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OBSERVATION OF STRONGLY FREQUENCY DEPENDENT LIFETIMES OF ACOUSTIC PHONONS IN CaF₂

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Abstract.—A strongly frequency dependent lifetime of acoustic phonons in CaF₂ at low crystal temperature is observed. The phonon lifetime decreases proportionally to \( v^{-5} \) indicating spontaneous phonon decay. Furthermore, we report measurements of the spectral distribution of phonons generated by nonradiative transitions.

Various attempts have been made to detect spontaneous decay of high-frequency acoustic phonons without, however, conclusive results.¹ In this paper we report experimental evidence of spontaneous decay of acoustic phonons. Our experiment was performed on CaF₂ at low crystal temperature.

The principle of our experiment is shown in Fig.1. Uniaxial stress is applied to a CaF₂ crystal doped with 0.003 mole% Eu²⁺ ions. The stress leads to a splitting of the lowest excited state level of Eu²⁺ into a doublet with variable energy separation \( h\nu \) where \( h \) is Planck's constant. This system allows tunable phonon detection up to a frequency \( \nu \) of 3.2 THz.² For phonon generation the crystal is optically pumped with radiation of a nitrogen laser. The radiation is absorbed in broad bands of the Eu²⁺ ions. Nonradiative transitions lead to population of the stress split energy levels and, simultaneously, to generation of phonons with a broad frequency distribution. An additional phonon source is due to one-phonon spin-lattice relaxation in the doublet levels. Phonons are detected by the phonon induced fluorescence radiation \( S₂ \) (Fig.1). Since the spin-lattice relaxation time is shorter than 1 ns the relative population of the doublet levels is in equilibrium with phonons of the frequency \( \nu \). Therefore, we can directly measure phonon occupation numbers by observing \( S₁ \) and \( S₂ \) fluorescence. In addition, from the decay of
the $S_2$ signal phonon lifetimes are obtained.

Experimental results for phonon lifetimes are shown in Fig.2. We observe an almost frequency independent phonon decay time in the range up to 1.5 THz, but, a strong decrease as $v^{-5}$ at higher frequencies. A $v^{-5}$ dependence is predicted by theory for the case of phonon splitting by anharmonic three phonon processes. We compare the lifetimes with theoretical values calculated for an isotropic dispersionless solid. Regarding that due to impurity scattering transverse and longitudinal phonons at the same frequency are in an equilibrium, and assuming that only longitudinal phonons can decay spontaneously, whereas transverse phonons cannot decay due to energy and momentum conservation, we find that our experimental decay times in the high-frequency range are in agreement with theory. In the low frequency range, the decay times are an order of magnitude longer than the ballistic times of flight ($\tau_T^L$ and $\tau_F^L$ in Fig.2) out of the optically excited volume, indicating a diffusive propagation of the phonons. The reason for a frequency independent lifetime is not known, the low frequency lifetime may be due to inelastic scattering of phonons at impurities.

In order to obtain information on the phonon spectrum, which is generated by the nonradiative transitions, we have determined phonon occupation numbers from the ratio $S_2/S_1$ of the fluorescence intensities. In Fig.3a experimental phonon occupation numbers $p$ are plotted for different phonon frequencies. We find that immediately after laser pulse excitation phonons with a nonthermal spectrum are contained in the crystal. In Fig.3a we have also drawn occupation numbers for a Planckian spectrum of $T=10$ K. This temperature is estimated using specific heat data for the case, that fast thermalization of the absorbed laser energy occurs. In the low frequency range, the observed occupation numbers are up to two orders of magnitude lower than for the 10 K spectrum, while at higher frequencies the experimental occupation numbers are larger than for the
Planckian spectrum. Our result indicates that by the nonradiative transitions mainly high-frequency phonons are created.

Fig. 3. (a) Phonon occupation numbers $p$ immediately after laser pulse excitation ($t = 0$); (b) Phonon spectrum at different times.

From the occupation numbers the spectral energy density of the phonons is derived. In Fig. 3b the spectral energy densities of the phonons for two different times $t$ after laser pulse excitation are shown. At $t = 0$ we find a large phonon energy at high frequencies. At a later time we observe a strong decrease in the high-frequency range, while at lower frequencies the spectral phonon energy has increased. This confirms that high-frequency phonons decay spontaneously into phonons of lower frequencies.

In summary, we have studied the temporal development of a non-thermal phonon distribution in $\text{CaF}_2$ at low crystal temperature. The results of our measurements show, that phonon decay times are strongly frequency dependent in agreement with theory.

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References