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Propagation of high-frequency acoustic phonons in germanium and their interaction with electron-hole drops

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The propagation on nonequilibrium transverse acoustic phonons ($T \gtrsim 5$–10 K, $\nu_{max} \gtrsim 4\times10^{11}$ Hz) in Ge is investigated at 1.8 K by a technique in which heat pulses are applied and detected with a superconducting bolometer. If the sample pumping is pulsed, the signals registered by the bolometer reflect the picture of heat propagation in Ge (at $\mathbf{q}\|[100]$) at velocities smaller than those of transverse sound, and characterize the change from ballistic phonon propagation to diffuse propagation with decay followed by ballistic propagation. The bolometer-signal fall-off time ($\tau \sim 10$–18 μsec) is evidence that the phonons are produced by a relaxation mechanism connected with the cooling of the photoexcited carriers, since the phonons emitted following Auger recombination of the carriers in the EHD would produce signals with times of the order of the recombination time of the carriers in the EHD, i.e., $\tau \sim 40$ μsec.

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The problem of heat relaxation and heat dissipation is of essential interest both for the solution of the basic problem of the behavior of a system of nonequilibrium phonons in a crystal, and for practical applications, say heat dissipation in microelectronics. A natural need arises for tracing the behavior of nonequilibrium phonons, studying their propagation in a crystal and their passage through a crystal-liquid helium interface, their interaction with impurities or nonequilibrium carriers, etc.[1]

Few direct experimental observation s were made of the interaction of nonequilibrium phonons with nonequilibrium carriers.[2-4] The heat-pulse technique used in some studies makes it possible to observe separately in time the arrival of longitudinal and transverse mode in a ballistic regime (when the phonon mean free path Λ is less than the sample length l), and thus study directly the action of the medium on Λ and hence on the phonon propagation.

Absorption of longitudinal sound of high frequency $\nu \gtrsim 10^{11}$ Hz by electron-hole drops (EHD) in germanium, and hence the dragging of EHD by a flux of such phonons, was observed by Hensel and Dynes.[2] They have also shown that transverse phonons interact weakly with EHD, the TA-phonon absorption being not more than 7%. On the other hand, judging from the distribution obtained by Greenstein and Wolfe[5] for the EHD cloud in a crystal, it is seen that a narrow flare of this cloud stretches out in the [100] direction. This anisotropy of the distribution can be due either to anisotropy of the longitudinal phonons along the [100] axis, which is denied in Ref. 3, or anyhow to the dragging of the drops by the TA phonons, although theoretically the interaction of the TA phonons with the carriers can be due only to nonsphericity of the conduction band or to a complex structure of the valence band of Ge.

One of the tasks undertaken in the present study was to produce a larger volume of the liquid phase than obtained by Hensel and Dynes[2] who used for the excitation a cw laser of low power, 6 mW (volume excitation), since we wanted to register reliably the damping of the TA phonons. The phonon propagation direction was specially chosen to be the [100] axis of the crystal, since it is precisely in this direction that the heat propagates in practice only in the form of a transverse mode.[6]

Hensel and Dynes[2] cited surprisingly long EHD motion times under the influence of the LA phonons (up to 6 μsec). It might seem that this time can in fact not

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exceed the duration of the thermal pulse (0.5 μsec in Ref. 2), since the drops are stopped within 10^{-9} sec after the end of the pulse.[7] A hypothesis was advanced that the high-frequency phonons are localized in a "hot spot" and can propagate only by diffusion. This spot in fact might be the cause of the rather prolonged acceleration of the drop. Under real carrier-excitation conditions on a nonmetallized semiconductor surface (as, e.g., in the study of Greenstein and Wolfe[5]) the relaxation phonons have energies higher than when the heat pulses are generated by a metal film,[8] i.e., if equal energies are absorbed by a metal film and by a semiconductor, the phonons of the hot spot of the semiconductor with nonmetallized surface will have the higher energies, and the deviations of the phonon distribution functions from equilibrium will take different forms.

