Observation of optically excited near-zone-edge phonons in GaAs by diffuse x-ray scattering

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We report on the use of diffuse x-ray scattering as a probe of nonequilibrium phonon excitations generated optically in GaAs at $T \approx 1.5$ K. The measurements show a substantial athermal population of TA phonons near the [111] zone boundary consistent with a lifetime of microseconds.

Recently there has been considerable activity in the area of high-frequency phonon transport in solids.[1] Even though a great deal has been learned about nonequilibrium phonon populations through propagation experiments using superconducting thin-film detectors, a direct, *in situ* determination of nonequilibrium phonon distributions has been difficult.[2] In this Communication, we report on the use of diffuse x-ray scattering[3] as a novel and powerful probe of the wave vector and time evolution of athermal phonon populations in solids.

We apply the technique to the case of phonons generated during nonradiative energy relaxation and recombination of optically excited e-h pairs in the polar semiconductor GaAs. Previous[4] propagation experiments had shown that energy transport in this case occurs mainly through extremely short wavelength, dispersive TA phonons of low group velocity which had an energy relaxation time of microseconds. The thermalization of these TA-phonon modes is the rate limiting step at low temperatures. Spontaneous decay of these near-zone-edge TA phonons is forbidden because of the large dispersion,[5-7] and four-phonon processes are negligible because they require thermal population of small wave-vector phonons. This leaves impurity and isotope scattering. These processes are difficult to calculate for the nonlinear regions of the phonon dispersion curve. Even though previous experiments[4] were consistent with long lifetimes, they could not directly show the frequency (wave-vector) distribution of the athermal phonon population. The present experiments, indeed, show that the phonon distribution consists of near-zone-boundary TA phonons.

The optically excited TA-phonon mode propagating along [111] with displacements parallel to $[1\bar{1}0]$ in high-purity[4] single crystals of GaAs held at temperatures $T \sim 1.5$ K was studied. The intensity of the first-order thermal diffuse scattering is given by [8]

$$I(\vec{Q}) \propto \sum_j \frac{|F(\vec{Q})|^2 (\vec{Q}\cdot\vec{\epsilon}_{qj})^2}{h\nu_{qj}} \left(\frac{2}{e^{h\nu_{qj}/kT}-1} + 1 \right) ,$$

where ν_{qj} and $\vec{\epsilon}_{qj}$ are the $\vec{q}$-dependent frequency and eigenvector of the jth phonon mode, $\vec{Q}$ is the scattering vector, and $F(\vec{Q})$ the structure factor.[8] At low temperature this scattering comes almost entirely from the acoustic modes because of their lower frequency. By mounting the crystal so as to give a $(11\bar{2})$ scattering plane, the TA mode can be studied [because of the $(\vec{Q}\cdot\vec{\epsilon})^2$ dependence of the scattering] by scanning from the $(4\bar{4}0)$ to the $(5\bar{3}1)$.

The experimental geometry is sketched in Fig. 1. A 12-kW Rigaku rotating anode x-ray generator, operating in the high brilliance mode with a copper

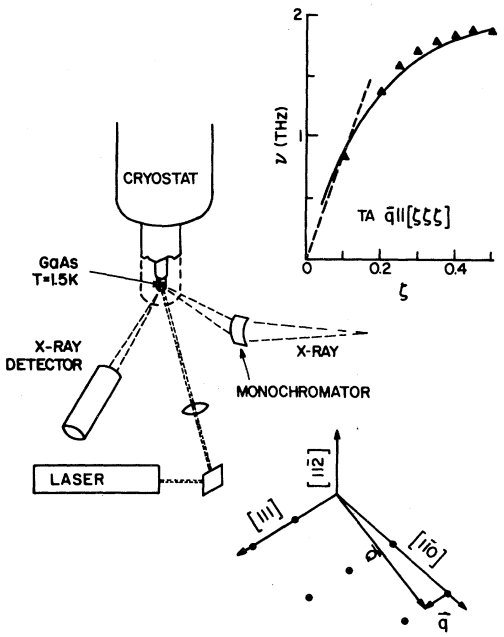

FIG. 1. Geometry of the experiment showing focused x-ray and laser beams scattering off a GaAs plate whose back surface is in contact with superfluid helium at $T \approx 1.5$ K. The scattering geometry to observe the TA mode with $\vec{q}\,\|\,[111]$ and $\vec{\epsilon}\,\|\,[1\bar{1}0]$ is shown at the lower right and the dispersion relation calculated from the x-ray measurements (solid curve) is compared with neutron (triangles) and acoustic (dashed curve) studies (Ref. 9) in the inset.

