Thermal boundary resistance

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The thermal boundary resistance present at interfaces between helium and solids (Kapitza resistance) and the thermal boundary resistance at interfaces between two solids are discussed for temperatures above 0.1 K. The apparent qualitative differences in the behavior of the boundary resistance at these two types of interfaces can be understood within the context of two limiting models of the boundary resistance, the acoustic mismatch model, which assumes no scattering, and the diffuse mismatch model, which assumes that all phonons incident on the interface will scatter. If the acoustic impedances of the two media in contact are very different, as is the case for helium (liquid or solid) in contact with a solid, then phonon scattering at the interface will reduce the boundary resistance. In the limiting case of diffuse mismatch, this reduction is typically over 2 orders of magnitude. Phonons are very sensitive to surface defects, and therefore the Kapitza resistance is very sensitive to the condition of the interface. For typical solid-solid interfaces, at which the acoustic impedances are less different, the influence of diffuse scattering is relatively small; even for the two limiting cases of acoustic mismatch and diffuse mismatch the predicted boundary resistances differ by very little ($\lesssim 30\%$). Consequently, the experimentally determined values are expected to be rather insensitive to the condition of the interface, in agreement with recent observations. Subsurface (bulk) disorder and imperfect physical contact between the solids play far more important roles and led to the irreproducibilities observed in the early measurements of the solid-solid thermal boundary resistance.

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I. INTRODUCTION

The idea that a thermal resistance might exist between liquid helium and a solid was first expressed as early as 1936. Kürti, Rollin, and Simon (1936) assumed such a thermal resistance to be small and therefore ignored it. Only a few months later, Keesom and Keesom (1936) recognized that the thermal resistance at the interface was "relatively very considerable," but they too allowed the idea to pass without further mention. In 1941, Kapitza reported his measurements of the temperature drop near the boundary between helium and a solid¹ when heat flows across the boundary. More than ten years later, Khalatnikov (1952) presented a model, an approximation to what is now known as the acoustic mismatch model, to explain that a thermal resistance, the thermal boundary resistance, occurs at boundaries to helium. In the presence of a heat flux across the boundary, this thermal resistance causes a temperature discontinuity at the boundary. This thermal boundary resistance (often called Kapitza resistance) is defined as the ratio of the temperature discontinuity at the interface to the power per unit area flowing across that interface. The acoustic mismatch model, essentially in its modern form, was first written down by Mazo and Onsager (Mazo, 1955), independent of Khalatnikov's earlier version. The thermal boundary resistance to helium predicted by this theory is of the order of 10^3 K/(W/cm^2) at 1 K, and varies as T^{-3} . This magnitude would be disastrous because it would severely limit attainable cooling powers and increase time constants for reaching thermal equilibrium, thus complicating the quest for experimentation at ever-lower temperatures. Fortunately, the experimentally observed Kapitza resistance was often as much as 2 orders of magnitude smaller near 1 K than that predicted using the acoustic mismatch model.^{2,3}

With more experimentation, patterns developed. Near 100 mK, the measured thermal boundary resistance was only about a factor of 3 below the prediction of the acoustic mismatch model, but above 100 mK the thermal boundary resistance decreased significantly with respect to the theory. However, results were generally irrepro-

¹In the initial experiment the boundary was to a bronze wire thermometer. In subsequent experiments the solid was a thin platinum film on glass, and after that, Kapitza performed a series of experiments on the thermal boundary resistance between copper and liquid helium.

²The term anomalous Kapitza resistance is often used to refer to the fact that the measured Kapitza resistance above 0.3 K is much lower than that predicted with the acoustic mismatch model. Although we are not yet able to calculate the Kapitza resistance quantitatively, as will be reviewed below, we can understand the disagreement qualitatively. It often occurs that a property is described as anomalous in the early phases of the development of a field. The descriptor is then dropped once the property is basically understood; the time has come for the anomalous Kapitza resistance, and we shall avoid the term.

³Some of the behavior of the Kapitza resistance at millikelvin temperatures, in ways unrelated to the contents of this review, is also sometimes referred to as anomalous. In this regime, as well, the Kapitza resistance can be orders of magnitude lower than that predicted using the acoustic mismatch model; see the review by Harrison (1979). ducible. In Fig. 1 are a few measurements of the Kapitza resistance between ⁴He and copper versus temperature between 0.1 and 2 K (Kapitza, 1941; Guan, 1962; Johnson and Little, 1963; Anderson, Connolly, and Wheatley, 1964; Folinsbee and Anderson, 1974). In order to highlight the deviations from the T^{-3} predicted temperature dependence, we have plotted the thermal boundary resistance as $R_{\rm Bd}T^3$. Also shown is the thermal boundary resistance as calculated using the acoustic mismatch model. The lower line, labeled diffuse



FIG. 1. A few examples of the measured Kapitza resistance between copper and ⁴He, multiplied by T^3 to remove some of the strong temperature dependence. Note the drop in $R_{Bd}T^3$ with temperature above 0.1 K. The upper solid line is the prediction of the acoustic mismatch model [500 $K^4/(W/cm^2)$]. The lower solid line is the prediction of the diffuse mismatch model [2.8 $K^4/(W/cm^2)$]. Both models will be discussed in detail in Sec. II. The prediction of the diffuse mismatch model should not be confused with the prediction of the "perfect match model," otherwise known as the phonon radiation limit, also discussed in Sec. II. The curves are as follows: a, Folinsbee and Anderson, 1974; b, Anderson, Connolly, and Wheatley, 1964 (the kink is not physics, but instead shows the typical irreproducibility of the Kapitza resistance from run to run, even on the same sample); c, Johnson and Little, 1963; d, Guan, 1962; and e, Kapitza, 1941. Below 0.1 K, the measured thermal boundary resistance is expected to approach the prediction of the acoustic mismatch model, if the sample were an insulator with a clean, smooth, and undamaged surface. The electrons in the copper contribute to the transport even at these lowest temperatures by providing a mechanism for phonon attenuation in the solid, due to the electron-phonon interaction. This is discussed in Secs. II and III. We know of no direct measurements of the Kapitza resistance at temperatures below 0.2 K of an insulator with a smooth, clean, and unstrained surface.

mismatch, will be discussed shortly (Sec. II.D).

