Propagation of short-wave nonequilibrium phonons and the dynamics of electron–hole droplets in germanium; sound excitation in liquid helium

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The propagation of short-wave nonequilibrium acoustic phonons and the motion of electron–hole droplets (EHD) under the action of phonon wind in germanium single crystals is studied by a method based on the detection of probing light diffraction from acoustic pulses generated in liquid helium as a result of energy transfer through the surface of an excited sample. The propagation of transverse phonons emitted into germanium by a thermal generator heated by short light pulses with energy fluence of 360–1200 erg/cm^2 is shown to be diffusive in nature for distances $\lesssim 1$ mm. The diffusion coefficient of these phonons is determined to be $D_{ph} \simeq 1.8 \cdot 10^3$ cm^2/s, and the critical densities of thermal flux into superfluid helium are determined to be $q_{2.15} \simeq 8$ and $q_{1.94} \simeq 20$ W/cm^2 at temperatures of 2.15 and 1.94 K respectively. The mean free path in resistive processes of transverse acoustic phonons generated by the relaxation of a nonequilibrium electron–phonon system of photoexcited germanium is measured to be $\Lambda_R \simeq 220$ μm. The energy stored in EHD is shown to convert to the energy of acoustic waves in liquid helium much more efficiently than the energy of nonequilibrium phonons resulting from optical pumping of the crystal.

INTRODUCTION

Information on the spatio-temporal evolution of the spectrum of nonequilibrium phonons generated by optical pumping of semiconductors is of primary importance for understanding kinetic processes in the phonon subsystem of excited crystals[1–5] and the nature of phonon wind,[6,7] which governs the dynamic behavior of a cloud of electron–hole droplets (EHD) and in some cases of excitons and nonequilibrium charge carriers.[3,6–11] Such information can be obtained by phonon spectroscopy methods.[4,12] However, most of these methods are highly specific in nature, and often cannot be used for one or another type of semiconductor or under particular experimental conditions. At the same time, qualitative information on phonon spectrum kinetics can be derived from investigations of the propagation of nonequilibrium phonons. In addition, if we are interested in the mechanism of phonon wind generation, certain conclusions can be drawn from comparisons of these investigations with data on the space-time dynamics of a cloud of nonequilibrium charge carriers.

The propagation properties of nonequilibrium phonons in a crystal depends on the relationship between relaxation times in normal and resistive processes and the time of ballistic phonon flight from the region of phonon generation to the observation point, as well as on the phonon filling factors. Phonon propagation regimes are highly diversified, and theoretical studies have been quite comprehensive.[5,13–21] The propagation of nonequilibrium phonons resulting from relaxation of charge carriers created by crystal photoexcitation has been studied experimentally in a number of papers,[22–36] in which many features of phonon subsystem kinetics important to an understanding relaxation processes going on in this subsystem were revealed and examined. However, the experimental data appear to be insufficient to construct a picture with full details. In particular, quantitative information on a phonon subsystem (phonon spectrum, relaxation times, and so on) created under specific experimental conditions at different stages in its space-time kinetics is needed. A similar group of problems, albeit each with their specific features, exists also when nonequilibrium phonons are injected into a crystal by a thermal phonon generator with a temperature much higher than the temperature of the crystal.[24,27,30,34,37–44]

Investigations of phonon subsystem kinetics should also address the mechanism for the generation of phonon wind that appears in the process of its relaxation at both comparatively low and high pump levels, when the formation of a phonon hot spot is possible.[18–20] Phonon wind is considered to mean flows of relatively long-wave acoustic phonons efficiently absorbed by free charge carriers, excitons, and EHD (for EHD, these are the phonons with a wave vector $q \lesssim 2k_F$ where k_F is the Fermi wave vector of carriers in the liquid phase). It is well known that as it undergoes spatio-temporal evolution, an EHD goes through at least three successive stages associated with different mechanisms of phonon wind generation.[1)] The first stage, discovered in Refs. 45–47, is due to the "primary" phonon wind emitted at the final stage of thermalization of light-produced charge carriers, when their kinetic energy becomes less than the Fermi energy of the carriers bound to EHD.[8,11,45,47–49] The third and final stage is cloud expansion because of mutual EHD repulsion, resulting from the fact that each EHD absorbs phonons emitted by other EHDs.[6,50,51] The second stage is most conspicuous in the dynamics of an EHD cloud. It was observed in most experimental work devoted to EHD motion under pulsed excitation.[11,40,45–47,50–56] Phonon wind most likely appears

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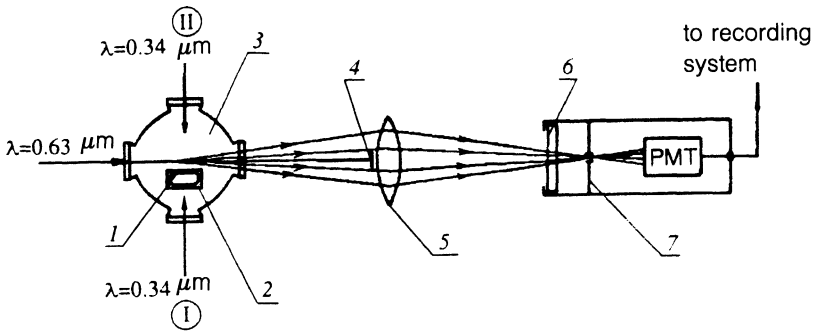

FIG. 1. Optical layout of the experiment. *1*) Germanium sample; *2*) sample holder; *3*) helium bath; *4*) beam blocker; *5*) collecting lens; *6*) filter; *7*) entrance aperture.

