Anharmonic Decay of Zone-Boundary Phonons Observed by a New Method of Phonon Detection

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A new method for detection of zone-boundary phonons is presented. Phonons generated by nonradiative transitions in thallium halide crystals are detected by phonon difference absorption with use of time-resolved far-infrared laser spectroscopy. Our experiments, performed at crystal temperatures between 3 and 20 K, give experimental evidence of strongly-temperature-dependent anharmonic lifetimes of transverse-acoustic zone-boundary phonons.

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In recent years several methods have been applied to study phonons in crystals at low temperatures.[1] In this Letter we report a new method that allows for the first time measurement of the temperature dependence of lifetimes of acoustic zone-boundary phonons. The experiments were performed on thallium halide crystals for which we found a strong increase of the decay rate of transverse-acoustic zone-boundary phonons in the temperature range from 3 to 20 K.

The principle of the new method is shown in Fig. 1. Phonon populations are probed by phonon difference absorption. In a phonon difference process a far-infrared photon (FIR in Fig. 1) and a transverse-acoustic (TA) phonon are annihilated and a longitudinal-acoustic (LA) phonon is created. The two phonons have the same wave vectors since the far-infrared photons have negligibly small wave vectors. The far-infrared absorption is a direct measure of the population difference of TA- and LA-phonon states. Nonequilibrium phonon populations are probed by measuring time-dependent absorption of far-infrared laser radiation. For a first experiment we have chosen thallium halide crystals. These crystals show strong phonon difference absorption;[2] therefore a high sensitivity for phonon detection was expected. We used the radiation of a HCN laser (at frequency 0.89 THz) for the probing of zone-boundary TA phonons at 0.95 THz (Fig. 1). By nonradiative transitions of optically excited impurity states mainly optical phonons (LO and TO in Fig. 1) are generated. By fast decay of these phonons a TA phonon population is obtained. For optical excitation we used second-harmonic radiation of a pulsed Nd-doped yttrium-aluminum-garnet laser. The photon energy (2.2 eV) of the pump-laser radiation is smaller than the gap energy of 3.4 eV.[3] An absorption spectrum of our crystals shows a broad impurity absorption band (center at 3 eV). By absorption of pump-laser radiation in the wing of the impurity band and by subsequent nonradiative transitions high-frequency phonons are generated. The nature of impurity absorption and the mechanism of the nonradiative processes are not known.

The configuration of the optical arrangement is shown in the inset of Fig. 2. A crystal[4] (5 mm thick) is held in the vacuum chamber of a temperature-variable optical cryostat and cooled by mechanical contact. A part of the crystal (area 1×3 mm^2) is excited with light pulses (20-ns duration, repetition rate 1 Hz). The radiation of a far-infrared cw HCN laser (100 mW power) is transmitted through the optically pumped volume and focused on a fast far-infrared InSb detector. The detector signals are recorded with a transient recorder and averaged over typically 200 sweeps.

An experimental signal curve is drawn in Fig.

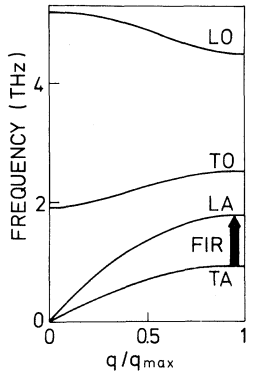

FIG. 1. Phonon dispersion curves of TlCl (schematically) and principle of phonon detection. Laser radiation at 0.89 THz is absorbed (see arrow) by difference processes of TA phonons (0.95 THz) and LA phonons (1.84 THz) near the S point $(\frac{1}{2},\frac{1}{4},\frac{1}{4})$ and equivalent points on the zone boundary (Ref. 2).