It was originally thought that the measured Kapitza resistance was lower than that expected from acoustic mismatch theory only at interfaces to liquid ⁴He. Then it was found that the same enhanced transport existed at interfaces to liquid ³He (Lee and Fairbank, 1959) and even to solid ³He and ⁴He (Anderson, Connolly, and Wheatley, 1964). Figure 2 shows results on liquid and solid ³He in contact with copper. These data show yet another surprising fact: Since the speed of sound in helium is a strong function of pressure, the acoustic mismatch model would predict a pressure dependence of the thermal boundary resistance. However, this dependence was found only near 0.1 K; around 1 K, the thermal boundary resistance is almost independent of pressure.

To understand the weakness of the pressure dependence, and at least qualitatively the temperature dependence, of the Kapitza resistance at temperatures between 100 mK and a few K, Challis, Dransfeld, and Wilks (1961) calculated the effects of the solid layer of adsorbed



FIG. 2. Measured Kapitza resistance between electropolished copper and ³He, multiplied by T^3 to remove some of the strong temperature dependence (adapted from Anderson, Connolly, and Wheatley, 1964). Note the similarity with the ⁴He data (Fig. 1). The pressure dependence is seen well below 1 K, but near 1 K the Kapitza resistance is nearly pressure independent. The acoustic mismatch prediction for the low-pressure curve is about 1200 K⁴/(W/cm²). The diffuse mismatch prediction for the low-pressure curve is about 2.7 K⁴/(W/cm²).

helium present at helium-solid interfaces due to van der Waals forces. They found the adsorbed layer to act as a phonon-wavelength-dependent reflection-reducing coating, analogous to antireflection coatings in optics. The boundary resistance was predicted to be unaffected by the layer at temperatures below 0.2 K, but to be reduced by the layer at higher temperatures. However, the predicted drop in the thermal boundary resistance was not enough to explain the observations.

Haug and Weiss (1972) and Peterson and Anderson (1973) independently explained the low magnitude of the thermal boundary resistance at the lowest temperatures (below or near 100 mK) by showing that thermal boundary resistance is lowered because of attenuation of phonons in the bulk solid near the interface, caused, for example, by electrons or dislocations. Their analysis is valid at higher temperatures as well, but additional effects usually dominate above 100 mK.

The common denominator in the experiments described so far was that they all involved a boundary to a quantum liquid or solid, and so quantum explanations were sought. Reynolds and Anderson (1976, 1977) measured the Kapitza resistance between copper and solid helium, hydrogen, deuterium, and neon, and observed that the magnitude of the transport in excess of that predicted using the acoustic mismatch model appeared to be proportional to the reduced quantum parameter⁴ of the solidified gas. Maris (1979) also suggested that quantum effects are likely to play a role in the Kapitza resistance. However, Goodstein et al. (1981) concluded from phonon reflection experiments that the Kapitza resistance was lower than predicted using acoustic mismatch theory even at some interfaces between nonquantum solids, specifically between sapphire and neon, argon, krypton, or xenon. We also suggest that the degree of quantum behavior is coincidentally large for exactly those elements for which the acoustic mismatch model predicts an extremely large thermal boundary resistance because the elements with a highly quantum nature tend to be light and soft. Therefore a given increase in the thermal transport will appear more significant for the elements with greater quantum nature, and there is less room for increased transport for elements with lesser quantum nature.

Later experiments showed conclusively that the condition of the interface is a critical factor determining the thermal boundary resistance; scattering at the interface somehow causes the drop in $R_{\rm Bd}T^3$ at interfaces to helium above 0.1 K. At "perfect interfaces," which were

⁴Reynolds and Anderson (1976 and 1977) define quantum behavior using the reduced quantum parameter $\Lambda = h / (\sigma m^{1/2} \varepsilon^{1/2})$, where *m* is the atomic or molecular mass, *h* is Planck's constant, and σ and ε are constants from a Lennard Jones-type potential. This parameter was introduced by de Boer (1948).

most closely approximated by crystals cleaved in situ in liquid helium (Weber et al., 1978a, 1978b), or laserannealed in situ in vacuum (Basso, Dietsche, and Kinder, 1984; Basso, Dietsche, Kinder, and Leiderer, 1984; Mok et al., 1986), the phonon transmission coefficients appeared to come very close to those predicted by the acoustic mismatch model, at least for phonons with frequencies less than 300 GHz (or thermal phonons with T < 3 K). These experiments focused attention on the state of the surface as crucial for the Kapitza resistance. For example, in the temperature range between 100 mK and a few K, coupling between helium and the solid through surface waves was considered in connection with surface roughness by Lapin (1969), Sheard and Toombs (1972a, 1972b), Khater (1978), and Shiren (1981a, 1981b); in connection with surface roughness and excitations in the adsorbed helium film by Nakayama (1985a, 1985b, 1986); and in connection with surface resonant states by van der Sluijs and van der Sluijs (1981a, 1981b, 1981c). The effect of coupling through tunneling states was calculated by Kinder (1981).

In 1959, Little extended the acoustic mismatch model to solid-solid boundaries. Experimental tests of this theory showed that, in contrast to the Kapitza resistance, the thermal boundary resistance between solids was often actually higher than that calculated from the acoustic mismatch model, but it was also poorly reproducible. The most thoroughly studied interfaces were those between sapphire and indium (Neeper and Dillinger, 1964; Wolfmeyer, Fox, and Dillinger, 1970; Park and Narahara, 1971a, 1971b; Schmidt and Umlauf, 1976). In these experiments, the indium was vapor deposited or ultrasonically soldered onto sapphire rods, the two rods were pressed together with additional indium in between, and the "sandwich" was annealed; a typical sample is shown schematically in Fig. 3. The measured thermal boundary resistance for different samples varied significantly, depending on the details of the sample preparation, but was typically a factor of 2 above the thermal boundary resistance as calculated from acoustic mismatch theory (see Fig. 4). The temperature dependence was also not as expected from acoustic mismatch theory; $R_{Bd}T^3$ showed a tendency to increase above 1 K. The magnitude and temperature dependence of the measured thermal boundary resistance suggested that the physical contact between the indium and the sapphire was not complete (Schmidt and Umlauf, 1976). Similar measurements were made by Nitsche and Schumann (1980; see also Sec. IV.B.2) of the thermal boundary resistance between sapphire and lead. The results were much closer to the prediction of the acoustic mismatch model, both in magnitude and in temperature dependence. Measurements by Sahling et al. (1981) of the thermal boundary resistance between aluminum and sapphire showed even closer agreement with the acoustic mismatch model, given a proper interpretation of the measured temperatures of the thermometers (see Sec. IV.B.2).

