Strong Phonon Conversion at the Helium-Solid Interface

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Tunable monochromatic phonon pulses (130 to 870 GHz) were scattered at solid surfaces covered with various amounts of helium. A strong phonon loss due to only 3 atomic layers was observed which was independent of frequency and did not exhibit structure due to the helium dispersion. The results indicate strong frequency conversion at the interface, and thus suggest a new approach to the Kapitza-resistance problem.

The thermal boundary resistance between liquid helium and solids, the well-known Kapitza resistance, is still not understood.[1] The magnitude of the thermal resistance is much smaller than predicted by the acoustic mismatch theory of Khalatnikov.[2] Several refinements of the theory remain unsatisfactory as yet, and heat-transfer mechanisms other than direct transmission of phonons have been proposed.[1] More recently, in experiments with high-frequency phonons (> 100 GHz or 5 K), Anderson and Sabisky[3] have apparently found an almost complete transmission through the interface. A strong phonon loss due to the He bath was also reported by Trumpp, Lassmann, and Eisenmenger.[4] By scattering heat pulses at surfaces covered with helium, Guo and Maris[5] found that the "transmission" was more effective for transverse than for longitudinal phonons.

We wish to report here on measurements where we employed monochromatic phonon pulses with variable frequency[6] rather than heat pulses, by using superconducting tunneling junctions as phonon generators and detectors. A frequency range extending from below the roton minimum to far above the dispersion curve of superfluid He[7] was investigated (130 to 870 GHz). The frequency of the backscattered phonons was discriminated by using the frequency threshold of the detector junction.[8] No indications of a direct transmission of phonons into bulk liquid He modes was found so that the previous interpretation[5] must be modified. Rather, effects due to inelastic processes in a few atomic layers near the interface were observed by our technique.

The experimental arrangement is shown in the inset of Fig. 1. The phonon-generator junctions were placed 0.2 mm apart on the same surface of a crystal. Mechanically polished disks of Al_2O_3, 6 mm thick, in c orientation, and of Ge, 4 mm thick, in [110] orientation were used. The phonons were scattered back from the far crystal surface to which a vacuum chamber was attached. The chamber contained either vacuum, He^4 gas, or bulk liquid. The pressure was measured by a Barocel capacitance manometer and corrected for thermomolecular flow.[9] The entire arrangement was immersed in a He bath to ensure rapid heat removal and constant conditions for the junctions.

The method of phonon generation was described previously.[6] The pulses emitted by the generator junction consist of two contributions, the monochromatic "bremsstrahlung" phonons with tunable frequency $(eV - 2\Delta_G)/h$, where V is the bias volt-

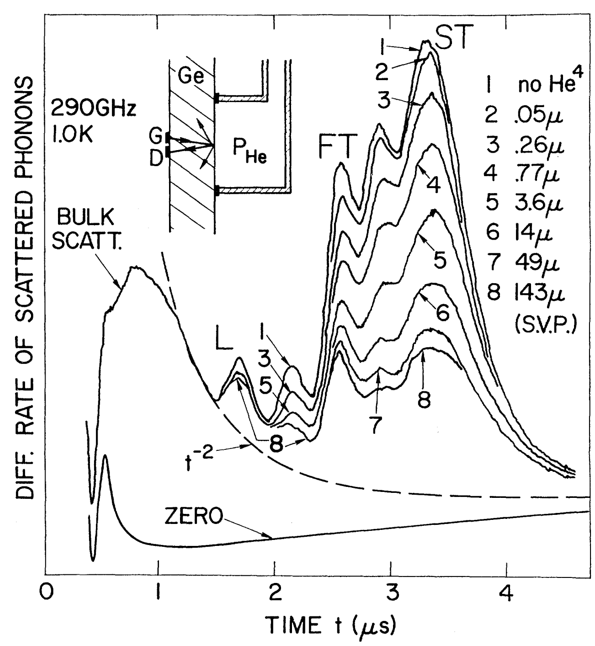

FIG. 1. Phonon echoes scattered from solid surface without helium (trace 1), with He^4 films (traces 2–7), and with bulk liquid at surface (trace 8). L, longitudinal; FT, fast transverse; ST, slow transverse echo. Inset shows sample with generator and detector junctions and vacuum chamber.

age and $2\Delta_G$ the energy gap, along with the "recombination" phonons with the fixed frequency $2\Delta_G/h$. The upper frequency limit of the "bremsstrahlung" phonons is also given by $2\Delta_G/h$, because of the onset of reabsorption. In most experiments, Sn junctions were used with $2\Delta_G/h = 290$ GHz (~14 K). In some experiments, frequencies up to 870 GHz (~42 K) were generated by using PbBi junctions. The use of these junctions will be described elsewhere. The detector junction is sensitive only to frequencies exceeding its energy gap $2\Delta_D$. This threshold was 130 GHz (~6 K) for the Al:O junctions used.

