# Experimental study of the reflection of phonons at an interface between dielectric crystals and liquid helium, gaseous helium, and solid neon\*

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Experimental measurements have been made of the reflection coefficient of phonons incident on a boundary between several dielectric crystals and liquid helium, gaseous helium, and solid neon. The measurements were made using the heat-pulse technique. A brief discussion of these results is given in terms of current theories of the Kapitza resistance.

# I. INTRODUCTION

Since the original discovery by Kapitza<sup>1</sup> of a thermal boundary resistance (Kapitza resistance) between helium II and solids, many experimental and theoretical investigations of this effect have been made.<sup>2</sup> Despite these efforts an understanding of the heat-flow mechanism has not yet been achieved. The theory of Khalatnikov<sup>3</sup> considers that the Kapitza resistance arises from the large acoustic mismatch between liquid helium and solids. This mismatch should lead to very small transmission probabilities for phonons incident on the boundary and hence to a large Kapitza resistance. A typical value of the transmission probability for a phonon coming from the solid side is 0.005. At temperatures above 1 °K the measured Kapitza resistance is usually one or two orders of magnitude smaller than Khalatnikov's theory predicts.<sup>2</sup> It therefore appears that the actual transmission probability for a phonon is much greater than the value given by the acousticmismatch theory.

To try to understand the origin of this discrepancy, a number of attempts have recently been made to make direct measurements of the phonon reflection or transmission coefficients.<sup>4-10</sup> Ideally, one would like to know the dependence of the transmission coefficient on the energy, polarization, and angle of incidence of the incoming phonon; the ambient temperature; the condition of the solid surface; and the pressure above the helium. At least partial information is now available on the influence of most of these variables. Ultrasonic techniques<sup>4</sup> have shown that for phonons of energy less than 0.05 °K the transmission coefficient is close to the acoustic-mismatch value. Anderson and Sabisky<sup>5</sup> used their spin-phonon spectrometer to generate monochromatic phonons with energies up to 14 °K and found that the transmission coefficient was considerably greater than the acousticmismatch value and that it increased with increasing energy. Measurements at high energy

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 $(\epsilon = 14 ^{\circ} K)$  have also been made by Trumpp *et al.*<sup>6</sup> The measurements of Anderson and Sabisky and Trumpp et al. gave values of the transmission coefficients averaged over phonon polarizations and all angles of incidence. The dependence of the reflection coefficient on phonon polarization was studied by Guo and Maris,<sup>7</sup> who used a heat-pulse reflection technique. Their results showed that, at least for phonons of energy above several degrees Kelvin, the transmission near normal incidence was appreciably greater for transverse phonons than for longitudinal. The transmission probability into an unsaturated film was also measured and it was found that appreciable transmission occurred even when the pressure was less than  $10^{-2}$  of the saturated vapor pressure. Recently Kinder and Dietsche<sup>10</sup> performed phonon-reflectance experiments with a tunnel-junction phonon source instead of the simple heated film used by Guo and Maris. This enabled them to study the transmission coefficient for phonons of quite welldefined energy, propagation direction, and polarization. Their data cover the energy range 6-40 °K and include results for unsaturated films. In this paper we will describe our earlier experiments<sup>7</sup> in more detail and will also report on some additional experiments that we have performed since then.

## **II. EXPERIMENTAL METHOD**

The experiments were performed with essentially the same apparatus as described in Ref. 7 (see Fig. 1). Heat pulses of duration  $\sim 10^{-7}$  sec were generated by applying current pulses to a thin film of constantan. This film was typically 2000 Å thick, 0.2 cm wide, 1.0 cm long, and had a resistance of 15  $\Omega$ . The phonons radiated by the constantan travelled through the dielectric crystal, were reflected at the upper face, and were then detected by one of the superconducting bolometers.<sup>11</sup> The bolometer films were in the form of narrow strips parallel to the constantan film at a distance of 0.3 cm from center to center. The bolometer films were 0.015 cm wide and 1.0 cm long. For measurements between 1.5 and 2.0 °K aluminum bolometers were used. These had a thickness of 200-300 Å and a normal-state resistance of 10-30  $\Omega$ . Measurements between 3.0 and 3.5 °K used indium-tin-alloy bolometers of typical thickness 2000 Å and resistance 20-80  $\Omega$ . Detector currents of 0.5-1 mA were used for the aluminum detectors and 5-10 mA for the indium-tin.