anode and a 0.2×2-mm^2 spot focus, is used in conjunction with a vertically bent pyrolytic graphite monochromator to produce a $< 1 \times 1$-mm^2 spot of Cu $K\alpha$ x rays at the sample. The laser beam is focused or defocused to match the x-ray spot on the crystal in size and position. Two types of experiments were done. The first used a 1-W, cw, multicolor argon ion laser operating in the TEM_{00} mode to produce a spot of uniform intensity at the sample. The intensity of the diffuse scattering was measured as a function of $\vec{q}$ with the laser on and off. In the second experiment, a flash lamp pumped dye laser at 6000 Å was used to deliver ~ 5-kW pulses with a full width of 1 μsec at a frequency of 30 Hz. In the latter experiment, the output of the x-ray pulse-height analyzer was time delayed and fed into a 15-channel multicounter in which each channel was 1.5-μsec wide. In this way the intensity of the diffuse scattering was time arrayed over a range from -2 to 28 μsec with respect to the 1-μsec laser pulse. The GaAs sample in the form of a 1.7-cm-diam, 0.24-cm-thick disk was attached to the inner helium bath of a pumped helium Dewar using an indium O ring seal. The back surface was in contact with superfluid helium and the front surface was in vacuum. A tin bolometer was deposited around the periphery of the front surface of the sample to monitor the sample temperature. The bolometer indicated that during both the cw and the pulsed experiments the front of the sample was below 3.8 K. The level of the superfluid helium was monitored with temperature sensors and kept well above the sample. The Dewar was equipped with a curved glass window for optical access and a 5-mil Mylar window for x-ray access.

In order to separate the thermal diffuse scattering from general background and other types of scattering such as Compton scattering, preliminary x-ray measurements were made as a function of wave vector q at 10 and 298 K and as a function of temperature at $q = 0.5$ (zone boundary). Normalizing the data at $q = 0.5$ to the frequency observed in neutron scattering studies[9] and assuming that $\vec{\epsilon}$ for the TA mode and the background were independent of q, the disperions relation was calculated and found to agree well with neutron scattering and acoustic measurements as shown in the inset on Fig. 1. In addition, the variation of the intensity at the zone boundary agreed well with the calculated population factor.

The results of the cw experiments are plotted in Fig. 2 as the additional population resulting from optical excitation versus q. In a cw experiment one expects to observe a steady-state rise in effective temperature which would result in a thermal distribution. The data at small q were fit to give an effective local thermal temperature of 8.5 K. Using the known dispersion relation, the thermal tail was calculated as a function of q, and the results are shown as the solid curve in Fig. 2. As can be seen, the thermal contribution is negligible above $q \approx 0.2$, whereas the observed population at large q is orders of magnitude larger. The error bar represents the variation expected from counting statistics for the ratio of the total counts with laser on to laser off. The experimental results are reproducible (see Fig. 2) and the data were collected in a variety of ways in order to rule out systematic errors. There appears to be a peak in the distribution of large wave-vector phonons near $q = 0.4$ which would be in qualitative agreement with the argument that there is a balance between the group velocity, which approaches zero at the zone boundary, and the propagation (lifetime) of these phonons. Previous theoretical[7] conjectures have suggested that there might be a "window" of lifetime because of this balance.

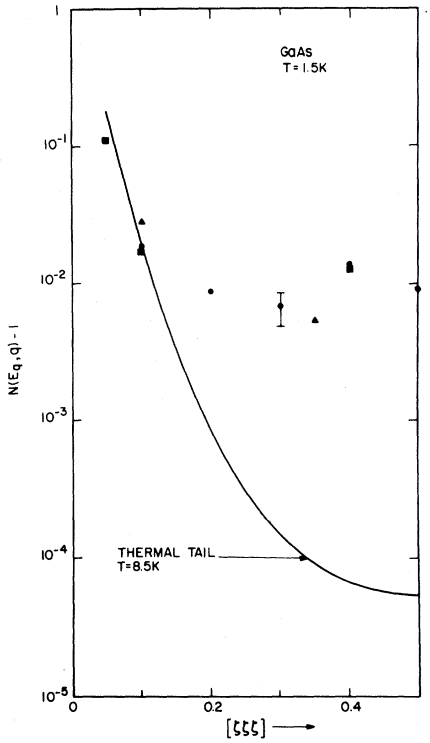

FIG. 2. Wave-vector dependence of the athermal phonon population resulting from cw optical excitation. The solid circles, triangles, and squares are for different experiment runs, and the error bar represents the counting statistics for the ratio of laser on to laser off. The solid curve is calculated for a thermal population using the dispersion relation in Fig. 1.

