Propagation of Large-Wave-Vector Acoustic Phonons in Semiconductors

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The spatial and temporal characteristics of phonon pulses generated in the interband energy relaxation process of photoexcited e-h pairs in GaAs and InP are reported. With use of a superconducting Pb tunnel junction as a phonon threshold detector, it is shown that the energy transport occurs via near-zone-edge transverse acoustic phonons and that over lengths of the order of several millimeters the pulse propagation shows all features of ballistic transport in a dispersive medium.

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Recently there has been considerable activity in the area of ballistic, high-frequency phonon propagation studies in a variety of dielectric solids.[1] Many years ago, Orbach[2] proposed that anharmonic decay can be used to produce long-lived, large-wave-vector transverse phonons by optical excitation. Such generation was first observed[3] in diamond whilst piezoelectric surface excitation has been studied in quartz.[4] In most materials the propagation[5] of large-wave-vector phonons is generally believed[1,5] to be severely affected by isotope and impurity scattering[6] and phonon degradation at the generator-sample interface.[7] In this Letter, we report first experiments on the propagation of large-wave-vector ($k \sim 0.6k_{max}$) transverse-acoustic (TA) phonons generated during the process of energy relaxation of photoexcited e-h pairs in bulk, high-purity samples of zinc-blende-type semiconductors such as GaAs and InP. These semiconductors are characterized by extremely "soft" TA branches[8] particularly in the $\langle 111 \rangle$ direction and relatively weak isotope scattering. The results show that the energy transport is dominated by short-wavelength (~ 10-Å) TA phonons which propagate macroscopic distances ~ 3 mm ballistically.

The photoexcitation experiments were done (1) with a cw krypton-ion laser (0.2 W at 799 nm) and (2) with a cw tunable oxazine 750 dye laser (~ 20 mW at 760 to 820 nm.) Light pulses of 20–100-nsec duration and few kilohertz repetition rate were produced with an electro-optic modulator. The peak power absorbed in the sample was typically a few milliwatts and could be attenuated with neutral density filters. The beam was focused to a spot size of 60 μm on the sample, thus giving relatively small peak power densities ($\ll 2$ W/mm^2). For the phonon detection we used Pb-oxide-Pb tunnel junctions with 0.25×0.25 mm^2 area and also small (~ 10^{-2} mm^2) superconducting Al bolometers. Our technique allowed *in situ* variation in distance and angle of the photopumped region relative to the fixed detectors simply by scanning the laser spot laterally on the opposite (or same) surface of the plane-parallel samples ("trans" and "cis" configurations; see inset of Fig. 1). In the "trans" geometry, the propagation direction changes with r. In addition an "edge-on" configuration was used with the detector close to a 90° sample edge and excitation spot scanning along the adjacent surface. With a [110]-cut crystal this configuration allowed the choice between the principal [100], [110], and [111] propagation directions. The experiments were done at $T = 1.2$ K (in He liquid) on high-purity samples of GaAs and InP which had previously[9] shown high-quality low-frequency ballistic phonon pulses with use of conventional heat pulse techniques. They were polished by a combined mechanical and chemical etch.

Figure 1 shows typical phonon signals detected

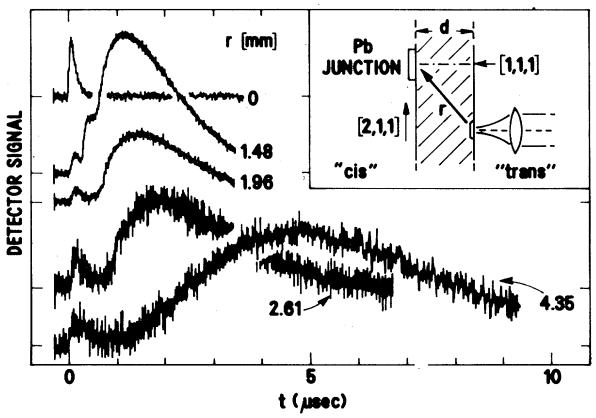

FIG. 1. Measured phonon signals for four propagation distances r. Thickness, $d = 1.48$ mm. The inset shows two of the geometrical configurations used (see text). On top: detector response under direct ($r = 0$; attenuated) laser excitation.