Heat-pulse echoes were first observed with the upper face of the crystal exposed to a vacuum. Some typical echo patterns obtained at 1.6 °K for a silicon crystal with  $\{100\}$  end faces are shown in Fig. 2. In Fig. 2(a) the first pulse I is caused by electrical pickup from the current pulse applied to the generating film. Pulses L and T are due to longitudinal and transverse phonons that have been reflected from the upper face of the crystal. The transverse pulse is much larger than the longitudinal because of the phonon focusing that occurs for transverse phonons in the  $\langle 100 \rangle$  directions in silicon. Pulse C is produced by phonons that have made one traversal of the crystal as longitudinal and one as transverse, mode conversion having occurred at the reflecting surface. The broad feature B arises from phonons that have been scattered in the bulk of the sample and have not reached the upper surface. This unwanted background was reduced considerably by cutting grooves approximately 0.1 cm deep and 0.1 cm wide between the constantan generator and the superconducting bolometers. The grooves and the generator and detector films were always along symmetry directions, i.e., the [100] direction on a (010) face, [112] direction on a (111) face, and [001] direction on a (110) face. The echo



FIG. 1. Experimental apparatus.

pattern after the grooves had been  $\operatorname{cut}^{12}$  is shown in Fig. 2(b).

The upper surfaces of the crystals were polished with either a series of diamond pastes (for sapphire and silicon) or AO Centriforce abrasives<sup>13</sup> (for calcium, lithium, and sodium fluoride) with successively decreasing particle sizes. The crystals, except for sapphire, were chemically etched to remove the damaged layer. Additional mechanical polishing was then done (again except for sapphire) with a solution of 0.3-µm alumina powder in distilled water. This step was repeated several times, each followed by chemical polishing, until a surface was obtained that appeared completely smooth when viewed through an optical microscope.

After measuring the amplitudes of the heat pulses with the upper face of the crystal exposed to a vacuum, helium gas or liquid was introduced into chamber A and the pulses remeasured. Solid neon could also be condensed on the crystal surface. The reduction in the pulse amplitude produced by adding helium or neon enabled the phonon reflection coefficient to be determined. Pulse amplitudes were measured either by direct measurement with an oscilloscope or by a Princeton Applied Research box-car integrator model 160. The resistance-versus-temperature characteris-



FIG. 2. Heat-pulse echo patterns obtained in a {100} silicon crystal at 1.69 K: (a) without a groove between heater and detector and (b) after a groove had been cut.

tics of the superconducting bolometers were measured by a dc method before any heat-pulse measurements were made. Care was then taken to use the bolometers only in the middle of their transitions where the sensitivity was nearly independent of the temperature change. The accuracy of the results for the reflection coefficient was typically  $\pm 0.02$  or 0.03. The heater temperature was calculated as a function of the ambient temperature and the power supplied using the method described by Weis.<sup>14</sup>

# **III. RESULTS**

#### A. Liquid helium

## 1. Dependence on phonon polarization

In our previous note<sup>7</sup> we reported that the reflection coefficient was always smaller for transverse phonons,  $R_T$ , than for longitudinal phonons,  $R_L$ . This result has been confirmed by the more recent measurements. The transmission coefficient for transverse phonons is usually about a factor of 2 greater than for longitudinal phonons. The extreme cases we have observed so far are the lithiumfluoride {110} data reported in Ref. 7 where  $R_T$ is only slightly less than  $R_L$ , and measurements on {100} silicon where the transmission coefficient of transverse phonons is more than three times that for longitudinal. These latter data are shown in Fig. 3.

In their experiments on germanium and sapphire Kinder and Dietsche<sup>10</sup> have also found about a factor of 2 difference between the transmission of transverse and longitudinal phonons. In one experiment on silicon Swannenburg and Wolter<sup>9</sup>



FIG. 3. Reflection coefficient as a function of source temperature for longitudinal L and transverse T phonons reflected at a {100} silicon surface covered with liquid helium at 1.76 °K.

obtained equal transmission coefficients for longitudinal and transverse phonons.