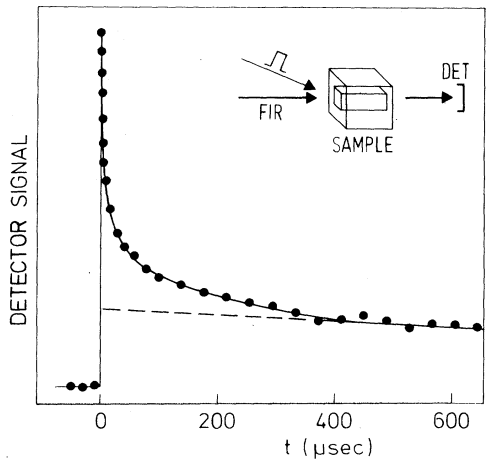

FIG. 2. Detector signal observed after optical pumping of a TlCl crystal at 3 K. The dashed curve is expected in the case of instantaneous thermalization. The inset shows the configuration of the optical beams.

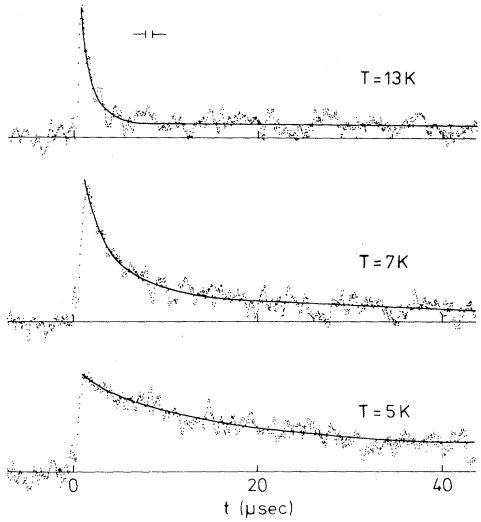

FIG. 3. Detector signals for different crystal temperatures. The time resolution of the far-infrared detector (0.7 μs) is indicated.

2. We find that by optical pumping (at time $t=0$) the far-infrared transmission is reduced (a positive detector signal in the figure corresponds to a reduction of transmission), indicating the occupation of TA-phonon states. The signal curve is strongly nonexponential: There is an initial fast decay (duration of a few microseconds) followed by a slow decay (200 μs) and a final very slow decay (not shown in the figure) with decay time $\tau_d \simeq 1.5$ ms. We found that τ_d was dependent on the size of the probe volume, indicating that the very slow decay is due to heat diffusion out of the probe volume. Our results show that a nonequilibrium phonon population exists for a time of a few hundred microseconds and that after this time a thermal equilibrium (at about 5 K) is reached in the optically pumped volume. From τ_d we estimate that the mean free path of thermal phonons at a temperature of 5 K is of the order of 10^{-4} cm. This shows that our crystal contained a large concentration of impurities which are responsible for strong impurity scattering of the high-frequency phonons.

A comparison with transmission experiments[2] at variable sample temperature shows that the reduction of sample transmission (10% at 2 mJ pump pulse energy) corresponds to a nonthermal occupation number of 10^{-3} in the probe volume. We found that the occupation number was linearly dependent on the optical pump energy, which we have varied by two orders of magnitude (up to 50 mJ). From the change of transmission at times when thermal equilibrium has been reached (Fig. 2) we estimate, using specific heat data,[5] that a few percent of the optical pump energy was absorbed in the crystal.

We have measured the temperature dependence of phonon decay (Fig. 3). We find that the phonon decay is strongly dependent on crystal temperature. At 5 K the decay is nonexponential (similarly at 3 K). The experimental curve can be described by a sum of two exponentials where the decay time of the fast decay is $\tau \simeq 5$ μs and that of the slow decay is about 50 μs (compare Fig. 2). With increasing temperature τ decreases and reaches a value of 1.5 μs at 13 K. Our experiment indicates, therefore, that the probed phonons decay at higher temperatures by interaction with thermal phonons. The slope of the signal curves at higher temperatures shows that thermal equilibrium is almost reached after the fast decay.