In order to ensure good physical contact, sandwiches



FIG. 3. Experimental geometry used for measuring the thermal boundary resistance between indium and sapphire (Schmidt and Umlauf, 1976). The indium is vapor deposited or ultrasonically soldered onto each sapphire cylinder, and the coated cylinders are vacuum melted together with a (few mm thick) disk of indium between the cylinders.

of epoxy between flat and polished pure metals were produced (Matsumoto, Reynolds, and Anderson, 1977). At about 100 mK, the measured thermal boundary resistance at the two copper-epoxy interfaces agreed with that predicted by the acoustic mismatch model. Above 100 mK, however, the total thermal resistance of the sandwich contained large contributions from the phonon scattering in the glassy epoxy, and the transmission at the interfaces could not be deduced accurately. This work established that solid-solid interfaces can be made such that the thermal boundary resistance approaches an agreement with acoustic mismatch theory, at least at low enough temperatures.

The solid-solid thermal boundary resistance involving thermal phonons with temperatures well above 1 K was studied by Weis (1969). His boundaries consisted of thin metal films vapor deposited onto dielectric substrates. The film was used as both heater and thermometer, and the substrate was typically kept at 4 K. In the time span of < 100 nsec, the film was Joule heated to a temperature between 15 and 300 K, after which its temperature was allowed to stabilize and was measured. During this time



FIG. 4. Thermal boundary resistance between indium and sapphire multiplied by the cube of temperature to remove the strong temperature dependence (adapted from Schmidt and Umlauf, 1976). \times , normal indium; \bullet superconducting indium. The two sets of data labeled *B* are for a roughened sapphire surface. *A* and *C* are for smooth sapphire surfaces with different indium thicknesses. Acoustic mismatch theory predicts a flat line at 20.4 cm²K⁴/W.

phonons radiated into the substrate, but did not have time to be scattered back into the heater or to thermalize in the substrate;⁵ therefore the substrate temperature remained at 4 K. In Fig. 5 is a plot of data for a gold/sapphire boundary obtained with this technique. The film temperature is plotted versus film power; the calculated film temperature, based on the power density and the acoustic mismatch model, is shown as the solid curve. To within the accuracy of the technique, theory and experiment could agree at temperatures as high as 100 K.

Using a steady-state technique Swartz (1987) measured the thermal boundary resistance from 0.6 to 200 K between metal films and the dielectric substrates onto which the films were deposited. In Fig. 6 is a plot that again demonstrates that at least below ~ 40 K a very close agreement with the acoustic mismatch calculation was obtained. At higher temperatures, significant deviations were found with this technique.

Although the experiments by Weiss and by Swartz



FIG. 5. Temperature of two thin gold films deposited onto sapphire as a function of the power input per unit contact area. The temperature of the sapphire is 4.2 K. For comparison, the solid line is the prediction of the acoustic mismatch model assuming the phonon dispersion in the gold matches that of a one-dimensional linear chain. Adapted from Herth and Weis (1970).

confirmed the acoustic mismatch model over a wide range of phonon frequencies and for a large number of solid-solid interfaces, they raised another problem: From phonon reflection experiments it is well known that phonons in the frequency range above a few hundred GHz are strongly scattered at all but the most carefully prepared surfaces (see, for example, Eisenmenger, 1986). There was good reason to believe that similar scattering would occur at the solid-solid interfaces studied by Weis and by Swartz. In view of the dramatic influence of surface preparation on the Kapitza resistance, should one not expect similar effects in the solid-solid work above a few K, where the heat is carried predominantly by phonons with frequencies exceeding 100 GHz? (See Fig. 6 for the conversion from phonon frequency to temperature.)

In order to estimate the effect of diffuse scattering on the thermal boundary resistance, Swartz constructed a model, the diffuse mismatch model, which represents the limit of strong diffuse scattering. He assumed that all phonons striking the interface are scattered once (elastically) and are then emitted into the adjoining substances with a probability proportional to the phonon density of states in the respective substances. This diffuse mismatch model leads to the lower line in Fig. 1, more than 2 orders of magnitude lower than the upper line corresponding to the acoustic mismatch model for the Kapitza resistance. For solid-solid interfaces, the effect of diffuse scattering is minute, of the order of 10%; in some cases the diffuse mismatch model leads to a slightly higher boundary resistance than that predicted using the acoustic mismatch model, and in some cases the diffuse mismatch model leads to a slightly lower boundary resistance than that predicted using the acoustic mismatch model. Consequently, the good agreement with the acoustic mismatch model in the work of Weis and Swartz

⁵This assumption may not be true for the very high frequency (several THz) phonons emitted from the film at temperatures above ~50 K, in which case the phonon scattering time can drop well below a μ sec, especially if the dielectric is damaged; see Secs. IV.B.4 and IV.C.2. This causes the film to overheat because of the backscattered phonons, causing a temperature rise in the crystal near the film.

was fortuitous in the sense that the boundary resistance is quite insensitive to diffuse scattering at solid-solid interfaces.

An understanding of thermal boundary resistance cannot be complete without studying the evidence provided by investigations of both Kapitza boundaries and solidsolid boundaries. Each type of boundary provides its own set of experimental challenges and its own set of advantages, and each can supply clues helpful to the understanding of both. Kapitza boundaries can be studied only at temperatures below 4 K. They are extremely sensitive to surface (and subsurface) conditions and thus