Figure 1 shows the rate[6] of backscattered phonons at 290 GHz as a function of time in a Ge crystal at 1.0 K, for various He^4 pressures at the reflecting surface. After a pulse of 0.1 μsec duration was generated at $t=0$, a bulk scattering signal was first observed. This decreased as t^{-2} when the scattering zone propagated away from the junctions. The dashed line is the appropriate extrapolation for times when the echoes from the far surface entered the detector.

The echoes due to longitudinal (L), fast transverse (FT), and slow transverse (ST) phonon modes were readily identified from the known ultrasonic velocities. The two additional pulses were probably due to mode conversion, resulting from the diffuse scattering at the far crystal surface. That the scattering was indeed diffuse rather than specular is also seen from the tails of the echoes which could be largely suppressed by an annular metal coating, such that the phonons traveling at oblique angles (having longer round-trip times) were absorbed by the metal rather than being diffusely scattered back into the detector.

On introducing He, the echoes strongly decreased. Strikingly, the magnitude of the decrease was different even for the two transverse modes. In total, the L echo decreased to 70% of its original height, the FT echo to 45%, and the ST echo to 32%. With Al_2O_3, the L echo decreased to 90%, and the T echo to 30%. With the heat-pulse technique, Guo and Maris[5] found only a decrease to about 70% for the T phonons in Al_2O_3.

By measuring the emission of second sound from heat pulses, Swanenburg and Wolter[10] found equal intensities for both L and T modes. This is not inconsistent with our results however, because their L pulse corresponds to the sum of the L and the mode-conversion echoes (L to FT) in our case. The latter was in fact strongly decreasing. This indicates that only those L phonons which are undergoing mode conversion, i.e. which give rise to a transverse motion of the surface, can strongly interact with the helium.

It is important to notice that the echoes decreased already at pressures below the saturated vapor pressure when only an unsaturated film was present at the surface. The film thickness is usually calculated by[3] $d=d_0[\alpha/T\ln(P_0/P)]^{1/3}$, with P being the gas pressure, P_0 the saturated vapor pressure, T the temperature, α a material parameter, and d_0 the thickness of a monolayer at a density of bulk He^4. Since the result is relatively insensitive to α, we can use for Ge the value for Si, 36 K, together with $d_0 = 3.6$ Å.[11] Accordingly, Fig. 2 shows the relative height of the ST echo as a function of the film thickness.

Obviously 2 or 3 atomic layers are already sufficient to cause the effect while increasing the film thickness has very little influence, and filling the chamber with bulk liquid has no effect. Thus it is clear that the effect is not merely due to an enhanced direct transmission. This would simply lead to reflection at the film surface and retransmission into the solid with the same high transmission coefficient. Similarly, other direct, i.e. frequency-conserving, processes such as conversion to and delayed reconversion from surface waves, or just an enhancement of the diffusivity of the backscattered phonons, would not allow the phonons to leave the system. Thus, if the echoes were decreased by such a process, their tails would be increased rather than also decreased as observed (see Fig. 1).

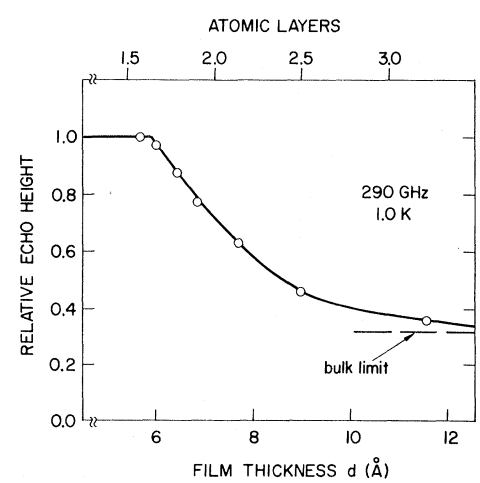

FIG. 2. ST echo height versus absolute helium film thickness. Upper scale shows thickness in atomic layers. At 3 atomic layers the bulk limit is almost reached.

