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HEAT PULSES IN NaF: ONSET OF SECOND SOUND*

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In NaF, NaI, and LiF crystals of modest chemical or isotopic purity, only ballistic and diffusive propagation of heat is observed. In very pure NaF an additional pulse occurs. This pulse shows the behavior of incipient second sound.

In pure dielectric crystals at low temperatures, heat pulses can propagate ballistically as transverse and longitudinal excitations with the speed of sound. If at increasing temperatures momentum-conserving phonon collisions (N processes) become frequent, while the momentum-destroying collisions are still infrequent, the energy will propagate as a collective temperature pulse with a characteristic velocity determined by the interaction of all the phonon modes in the crystal. This so-called "second sound" has been observed in solid helium.[1] In many respects, solid helium is an extraordinary solid. In particular, it is a highly anharmonic crystal, indeed unstable in the harmonic approximation. One may, therefore, ask whether the conditions for second sound can also be met in more ordinary crystals with highly harmonic binding forces. In this Letter, we report on our search for second sound in such crystals.[2]

In order to enhance the strength of the momentum-conserving relative to the momentum-destroying processes, we used crystals of high chemical and isotopic purity, namely NaF, NaI, and LiF. The two sodium salts are naturally isotopically pure, the LiF was isotopically enriched[3] (99.99% Li^7F). The crystals were seed-pulled under purified argon after the starting material had been thoroughly outgassed by heating it in vacuum.[4] The samples were cleaved or string-sawed, ground, annealed near the melting temperature, and polished. Thin zigzag-shaped Manganin heaters (50-Ω) and lead detectors (300 Ω at room temperature) approximately 6 mm × 6 mm were evaporated onto opposing faces. Heat pulses of 0.1 to 1.0 μsec duration and up to 150-W power were generated with a repetition rate of 10^3 sec^{-1}. The temperature rise of the bolometer, recorded using a wide-band amplifier and a boxcar integrator, never exceeded 1°K. The average temperature of the crystal, mounted in a standard conduction cryostat, was adjusted by an auxiliary heater.

Figure 1(a) shows heat pulses in NaF of average purity, with a maximum thermal conductivity $K_{max} = 70$ W/cm °K at 12°K (5 mm × 5 mm cross section, sandblasted surfaces). At low temperatures, the leading edges of the pulses arrive with the speed of longitudinal and transverse sound, respectively. At higher temperature the energy in the ballistic pulses decreases, and an increasing amount arrives diffusively, with no well-defined time of arrival. Such a behavior has been previously reported for a variety of crystals.[5,6] We have also observed it in NaI ($K_{max} = 22$ W/cm °K at 5°K and in the isotopically enriched LiF. It appears that defect or umklapp scattering dominates over N-process scattering in all these crystals.

By repeated seed pulling, the impurity concentration in NaF can be decreased markedly.[4] This causes an increase of the thermal conductivity ($K_{max} = 150$ W/cm °K at 14.5°K). In such a crystal, the heat pulses show a striking new feature

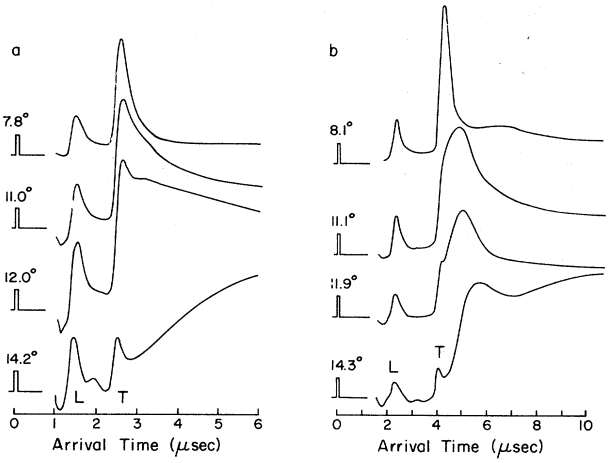

FIG. 1. Heat pulses in NaF in the ⟨100⟩ direction at temperatures in the vicinity of the thermal conductivity maximum. (a) Singly grown NaF, separation between heater and bolometer $l = 7.8$ mm. (b) Triply grown NaF, $l = 12.7$ mm. Crystal faces approximately 12 mm $\times$ 12 mm. The letters L and T mark the longitudinal and transverse pulses, respectively. The pulse amplitudes cannot be compared for different traces, because the amplifier gain was increased with increasing temperature.

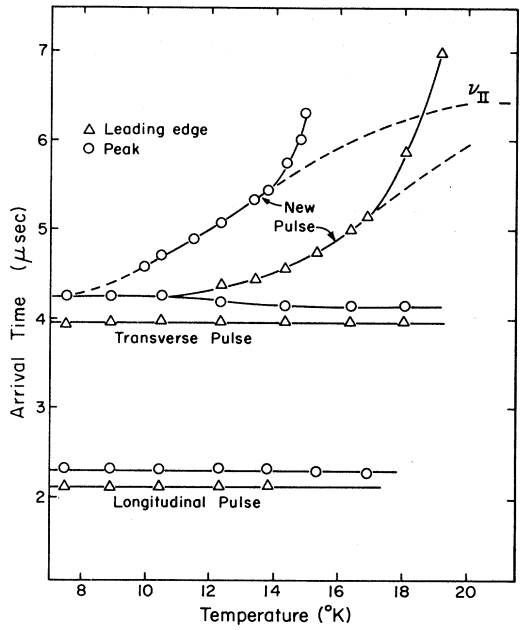

FIG. 2. Arrival times of leading edges and peaks in triply grown NaF, $l = 12.7$ mm. The dashed curves indicate the arrival times expected in the case of relatively weaker momentum destroying phonon collisions. These dashed curves are similar to those observed in solid helium. v_{II} marks the arrival time expected for the fully developed second-sound peak ($v_{II} = 2.0 \times 10^5$ cm/sec).