# 2. Dependence on angle of incidence

According to the acoustic-mismatch theory<sup>2,3</sup> the transmission coefficient for a transverse wave should be zero at normal incidence but should be finite for other angles. The finite transmission of transverse phonons observed above might therefore arise because not all of the phonons that are detected have been reflected at normal incidence. This occurs as a result of the finite size and separation of the generating and detecting films. Also the reflection at the upper surface may not be specular, so that phonons can be reflected from any point on the upper surface and still reach the detector. These effects were minimized by making measurements on the peak of the pulse. As described previously,<sup>7</sup> this has the effect that the main contribution comes from those phonons that have travelled by nearly the shortest path. To investigate the importance of the angle of incidence four films were deposited on the lower  $\{100\}$  face of a silicon crystal as shown in Fig. 4. The ambient temperature was 1.6 °K and the heater temperature 4 °K. Using this arrangement, the reflection coefficients of transverse phonons into liquid helium were found to be 0.30, 0.57, and 0.76 for combinations of generating and detecting films of 3 to 2, 3 to 1 (or 4 to 2), and 4 to 1. Thus these results show that transmission of transverse phonons is greater near normal incidence. We should emphasize, however, that only one experiment of this type was performed. Thus we cannot be sure that this result is true in general (e.g., for all materials and for all orientation faces). We have made no measurements for longitudinal phonons.



FIG. 4. Arrangement of films for studying the effect of the angle of incidence. Films 1 and 2 are aluminum detectors and 3 and 4 are constantan generators. The thickness of the films is not drawn to scale.

#### 3. Dependence on heater temperature

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In all cases that we have studied the reflection coefficients show a small monotonic decrease as the temperature of the source is raised. The data of Fig. 3 are typical results for measurements of this type. The source radiates a broad energy distribution of phonons peaked at about  $\epsilon_m = 3k_B T_s$ . Because of the broad distribution one cannot work backwards to obtain precise information about the energy dependence of R. However, the monotonic decrease of R as a function of  $T_s$  indicates that for the energies involved here (roughly 5-35 °K) the reflection coefficient decreases with increasing frequency in a fairly smooth way. There is certainly no evidence to suggest that  $R(\epsilon)$  has any resonant structure associated with critical points of the dispersion curve for excitations in liquid helium. These results are consistent with the conclusions of Anderson and Sabisky<sup>5</sup> who found no significant structure in  $R(\epsilon)$  up to  $\epsilon = 14$  °K, and the more recent measurements of Kinder and Dietsche<sup>10</sup> which extend to  $43^{\circ}$ K.

# 4. Dependence on ambient temperature

An experiment was performed using aluminum and indium-tin detectors with the constantan generator placed symmetrically between them. With this arrangement it was possible to obtain data at ambient temperatures of 1.85 and 3.35 °K on the same run. For transverse phonons incident on a {110} face of silicon covered by liquid helium the reflection coefficient was the same at the two ambient temperatures to within the experimental uncertainty of ±0.03. The source temperature was 6.8 °K.

The same crystal was used to look for a discontinuous change in R near the  $\lambda$  temperature. In this experiment the detector was indium-tin whose transition was lowered by the application of a magnetic field. The results are shown in Fig. 5. Within the experimental uncertainty no discontinuity is present at  $T_{\lambda}$ . We have not made a similar detailed study for longitudinal phonons.

#### 5. Surface roughness

To investigate the effect of surface roughness an experiment was performed with a  $\{111\}$  silicon crystal at an ambient temperature of 1.83 °K. With the top surface of the crystal exposed to a vacuum, the intensity ratios for the longitudinal, conversion, and transverse heat-pulse echoes changed from 1:0.45:2.8 for a polished surface to 1:2.6:8.2 for a surface roughened with emery paper. These ratios were measured in separate helium runs, but the same generating and detecting films were used in each run. The source temperature was 5.5 °K. The large increase in the relative amplitude of the conversion mode for the rough surface is as expected. For transverse phonons the reflection coefficient when liquid helium was added was found to be 0.4 for the smooth surface and 0.7 for the rough surface.