upon relaxation of phonon subsystems formed via thermalization of photoexcited electrons and holes. This is the mechanism of generation that makes the major contribution to the phonon wind responsible for the formation of spatial structure in a steady-state EHD cloud.[3,8,57–60] However, there appears to be no definite answer yet to the question as to what processes are responsible for the transformation of the energy of high-frequency acoustic phonons to phonon wind at distances of the order of EHD cloud dimensions (0.2–3 mm) in times of 0.5–10 μs (the duration of the second stage of cloud dynamics).[3,7,8,58]

There is one more point that has received insufficient attention, in our opinion. The second stage of EHD cloud dynamics is observed most clearly either in samples in which the excited surface is in vacuum (no contact with liquid helium),[45–47] or under such an intense pumping[11,40,52–56] that a film of gaseous helium, which prevents nonequilibrium phonons from leaving the sample and entering the liquid helium, is formed on the sample surface at the laser focus in an extremely short ($\leqslant$0.1 μs) time.[2)] The formation of a vapor bubble at the sample surface being excited also results in a considerable increase in steady-state EHD cloud dimensions.[60,62,63] In this connection, a number of questions arises, having to do with the role of phonon reflection from the sample surface in the transformation of high-frequency phonon into phonon wind, the effect of vapor film formation on the lifetime of a phonon hot spot and its dependence on the pump intensity observed in Refs. 27 and 56, etc.

A vapor film is formed if the thermal flux density from a solid body to liquid helium exceeds a critical value that depends on the experimental conditions (temperature, pressure, pump pulse length, and other factors).[64–70] The occurrence of a vapor film is accompanied by sound wave excitation in liquid helium. These waves were observed to appear during pulsed heating by thermal generators[71–74] and optical pumping of semiconductors[75,76], as well as blowing of EHD onto the sample surface under the action of phonon wind[61] (as a result of rapid energy release in the process of radiationless surface recombination of electrons and holes[77]).

In the present paper, propagation of nonequilibrium acoustic phonons and EHD motion in germanium are studied. The method used is based on the detection of probing light diffraction from pulses of first sound generated in liquid helium as a result of energy transfer through an excited sample surface.[61,75,76] Nonequilibrium phonons were produced both by a thermal phonon generator heated by laser pulses and by the thermalization of photoexcited charge carriers. The experiments were performed on samples entirely immersed in superfluid helium and samples with an excited surface in vacuum. Mean free paths of acoustic phonons detected in resistive processes and critical thermal flux densities into liquid helium are determined. It is shown that for identical energies stored in the electron and phonon subsystems after completion of the charge carrier thermalization process in a crystal excited by a short laser pulse, the density of the electron-excitation energy flux induced by phonon wind considerably exceeds the flux density of energy carried by nonequilibrium phonons at distances $d > \Lambda_R$ (Λ_R is the phonon mean free path in resistive processes).

EXPERIMENTAL TECHNIQUE

A mechanically polished germanium disk (with residual impurity concentration less than 10^{12} cm^{-3}) of diameter 10 mm and thickness d (the disk axis is parallel to the $\langle 111 \rangle$ direction) was either placed directly in liquid helium or mounted in a special holder in such a way that one of its planes was in vacuum and the other was immersed in liquid helium (Fig. 1 and insert in Fig. 2). To investigate

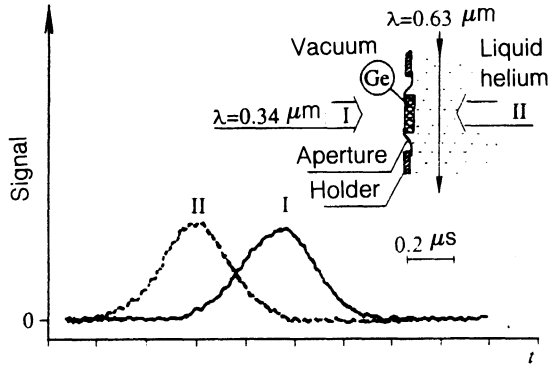

FIG. 2. Signals proportional to the intensity of light diffracted from the pulses of first sound excited in liquid helium at the surface illuminated by laser radiation (II) and at the unilluminated surface (I) of the sample. When recording the pulse I, the pump pulse energy was $W \simeq 120$ erg. The laser spot diameter at the sample was $\simeq$3 mm. The experimental geometry is shown in the insert.

the propagation of nonequilibrium phonons emitted into germanium by a thermal generator, we used samples with an aluminum film 0.55 μm thick evaporated on the disk plane that remained in vacuum during the experiment. Germanium surface photoexcitation was accomplished either by a copper vapor laser (wavelength $\lambda \simeq 0.51$ μm, pulse length $t_p \simeq 10$ ns, pulse repetition frequency $f \simeq 5$ kHz, maximum pulse energy $W_{max} \simeq 120$ erg) or by a nitrogen laser ($\lambda \simeq 0.34$ μm, $t_p \simeq 10$ ns, $f \simeq 50$ Hz, $W_{max} \simeq 120$ erg). The latter was also used for pulsed heating of the aluminum film, serving as a thermal generator of phonons. The diameter of the excitation light spot on the sample was 3–5 mm.

The probing light beam from a CW He–Ne laser ($\lambda = 0.63$ μm passed through liquid helium parallel to the sample surface at a distance much greater than the size of a gaseous helium bubble formed as a result of energy transfer through this surface[61] (Figs. 1 and 2). This beam was focused in such a way that the beam waist ($\simeq 100$ μm in diameter) was in front of the center of the germanium disk. Probing light diffracted (scattered at small angles) by sound pulses propagating in the liquid helium was collected onto an entrance aperture in front of the PMT used for detection. The direct laser beam was blocked by a screen set in front of the collecting lens (Fig. 1). A set of optical filters in front of the PMT entrance aperture prevented stray illumination of the PMT photocathode by exciting laser radiation scattered by optical elements of the installation, recombination radiation of the sample, and light from other sources in the room.