with a superconducting gap $2\Delta = 2.8$ meV (0.7×10^{12} Hz) threshold Pb tunnel detector in the "trans" configuration. The well-defined phonon pulse is accompanied by direct optical luminescence pickup (~ 300-nsec decay time) at $t = 0$. For comparison the direct detector response under attenuated light pulse excitation is also shown ($r = 0$). The most important observations are (1) pulse peak delay and pulse width scaled linearly with r over almost two decades, (2) signals without significant diffusive scattering were observed for distances r up to 3 mm, and (3) the pulse shapes were independent of the excitation power within the range 0.02 to 2 W/mm^2. For $\vec{r} \| [111]$ a small LA-phonon contribution in the leading edge of the signal was found as one expects from LA-phonon focusing properties.[9]

Measurements with superconducting Al bolometers gave qualitatively similar pulse shapes. With these detectors we also investigated low-frequency (~ 3×10^{11} Hz), ballistic phonon pulses propagating through the same sample and generated via direct optical excitation of evaporated Constantan metal heater films (700 Å thick). Sharp LA- and TA-phonon peaks characteristic for ballistic, nondispersive propagation with much narrower widths were found in this case.

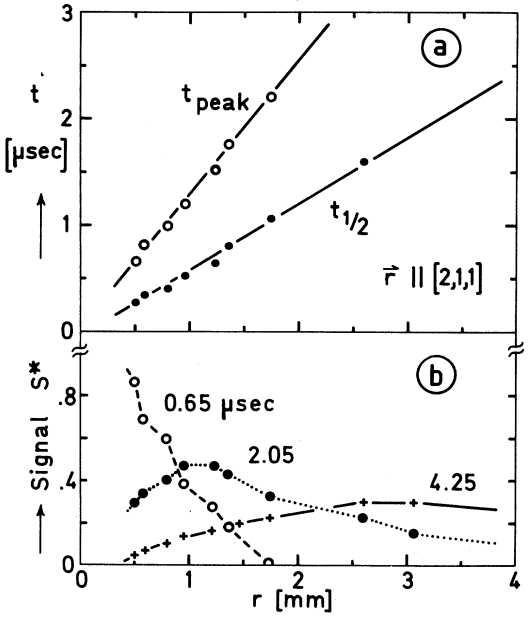

FIG. 2. (a) Measured delay time for the signal peak (t_{peak}) and the leading-edge half-height ($t_{1/2}$) as a function of r in "cis" configuration. (b) Detector signal (corrected for Lambert's law) as a function of r at three different times after laser pulse excitation.

By moving the light spot from the metal film on to the semiconductor surface we could measure precisely the relative delays of the pulse-leading-edge intersection times, and we found in the latter case a slightly slower (~ 15–20%) maximum group velocity than the known low-frequency TA-phonon velocity, which we saw in the down-conversion metal film experiment. The relative TA to LA intensity in the direct semiconductor excitation experiment was typically 1 to 2 orders of magnitude greater than in the metal film experiment.

The length dependence of the observed pulse phonon delay is shown in Fig. 2(a). Here the pulse-peak arrival time (t_{peak}) and leading-edge half-height time ($t_{1/2}$) are plotted for $\vec{r} \| [211]$ in the "cis" configuration. The solid lines in Fig. 2(a) indicate group velocities of 0.78×10^5 and 1.55×10^5 cm/sec for pulse-peak (V_{peak}) and half-point ($V_{1/2}$) propagation, which are considerably smaller than the low-frequency TA ballistic velocity of 2.8×10^5 cm/sec.

Figure 2(b) shows the observed phonon signals S (corrected for Lambert's law) as a function of *distance* r for three different elapsed times after pulsed excitation at $t = 0$. This plot shows directly that the phonon energy density has a peak which moves with increasing time away from $r = 0$ in a quasiballistic fashion. This is clearly different from diffusive phonon transport, where the excitation region would always have the maximum

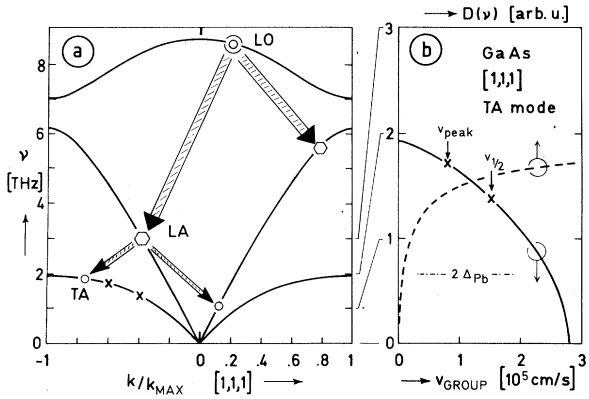

FIG. 3. (a) Decay scheme of a near zone-center optical phonon indicating zone-edge TA-phonon final states compatible with energy- and momentum-conserving three-phonon processes. (b) Acoustical phonon group velocitites (solid line) and density of states (dashed line) from neutron scattering data according to Ref. 8. The crosses denote measured peak and half-height velocities.

energy density after pulsed excitation.