This result is interesting because the Kapitza resistance is usually found to be *smaller* for rough surfaces,<sup>2</sup> implying that it is *easier* for phonons to go from the solid to the liquid when the surface is damaged. It is possible that in the present experiments with a roughened surface a significant fraction of the phonons are reflected from a damaged layer beneath the surface and never actually reach the true surface. Addition of liquid helium would then cause a smaller effect than for a polished surface with no damaged layer behind it.

# B. Helium gas

In Ref. 7 we presented results for the reflection coefficient at a  $\{111\}$  face of a silicon crystal exposed to He<sup>4</sup> gas. At an ambient temperature of 1.88 °K a reduction in the amplitude of the reflected heat pulses was first observed when the pressure was increased above about 10<sup>-3</sup> of the saturated vapor pressure  $P_{SVP}$ . The reflection coefficient dropped monotonically with increasing P and became constant above  $0.5P_{SVP}$ , showing no change even when P reached  $P_{SVP}$  and there was bulk liquid above the crystal surface. We have since made more measurements of this type. Figure 6 shows results obtained for transverse phonons incident on a  $\{110\}$  silicon surface. By using aluminum and indium-tin detectors with the constantan generator placed symmetrically between them, it was possible to obtain data at ambient temperatures of 1.85-3.35 °K on the same run. The temperature of the source was



FIG. 5. Reflection coefficient for transverse phonons at a  $\{100\}$  silicon surface covered with liquid helium as a function of ambient temperature. The source temperature was 6.8 °K.



FIG 6. Reflection coefficient as a function of heliumgas pressure for transverse phonons incident on a  $\{110\}$ silicon surface at ambient temperatures of 1.85 and 3.35 °K. The temperature of the source was 6.8 °K.

6.8 °K. The results show a significant variation with ambient temperature except when P is nearly equal to  $P_{SVP}$ , in which case the results are independent of ambient temperature to within experimental error.

When helium gas is introduced into the space above the crystal, a film of helium will be formed on the upper crystal face. The fact that the reflection coefficient when bulk liquid helium is present (the point  $P = P_{SVP}$ ) is independent of ambient temperature suggests that the difference between the results at 1.85 and 3.35 °K when P is less than  $P_{SVP}$  may simply be due to differences in film thickness. To test this we may estimate the film thickness using the relation<sup>15</sup>

$$-k_B T \ln(P/P_{\rm SVP}) = \alpha/d^3 , \qquad (1)$$

where  $\alpha$  is a constant characterizing the van der Waals interaction between helium and the solid



FIG. 7. Reflection coefficient data of Fig. 6 replotted as a function of  $T\ln(P/P_{SVP})$ . The ambient temperature is 1.85 °K for the triangles and 3.35 °K for the circles.

surface, and d is the thickness of the adsorbed helium film when the pressure is P.  $\alpha$  may be conveniently expressed in units of  $d_0^3 \,^{\circ}$ K, where  $d_0 = 3.6$  Å is the interatomic distance in liquid helium. If the reflection coefficient is a function only of d, a plot of R vs  $T \ln(P/P_{SVP})$  should lead to the same curve at all temperatures. A plot of this type is shown in Fig. 7, which uses the data from Fig. 6. The good agreement between the 1.85 and the 3.35 °K data that results when they are plotted in this way suggests that the reflection coefficient is indeed a function of the film thickness and does not depend significantly upon P and T separately.<sup>16</sup>

Figure 8 shows results for  $\{111\}$  calcium fluoride for source temperatures 5.0 and 6.4 °K. The ambient temperature was 1.76 °K. Measurements on CaF<sub>2</sub> are particularly interesting, since Anderson and Sabisky<sup>15</sup> have recently calculated  $\alpha$  for this crystal and obtained the result  $22.8d_0^3$  °K. This value is in excellent agreement with the value of  $\alpha$  they determined experimentally by film-thickness measurements. Using this value of  $\alpha$  in Eq. (1), we have calculated the film thickness as a function of  $P/P_{SVP}$ . The results (see Fig. 8) show that even with a film only  $3d_0$  thick the reflection coefficient is nearly equal to the value for the bulk liquid. Using their calculated value for silicon ( $\alpha = 36.3d_0^3$  °K) we have also worked out the thickness scale shown on the upper abscissa of Fig. 7.