For an analysis of the phonon decay, we have plotted in Fig. 4 the experimental decay constant τ^{-1} for different sample temperatures. We can describe the decay constant (dashed line in Fig. 4) as a sum of a temperature-independent decay constant τ_0^{-1} and a temperature-dependent part $A\bar{n}$, where $\bar{n}(\delta, T)$ is the Bose factor for phonons at frequency δ (0.3 THz) and A (8×10^5 s^{-1}) is a constant. Our analysis suggests, therefore, that the purely temperature-dependent decay is in-

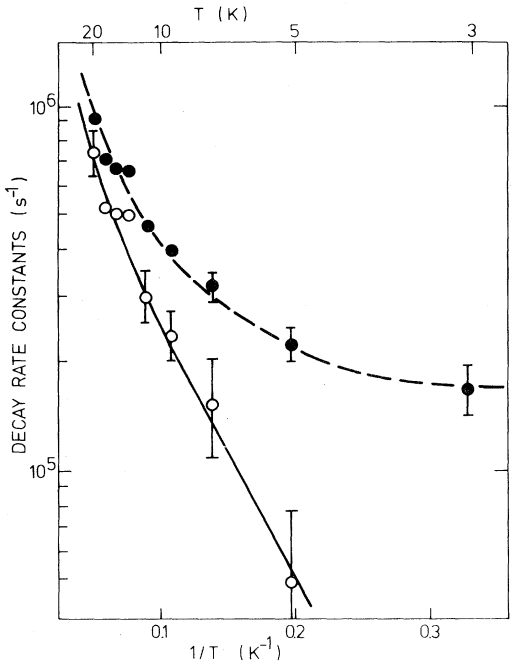

FIG. 4. Decay constants for TA zone-boundary phonon population, experimental values (points) and purely temperature-dependent portion (circles). The curves are discussed in the text.

duced by anharmonic interaction with thermal phonons of frequency δ. It is interesting that δ is the frequency of TA phonons near the $M(\frac{1}{2}, \frac{1}{2}, 0)$ point of the Brillouin zone.[6] According to the dispersion curves, anharmonic three-phonon processes in which TA phonons at the S point (Fig. 1) undergo up conversion to phonons of higher frequency are possible with respect to energy and wave-vector conservation.[2,6] A temperature-dependent decay similar to the purely temperature-dependent part has been calculated for TA phonons in an isotropic crystal.[7]

At low crystal temperature the decay time ($\tau_0 \simeq 6$ μs) of the fast decay is independent of temperature (Fig. 4). We think that τ_0 is an average decay time of phonons with frequencies of the probed TA phonons because elastic impurity scattering leads to mode conversion and therefore to a quasiequilibrium of the populations of phonons with the same frequency, but with different polarizations. We guess that the phonon decay is determined by the proportion (estimated to be 10^{-2}) of rapidly decaying LA phonons in the phonon mixture and we obtain for LA phonons (at 0.95 THz) a lifetime of the order of 10^{-8} s that is in an agreement with an estimate obtained from the theory of anharmonic decay of LA phonons.[8] The nonexponential slope of the experimental curve indicates that the decay of LA phonons by down conversion is complicated by phonon bottleneck effects.

We have also performed experiments on TlBr crystals (using the HCN laser) and found very similar results.[9] We point out that the new method of phonon detection is very sensitive and allows for detection of nonpropagating phonons. Our experiment indicates long-lived TA zone-boundary phonons in thallium halide crystals at low temperature. Long-lived TA phonons (propagating in well-defined directions) were recently detected by time-of-flight experiments in very pure crystals of crystalline quartz[10] and in semiconductors.[11]

In summary, with use of HCN laser radiation, zone-boundary phonons in thallium halides at various crystal temperatures were detected by phonon difference absorption. This intrinsic lattice absorption occurs in almost every crystal as a result of anharmonicity or nonlinear dipole moments. Therefore, with use of other far-infrared lasers, the new method should be suitable to detect acoustic phonons in other ionic and also in homopolar crystals.

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