In a number of experiments, probing laser beam attenuation resulting from this scattering was recorded instead of light scattered by sound pulses. In this case, an aperture blocked the diffracted light in front of the collecting lens, instead of a screen. In several experiments in which high spatio-temporal resolution was not needed, He–Ne laser radiation with $\lambda \simeq 3.39$ μm was used as the probing light. It was detected by a Ge:Au photoresistor cooled to liquid nitrogen temperatures. Electric pulses produced by scattered light incident on the PMT or photoresistor were first amplified and then sampled and recorded on a two-coordinate recorder. The temporal resolution of the installation operating with a PMT was $\simeq 20$ ns.

The time of EHD motion or nonequilibrium phonon propagation through the sample was measured from the time shift of a signal pulse relative to a reference pulse (pulses I and II respectively in Fig. 2). The signal pulses resulted from diffraction of probing light from the pulses of first sound[3)] generated at the germanium disk plane facing the probing laser beam, its opposite plane in vacuum or in liquid helium having been excited by the laser beam I (Figs. 1 and 2) or the thermal generator evaporated on this plane having been heated by this beam. The reference pulse was produced when the laser beam II excited the sample plane nearest the probing beam (Figs. 1 and 2). With the help of neutral density optical filters, the energy in the laser pulse II exciting a pulse of first sound was selected in such a way that the pulses I and II had approximately equal amplitudes (Fig. 2). As a result, the sound pulses I and II

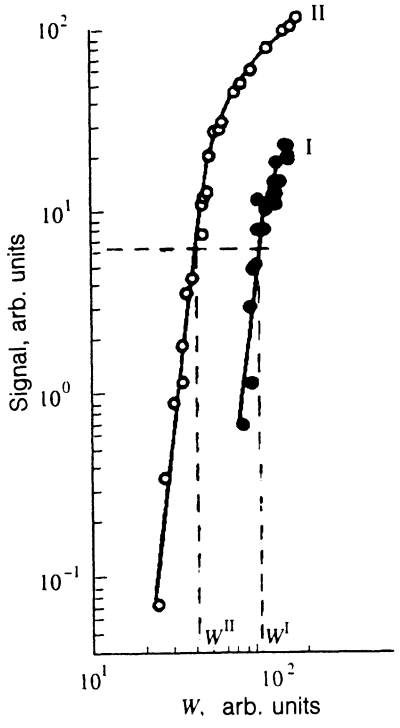

FIG. 3. Signal amplitudes of probing light diffraction from pulses of first sound excited at the illuminated (II) and unilluminated (I) planar facets of a sample of thickness 130 μm as functions of the laser pulse energy; $T = 1.7$ K.

were low-intensity shock waves propagated in liquid helium with identical velocities,[76] so their transit times from the sample surface to the probing beam were the same. The measurements were made over the temperature range 1.7–2.15 K. The pressure above the superfluid helium surface was equal to the saturated vapor pressure.

In certain of the experiments, a superconducting bolometer made of granular Al evaporated on the disk plane immersed in liquid helium was used to measure the time needed for nonequilibrium phonons generated by excitation of the *in vacuo* sample surface to traverse the sample.

To measure germanium fluorescence spectra, a standard installation containing a wide-aperture MDR-2 monochromator with a 300-line/mm grating and a germanium photodiode as a detector was used. To detect EHD motion in bulk germanium, the spatio-temporal distribution of probing light with $\lambda = 3.39$ μm scattered by EHD[45] was measured. These measurements were made on samples of size $3 \times 5 \times 15$ mm^3 whose working half was in vacuum.[45]

RESULTS AND DISCUSSION

First we present the results obtained on thin germanium samples ($d < 200$ μm) immersed entirely in superfluid helium. In Fig. 3, the intensity of probing light diffracted from the pulses of first sound excited at the sample facet irradiated by pump light and at the opposite facet is presented as a function of pump pulse energy. The diffraction signal magnitude is proportional to the diffracted light

intensity, which in turn is proportional to the square of the relative variation of the liquid helium refractive index in a sound pulse. As seen from Fig. 3, a significant increase in the pump pulse energy is required to excite sound pulses at the sample side opposite the illuminated one.

The transit time of excitation resulting from pumping was measured in accordance with the technique described above for a sample of thickness $d=180$ μm. It was shown that the excitation energy is transferred to the nonilluminated surface of the sample at the velocity of transverse sound. Most probably, this means that sound excitation in helium in this case is governed by the energy flux density of transverse acoustic phonons propagating in the ballistic mode from the excitation region of the sample to its nonilluminated surface. If so, the difference in the amplitudes of sound pulses excited at the illuminated and the opposite surfaces of the sample (compare the diffraction signals II and I in Fig. 3 for equal energies W in a laser pulse) should be related to the attenuation of the phonon flux, as it propagates deep in the sample, due to resistive scattering of nonequilibrium phonons.