FIG. 6. Thermal boundary resistance between Rh:Fe and sapphire plotted log-log vs temperature. The surface of the sapphire substrate was as received (Syton-polished by the supplier). The substrate was sequentially soaked in heated acetone, in a heated 1:1:1 solution of NH₄OH:H₂O₂:H₂O, and in distilled H₂O, immediately before loading into a dc sputtering chamber for the Rh:Fe deposition. The solid line represents the acoustic mismatch prediction, the dashed curve the diffuse mismatch prediction. Note the agreement over 6 orders of magnitude of the thermal boundary resistance. The small rise in the boundary resistance relative to the solid curve below a few kelvin is due to the effects of incomplete electron-phonon thermalization (the electrons are hotter than the phonons during the measurement at temperatures below or about 1 K). See the caption of Fig. 32 for more information. The enhancement of the measured resistance above 50 K is caused by substrate damage in the sapphire, as discussed in Sec. IV.C.2 (from Swartz and Pohl, 1987). The frequency scale at the top of the graph marks the dominant phonon frequency $v_{dom} = (90 \text{ GHz/K})T$ (Klitsner and Pohl, 1987), i.e., the frequency of the phonons that carry most of the heat at a given temperature T, in the Debye approximation.

afford a rich set of opportunities for the careful experimenter. That sensitivity, though, also makes interpretation of the data extremely difficult and controversial. In situ laser annealing and in situ cleaving have proven to be powerful tools, providing a clean surface onto which well-characterized defects can be introduced. Lowering the temperature (thus lowering the average frequency and raising the average wavelength of the phonons that dominate thermal transport) has proven to be another way to clean the slate so that the effects of added structure at the surfaces can be studied (Klitsner and Pohl, 1987). Solid-solid interfaces have the technical difficulty that perfect physical contact cannot generally be expected. They are, however, expected to be less sensitive to details at the interface than Kapitza boundaries. Solidsolid interfaces can be studied over the entire temperature range from millikelvins to hundreds of kelvins. At the high temperatures, effects of dispersion, realistic phonon density of states, and inelastic scattering can also be studied.

The main purpose of this review is to show that the physics of phonon transmission at both types of boundaries can be qualitatively understood within a single framework. At low enough temperatures (usually well below 1 K), at sufficiently defect-free (i.e., "smooth") interfaces, phonons are not scattered and therefore behave according to the rules of classical continuum acoustics; under these circumstances, the acoustic mismatch model is a realistic model. At high temperatures (usually above 1 K) and at sufficiently imperfect interfaces, phonons will not usually behave ideally; they will instead scatter. In this case the diffuse mismatch model leads to a more realistic description of the measurements. Various sources of the scattering will also be reviewed.

We shall not discuss the problems of thermal boundary resistance at millikelvin temperatures, spin-spin thermal transport, or thermal boundary resistance to sinters. These issues are extremely important in the general understanding of thermal transport at interfaces in the regime of temperatures below 100 mK. A review of these issues is given by Harrison (1979). We shall consider only interfaces in which the thermal transport on at least one side is dominantly due to the lattice.

There have been several reviews of the problem of the thermal boundary resistance. The classic first studies of the many fascinating properties of helium were reviewed by Keesom (1942) and Atkins (1959). A very complete review of the early work on the Kapitza resistance, of the original derivation of Kalatnikov, and of some of the first experimental and theoretical attempts to understand the observations was written by Pollack (1969). Later reviews of thermal boundary resistance stressing mostly the Kapitza problem have been written by Challis (1961a, 1974), Frederking (1968), Cheeke (1970b), and Wyatt (1980, 1981). For the solid-solid thermal boundary resistance, Little (1959) reviewed the early data and extended the acoustic mismatch model to solid-solid interfaces. Reviews of the solid-solid interface thermal boundary resistance have been written by Anderson (1976, 1981), and Cheeke, Ettinger, and Hebral (1976). Reviews of the use of heat pulses in the study of the interaction of phonons with interfaces are given by Kinder, Weber, and Dietsche (1980), Goodstein *et al.* (1981), Eisenmenger (1986), and Wybourne and Wigmore (1988).

In Sec. II of this paper, the thermal boundary resistance will be carefully defined and many of the subtleties of the definition will be discussed. The problem of what resistance is measured in an experiment will be considered in some detail, and the fundamentals of the theories of thermal boundary resistance will be reviewed. The reader will be referred to original papers for details of calculations. In Sec. III Kapitza resistance measurements and interpretations will be reviewed. Section IV contains the work on solid-solid thermal boundary resistance. Section V contains the conclusions.

II. MODELS FOR THE THERMAL BOUNDARY RESISTANCE

In this section, we shall review the simplest models and definitions needed for a fundamental understanding of the thermal boundary resistance. In Sec. II.A we shall discuss the definitions and the subtleties involved in such things as measurement of temperature in nonequilibrium systems (i.e., in the presence of a heat flow and in systems where the carriers may have long mean free paths). We shall show that, even independent of experimental difficulties the measurement of the thermal boundary resistance is much more subtle than measuring the temperature on two sides of an interface and dividing the measured ΔT by the power per unit area across the interface. As these subtleties have led to much confusion in the literature we have tried to discuss them very thoroughly, using simple analogies to optics and blackbody radiation whenever possible. At the end of this section, we shall critically analyze several experimental arrangements. In Sec. II.B we discuss briefly the modelindependent formalism used to calculate a boundary resistance, and in Secs. II.C and II.D we discuss the acoustic mismatch model and the diffuse mismatch model, respectively. In Sec. II.E we discuss the phonon radiation limit.

A. Definitions and subtleties of the measurement

In order to induce a heat flow \dot{Q} in a material, we must impose a temperature gradient ∇T . The ratio of the heat flow per unit area A to the temperature gradient is defined as the thermal conductivity:

$$\Lambda = \dot{Q} / (A \nabla T) . \tag{2.1}$$

At low temperatures (in the boundary scattering regime), the thermal conductivity is not well defined; it depends on the geometry of the crystal, on the condition of the surfaces (Casimir, 1938), and even on the condition and geometry of the thermometers used for the measurement (VanCleve, Klitsner, and Pohl, 1986; Klitsner *et al.* 1988). We shall see that all the insight necessary to interpret thermal conductivity measurements at low temperatures is needed as well in the interpretation of data on the thermal conductance at an interface.

In order to induce a heat flow Q across a boundary between two materials, there must be a temperature difference ΔT between the two sides of the interface (in addition to the temperature gradients in the adjoining materials). The nature of this "temperature discontinuity" is subtle, because both the presence of the interface and the presence of a heat flow make uncertain even the definition of a temperature. This sublety will be addressed later in this section. The thermal boundary conductivity $h_{\rm Bd}$ is defined as the ratio of heat flow per unit area A across the interface to the temperature discontinuity ΔT at the interface:

$$h_{\rm Bd} = Q / (A \Delta T) . \tag{2.2}$$

The calculation of the thermal boundary conductivity is very similar to that of the thermal conductivity; the thermal boundary conductivity is determined by the number of carriers (phonons) incident on the interface, the energy carried by each phonon, and the probability that each phonon is transmitted across the interface. For the thermal conductivity, the hard part is to determine the mean free path, whereas for the thermal boundary conductivity the hard part is to determine the transmission probability.