In order to set an upper limit on the lifetime of the optically excited phonons at $q = 0.4$, a series of experiments using a pulsed dye laser were done. In preliminary experiments it became apparent that there was a limitation on the peak pulse power of the dye laser which could be used. In experiments using only two counters, one gated during the laser pulse and a second counter gated 130 μsec later, the curve of the number of counts in the first counter versus

the second initially had a slope of 1.4 on a clean GaAs surface. After 20–30 min the slope approached that observed with the laser blocked, namely, 1. Visual examination revealed a damage spot on the surface of the crystal. Therefore the laser power was reduced to 1 kW per pulse and the laser focused less tightly (approximately equal to a few millimeters) on the sample. In addition, the Dewar was translated every 15 min so that the laser and x-ray beam would be illuminating a virgin part of the crystal. The results shown in Fig. 3 were obtained under these conditions. At the top of the figure the gating sequence for the laser and the multicounter is sketched and at the bottom of the figure the number of counts versus channel number (or time) are shown. The total counts in the counter that was gated during the laser pulse is four standard deviations higher than the average for the other channels. Experiments with the laser blocked showed no statistically significant deviation in any of the channels.

The time-dependent results set an upper limit on the phonon lifetime of $1-2$ μsec for $q \sim 0.4$. Earlier propagation experiments[4] had revealed an average lifetime ~ 2 to 3 μsec at low excitation levels and considerably shorter (~ 1 μsec) values at high pulse powers (> 10 W/mm^2). Our results are consistent with earlier measurements. In our present experiments the average value of the phonon occupation number at $q \sim 0.4$ was found to change by a factor of 30 on changing the *relative* power densities between the pulse and cw experiments by a factor of 100. This would be consistent with a reduction of lifetime by a factor of 3 with increasing power. It is to be noted that for $q \sim 0.4$, the group velocity of these phonons is $\sim 2 \times 10^5$ mm/sec. Since the x-ray spot size is ~ 1 mm in diameter, the escape time is much longer than the measured decay time and therefore it should play no role in interpreting the experimental results. Measurements with narrower channel widths, to determine details of the decay time, should be possible with a more powerful (synchrotron) x-ray source.

In summary, we have shown that diffuse x-ray scattering can be used for studying the wave-vector dependence of athermal phonon distributions in solids. For the case of optically excited GaAs we have shown that this wave vector is centered around large wave vectors near the zone boundary for the [111] direction. The results of the pulsed experiments are consistent with lifetimes of the order of microseconds. Having shown the feasibility of studying nonequilibrium phonon populations using combined time resolved x-ray and optical techniques, we hope to extend these studies by using the extreme brilliance and pulsed nature of synchrotron radiation. Such experiments can serve as powerful detailed probes of the temporal, spatial, and wave-vector dependence of nonequilibrium phonons in solids. These experiments can also shed light on other nonequilibrium physical phenomena, such as laser annealing of semiconductors.[10]

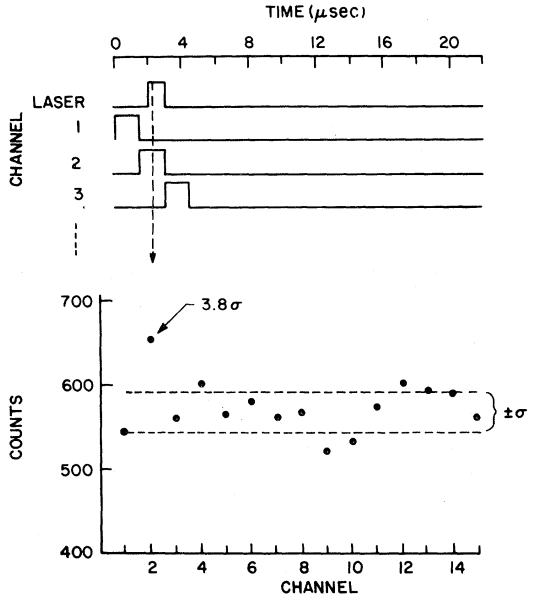

FIG. 3. Results of pulsed experiments at $q = 0.4$. A counting rate 3.8σ above ambient is observed in the channel which coincides with the laser pulse. When corrected for background this corresponds to $N(\epsilon,q) = 1.36$. The pulsed and cw experiments are consistent with a phonon lifetime of microseconds.

ACKNOWLEDGMENTS

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[1]See, for example, J. Phys. (Paris) 42, C6 (1981).
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