Data like that presented in Fig. 3, obtained from samples of different thicknesses, make it possible to determine the mean free path of nonequilibrium phonons in resistive processes. It is quite clear that sound pulses excited at the illuminated and the opposite sides of the sample will have equal amplitudes if the flux densities of phonon energy incident on these surfaces are equal. Assuming that the total energy of phonons produced by pumping the sample (that can efficiently excite sound in helium) is proportional to the energy in a laser pulse, we obtain, by equating the phonon energy flux at the two sample surfaces, an expression for one-dimensional phonon propagation:[4)]

$$\frac{W^{II}}{W^{I}}=\frac{\alpha}{1-\alpha}\exp(-d/\Lambda_R), \qquad (1)$$

where W^{I} and W^{II} are the pump pulse energies for which sound pulses of equal amplitudes are excited at the nonilluminated and illuminated sides, respectively (see Fig. 3), α is the fraction of phonons generated in the excitation region with quasimomentum directed toward the nonilluminated surface of the sample, $\Lambda_R=s_T\tau_R$ is the mean free path of nonequilibrium acoustic phonons in resistive processes, τ_R is the corresponding time of flight, and s_T is the transverse sound velocity. The exponential in formula (1) gives the probability that a nonequilibrium phonon does not experience resistive scattering during ballistic propagation through the sample.

The ratio W^{II}/W^{I} is presented in Fig. 4 as a function of the thickness of the samples investigated. The slope of the straight line yields the phonon mean free path, $\Lambda_R\simeq 220$ μm. Extrapolating this line to $d=0$, we find that $a=0.4$. As should be expected, about half the phonons generated enter the sample at distances $x\ll\Lambda_R$ from the excited surface.[5)] This result is extremely important for a correct explanation of causes for strong suppression of the second stage of the EHD cloud dynamics (see Introduction) in samples entirely immersed in liquid helium at moderate

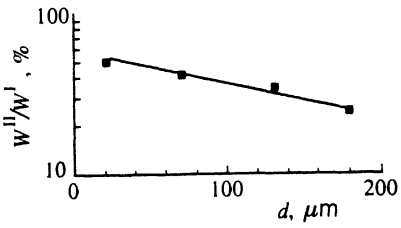

FIG. 4. Ratio W^{II}/W^{I} as a function of sample thickness; $T=1.7$ K. The slope of the straight line gives the mean free path of nonequilibrium acoustic phonons in resistive processes, $\Lambda_R\simeq 220$ μm.

pump intensities,[51] and probably also for explaining the dependence of the phonon hot spot lifetime on the excitation level observed in Refs. 27 and 56.

We now turn to the results obtained on samples with one plane in vacuum (the one excited by beam I; see insert in Fig. 2). Two sets of experiments were carried out. In one, we studied the propagation of nonequilibrium phonons emitted into germanium by a thermal generator heated by laser pulses. In the other, the laser beam was focused directly on the germanium surface. The results of these two types of experiments turned out to be substantially different (Fig. 5). First we discuss the data obtained using a thermal generator of nonequilibrium phonons.

As can be seen from Fig. 5, the recorded propagation time of nonequilibrium phonons decreases with increasing laser pulse energy, and at high energies absorbed by the heat generator in a single laser pulse, W^{abs}, it coincides with the transverse sound propagation time ($s_T=3.07\cdot 10^5$ cm/s). At $T=2.15$ K (open hexahedra), the critical thermal flux density into liquid helium, corresponding to formation of a vapor film at the sample surface with accompanying generation of first sound, is lower than at $T=1.94$ K (filled hexahedra). The dependence of t on W^{abs} at $T=2.15$ K was therefore measured over a wider range of

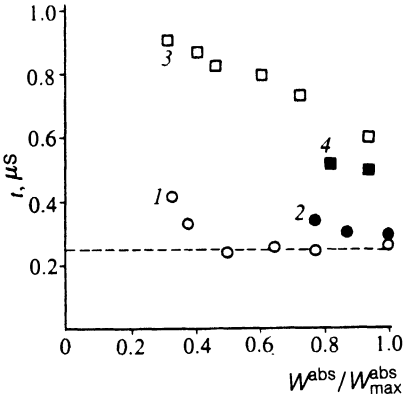

FIG. 5. Excitation propagation time through a sample of thickness $d=0.72$ mm as a function of the energy absorbed. W^{abs}_{max} is the maximum energy absorbed by the aluminum film in a single laser pulse. 1,2 Phonons are emitted by the thermal generator; *3,4*—excitation of the germanium surface; *1,3*—$T=2.15$ K; *2,4*—$T=1.94$ K. The dashed line shows the transverse sound propagation time ($s_T=3.07\cdot 10^5$ cm/s).

laser pulse energies. For the same reason, the recorded phonon propagation time decreases with increasing temperature.

The results can be satisfactorily explained in terms of diffusive propagation of phonons emitted by an instantaneous source with allowance for the threshold properties of sound excitation in liquid helium. Let us assume that the nonequilibrium phonons detected have approximately the same frequencies and, as a consequence, their propagation can be described by a single diffusion constant D_{ph}. Then under the assumption that all of the energy absorbed by the thermal generator is converted into phonons characterized by the diffusion coefficient D_{ph}, we have an expression for the one-dimensional phonon energy flux density at time t and distance d from an instantaneous source operating at time $t=0$ at the surface of a crystal occupying a half-space:

$$j=\frac{W^{abs}}{2\sqrt{\pi\sigma t}}\frac{d}{\sqrt{D_{ph}t}}\exp\left(-\frac{d^2}{4D_{ph}t}\right), \qquad (2)$$

where σ is the area of the light spot at the surface of the thermal generator. Setting j equal to the critical thermal flux density into liquid helium, we can derive an expression describing the relation between the recorded propagation time t and the luminous energy W^{abs} absorbed by the thermal generator. However, for experimental data processing, it is more convenient to use an equation expressing this relation in dimensionless form. To derive this equation, we write down an expression for the maximum phonon energy flux density at distance d from the source,

$$j_{max}=\frac{3\sqrt{6}W^{abs}}{\sqrt{\pi\sigma t_m}}\exp(-3/2), \qquad (3)$$

where $t_m=d^2/6D_{ph}$ is the time it takes for the diffusive flux at the nonilluminated surface to reach its maximum. Denoting by W_m^{abs} the absorbed energy for which $j_{max}=q_T$[6)] and equating the right-hand sides of (2) and (3), we obtain the desired equation,

$$\left(\frac{W}{W_m}\right)^{2/3}=\frac{t}{t_m}\exp\left[-\left(1-\frac{t_m}{t}\right)\right]. \qquad (4)$$

We have taken into account in (4) that $W^{abs}/W_m^{abs}=W/W_m$, where W and W_m are the laser pulse energies for which the measured nonequilibrium phonon propagation time is t and t_m, respectively.