We have been careful to use the term conductivity, because the thermal boundary conductivity is geometry independent; doubling the area of the interface doubles the thermal boundary conductance, not the thermal boundary conductivity. Typically, the quantity used to describe the thermal transport across an interface is called a thermal boundary resistance (not a thermal boundary resistivity) and is defined as the inverse of the thermal boundary conductivity. For historical reasons, the word resistance is used because resistivity usually describes a bulk property. Nevertheless, we believe that the term resistivity would be more appropriate because the thermal boundary resistance is a geometry-independent property. As the terminology is generally accepted, we shall not attempt to change it.

There is another reason why we chose to introduce the thermal boundary conductivity first and not its inverse. Even interfaces between two identical materials will, given the above definitions, have a finite thermal boundary conductivity and therefore a nonzero thermal boundary resistance, even though there is nothing *resistive* occurring at the interface. This can be understood by considering the blackbody radiation analog shown in Fig. 7. Two black cavities are connected by a small hole. One cavity is held at a temperature T_2 and the other at a lower temperature T_1 . We know that the heat flow between them is given by

$$\dot{Q} = \sigma A (T_2^4 - T_1^4) , \qquad (2.3)$$



FIG. 7. Blackbody picture to demonstrate that the thermal boundary conductance is finite even in the absence of any resistive mechanisms. Two black cavities, held at different temperatures T_1 and T_2 , are connected via a small aperture with area *A*. All phonons incident on the aperture transmit across the aperture. The energy flux across the aperture is finite and given by $4A\sigma T^3\Delta T$, for small temperature differences $\Delta T = T_2 - T_1$. σ is the Stephan Boltzmann constant.

where σ is the Stefan Boltzmann constant and A is the area of the aperture. The hole has no *resistance*, yet there is a finite flow of energy across it, determined by the Planck density of photons at a given temperature. Thus the fact that the conductance through the hole is finite is not surprising; and that the resistance is not zero, although seemingly counterintuitive, follows from its definition as the inverse of the conductance. Conceptually, then, it is the thermal boundary conductivity which is the fundamental quantity and which is most easily understood.

In this example, there is no interface except the imagined one between the two cavities, and there is no temperature discontinuity at that interface. However, the photons incident on the interface from above have a frequency distribution characteristic of the temperature T_1 , and the photons incident on the interface from below have a frequency distribution characteristic of the temperature T_2 . The temperature difference used in the definition of the thermal boundary conductivity (or resistance) is that between the photons (phonons) incident on the respective sides of the interface.

The physics of phonon transport in bulk matter at low temperatures, in the absence of scattering in the bulk, is virtually identical to the physics of the blackbody photon radiation. In fact, one can define an acoustic analog of the Stefan Boltzmann constant σ_{phonon} . At low temperatures, in isotropic condensed matter,

$$\sigma_{\rm phonon} = \frac{\pi^2 k_B^4}{120\hbar^3} \sum_i \frac{1}{c_i^2} \ . \tag{2.4}$$

Here, k_B is Boltzmann's constant, \hbar is Planck's constant divided by 2π , and c_i are the speeds of sound, one longitudinal and two transverse.

The calculation of the heat transport between two solids is complicated by the solids' having a different σ_{phonon} ; furthermore, the transmission probability will generally depend on the side from which the phonon hits the interface, the angle of incidence, the phonon frequency, the phonon polarization (longitudinal or transverse), and the temperature of both sides of the interface. If the transmission probability were to depend only on the side from which the phonon was incident, then it would be easy to write down the net heat transport \dot{Q} ,

$$\dot{Q} = A \left(\sigma_2 T_2^4 \alpha_{2 \to 1} - \sigma_1 T_1^4 \alpha_{1 \to 2} \right) , \qquad (2.5)$$

where $\alpha_{1\rightarrow 2}$ is the transmission probability from side 1 to side 2, and σ_i is the phonon Stefan Boltzmann constant of side *i*. From this simple (and unrealistic) expression, using the fact that there can be no net heat flow if the temperatures of the two sides are equal (the second law of thermodynamics), it can be seen that $\sigma_1 \alpha_{1 \rightarrow 2} = \sigma_2 \alpha_{2 \rightarrow 1}$. That is, in thermal equilibrium the number of phonons leaving one side is the same as the number of phonons returning from the other side. This result is still true if the unrealistic assumption is removed, but generally even more can be said: in thermal equilibrium, the number of phonons of a given phonon state (polarization and wave vector) leaving one side is the same as the number of phonons returning from the other side into that state. This is simply a statement of the principle of detailed balance. That principle is a very powerful tool and can be used to simplify transport calculations significantly, including the calculation of the thermal boundary resistance.

Using that principle, it suffices to calculate the gross heat transfer from one side of the interface at the two incident phonon temperatures and to subtract to find the net heat transfer.⁶ The heat transport from the other side of the interface need never be considered, and the second law of thermodynamics and the law of detailed balance are automatically satisfied. In nearly any experiment, the net heat flow across the interface is many orders of magnitude smaller than the gross heat flow. It is thus justified to use the equilibrium phonon density and the principle of detailed balance.

The fact that the thermal boundary resistance is nonzero for the interface between identical materials, or for that matter even for imagined interfaces, has led to some controversy and misunderstanding. For example, it might be asked whether, if one imaginary interface has a nonzero thermal boundary resistance, then must not all matter, which can be thought of as having an infinity of

⁶We assume for purposes of this discussion that there are no anharmonic effects (temperature dependence and not just frequency dependence). Otherwise, although the second law and the law of detailed balance are still true, the gross flow from one side will depend on the temperature and dynamics of the other side, and the calculation does not simplify.

imagined interfaces, have infinite thermal resistance?⁷ This absurd query serves to point out the subtle nature of the definitions. The fundamental property of an interface is the thermal boundary conductivity, which is defined, as above, as the ratio of the heat flux across an interface per unit area to the temperature difference between the distributions of phonons *incident* on the two sides of an interface.

