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Absence of Anomalous Kapitza Conductance on Freshly Cleaved Surfaces

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The reflection coefficient of 290-GHz phonon pulses between LiF or NaF and liquid helium was measured with surfaces prepared *in situ* by cleaving at 1 K. Nearly 100% reflection was found, thus demonstrating the absence of the anomalous Kaptiza conductance in contrast to all previous experiments.

The process of heat transfer between solids and liquid helium has received great interest in recent years.[1] The Khalatnikov model[2] of phonon transmission under strong acoustic mismatch can explain the results of both thermal conductivity experiments below 0.1 K on vacuum-annealed surfaces[3] and the high reflection coefficient observed in phonon experiments below 20 GHz.[4] At higher temperatures or higher phonon frequencies, however, the energy transfer was too large by about two orders of magnitude in all experiments so far. This phenomenon is commonly referred to as the anomalous Kapitza conductance. Its origin is still unknown, although several theoretical models have been suggested.[1,5]

Before proposing specific models, it is important to know whether the effect is intrinsic to an ideal solid surface or it is caused by irregularities. This question has not been answered yet. Although in some experiments cleaved surfaces were used, all of these had been exposed to air. In one attempt to use ultrahigh-vacuum (UHV) conditions with ion-bombarded surfaces, only 10^{-8} Torr was maintained for several hours before the measurement.[6] Nevertheless, the Kapitza conductance appeared to be reduced. We wish to report here on the first experiments on surfaces prepared by cleaving at 1 K *in situ*. Surprisingly, the anomalous Kapitza conductance is almost absent on these nearly ideal surfaces.

The experimental setup was similar to that used in our previous work,[7] except for the crystal holder shown in the inset to Fig. 1. A crystal of LiF or NaF was sealed into a vacuum chamber so as to form a window. Onto the outside surface of the crystal, superconducting tunneling junctions were evaporated as generator (Sn) and detector (Al) of monochromatic phonon pulses of 290 GHz.[8] The chamber was immersed in the helium bath. By this geometry, stable conditions were maintained for the junctions.

Inside the chamber, a razor blade had been carefully adjusted to a (100) cleavage plane of the crystal far from the junctions. The blade could be actuated by a screw which also served as a baffle to the filling line on top of the chamber. In a typical run, the chamber was pumped at room temperature for 1 h with the screw out of position, and then cooled down with the screw in position. At 1 K, the seal was tested by a helium-leak detector. In the absence of a helium leak, an excellent vacuum (better than 10^{-12} Torr) is expected.

Figure 1 shows the results of an experiment with LiF. The dash-dotted trace is the detected phonon signal versus time before cleaving. Two reflected pulses are seen on top of a background due to bulk scattering in LiF. The direction of the group velocity as determined by the junction geometry was about 3° away from the [100] axis.

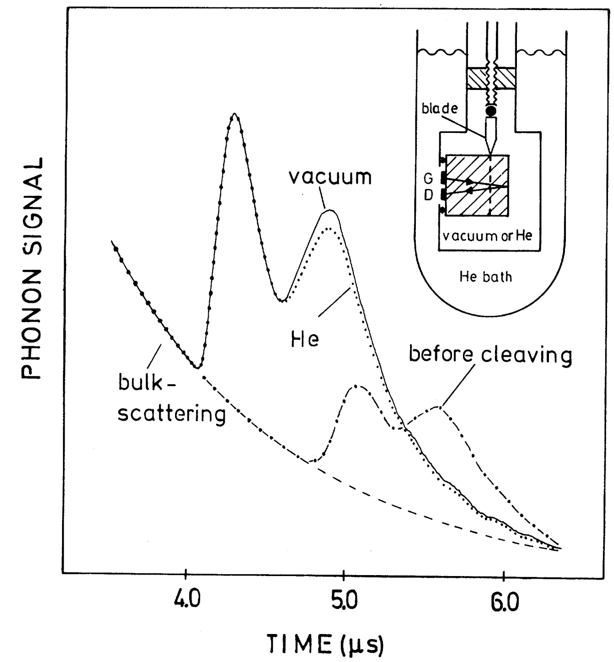

FIG. 1. Phonon pulses reflected from LiF surfaces: dash-dotted line, before cleaving in vacuum; solid line, freshly cleaved at 1 K; dotted line, with helium (the effect is surprisingly small).

Therefore, the first pulse is the fast transverse (FT) mode whose polarization is purely parallel to the reflecting surface. The second pulse is mainly a slow transverse (ST) mode with an oblique direction of phase velocity (20°) and polarization. The longitudinal (L) mode was not observed in LiF because of phonon focusing.

To cleave the crystal, the blade was moved by about one turn of the screw (0.3 mm) while the phonon signal was monitored on an oscilloscope. Successful cleaving manifested itself by a sudden shift of the pulses to shorter times of flight. This is shown in Fig. 1 by the solid trace; in this case, the freshly cleaved surface was still in vacuum.

After stability and reproducibility have been checked to be much better than the thickness of line on the recorder, pure helium was taken out of a storage vessel and slowly introduced into the chamber through the thread of the screw. After 15 min, the screw was taken out and the chamber was totally filled with liquid helium. During this procedure, the helium-bath temperature was monitored and kept below 1.1 K. Thereafter, the original temperature was restored by using the detector junction characteristics for temperature regulation.[9] The result with helium in the dotted trace of Fig. 1. Within an accuracy of at least 1%, no change was observed on the FT pulse, and only a 6% change on the ST pulse was observed. This is in contrast to the many previous experiments on less ideal surfaces of various crystals in which changes up to 70% (i.e., reflection coefficient 30%) were observed.[1] In particular, the purely parallel-polarized FT mode shows a behavior in perfect agreement with the Khalatnikov model, which predicts a null effect in this case. On the other hand, the strong energy transmission observed for transverse waves on the less ideal surfaces is a characteristic feature of the anomalous Kapitza conductance.[10] In this sense, the anomalous Kapitza conductance is obviously absent on our more ideal surfaces.