The fitting parameters W_m and t_m were determined by processing experimental data with the help of Eq. (4) (see Fig. 6). For a sample of thickness $d=0.72$ mm, we have $t_m\simeq 4.8\cdot 10^{-7}$ s. Substituting this value of t_m into the foregoing formula relating t_m and D_{ph}, we obtain $D_{ph}\simeq 1.8\cdot 10^3$ cm/s, for the diffusion coefficient of nonequilibrium transverse phonons detected in our experiment and with the aid of the formula

$$D_{ph}=(1/3)\Lambda_R s_T,$$

we find their mean free path in resistive processes, $\Lambda_R\simeq 180$ μm. This value of Λ_R is not much different from the nonequilibrium phonon mean free path measured in the preceding experiments on thin samples of photoexcited germanium. Thus, phonons with approximately the same frequencies were observed in these two types of experiments.

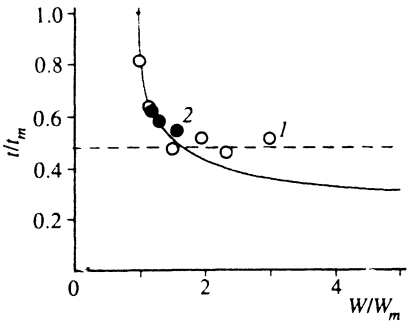

FIG. 6. The results of processing the dependences 1 and 2 presented in Fig. 5 using expression (4). The notation for the dots is the same as in Fig. 5. The solid curve shows the theoretical dependence. The dashed curve shows the transverse sound propagation time.

The proportionality factor between W^{abs} and W must be known to determine the critical thermal flux densities into liquid helium. This factor was found by measuring the incident and reflected luminous flux at the thermal generator; it turned out that $W^{abs}\simeq 0.3W$. Upon substituting $W^{abs}\simeq 0.3W_m$ in expression (3), we obtain $q_{1.94}=20$ W/cm^2 and $q_{2.15}=8$ W/cm^2 for the critical thermal flux densities into superfluid helium at $T=1.94$ K and $T=2.15$ K, respectively. These values of q_T are appreciably greater than the critical thermal flux density typical of steady-state heat exchange.[64,65] However, for nonsteady heat transfer from a solid body to liquid helium, the critical thermal flux density is related to the time elapsed since the onset of the process. The form of this dependence is dictated by the mechanism of vapor film formation at the sample surface and the regime of heat propagation in liquid helium. For both normal and superfluid helium, the experimental results are quite well described by one and the same relation,

$$q_T t^{0.5}=C_T, \qquad (5)$$

although physical processes in He I and He II, which are represented by this formula, are naturally different.[67–70,73,74] Substituting $t=t_m$ and the values of q_T given above into (5), we find numerical values of the constant C_T: $C_{1.94}\simeq 1.4\cdot 10^{-2}$ W$\cdot$s$^{1/2}$/cm^2 and $C_{2.15}\simeq 5.5\cdot 10^{-3}$ W$\cdot$s$^{1/2}$/cm^2. The calculated value of $C_{1.94}$ is close to the value of $C_{3.8}$ for normal helium measured in Ref. 74. However, it is about one-third the value of $C_{1.8}$ found in Ref. 70 for He II. Note that the critical thermal flux density measured from the threshold energy of first sound pulse excitation, as is done in this work, is several times the lowest energy flux density at which a gaseous helium film is observed to form at the sample surface.[61] This probably means that rapid expansion of this film is required to excite pulses of first sound.

We now turn to a discussion of the results obtained with surface optical pumping of germanium samples whose excited surface is in vacuum. As can be seen from Fig. 5,

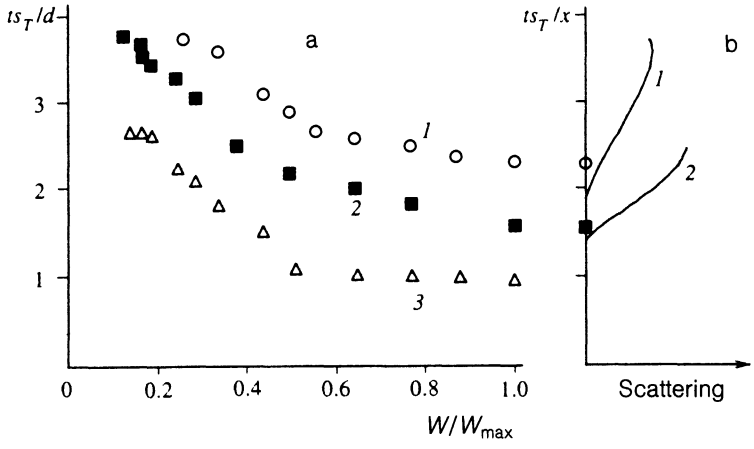

FIG. 7. a) Propagation time of excitations produced with germanium optical pumping through samples of different thicknesses d as functions of laser pulse energy. *1*—$d=0.5$ mm; *2*—$d=0.72$ mm; *3*—1 mm; $T=2.1$ K. (b) Kinetics of signal build-up for light scattering from EHDs at different distances [($x=0.99$ mm (*1*) and 0.72 mm (*2*)] from the excited surface ⟨111⟩ of Ge sample 5 mm thick; $W=W_{max}\simeq 120$ erg.

