the exciton cloud to the surface (when the drift velocity is $v = 10^5$ cm/s). In this case the inertia of the FE exciton of the cloud includes a contribution also from the finite time $(w_0 + w_{BF})^{-1} \lesssim 1\ \mu$s, describing the creation of free excitons by dissociation of bound excitons localized in a spatial region (r_0) initially occupied by the exciton cloud.

In all probability the inertia of the exciton cloud is responsible for the bipolar nature of the $\Delta I^F(t)$ luminescence signal usually observed for samples with an ordinary surface washed with superfluid He (Figs. 1c, 2, and 3). The initial positive peak $\Delta I^F(t)$ can be explained by the fact that the phonons arriving in the exciton cloud in a short time $t \ll \tilde{t}$ cause $BE \rightarrow FE$ dissociation which increases the number of free excitons; moreover, after a time $t \sim \tilde{t}$ the drag process becomes dominant and quenches the luminescence ($\Delta I^F < 0$).

We shall now consider the behavior of free and bound excitons during short time intervals. If $t \ll \tilde{t}$, the change in the luminescence intensity is

$$\Delta I^F(t) \propto (N_0^B w_{BF} - N_0^F v/r_0)\,t, \tag{14}$$

where N_0^B and N_0^F are, respectively, the numbers of bound and free excitons in the exciton cloud before the arrival of nonequilibrium phonons. The first term in Eq. (14) reflects an increase in the signal $\Delta I^F(t)$ due to the $BE \rightarrow FE$ dissociation, whereas the second term describes the drift of free excitons in an exciton cloud toward the surface under the influence of the phonon wing. Assuming that in the absence of phonons ($T = 1.7$ K) we have $w_{BF} = 0$, $w_{FB} = \tau_0^{-1}$, $N_0^B = N_0^F/w_0\tau_0$, and also using Eq. (7) for τ and Eq. (8) for β, we find that the condition $\Delta I^F(t) > 0$ in the $t \ll \tilde{t}$ case is

$$2(\tau_0/\tau)^{1/2}\beta + \tau_0/\tau < 1. \tag{15}$$

On the other hand, the solution of the steady-state problem of Eq. (13) shows that the condition $\Delta I^F < 0$ in the case when $t \gg \tilde{t}$ is

$$(\tau_0/\tau)^{1/2}(\beta + (\beta^2+1)^{1/2}) > 1. \tag{16}$$

Variation of the parameters τ_0/τ and β can ensure that the inequalities of Eqs. (15) and (16) are satisfied simultaneously (for example, if $\tau = 2\tau_0$ and $\beta = 0.5$, we find that $v = 0.7 \times 10^4$ cm/s). In this case after a short "positive" rise of $\Delta I^F(t)$ there is a change of sign and a tendency to assume a steady-state value $\Delta I^F < 0$. However, if the drag is strong so that $\beta \gg 1$ ($v \gg 10^4$ cm/s), the condition (15) may not be satisfied and the signal $\Delta I^F(t)$ will be negative even for short times. This $\beta \gg 1$ case is clearly realized for short distances between h and the exciton cloud ($R = 0.3$ mm—see Fig. 4a).

In addition to the inertia of the exciton cloud, the observed profile of the $\Delta I^F(t)$ pulses manifests also the characteristic features of the kinetics of nonequilibrium phonons in the exciton cloud region, which are associated with the kinetics of phonon injection into a crystal from the heater h and with the propagation of phonons in the bulk of the sample from h to the exciton cloud. It is clear from Fig. 3 that an increase in the heating energy density W results in a strong lengthening of the trailing edge of negative $\Delta I^F(t)$ pulses caused by low-frequency ($\hbar\omega < 1.8$ meV) phonon drag. The lengthening is due to a time delay in the emission of low-frequency phonons from a strongly heated crystal-surface region adjoining the film heater h (Ref. 15). This is supported by the experimental results obtained in the main geometry, but with an additional 0.6 mm cut in a crystal,[16] which interrupts a ballistic phonon flux between h and the exciton cloud (inset in Fig. 6). Figure 6 shows the $\Delta I^F(t)$ signals measured in the absence (a) and in the presence (b) of a cut in the path between a strongly heated region h and the exciton cloud. The absence of the $\Delta I^F(t)$ signal from Fig. 6b demonstrates that the whole $\Delta I^F(t)$, pulse observed in the usual scheme (Figs. 1–3), including the extended pulse tail, is induced by the phonons arriving in the exciton cloud from h along the direct line. It should be noted that a considerable extension (lasting microseconds) of the period of emission of low-frequency phonons in the case of strong heating in the region of a film heater had been observed earlier for Si by the method of bolometric detection of phonons.[17]