To our knowledge, there is only one published mention of the form of the phonon spectrum following relaxation of photoexcited carriers in indium antimonide.[9] Kazakovtsev and Levinson[10] considered ballistic and diffuse propagation of phonon pulses, accompanied by degradation of the frequency spectrum in spontaneous phonon decay. It would be of interest to study experimentally the transition from ballistic to diffuse propagation of the phonons, by creating nonequilibrium phonons via relaxation of photoexcited carriers, bearing in mind also the possible observation of the phonons emitted when Auger carriers are cooled in the act of Auger recombination of electrons and holes into EHD.

EXPERIMENTAL PROCEDURE

Germanium samples with $N_D - N_A \sim 5.10^{12}$ cm^{-3} were cut in the form of cylinders with polished end faces and ground lateral faces for optical excitation. One of the end faces of the sample was coated with a 3000 Å gold film, and on the opposite end face was sputtered a dielectric SiO layer ~3000 Å thick and a hairpin-turn structure of granulated aluminum[11] of thickness 300–400 Å (see the inset in Fig. 1). The bolometer, made of granulated aluminum, had a superconducting transition temperature in the interval 1.8–2.1 K (Fig. 1), unlike pure aluminum bolometers, which have $T_{cr} = 1.4$ K.[12] The thermal pulses were excited by heating the gold film spittered on the germanium with a YAG–Nd^{3+} laser ($\lambda = 1.06$ μm) at a repetition frequency 12.5 Hz and a pulse power up to 3×10^5 W (channal A). The laser pulse duration was ~10 nsec. The bolometer measured 2×2 mm, and the unfocused laser-beam spot area was ~2 mm^2, so that the solid angle subtended by the bolometer on the source was ~16°, corresponding approximately to the TA-phonon focusing for the [100] direction.[3]

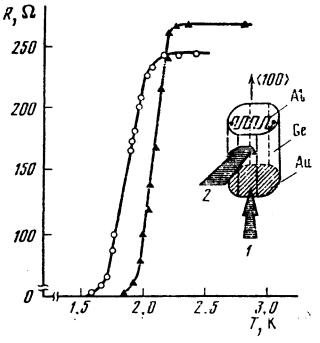

FIG. 1. Plots of the superconducting transition of a film of granulated Al: ▲—$i_{bol} = 1$ μA, $T_{cr} = 2.10$ K; ○—$i_{bol} = 70$ μA, $T_{cr} = 1.85$ K. Inset—geometry of the heated metallic film and of the bolometer on the Ge sample relative to the laser beams ($\lambda = 1.06$ μm): 1—channel A, 2—channel B.

The bolometer working current and the temperature T_0 of the helium bath were chosen with an eye towards obtaining a linear response in the detection of the thermal pulses in a sufficiently broad excitation energy range (1–10^3 erg). In particular, the results shown in Figs. 2–4 were obtained at $i_{bol} = 70$ μA and $T_0 = 1.8$ K, so that $R_{bol} = 100$–120 Ω. The signal from the bolometer was fed to a broadband preamplifier, and next through a strobing integrator to an x-y recorder. Using a beam splitter, the laser beam could also be focused on the lateral face of the sample (channel B) to generate the electron-hole drops.

EXPERIMENTAL RESULTS AND DISCUSSION

1. It is known that the emissivity of a gold film is larger for TA phonons than for LA phonons in a ratio $2(v_l/v_t)^2 = 8$,[13] where v_l and v_t are the velocities of the LA and TA phonons in gold. Since we have investigated phonon propagation along the [100] direction, for which the ratio of the focusing of TA and LA phonons in germanium is 40 : 1,[14] the amplitude of the LA phonons exceeded the noise level by only two or three times.