We would like to emphasize that the linear length dependence was also observed in the "trans" configuration for three discrete propagation distances (0.28, 1.48, and 2.9 mm) and $\vec{r} \parallel [111]$. It was also observed for distances up to 3 mm in the "edge on" configuration and $\vec{r} \parallel [111]$, [110], [100], and [211]. The peak velocities of the observed phonons were found to change with orientation in a manner consistent with acoustic dispersion. These results preclude explanation of our data in terms of strong volume backscattering[10] of focused low-frequency phonons.

We interpret our data in detail as follows. The photoexcitation creates e-h pairs in a distance of less than 1 μm. These pairs can diffuse a typical distance ~10 μm before recombining. The bulk of the energy goes into nonradiative capture via multiphonon emission[11] involving primarily LO phonons. The LO phonons quickly decay in turn to LA phonons which in turn decay into the lowest TA branch via the Orbach mechanism[2] as shown in Fig. 3(a). Figure 3(b) shows a plot of the group velocity of this branch and its density of states $D(\nu)$ as a function of frequency. The arrows show the experimentally found velocities $V_{1/2}$ and V_{peak}. Thus the frequencies of the dominant phonons in our pulse lie in the region of 1.5×10^{12} Hz.

In Fig. 4 we show normalized plots of the time-integrated signal strengths (corrected for Lambert's law) for "cis" (starting at $r = 0.2$ mm) and "trans" (starting at $r = 1.48$ mm, thickness of crystal) configurations. The absolute magnitude for "cis" signals was much smaller (~3%) than the corresponding "trans" signal for the same distance r. The data suggest a mean free path of 2.5 ± 0.5 mm, which corresponds to a frequency down-conversion time of >2 μsec.

One important source of loss we would like to discuss is the scattering due to naturally occurring isotopes. Isotope scattering is elastic (preserves frequency) but causes momentum randomization and hence mode conversion. Mode-converted phonons could, of course, anharmonically decay to lower frequencies. The isotope scattering[6] rate (τ_{iso}^{-1}) is proportional to $\nu^2 D(\nu)$. $D(\nu)$ has to be calculated with care for the dispersive regions of the phonon branch. For simplicity we have made order of magnitude estimates of the scattering time assuming an isotropic model but taking into account the large difference in $D(\nu)$ for the two branches. For a frequency of 10^{12} Hz the mode-conversion time from TA to LA modes in GaAs is calculated to be ~2 μsec while the TA to TA scattering time ~0.22 μsec. Our data suggest that a more careful theoretical treatment of isotope scattering is necessary,[12] when λ phonon is comparable to the isotope site correlation length. Also the highly anisotropic nature of zone-edge TA mode-dispersion curves may modify the scattering rates.

In summary, we have observed the propagation of near-zone-edge TA phonons generated during e-h recombination in bulk GaAs and InP. This simple, bulk method when combined with epitaxial multilayered filters[13] should allow time- and space-resolved phonon spectroscopy at well-defined frequencies and at a wavelength of a few angstroms in technologically important semiconductors.

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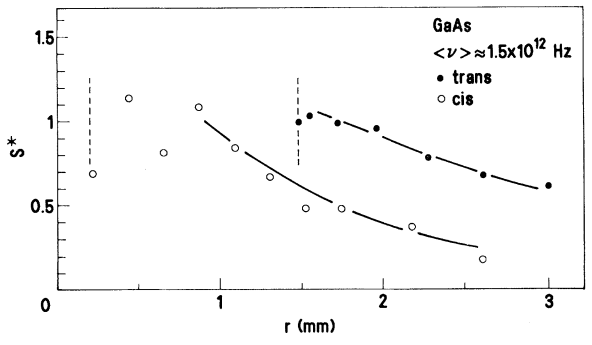

FIG. 4. Integrated TA-phonon signal $S^* \equiv r^2 \int_0^\infty S(t)\, dt$ as a function of distance r for two configurations. See text.

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