# C. Solid neon

In Ref. 7 we reported that the reflection coefficients at an interface between  $\{110\}$  lithium



FIG. 8. Reflection coefficient as a function of heliumgas pressure for transverse phonons incident on a  $\{111\}$ calcium-fluoride surface for source temperatures 5.0 and 6.4 °K.

fluoride and solid neon were consistent with the acoustic-mismatch theory. We have since made measurements on  $\{110\}$  silicon which are also consistent with the acoustic-mismatch theory. Both in LiF and Si the results are insensitive to the source temperature. This is also consistent with the acoustic-mismatch picture.

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We also reported earlier that in one experiment with a chemically polished  $\{111\}$  silicon surface the transmission into liquid helium was greater than into solid neon. We have since repeated this experiment with transverse phonons incident on a  $\{110\}$  silicon surface and obtained  $R_T = 0.58$ for liquid helium and 0.74 for neon. These data were at an ambient temperature of 1.83 °K and with source temperatures between 5 and 7 °K. The only other data on neon that we have obtained so far are the LiF results described in Ref. 7. In this case  $R_T$  was roughly equal for helium and neon and  $R_L$  was significantly *smaller* for neon than for helium.

## **IV. DISCUSSION**

The results presented here together with the previous data<sup>4-10</sup> constitute a reasonably complete set of information about the general features of the reflection coefficient of phonons at interfaces between dielectric crystals and liquid and gaseous helium. There remain, however, at least two important questions that are still unsettled. The reflection coefficient from bulk liquid helium at a given phonon energy has been found to be essentially independent of temperature between 1.85 and 3.35 °K. It would be very interesting to see if the coefficient remains the same even at temperatures below 0.5 °K where there are very few thermal excitations in the liquid.

The second question concerns what happens to the energy that is absorbed by the helium. We have assumed throughout that the reduction in the amplitude of the reflected heat pulses is caused by a transmission of energy into the helium. We have no information about the form in which the energy appears in the helium, e.g., as rotons, phonons, or some other excitation. One might even wonder if the addition of helium can simply alter the way in which the phonons are reflected at the surface, so that all phonons stay in the solid but fewer find their way back to the detector.<sup>17</sup> It is known, however, that "transmission coefficients" measured by the reflection technique used in this paper are of the same order of magnitude as those deduced from conventional Kapitza studies of heat flow between solids and liquid helium.<sup>18</sup> In these latter experiments energy definitely does cross the interface and thus it seems likely that it does in the present experiments also. It would be very interesting to make a more detailed study of this problem, perhaps by the method suggested in Ref. 9.

As has been repeatedly emphasized<sup>2</sup> there still is no satisfactory theory of the Kapitza resistance at temperatures ~1 °K or higher. It is, therefore, hardly surprising that the reflection coefficient data are not consistent with any of the current theories. The simple acoustic-mismatch model<sup>3</sup> gives transmission coefficients much smaller than observed. There is no evidence for any enhanced transmission of phonons with energy near to the roton minimum.<sup>5, 10, 19</sup> The lack of reproducibility in Kapitza-resistance studies<sup>2</sup> suggests that surface damage or contamination plays a role. However, at least as developed so far the models in which surface damage plays an essential role appear to be unsatisfactory, since they cannot explain how the transmission into helium can be greater than into neon.<sup>18</sup>

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mation when applied to monolayer or thinner films.

- <sup>16</sup>We believe that the small plateau regions evident in Figs. 6 and 8, and Fig. 3 of Ref. 7 are not genuine but arise from slight drifts in temperature of the crystal leading to changes in sensitivity of the detector.
- <sup>17</sup>It is also possible that after the helium is added the reflected phonons have a different distribution of frequencies. If the detector is less sensitive to the new frequency distribution, a reduction in pulse amplitude will be observed. This was proposed by Kinder and Dietsche (Ref. 10).
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