Figure 2 shows arrival plots of TA phonons generated by the gold film against the laser emission energy (channel A), i.e., as a function of the heater temperature T^*. Since the film heating time (~10 nsec) is much less than the time τ_{therm} of its thermal relaxation (see, e.g., Ref. 15, where τ_{therm} is ~50 nsec for various metallic films of this thickness), it follows that

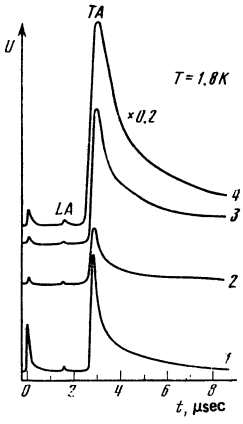

FIG. 2. Heat pulses in germanium, detected with an aluminum bolometer and produced by laser heating of a gold film. The parameter is the energy absorbed by the gold film: 1—4 erg; 2—1.1 erg, 3—4 erg, 4—13 erg. Curve 1 was obtained at the preamplifier $R_{in} = 50$ Ω and $\tau_s = 50$ nsec; the remaining curves were obtained at $R_{in} = M\Omega$ and $\tau_s = 200$ nsec.

T^* of the heater can be estimated in the adiabatic approximation from the relation

$$N\Delta t = m\int_{T_0}^{T^*} c(T)\,dt,$$

where N is the absorbed power, Δt is the pulse duration, c is the heat capacity of the gold, and m is the heated film mass. Assuming that the absorption coefficient of gold at $\lambda = 1.06$ μm is ~1%,[16] it is easy to estimate the heater temperature: $T^* \approx 5.5$ K at an absorbed energy 1.1 erg 11 (Fig. 2, curve 2), and $T^* \approx 10$ K at 13 erg (Fig. 2, curve 4).

The leading front of the detected pulse is determined by the operating speed of the receiver (bolometer) and by the time constant of the recording, which is ~50 nsed under our condition. The heat pulse (bolometer signal) has a distinct shape (curve 1): an appreciable fraction of the heat is contained in a narrow peak that lags the excitation pulse by a time $t = l/v_t$, where l is the sample length (10 mm) and v_t is the transverse-sound velocity for [100]. The duration ~350 nsec of this peak is governed mainly by the dispersion of the TA phonons in the frequency band 10^{10}–10^{12} Hz (at $T^* \approx 5.5$ K); part of the heat, however, arrives in the form of a long time that is delayed in time—these are the phonons whose mean free path is less than the length of the sample and which reach the detector either by diffusion after numerous scattering acts, or acquiring after their decay a frequency different from that of the injected phonons.[10] It is seen from Fig. 2 that the fraction of these phonons increases rapidly with increasing T^*.

It is more convenient to determine the absorption of the transverse phonons by the electron-hole drops under conditions close to ballistic, for only then do the signals arriving at the bolometer take the form of sharp peaks. The EHD were generated in the sample by the same laser (channel B) as the thermal pulses, at a distance 2 mm from the end face on which the bolometer was deposited. The laser beam was focused by two cylindrical lenses into a 0.5×2 mm strip, and the energy input prior to the attenuation by the filters was 10% of the maximum energy in channel A, i.e., ~1.5×10^3 erg.

Figure 3 shows the bolometer phonon-detection signals when EHD are generated in the sample. Approximately 0.5 μsec after the laser pulse we see signal I from the phonons produced by relaxation of the photoexcited carriers from channel B, and 2.7 μsec later the bolometer registers the phonons (signal II) from the heated gold film (curves 2–5). It shows also the signal produced by the phonons generated only in channel A, i.e., in the absence of EHD in the sample (curve 1). If we subtract from the amplitude of signal II the residual signal from channel B, then it is clear that the amplitude of signal II decreases in comparison with curve 1. It can be assumed that the procedure of subtracting the residual signal from channel B is correct if the detector is linear in this response region.

When the power in channel B increases, i.e., when the volume of the liquid phase is increased, the

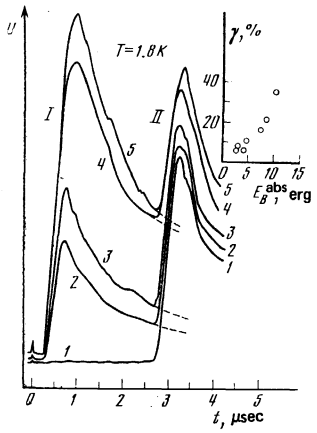

FIG. 3. Heat pulses in Ge, detected with an aluminum bolometer when EHD are excited in the sample: I—heat pulses propagating from a sample where the EHD are generated (channel B) to the bolometer within ~ 0.5 μsec [curves 2 (E_B^{abs} = 2.7 erg), 3 (E_B^{abs} = 4.1 erg), 4 (E_B^{abs} = 7.7 erg), 5 (E_B^{abs} = 10 erg)]; II—heat pulses of transverse phonons from a heated gold film (channel A). For comparison, the bolometer signal in the absence of EHD is shown by curve 1. Inset—dependence of the damping of the transverse phonons from channel A on the excitation energy in channel B, i.e., on the volume of the liquid phase.