While the problem of merely defining the temperature on either side of an interface is nontrivial, the problem of experimentally measuring that temperature without the thermometer's affecting that temperature is even more challenging. Consider the blackbody analog again, but separate the two cavities with a mirrored tube, as in Fig. 8. Consider an imagined interface at the center of the tube. The photons traveling in an upward direction have a Planck distribution with characteristic temperature of the lower cavity, and photons traveling downward have a distribution with the temperature of the upper cavity. The mirrored tube does not affect these distributions; therefore, the two temperatures in the definition of the thermal boundary conductivity at the imagined interface are the temperatures of the two blackbody cavities. These two temperatures are by no means the temperatures that would be measured by infinitesimally sized thermometers placed inside the tube on the two sides of the interface (see Fig. 8). In fact, both of those thermometers would measure the same temperature, namely, the fourth root of the average of the fourth power of the temperatures of the two cavities:

$$T_{\text{ideal thermometer}} = \left[\frac{1}{2}(T_1^4 + T_2^4)\right]^{1/4}.$$
 (2.6)

This follows from calculating the temperature at which the thermometer must equilibrate so that the net radiation from the thermometer to the colder end equals the net radiation from the hotter end to the thermometer. The calculation is independent of the position of the thermometer in the tube. Therefore, even in the presence of a heat flux, two infinitesimal thermometers on opposite sides of the imagined interface would read *identical* temperatures. Using these thermometers, then, one would calculate an infinite thermal boundary conductivity for the interface, and thus a vanishing thermal boundary resistance. The problem with these thermometers is that they do not discriminate between photons traveling upward and photons traveling downward.



FIG. 8. Blackbody cavities separated by a perfectly specular tube. The placement of the thermometers shown is faulty; ideal thermometers would measure a zero ΔT . If the ideal thermometers in the tube are replaced with real thermometers, then the real thermometers will measure a nonzero ΔT , depending only on the extent of the photon scattering at the thermometers. If water is added to the lower cavity, so that the water level is between the thermometers, then the ΔT measured depends on both the transmission coefficients and the geometry of the thermometers; the thermal boundary resistance could not be correctly deduced. If the thermometers were placed inside the cavities in the positions outlined, even if the thermometers were not ideal, the correct ΔT would be measured in all cases.

If the lower cavity were filled with water to a level between the two infinitesimal thermometers of Fig. 8, there would be a temperature difference measured between them, and that temperature difference would be due entirely to the reflection of photons at the interface. Simons (1974) calculated the temperature that would be measured by such infinitesimal thermometers adjacent to an interface between two solids, for the phonon analog of this picture. He used the same assumptions concerning the phonon transmission properties at the interface as in the acoustic mismatch model. The integrals written down and estimated by Simons were later evaluated numerically for several interfaces by Katerburg, Reynolds, and Anderson (1977). Phillips and Sheard (1976) used path-integral techniques to solve the Boltzmann equation for the phonon transport at interfaces, again attempting to fix the "problems" associated with the prediction of a nonzero temperature discontinuity at an interface between identical solids. Only a one-dimensional model was solved, due to the complexity caused by the formulation of the problem in terms of adjacent infinitesimal thermometers. This further justifies using the temperatures corresponding to the distributions of the incident phonons; apparently, for no other definition is the calculation of a thermal boundary resistance reasonably tractable.

To illustrate the experimental subtleties of ther-

⁷The fact that thermal boundary resistances do not simply add is not just a consequence of the definition. In general, the way to calculate a thermal resistance is to assume a temperature difference at the ends of the system in question and then to calculate the heat flux. Consider a sandwich of one material between two identical materials. An incident phonon will set up standing waves between the two interfaces, and the transmission will depend on the ratio of the film thickness to the phonon wavelength. Thus for *no* definition of a thermal boundary resistance do series thermal resistances add.

mometry in systems with ballistic carriers, consider what happens if the infinitesimal thermometers in Fig. 8 on each side of the imagined interface are replaced by real thermometers with nonzero size. The thermometers affect the measurement by scattering the photons (phonons), and thus the measured thermal boundary conductivity depends on the details of the thermometers. (This is the same subtlety as in the measurement of thermal conductivity in the boundary scattering regime.) Now, if the lower cavity in this picture is filled with water to a level between the thermometers, the measured temperature difference between the two thermometers will be due to the scattering from the thermometers as well as the reflection from the interface; the transmission probabilities at the interface are therefore nearly impossible to deduce. Consider the alternate placement of the thermometers in the outlined positions inside the cavities in Fig. 8. Thermometers in these positions measure exactly the temperatures of the cavities without affecting those temperatures, largely independent of the size or geometry of the thermometers. Thus, for this geometry, there exists an experimentally simple way to measure the appropriate temperatures (the temperatures of the incident phonons) without the measurements' affecting those temperatures. A similar discussion of the effects of thermometry on the interpretation of thermal boundary resistance data and on the correspondence between the calculations [specifically the calculation by Simons (1974) versus the acoustic mismatch calculation] and the measurements is given by Katerberg, Reynolds, and Anderson (1977).

In a real blackbody experiment, the mirrored tube is never perfect; scattering from the surface of the tube will increase the temperature difference between the black bodies, just as a radiation shield would. In phonon systems, the analog of the mirrored wall is a crystal surface or a surface of the container which confines helium. In addition, phonons (especially those characteristic of temperatures above a few kelvin) can scatter in the interior of the crystals and the helium. Scattering in the bulk and at the walls can affect the thermal boundary resistance measurement very significantly; the farther the thermometers are from the interface, the greater is the effect of this scattering on the measurement. There seem to be mutually conflicting requirements: (1) the thermometers must be placed where they measure the temperature of the distribution of phonons incident on the interface without affecting this distribution, and (2) the thermometers must be placed as close to the interface as possible in order to minimize the effects of unwanted phonon scattering. Thus the geometry used to measure a thermal boundary resistance is an important factor; an inappropriate geometry will produce a boundary resistance that does not correspond to the accepted definition, or that has been affected by thermal resistances in series with the thermal boundary resistance. Below we shall consider the possible geometries that satisfy these requirements. We shall start by comparing the thermal boundary resistance to the thermal resistance of bulk solids and liquids, with the motivation that we must determine how close to the interface the thermometers must be positioned.