We observed (§1), in the main experimental geometry, a strong ballistic component in the propagation of low-frequency $\hbar\Omega \geqslant 3.8$ meV phonons in silicon. The observation

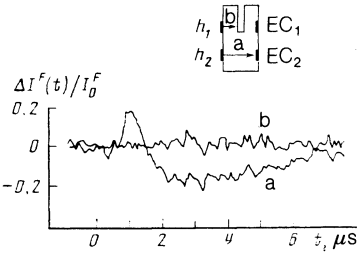

FIG. 6. Free-exciton $\Delta I^F(t)/I_0^F$ luminescence pulses obtained for different positions of the heater (h) and the phonon detector in the form of an exciton cloud (EC) relative to a cut (the experimental geometry is shown in the inset). The surface was chemically etched and the energy density was $W = 10\,\mu$J/mm^2.

(see Figs. 5b and 5c) of an $\Delta I^F(t) > 0$ signal in the experimental geometry of Fig. 5 (inset) indicated a fairly strong pulse scattering of $\hbar\Omega > 3.8$ meV phonons (most probably phonons of frequencies exceeding greatly the 3.8 meV threshold). These results are in agreement with the current ideas on the isotopic nature of the scattering of acoustic phonons in pure silicon at low temperatures. In fact, a theoretical estimate of the mean free path of phonons

$$\bar{l} = \tau^*(\omega)s = (\eta\omega_D)^{-1}(\omega_D/\omega)^4 s,$$

gives $\eta = 9.3 \times 10^{-4}$ (Ref. 18) for the scattering on the Si isotopes (4.7% of ^{29}Si and 3.05% of ^{30}Si) and this yields the value $\bar{l} = 4$ mm for phonons of 4-meV energy, which is generally in agreement with these observations.[2)]

§3. CONCLUSIONS

We reported the first experimental investigations of the influence of thermal phonon pulses on the luminescence of excitons in silicon crystals. The great variety of the effects of thermal pulses observed in the exciton luminescence yielded new data on the main properties of terahertz and subterahertz acoustic phonons in silicon and on their interaction with exciton states in Si.

1. The drag of excitons by a directional flux of nonequilibrium acoustic phonons was observed for the first time in silicon. The drift of excitons was due to the interaction with subterahertz phonons ($\hbar\omega < 1.8$ meV) and in the experimental geometry adopted by us resulted in the displacement of excitons to the surface of silicon accompanied by the quenching of the exciton luminescence because of the fast ($w > 10^4$ cm/s) surface recombination.

2. Our experiments revealed clearly a characteristic behavior, depending on the state of the Si surface, of subterahertz acoustic phonons at the boundary between silicon and superfluid helium. In the case of an atomically clean freshly cleaved Si surface we observed for the first time a strong reflection of phonons from the boundary, whereas in the case of an ordinary surface with an oxide film a considerable fraction of phonons escaped from the crystal into He.

3. Our experiments revealed important features of the frequency-dependent propagation of nonequilibrium acoustic phonons in pure silicon crystals. A ballistic component in the propagation of phonons of such a high frequency as $\hbar\Omega > 3.8$ meV was observed for the first time in silicon and this observation was in agreement with the concept of the dominant isotopic mechanism of the scattering of acoustic phonons in pure silicon at helium temperatures. The existence of a strong time delay in the emission of low-frequency ($\hbar\omega < 1.8$ meV) phonons from the surface hot region, adjoining the film heater, was observed when the heating power was sufficiently high.

The results of the present study demonstrated the very promising nature of the luminescence method for the investigation of the properties of nonequilibrium acoustic phonons in silicon crystals. The spectrally selective potentialities of the method are important: they are related to the presence of two spectrally dependent factors in the interaction of phonons with an exciton gas (drag of free excitons and dissociation of bound excitons). The method is promising for the study of the nature of the propagation of phonons in silicon and particularly the reflection of phonons from the interface between silicon and liquid helium (problem of the anomalous thermal Kapitza resistance).

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[1)] The results were reported briefly in a review paper presented at the All-Union Conference on Luminescence, held in Tallinn in 1987 (Ref. 1).
[2)] This estimate was provided by I. B. Levinson.

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