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To cite this version:
H. Lengfellner, K. Renk. DETECTION OF ACOUSTIC ZONE-BOUNDARY PHONONS BY PHONON DIFFERENCE ABSORPTION. Journal de Physique Colloques, 1981, 42 (C6), pp.C6-259-C6-261. <10.1051/jphyscol:1981675>. <jpa-00221611>

HAL Id: jpa-00221611
https://hal.archives-ouvertes.fr/jpa-00221611
Submitted on 1 Jan 1981

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DETECTION OF ACOUSTIC ZONE-BOUNDARY PHONONS BY PHONON DIFFERENCE ABSORPTION

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Abstract.— A new method for detection of zone-boundary phonons is reported. Phonons are detected by phonon difference absorption of far-infrared laser radiation. We present experimental results obtained for TlCl and TlBr crystals. Phonons are generated by nonradiative transitions in optically excited crystals. Our experiments indicate long-lived zone-boundary phonons at low crystal temperature and a strong decrease of the phonon lifetime with increasing temperature.

1. Experiment.— Phonon difference absorption is used to detect zone-boundary phonons in TlCl and TlBr crystals. The principle of this new method of detection of high-frequency phonons has recently been published\(^1\) and is characterized in Fig.1. The population of transverse acoustic phonon states is determined by far-infrared phonon difference absorption. In a phonon difference process a transverse acoustic phonon (TA in Fig.1) is annihilated and a longitudinal acoustic phonon (LA) is generated. The two phonons have the same wave vector. The absorption coefficient is proportional to the difference of the occupation numbers of TA and LA phonon states.

Fig.1. Principle of phonon detection.

Phonons are generated by radiationless processes induced by optical excitation of the crystal. The radiationless processes lead to generation of high-frequency phonons which decay rapidly in TA phonons of the lower phonon branches. The optical absorption is due to impurity states of unknown origin. For the optical excitation second harmonic radiation of a Nd:YAG laser is used.

The phonons are detected by time-resolved far-infrared laser spectroscopy. The radiation of a HCN laser with emission at a frequency of 0.89 THz is transmitted through the optically excited
crystal volume (typically 2 mm diameter and 4 mm thickness). The transmitted laser radiation is detected with a fast InSb detector (time resolution 0.7 μs). The phonon induced change of sample transmission leads to a time dependent detector signal which is analyzed by a transient recorder and averaged with a computer.

The far-infrared laser radiation is absorbed mainly by phonons at the zone-boundary as has been shown by far-infrared absorption experiments at different crystal temperatures. The TA phonons involved in the absorption process have frequencies near 1 THz.

2. Results.— We find that the optical phonon generation leads to a reduction of sample transmission. Experimental signal curves obtained for different crystal temperatures are shown in Fig. 2. At low crystal temperature \( T \leq 5 \) K we find after the optical excitation a signal that decreases with a nonexponential slope. An analysis of the decay curve at longer times (not shown in Fig. 2) indicates that a nonthermal phonon population is maintained for about 100 μs. After this time thermal equilibrium in the optically excited crystal volume is established. The decay curve (lower curve in Fig. 2) can be described by two exponential curves, where the fast decay has a time constant of about 5 μs. At higher temperatures the decay time of the fast decay decreases with increasing temperature, at 13 K (upper curve in Fig. 2) the fast decay occurs within 1.5 μs. After the fast decay, the signal curves reach an almost constant slope. Our results indicate that at higher temperatures thermal equilibrium is established very

![Fig. 2. Far-infrared absorption at 0.89 THz after optical excitation of a TlCl crystal.](image)
fast and that heat diffusion out of the excited volume occurs very slowly. We think that the fast signals are due to long-lived TA phonons. Our results indicate that the TA phonons have a lifetime of a few microseconds at low temperature and that the lifetime decreases strongly with increasing temperature.

The temperature dependence of the phonon lifetime is shown in Fig.3 for TlCl and TlBr crystals. We find that the lifetime for TlBr has a stronger decrease than the lifetime for TlCl.

Fig.3. Lifetimes of zone-boundary phonons in TlCl and TlBr.

3. Discussion.— According to nonlinear elasticity theory for isotropic dispersionless solids, high-frequency LA phonons have short lifetimes at low crystal temperature due to anharmonic spontaneous decay, while TA phonons are not allowed to decay spontaneously. At higher temperatures TA phonons can decay by the interaction with thermal phonons. Our results can qualitatively be explained by the nonlinear elasticity theory. We attribute the lifetime at low temperature to mode conversion of the TA phonons into LA phonons caused by impurity scattering. At higher temperatures we obtain an additional decay mechanism caused by the interaction with thermal phonons. Our results are in qualitative agreement with predictions of theory.