Consider the magnitude of the thermal boundary resistance relative to the thermal resistance per unit area of a length of bulk material. For an imagined interface in a dielectric, the transmission probability is unity, and the thermal boundary resistance is exactly equal to the thermal resistance per unit area of a length of the dielectric equal to one phonon mean free path (Little, 1959).⁸ Similarly, at a real interface between two solid dielectrics, for which the mean transmission probability is on the order of unity, the thermal boundary resistance is of the order of the thermal resistance of a mean-free-path-long section of either dielectric; this case is simply not very different from the case of the imagined interface. Another way to understand the comparison is that, for boundaries to dielectrics, the ratio of the thermal boundary resistance to the thermal resistance of a mean-free-pathlong section of dielectric is the inverse of the average phonon transmission probability from the dielectric side. At boundaries to superfluid helium, since the phonon transmission probability from the helium into a solid is several orders of magnitude less than 1, the length in the helium with thermal resistance comparable to the thermal boundary resistance is several orders of magnitude greater than the phonon mean free path in the helium.⁹ In addition, the predicted transmission probability for phonons at helium-solid interfaces is much less than unity, even from the solid side. At a boundary between a dielectric crystal and helium, when the transmission probability is indeed small, even in solid dielectrics the length with thermal resistance comparable to the thermal boundary resistance is large compared to the phonon mean free path in the dielectric. For temperatures under a few kelvin, if one side of the interface is a pure, wellannealed, crystalline metal (not superconducting), then, given the high thermal conductivity of such metals (for example, copper), the length in the metal with a thermal resistance per unit area comparable to the thermal boundary resistance is also large.

Given the comparison of the thermal boundary resistance with the resistance of a given length of bulk materi-

⁸This can be understood by considering the blackbody analogy. The temperature difference is that between the cavities; the phonons (photons) emitted by the upper cavity are thermalized in the lower cavity. All phonons contributing to the heat flux are thermalized precisely once in the problem. Thus the resistance is thermally equivalent to that of a section of bulk material in which the phonons scatter, on average, precisely once, i.e., to a section of bulk with length equal to one mean free path.

⁹The phonon mean free path in ³He, however, is extremely short at temperatures above ~ 0.2 K; therefore, the length in ³He with thermal resistance comparable to the thermal boundary resistance is not long.

al, we can start to discuss the placement of thermometers. The criteria for the thermometers are as follows: The thermometers must measure the temperature characteristic of the phonons *incident* on the interface without affecting these temperatures, and the thermometers must be placed close to the interface relative to the length in the bulk having a thermal resistance equivalent to the boundary resistance.

In pure metals, because electrons dominate the heat transport and strongly scatter the phonons, the phonon distribution is isotropic; the total phonon distribution and the distribution of phonons incident on the interface have the same temperature. Since electrons dominate the thermal transport, phonon scattering at the thermometer will not affect the temperature measurement. Thus, for pure metals, there are no obvious problems with thermometry, at least at temperatures below a few K; the position of the thermometer on the metal is not critical.¹⁰ In superfluid helium, although the thermometers may affect the measurement as much as a mean-free-path long section of dielectric, that resistance is small compared to the thermal boundary resistance. The position of a thermometer in the superfluid is also not critical.

In boundary resistance measurements, the positioning of thermometers usually is critical if one side of the interface is a dielectric. At temperatures below 1 K, in dielectric crystals, the mean free path is often determined by the condition of the surfaces and the size and positions of thermometers on the crystal, yet the thermometers must be placed within a phonon mean-free-path length of the interface. If the thermometers scatter phonons so strongly that they strongly influence the mean free path, there may be no reasonable place to put the thermometers. If the average phonon transmission probability from the dielectric is small, as is sometimes the case for boundaries to helium, then the problem is alleviated. We have implicitly assumed that the phonon mean free path is large in the dielectric (for example, on the order of the width of the sample). When that is not the case, for example, if the dielectric is amorphous or at temperatures above 1 K even in crystals, the length of dielectric with thermal resistance equivalent to the thermal boundary resistance will diminish. The same problem arises for any interface above a few kelvin, because the thermal resistivity per unit length of the bulk material increases rapidly with temperature, and the boundary resistance decreases rapidly with temperature. If the material is noncrystalline, a disordered, impure metal, or a superconductor, then the bulk thermal resistance per unit length can be large compared to the thermal boundary resistance even below 1 K. For these cases, the positioning of the thermometer so that the above two criteria are satisfied becomes rather difficult. Often complicated subtractions of bulk thermal gradients from the measured ΔT between the thermometers become necessary.

A conventional geometry used for measuring a thermal boundary resistance is shown in Fig. 9. If one side of the



FIG. 9. Conventional geometry for measuring the thermal boundary resistance. If both sides are dielectric crystals, then this geometry is precisely analogous to the geometry of Fig. 8, with the thermometers placed inside the tube. If one side is metallic, then the electronic thermal transport causes the phonon distribution to be much more isotropic, and only the thermometer on that side measures the temperature that corresponds to the accepted definition. In either case, the measured ΔT does not correspond to the temperature drop used in the definition of the thermal boundary resistance.

¹⁰Unfortunately, a complication can arise due to the transition from phonon-dominated heat transfer across the interface to the electron-dominated heat transfer in the bulk metal; the capacity for transport between the two systems of carriers is finite. At low enough temperatures, a thermal bottleneck arises due to this finite electron-phonon thermal transport. Although we shall not further address electron-phonon thermal transport in this review, the problem can strongly affect or even dominate measurements of thermal boundary resistance in certain regimes (usually at temperatures well below 1 K and/or in very small structures, such as thin films), and its understanding, at least at a phenomenological level, is critical in order to avoid or account for such effects. For more information, see, for example, Anderson and Peterson (1972), Perrin and Budd (1972a, 1972b), Perrin (1975, 1976), Murmann and Heber (1977), Roukes (1985), Roukes, et al. (1985), Yoo and Anderson (1986), and Swartz (1987), and references therein.

interface is a pure, well-annealed metal, then, for temperatures below a few K, the placement of the thermometer on that side of the interface can be accomplished without introducing error, as explained above. On the dielectric side of the interface it is essential to put the thermometer well within a phonon mean-free-path length of the interface, in order to eliminate a complicated subtraction of the temperature gradient. However, in that case scattering at the thermometer itself influences the mean free path. Even worse, the thermometer does not measure the distribution of phonons incident on the interface, but a combination of the distributions of phonons incident on the interface and of phonons coming from the interface. That is not the appropriate phonon distribution. As a result, the experimenter will measure a thermal resistance smaller than the actual thermal boundary resistance.