the time for an excitation to propagate through the sample in this case is much greater than the propagation time of nonequilibrium phonons emitted by the thermal generator into the crystal. Moreover, in contrast to the nonequilibrium phonon propagation time (open and filled hexahedra in Fig. 5), the propagation time measured with optical pumping of germanium (open and filled squares) decreases with falling temperature. The propagation time as a function of the pump pulse energy is presented in Fig. 7a for three samples of photoexcited germanium. The excitation propagation time is seen to decrease with increasing pumping level. For the two thicker samples, it is appreciably greater than the transverse sound propagation time, and only for the thinnest of the samples investigated ($d=0.5$ mm) is it comparable at high excitation pulse energies.

The observed behavior of the propagation time as a function of sample thickness, temperature, and pump intensity is typical of EHD moving to the unilluminated surface of the sample under the action of phonon wind (decreasing propagation time with falling temperature is associated with an increase in the EHD mobility.[3,8,45–47,52,53] In this case, the excitation of sound pulses in helium is related to the rapid release of energy stored in EHDs via the nonradiative recombination of electrons and holes that are bound in droplets at the nonilluminated surface of the sample reached by liquid helium.[61,77] We published the results of one of the experiments proving the feasibility of sound generation in helium as a result of the release of electron excitation energy propagating through a germanium crystal in the form of EHD in Ref. 61. Here we present additional evidence for the existence of this effect.

The results of measuring signal build-up kinetics for light scattering by EHD are given in Fig. 7b. Germanium samples of size $3\times 5\times 15$ mm^3 were used. The working half of the samples was in vacuum (its height was 7–8 mm), while the other half was immersed in liquid helium. EHDs were created by exciting one of the narrow facets of a sample, and they were driven deep into the sample by the phonon wind. The sample thickness in the direction of droplet motion was 5 mm. Comparison of the data presented in Figs. 7a and 7b shows that for identical pump intensities ($W=W_{max}$), the propagation times given by light diffraction by sound pulses in helium and by light scattering by EHDs are essentially identical. Note that the minimum pump intensity for which detection of sound pulses excited at the unilluminated side of the sample was still possible (Fig. 7a) also indicates that excitation energy is transported by liquid-phase droplets. In contrast, when the nonequilibrium phonon propagation time was measured by the method used, the pump threshold intensity depends on the sample thickness (for $d\lesssim\Lambda_R$ and $d>\Lambda_R$), as seen from Fig. 4 and expression (3). The aforesaid is not true for the results obtained with a sample of thickness $d=0.5$ mm in the pump energy range $W\gtrsim 0.4W_{max}$ (Fig. 7a). In this case, sound at the unilluminated sample surface is probably excited by nonequilibrium phonon flow propagating to this surface. This conclusion follows from the data obtained in Ref. 47: in the pump intensity range investigated, the average EHD velocity at distances $x\sim 0.5$ mm is markedly lower than the transverse sound velocity.

The results of another experiment are presented in Fig. 8. For a thin sample, a rising EHD recombination emission intensity gives way to a sharp drop due to a high surface recombination rate at times corresponding to the time required for a droplet to move to the unilluminated side of the sample (compare with data presented in Fig. 7a).

The excitation of sound in liquid helium at the unilluminated surface of photoexcited germanium samples of thickness $d>0.5$ mm is thus associated with excitation energy transport by electron–hole droplets, in spite of the fact that under our experimental conditions, the fraction of excitation energy stored in EHDs is several times lower than that in the phonon subsystem. The question arises as to why nonequilibrium phonons, which reach the unilluminated surface of an optically pumped germanium sample before EHDs, do not excite sound, whereas phonons injected into the crystal by a thermal generator produce sound in liquid helium even at energies W^{abs} much less than the energy transferred to the germanium phonon subsystem via photoexcited charge-carrier relaxation. The answer can be found by comparing spectra of nonequilibrium phonons emitted into a crystal by the thermal generator and phonons produced by semiconductor optical pumping. As already mentioned in the Introduction, the spectral distribution of nonequilibrium phonons in a photoexcited

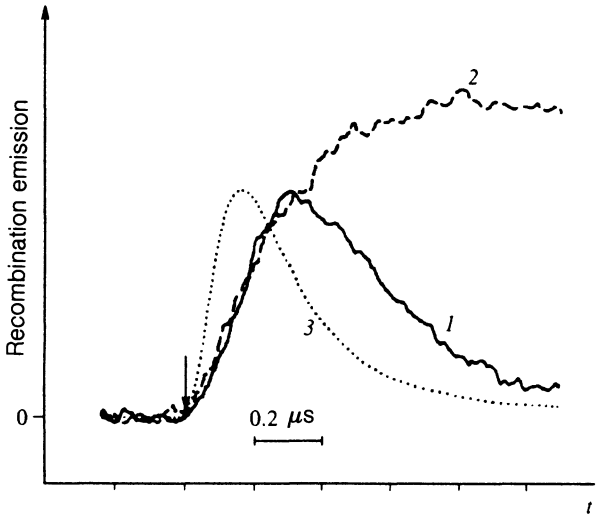

FIG. 8. EHD fluorescence intensity as a function of time elapsed from the excitation pulse (its position is marked by the arrow) for samples of thickness 0.72 mm (*1*) and 5 mm (*2*); $W \simeq 120$ erg, $T = 1.9$ K. Curve *3* is the signal recorded by a photodetector illuminated by the laser pulse.