[Fig. 1 (b)]. An additional heat pulse appears between the transverse and the diffusive pulse. The velocity of this new pulse decreases with increasing temperature. Eventually, a temperature is reached at which this pulse disappears into the diffusive signal. This behavior was observed for several different sample geometries with path lengths varying between 0.5 and 1.3 cm, for pulse propagation directions along ⟨100⟩, ⟨110⟩, and ⟨111⟩, and even for reflected pulses. A dependence on the heat pulse length t_p was not observed. Figure 2 shows the arrival times of the peaks and of the leading edges of the pulses as a function of temperature. It shows the slowing down and the broadening of the new pulse as the temperature increases from 10°K. A rapid increase in the arrival time of the peak occurs at high temperatures, when the new pulse appears more like a kink on top of the large diffusive signal, rather than like a separate pulse. In Fig. 2 this behavior sets in above ~14°K.

A fully developed second-sound pulse in an isotropic crystal travels with a velocity $v_{II} = v_D/\sqrt{3}$, (v_D = Debye velocity), and with a width comparable to that of the ballistic pulse.[1] We now consider the transition from second to first sound. As the temperature is decreased and τ_N becomes comparable with t_p, the second-sound pulse speeds up and broadens, as observed in solid helium[1] and indicated by the dashed curves in Fig. 2. As τ_N is further increased (decreasing temperature) the pulse should continue to speed up and eventually disappear into the ballistic pulses. Because of the higher energy content of the transverse branches relative to the longitudinal branch and since the transverse-transverse interactions should be stronger than longitudinal-transverse interactions, one would expect that most of the second sound finally ends up as transverse ballistic phonons at low temperatures. The new pulse in our purest NaF crystal undergoes such a transition as seen in Fig. 2 and hence appears to be what might be termed incipient second sound.

Since even in this very pure crystal only an incipient and not a fully developed second sound was observed, it appears that the strength of the N processes relative to the resistive scattering processes (τ_R/τ_N) is considerably smaller than in solid helium. This agrees with the estimate made for Li^7F on the basis of thermal-conductivity measurements.[1] We may expect a residual impurity concentration of order 10^{-6} mole ratio in the purest crystal[4]; hence, part of the resistive scattering may not be intrinsic.

We conclude that an incipient second sound has been observed in our purest NaF, and hence that heat propagation in the form of second sound is not a special property of solid helium.

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TEMPERATURE DEPENDENCE OF THE WAVELENGTH-MODULATION SPECTRA OF GaAs†

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Wavelength-modulation spectroscopy is used to obtain the temperature dependence of the reflectivity spectrum for GaAs. Results are given in the regions of the E_1 doublet and the major E_2 peak at 5, 80, 150, 225, and 300°K. The theoretical temperature dependence in these regions is obtained through use of Debye-Waller factors and thermal expansion coefficients in an empirical pseudopotential calculation of the Λ_3-Λ_1 and Σ_2-Σ_1 energy splittings.

We have measured the derivative of the reflectivity for GaAs using a wavelength-modulation technique. Results are given in the vicinity of the E_1 and E_2 reflectivity peaks at 5, 80, 150, 225, and 300°K (Fig. 1). This is the first report of a derivative spectrum which has been accurately measured over a wide temperature range, and of a successful theoretical calculation of the temperature dependence of the reflectivity structure at and above the fundamental gap.

Wavelength modulation is achieved through the vibration of a mirror in the optical path inside the spectrometer. A two-beam method with appropriate electronics is used to eliminate the background noise and to yield a derivative reflectivity spectrum $R'(\lambda)/R(\lambda)$ of the sample. This output is converted to the functional form $R'(\omega)/R(\omega)$. The sample is a single crystal of n-type GaAs with a carrier concentration of 10^{16} cm^{-3}. After the sample is freshly polished and etched, it is mounted within an optical Dewar, in which the temperature can be varied continuously from 4 to 300°K, with an accuracy of ±1°K. The detailed construction of our wavelength-modulation spectrometer and associated experimental equipment is described elsewhere.[1]

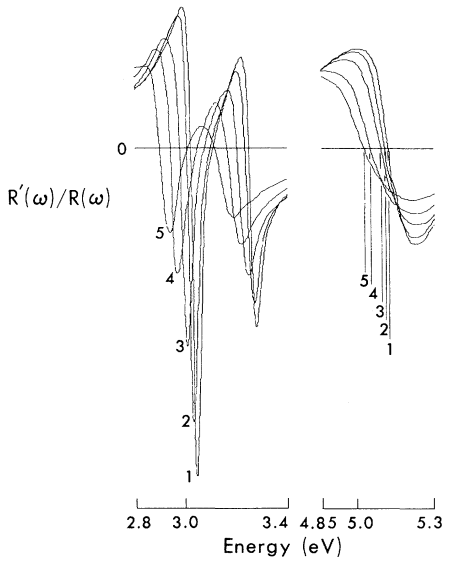

FIG. 1. Plots of $R'(\omega)/R(\omega)$ in the regions of the E_1 doublet peak and the E_2 major peak. Plots 1 through 5 refer to temperatures of 5, 80, 150, 225, and 300°K, respectively.