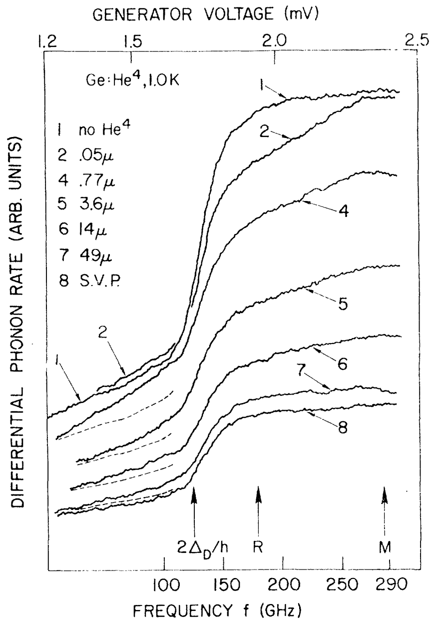

FIG. 3. Frequency dependence of ST echo between 130 and 290 GHz. The dashed traces indicate the decreased background of "recombination phonons." The actually measured traces show an additional contribution due to the up-conversion of "bremsstrahlung" phonons. No structure is seen due to the roton minimum (R) or the phonon maximum (M) of the helium dispersion.

The only direct process by which a phonon can indeed leave the system is the direct desorption of a He atom from the film surface. In that case, however, an onset frequency is expected[12] corresponding to the energy of vaporization, 7.17 K (150 GHz), plus a contribution of the order $kT \times \ln(P_0/P)$. No indication of this was observable, as one can see from Fig. 3 where the ST echo height is displayed as a function of the frequency of the "bremsstrahlung" phonons.

Thus, no direct processes can be responsible for the signal decrease in the case of thin films. Rather, one has to consider indirect, i.e. inelastic, processes which involve conversion of the frequency of the incident phonons. Evidence for such processes can be obtained by analyzing the frequency distribution of the backscattered (or reemitted) phonons. A true frequency analyzer for this purpose is not available yet. However, one can obtain similar information by using the frequency threshold of the detector junction. At frequencies just below $2\Delta_D$, the "bremsstrahlung" phonons are not detectable. If some of the inelastic processes due to the helium film lead to the backscattering (or reemission) of phonons with higher frequencies, however, these will show up as a detector signal.

One can in fact see this from Fig. 3 after accounting properly for the decreasing background of "recombination" phonons. The latter have the fixed frequency $2\Delta_G/h = 290$ GHz so that they decrease with the same ratio as the signal at 290 GHz. Therefore, this ratio can be used to calculate the decreased background from the vacuum trace 1, for each film thickness. The result is plotted as the dashed traces in Fig. 3. As compared to these, the actually measured traces are enhanced, thus demonstrating that additional phonons with frequencies higher than $2\Delta_D/h$ were indeed detected. Their rate was largest for trace 4 (about 10 times the noise), and equally large also for a trace taken at $P = 0.26$ mTorr (not shown). Thus we have obtained experimental evidence for relatively strong frequency-up-conversion processes.

The up-conversion processes naturally also imply the presence of down-conversion processes. For frequencies above $2\Delta_D$, all the up-converted phonons will be detected in the same way as the original ones. However, the down-converted phonons will not be detected if they have frequencies below $2\Delta_D/h$. Thus, the strong loss of phonons at the frequencies above $2\Delta_D/h$ can be indeed also understood by the inelastic processes.

In the case of very thin films (trace 2), the spread of the backscattered (or reemitted) spectral distribution seems still to be narrow, so that at low or high frequencies away from the threshold, the signal has not yet changed whereas near the threshold the spread already manifested itself by the change in signal amplitude. For thicker films, the spread seems to become larger and down conversion dominates, leading effectively to thermalization. Simultaneously the gas pressure becomes larger so that the heat conduction to the gas is no longer negligible. In fact, the signal decrease observed with the heat-pulse technique[5] shows that heat is conducted into the gas, because the bolometers used were not particularly sensitive to frequency-conversion effects. However the total decrease observed with bolometers seems to be smaller than what we observe with junction detectors, so that even in the case of bulk liquid there seems to be some of the energy returning to the solid as thermal phonons. We plan to clarify this point by detecting the backscattered phonons simultaneously by a bolometer and a junction detector.

Since the signal shows no further change after about 3 atomic layers are completed, it is very likely that the inelastic processes occur in these few atomic layers regardless of whether gas or bulk liquid is adjacent. This is also consistent with the fact that we have observed no structure at the frequencies of the maximum phonon (M in Fig. 3) and minimum roton (R) energies, in the case of bulk liquid (trace 8), whereas one would expect singularities[13] in the transmission coefficient if the phonons were directly transmitted into bulk liquid-helium excitations. Also, the total signal decrease was found to be independent of frequency up to 870 GHz which is far above the dispersion curve of liquid helium.

Thus our measurements indicate that indirect, i.e. frequency-conversion, processes are involved in the mechanism responsible for the enhanced heat conduction through the interface observed above ~1 K when the higher-frequency phonons become more important. To understand the Kapitza problem, one should therefore consider the microscopic nature of the first few atomic layers at the interface which are compressed by the Van der Waals force.

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