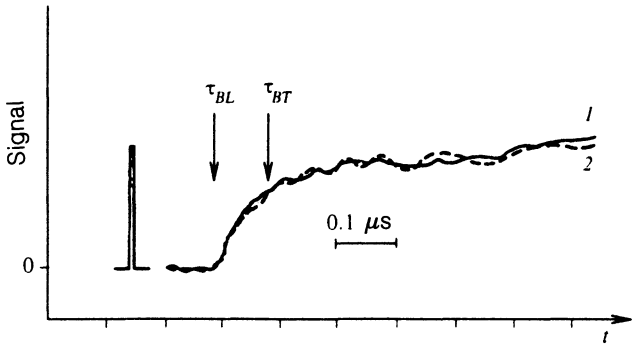

FIG. 9. Kinetics of signal build-up for nonequilibrium phonons detected by bolometer under the conditions of germanium ⟨111⟩ photoexcitation (*1*) and thermal generator laser heating (*2*); $d = 0.72$ mm, $T = 1.8$ K. The energies absorbed W^{abs} are the same in both cases. The times of ballistic propagation of longitudinal (τ_{BL}) and transverse (τ_{BT}) acoustic phonons are marked by the arrows. The rectangular pulse shows the position of the pump pulse.

crystal depends critically on experimental conditions whose effect on the phonon subsystem properties and behavior have been studied theoretically in great detail.[5,13–21] We shall not discuss this problem in detail, restricting ourselves to brief qualitative considerations directly related to our experiments.

Note above all that the maximum excitation pulse energy in our experiments was a factor of about 50 less than required, according to the estimates by Kazakovtsev and Levinson,[18] for phonon hot spot formation. This means that the temperature of the phonon subsystem of a photoexcited crystal has no time to be established during the interval needed for phonons to propagate to the observation point, and the principal mechanism for frequency degradation of short-wave acoustic phonons resulting from the decay of optical phonons generated by the relaxation of light-produced charge carriers is their spontaneous decay. The phonon frequency distribution produced under these conditions is shifted to higher frequencies relative to the Planck spectrum of phonons emitted by a sufficiently thick metal film (thermal generator), and is depleted of low-frequency phonons.[24,34] In addition, short-wave phonons accumulated in lower nondecaying acoustic branches have a long lifetime at low temperatures,[78] and have no chance to relax during the observation time. Therefore, a considerable fraction of the energy of the phonon subsystem in optically excited crystals is accounted for by high-frequency phonons. In sufficiently pure germanium at low temperatures, the phonon diffusion coefficient

$$D_{ph} = (1/3)\Lambda_R \tau_R = s\tau_R^2$$

is dominated by scattering from isotopic impurities. The probability of this scattering is given by (see, for instance, Refs. 3, 5 and references therein)

$$\tau_R^{-1} = A\omega^4 \tag{6}$$

For germanium, $A \simeq 2.5 \cdot 10^{-44}$ s^3.[38] As a consequence, the phonon diffusion coefficient increases rapidly with decreasing frequency ω, and low-frequency phonons reach the sample's unilluminated surface faster. Moreover, the maximum energy flux density is proportional to the phonon diffusion coefficient, as can be seen from (3). Therefore, sound in helium is most likely excited by a flux of relatively low-frequency phonons incident upon the sample surface immersed in liquid helium. With due regard for the difference between the spectra of nonequilibrium phonons generated in the sample by the thermal generator and by optical pumping, we may expect that sound will not be generated at the unilluminated surface of the photoexcited sample in the pump level range investigated.

These arguments are confirmed by the proximity of the values of the phonon mean free path Λ_R [and consequently the frequencies of phonons responsible for sound generation in helium, according to (6)] determined from the experimental data for thin samples of photoexcited germanium (Fig. 4) and from the experiments with a thermal generator (Fig. 6). One more argument in favor of the aforesaid was obtained in the experiments with a superconducting bolometer placed at the sample surface immersed in liquid helium, serving to detect nonequilibrium phonons. The shapes of the leading edges of phonon pulses detected by the bolometer following germanium photoexcitation (curve *1*) and laser heating of the thermal generator (curve *2*) are compared in Fig. 9. The fact that the edge shapes are the same at times corresponding to the phonon propagation times suggests that nonequilibrium phonons resulting from photoexcitation of the sample that reach its unilluminated surface within this time interval have a frequency spectrum similar to that of the phonons responsible for sound excitation in helium in the experiments with a thermal generator.

The phonon frequency can be estimated from the measured mean free path Λ_R using Eq. (6). Such an estimate

shows that transverse phonons with $\Lambda_R = 220$ μm have an energy $\hbar\omega \simeq 3.2$ meV, which is about an order of magnitude higher than the limiting energy of transverse phonons that contribute to the phonon wind that drags the droplets in the ⟨111⟩ directions,[8] i.e., nonequilibrium phonons responsible for sound excitation in helium do not participate in EHD dragging. We shall not discuss here the mechanisms for phonon wind generation,[8] since the experimental results presented here do not provide any direct guidance on this question. However, we shall make some remarks concerning the effect of gaseous helium film formation at the surface of an excited sample on the generation of phonon wind, which is responsible for EHD motion in the second stage of droplet cloud dynamics (see Introduction).

Above all, note that the vapor film did not cover the whole illuminated sample surface at the pump intensities used in our experiments (the cases in point are the excitation by the beam II (see insert in Fig. 2) or samples entirely immersed in liquid helium). It had an area close to the laser spot area at this surface. This was also true of the vapor film formed at the unilluminated surface of the sample when nonequilibrium phonons or EHDs were incident upon it. In this case, sample overheating relative to the helium bath is negligible (as evidenced by the existence of EHD itself) and is local. By "local overheating" we mean the presence of nonequilibrium phonons which have not necessarily come to thermal equilibrium (quite the contrary, as noted earlier) in some fraction of the sample volume that increases with time.