The situation is a little more complicated for the ST mode whose polarization has a component perpendicular to the surface. The observed change of 6% is roughly a factor of 10 larger than predicted by the Khalatnikov model. However, one can still make the assumption that the transmission is enhanced, according to some modifications of the Khalatnikov model, by widening of the "critical cone"[11] or by surface roughness.[12] These mechanisms would not affect the first pulse because it has no normal component of the polarization. We have tested this assumption by a comparison of L and T modes.

Both L and T modes could be observed in NaF. The traces shown in Fig. 2 were taken in the same way as described above. The background of bulk scattering is almost absent in NaF. In spite of this, the reflected pulses exhibit long diffusive tails. One should, therefore, not ascribe

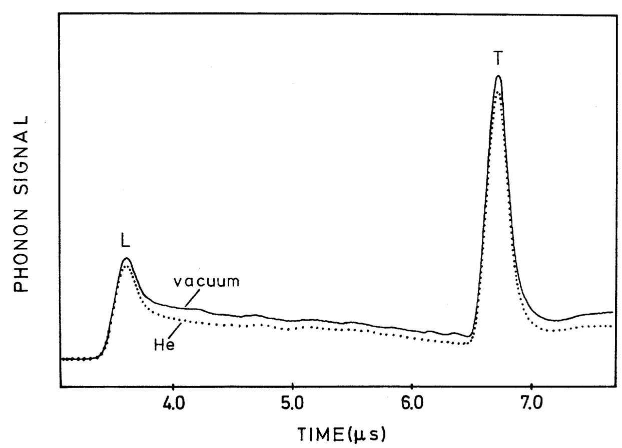

FIG. 2. Longitudinal (L) and transverse (T) echoes from a freshly cleaved NaF surface. The helium effect is not larger on the L phonons than on the T phonons, but is largest for the diffusive tails.

these to bulk scattering. Rather, diffuse scattering from the surface is expected because the phonon wavelength was about 10 nm and microscopic steps were resolved on the surface with an electron microscope. The L and T modes both were almost pure modes with angles of 13° (L) and 4° (T) between the phase velocity and the [100] direction. Therefore, the T mode had only a very small normal component of the polarization, as compared to the L mode.

The effect of helium is shown by the dotted trace of Fig. 2. The change is about (3–4)% on the L pulse and 6% on the T pulse. This behavior cannot be explained by the modified Khalatnikov models, because the T mode should show almost no effect in comparison with the L mode. In contrast, the effect on the T mode is rather larger. An even larger relative change with helium is seen for the diffusively scattered tails of the pulses. Therefore, although small, a residual "anomalous" energy-transmission mechanism still persists even on these nearly ideal surfaces. This residual effect seems to depend on the quality of the cleavage. In fact, electron micrographs of the LiF surface of Fig. 1 revealed much fewer steps than those of the NaF crystal of Fig. 2. Also, some further experiments on less perfectly cleaved LiF show a greater helium effect (up to 10%). Note that all these surfaces were atomically clean. This suggests that the irregularities which cause diffusive surface scattering are also responsible for the anomalous Kapitza conductance.

The effect of exposing the surface to air for one day is demonstrated in Fig. 3. The same crystal

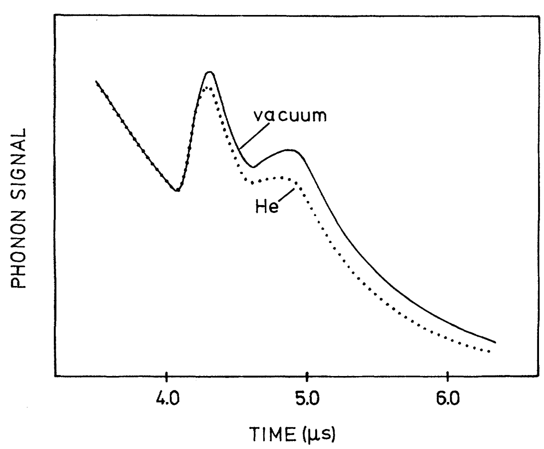

FIG. 3. After the LiF surface of Fig. 1 was exposed to air at room temperature. The helium effect is greatly enhanced.

surface as that of Fig. 1 was used. Under vacuum conditions, the echo pulses are somewhat diminished and broadened, as compared to Fig. 1. This indicates that the reflection was less specular but more diffusive. The dotted trace of Fig. 3 was taken with bulk helium. The relative change is 12% for the pure T mode and 22% for the oblique mode.

We have done further experiments where a freshly cleaved sample was simply warmed up to intermediate temperatures and then cooled down again. The first enhancement of the helium effect was observed after warming to 20 K. The further increase was rather gradual. The greatest helium effect was obtained after "cleaning" the surface with acetone.

We conclude that the anomalous heat transfer which cannot be accounted for by the Khalatnikov mechanism is caused by irregularities and is not intrinsic to ideal surfaces. More information should be obtained by studying well-characterized nonideal surfaces.

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