amplitude of the transverse sound decreases even more significantly. The inset of Fig. 3 shows the dependence of the attenuation of the transverse sound amplitude as a function of the pump energy in channel B, i.e., of the volume of the liquid phase. At the maximum energy in channel B the damping of the transverse phonons with $\nu_{max} \sim 5 \times 10^{11}$ Hz is ~30%.

2. To compare the picture of the phonon propagation in the two different cases—when the phonons are generated by a heated metallic gold film and when the carriers are excited on a germanium surface, the gold film was removed from part of the sample end face and the laser beam could be focused either on the gold film or on the Ge surface. Figure 4 shows the phonon detection signals obtained in this experiment. We note that the distance between the excitation points on the sample end face was not more than 2 mm in these two cases. When the maximum temperature to which the germanium is heated is estimated, it must be borne in mind that the reflection coefficient of Ge at $\lambda = 1.06$ μm is

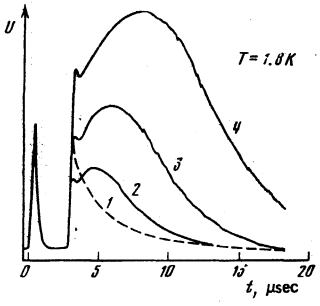

FIG. 4. Heat pulses detected in Ge by a bolometer, following direct irradiation of the Ge surface with a laser at various energies absorbed by the samples: 2—220 erg, 3—420 erg, 4—1100 erg. The dashed curve 1 shows for comparison the bolometer signal when the Au film is heated to $T^* = 7$ K (absorbed energy ~ 6.5 erg).

~30%, i.e., at equal incident energy the germanium absorbs ~70 times more energy than the heated gold film. The bolometer signals differ radically in form in the case of heating of the gold film from the case of direct heating of the germanium surface. (Compare curves 1 and 3 of Fig. 4 at an absorbed-energy ratio ~35.)

It is of interest to note that when the germanium surface is excited the heat propagates only in the form of transverse phonons, and a very insignificant part of them propagates ballistically. The bolometer signals have in this case a broad maximum at delay times up to 8 μsec relative to the laser pulse. The signal fall-off time constant varies between 7 and 18 μsec. The variation of the fall-off times with changing excitation level and the absolute values of these times indicate incontrovertibly that the phonons emitted when the carriers are thermalized via Auger recombination in the drops do not make a noticeable contribution to the signal registered by the bolometer. In fact, when phonons emitted by hot Auger carriers are registered the signal fall-off time constant should be 35–40 μsec, i.e., equal to the lifetime of the drop.

Thus, at an absorbed energy 220 erg the photoexcited carriers relax with emission of predominantly transverse phonons having an energy such that their propagation is not ballistic.

A rough estimate of the heat-propagation velocity (from the maximum of curve 4 of Fig. 4) yields a value 1.2×10^5 cm/sec; knowing the transverse sound velocity $v_t=3.6\cdot10^5$ cm/sec, we obtain the ratio $\Lambda/l\sim1/3$ of the mean free path to the sample length[17] and a phonon mean free path ~3 mm. We see therefore that the assumption that the phonon propagation is diffuse is absurd. Using the known relation for the relaxation time [see. e.g., Eq. (4.49) of Ref. 18], we can estimate the mean free path of the phonons emitted upon relaxation of the photoexcited carriers:

$$\tau^{-1}=2.8\cdot10^{-23}T^3\omega^2+2.57\cdot10^{-44}\omega^4+v/l_{r_b}$$

where T is the lattice temperature, ω is the phonon frequency, v is the speed of sound in the crystal, and l_b is determined in our case by the transverse dimensions of the sample. The first term of the equation takes into account the phonon decay, the second the scattering of the phonon by the isotopic impurity, and the third is small enough to be neglected.