An expanding cloud of nonequilibrium phonons is a source of phonon wind, i.e., relatively long-wavelength phonons emitted in the process of phonon subsystem relaxation, interacting efficiently with EHDs. The rate of phonon wind generation τ_{ph}^{-1} was obtained from the measurements of the EHD cloud dimensions as a function of time elapsed after the excitation pulse (or excitation turn-on) in the second stage of droplet cloud dynamics,[40,45,56,57] as well as by the method based on spatial filtering of ballistic phonons detected by a superconducting bolometer.[27] In the experiments conducted on samples entirely immersed in liquid helium, the relaxation time τ_{ph} was observed to increase with pump level.[27,56] Such behavior of τ_{ph} is possible when a phonon hot spot is formed.[20] However, it seems to us that the observed increase in relaxation time is associated with the formation of a gaseous helium bubble at the laser focus. In these experiments, the minimum pump density was much higher than the highest excitation density used in the present paper, and the increase in τ_{ph} with pump level (its value was $\tau_{ph} \simeq 1.3$ μs at the highest pump level) is most likely related to an increase in the vapor film dimensions or a decrease in its formation time. It was found in Ref. 40 that $\tau_{ph} \simeq 1.1$–1.3 μs independent of the pump intensity, and judging by the experimental conditions, the gaseous bubble formed over the whole range of pump levels. In Ref. 57, a value $\tau_{ph} \simeq 2.6$ μs was obtained for the relaxation time. As was noted later by the same authors,[60] pumping of the sample was accompanied by helium boiling at the laser focus.

Our investigations of EHD cloud dynamics on samples with the working half in vacuum[45] showed that $\tau_{ph} \simeq 1.8$ μs, and does not depend on the pump level over the range $W = (0.2$–$1.0)\,W_{max}$ (the maximum excitation density was the same as in the present work), with a sample temperature in the range $T = 1.7$–4.2 K. The latter means that it is unlikely that relaxation of nonequilibrium phonons of the nondecaying transverse branches plays an appreciable role in phonon wind generation, because the relaxation rate of such phonons depends heavily on temperature.[78] In the experiments carried out under the same conditions but on samples entirely immersed in superfluid helium, we observed strong attenuation of the intensity of phonon wind responsible for the second stage of EHD cloud dynamics, and a sharp decrease (more than an order of magnitude) in the relaxation time τ_{ph}.[51] As noted in Ref. 51, this effect is related to the outflow of short-wavelength nonequilibrium phonons into the liquid helium through the illuminated facet of the sample. Although about half the phonons generated initially have a quasimomentum component directed away from the excited surface (the conclusions concerning the coefficient α drawn in the discussion of Fig. 4 are probably valid for phonons of all generations), most phonons return to the excited facet of the sample during diffusive propagation of high-frequency phonons, just as short-wavelength phonons emitted by a thermal generator into the substrate return and slow down its cooling.[41–43]

CONCLUSION

From studies of the propagation of nonequilibrium phonons injected into germanium by a thermal generator heated by short optical pulses and phonons generated by the thermalization of photoexcited charge carriers, we have measured the diffusion coefficient of transverse acoustic phonons and their mean free path in resistive processes. Nonequilibrium phonons with similar energies are shown to be detected for both techniques of phonon generation, although the energy flux density of such phonons produced with the help of a thermal generator is substantially higher than when optical pumping is used for identical energies supplied. At the same time, when the energies of optical pumping stored in the electron and the phonon subsystems are equal, the energy transported by EHDs from the excited surface of a crystal is much more efficiently converted at its opposite surface to acoustic energy in liquid helium than the energy of nonequilibrium phonons. The critical thermal flux densities q_T into superfluid helium at temperatures of 1.94 and 2.15 K has been determined, and the temperature-dependent constant C_T used in the description of the relation between q_T and the formation time of a vapor film at the sample surface under the conditions of nonsteady heat transfer from a solid body to liquid helium has been estimated.

The results of this and other work discussed at the end of the preceding section imply that the formation of a gaseous helium bubble on the sample surface at the laser focus has a significant influence on relaxation processes in the phonon subsystem of an excited crystal. The formation of the bubble most probably affects the feasibility of nonequi-

librium phonon energy conversion in collisions with the excited surface.[79,80] Further experimental investigations are needed for a definitive answer to the question of the part played by these effects in phonon subsystem kinetics.

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[1)]EHD cloud dynamics and a great variety of phenomena associated with exciton and EHD drag by phonon wind are considered in detail, in Reviews 3 and 8.

[2)]This question is considered in more detail in Ref. 8.

[3)]In superfluid helium, pulses of not only first, but also second sound are excited.[70–73,75] However, pulses of first sound are more convenient to use for our measurements because of the significantly higher magnitude of the scattering signal and an order of magnitude higher propagation velocity in the working temperature range.

[4)]Phonon propagation can be considered one-dimensional, because in our case the linear dimensions of the laser spot are much greater than the sample thickness.

[5)]In fact, somewhat less than one-half ($\alpha < 0.5$). This is probably associated with the outflow of phonons of preceding generations, which possess higher frequencies than those detected in the experiment, through the excited surface from the sample to the liquid helium. Correspondingly, these phonons have a significantly smaller mean free path Λ_R.

[6)]In essence, W_m^{abs} is the minimum energy absorbed by the thermal generator per laser pulse, for which sound in helium can be excited at the unilluminated side of a sample of thickness d.

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