The unknown parameter in this case is the phonon frequency, inasmuch as the phonons emitted when the carriers are cooled do not have a Planck spectrum. The most probable frequency ω can be assumed to be the one determined from the Debye temperature of the transverse acoustic phonons, ~100 K. In addition, in our case when the phonon concentration deviates considerably from equilibrium we assume that $T=100$ K. Putting $l_b\sim0.5$ cm and $T\sim100$ K we obtain $\tau^{-1}\sim10^9$ sec^{-1}. Hence $\Lambda=v_t\tau\sim5\cdot10^{-5}$ cm.

On the basis of the results we can therefore present the following schematic picture of the heat propagation in the sample. A short laser pulse of 1.06 μm wavelength produces nonequilibrium hot electron-hole pairs near the sample surface. The hot carriers, as they cool, generate optical phonons that decay in a time 10^{-11}–10^{-12} μsec into acoustic phonons. It can be assumed that as a result of this process the initial temperature of the bulk of the acoustic phonons is ~100 K. Phonons with this temperature decay, because of their anharmonicity, into phonons of lower frequency. According to estimates, in the case of phonons with temperature ~100 K it is precisely the short lifetimes which determine the mean free path ($\sim5\times10^{-5}$ cm). However, as the phonon frequency (or temperature) their lifetime increases, and when the phonon temperature is of the order of several dozen degrees their decay probability and the probability of their scattering by an isotopic impurity become equal and lead to a mean free path.

The small mean free paths limit the phonon diffusion from the excitation region into the interior of the sample and are the cause of the formation of the so-called hot spot[2] near the surface. The multistep decay process leads to a decrease of the frequency and to an increase of the phonon mean free path, as a result of which the hot spot cools and spreads. The only phonons that can leave the localization region and reach the bolometer are those with temperature ~8 K (a mean free path ~1 cm).

In our case of nonstationary phonon generation, when the form of the phonon distribution function is not known, we can only state that in the course of the carrier thermalization and phonon decay a small number of transverse phonons is produced immediately with frequencies ω^* low enough such that the mean free path is commensurate with the sample length l. This manifests itself in the fact that in Fig. 4 the bolometer signal begins to increase with a time delay equal to the time of travel of the phonons through the sample at the speed of transverse sound in germanium in the [100] direction. In the course of the phonon decay, the fraction of the phonons with frequencies ω^* increases for some time, goes through a maximum, and then begins to decrease. We assume thus that the observed signals (Fig. 4) are determined by the arrival of ballistic phonons at the bolometer, and the shape of the curves reflects mainly the phonon-decay kinetics, or in other words the cooling of the hot spot.

An analysis of the shape of the curves in Fig. 4 allows us to assume that the decay of a phonon into two having approximately equal frequency has a lower probability than the case when one of the produced phonons has the low frequency ω^*, for only in this case can we understand the asymmetry of the shape and the prolonged attenuation of the signal.

CONCLUSION

1. Damping of transverse phonons with $\nu\sim4.10^{11}$ Hz was observed in Ge at helium temperatures. This damping is due to the interaction of TA phonons with electron-hole drops. When the phonons propagate along the [100] axis of the crystal, at the employed nonequilibrium-carrier pump levels (absorbed energy

~10 erg) the damping was 30%.

2. It is shown that when the carriers are generated on a clean Ge surface and the photoexcited carriers relax, the propagating phonons are for the most part transverse, and their propagation in a sample 1 cm long is quasiballistic and attenuated.

3. The large cross section for scattering by an isotropic impurity and the high decay probability of the short-wave phonons cause their mean free paths to be small, so that a hot spot is produced and is localized near the excited surface of the crystal. The presence of a slowly cooling hot spot that generates ballistic phonons with relatively large wavelength ($\sim 2k_F$) seems to explain the prolonged motion of the EHD in Ge, with sufficiently low velocities $\sim 10^4$ cm/sec, which were observed in a number of experiments.[19,20]

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