As an example, consider the analysis of the thermal boundary resistance between helium II and sapphire. If we assume that the mean phonon transmission probability from the sapphire side is correctly predicted using the acoustic mismatch model, then the positions of the thermometers are not critical. A problem with this analysis arises if the thermal boundary resistance is not as high as that predicted using the acoustic mismatch model, i.e., if the mean transmission probability is not small, but is on the order of unity. Then the position of the thermometer on the sapphire is absolutely critical, as the thermometer impedes phonons about as much as the interface in this case, and because the thermometer measures not the temperature of the incident phonons but that of a combination of the incident phonons from the sapphire and the transmitted phonons from the helium. These subtleties were overlooked in the analyses of the Kapitza resistance data for sapphire (Gittleman and Bozowski, 1962) and for LiF (Johnson and Little, 1963). It was concluded that the mean transmission probability was nearly 100% for phonons in these crystals (Cheeke, 1970b). We estimate that, with a proper analysis of the experiment, the mean transmission probability from the sapphire or the LiF into the superfluid helium was closer to 70%, and not the reported value of nearly 100%.

A geometry that can be used to measure the thermal boundary resistance between a metal and a crystalline dielectric, which does not suffer from these complexities, is shown schematically in Fig. 10(a). Owing solely to its position, the thermometer on the dielectric side of the interface measures the temperature of the phonons incident on the interface from the dielectric side. Phonons from the interface do not affect the thermometer because those phonons cannot "see" the thermometer. A variation of that geometry for two dielectrics is shown in Fig. 10(b). However, for these geometries, there are still two problems: (a) producing interfaces between bulk solids for which there is intimate contact over the entire area is extremely difficult, and (b) these geometries would not be useful for temperatures exceeding a few K.

A practical design for the measurement of the thermal boundary resistance at a solid-solid interface which in-



FIG. 10. Experimental geometries for measuring thermal boundary resistance. (a) Between a metal and a crystalline dielectric. The thermometer on the dielectric side measures a temperature corresponding to the distribution of phonons incident on the interface. The phonon distribution on the metallic side is nearly isotropic if the metallic thermal conductivity is sufficiently high, and thus the thermometer on the metallic side measures the same temperature as the temperature of the phonons incident from the metallic side. (b) Between two dielectrics, or across a dielectric-metal-dielectric sandwich.

corporates these considerations is shown in Fig. 11 (Swartz and Pohl, 1986, 1987). Two closely spaced (~ 2 μ m) metal strips with widths $\sim 2 \mu$ m are vapor deposited onto a dielectric substrate. The temperature dependence of the electrical resistance of each metal strip allows the use of each strip as a thermometer. The interface studied is that between one metal strip and the substrate underneath it. Through that strip, a heater current I is passed. That current is also used to measure the resistance R and thus the temperature of the strip; the heat developed is $Q = I^2 R$. The thermal boundary resistance between the Joule-heated thermometer and the underlying substrate is given by the ratio of the temperature difference ΔT between the strip and the substrate, and the power flowing across the interface divided by the area of contact A. (A is the product of the length and the width of the metal strip.) The temperature of the dielectric substrate under the heated strip, needed to obtain ΔT , is determined by measuring the temperature of the second thermometer using a much smaller sensing current i, to avoid selfheating.

In the geometry of Fig. 11, the temperatures of the phonon distributions incident on the two sides of the interface are measured, as required for the measurement to correspond to the definitions (cf. Figs. 7-10). The temperature of the distribution of phonons incident from the metallic side of the interface is the same temperature as that of the electrons in the film [see, however, the discussion of electron-phonon coupling in Swartz (1987)]. This temperature is uniform in the film due both to the high electronic thermal conductivity in the film and to its thinness (about 0.5 μ m). Because of the proximity of the two metal strips, the temperature of the phonons incident on the heater film is the same as that of the phonons incident on the thermometer film, at least at low temperatures (in sapphire below 40 K), where the phonon mean free path in the substrate is much larger than the ther-



FIG. 11. Experimental geometry for measuring the thermal boundary resistance between vapor-deposited thin metal films and the underlying dielectric substrate over a broad temperature range. For adequate sensitivity, it was found that the thermometers had to satisfy $d \ln R / d \ln T > 0.1$ (Swartz, 1987), which for very pure metals holds only above about 10 K. For the alloy Rh_{99.5}Fe_{0.5}, on the other hand, this temperature sensitivity requirement is satisfied to well below 1 K (Rusby, 1975).

mometer spacing. At temperatures above 50 K, the difference between the temperature of the second thermometer and the temperature of the substrate underneath the first thermometer must be calculated by integrating the Laplace equation (time-independent heat equation). That temperature difference is small due to the close proximity of the thermometers and the large phonon mean free path in the substrate.

The small dimensions of the design have one further advantage: above a few K, the thermal boundary resistance is rather small and dropping as T^{-3} . Therefore, in order to generate a measurable temperature discontinuity at an interface, a large heat flux per unit area (thousands of watts per cm² at 100 K) is required. Such heat fluxes, if maintained for any significant period of time, inevitably induce large temperature gradients in bulk materials. If the area of the interface is a significant fraction of a cm², then the total heat required is large enough to overwhelm most cryogenic systems. The way to avoid the large power requirements is to reduce the area of the interface.

B. General considerations

We shall consider only the part of the thermal transport across the interface that is due exclusively to phonons. A phonon incident on the interface has only two options: either it is transmitted or it is not. Classically (or statistically) some fraction of the phonon energy is transmitted; the transmission probability for phonons will be called α . It will depend in general on the phonon mode, wave vector, and frequency, and on the temperature. For simplicity, we shall assume both sides of the interface to be isotropic and the transmission probability to be independent of temperature.¹¹ The assumption of isotropy allows us to write α as a function of the phonon frequency ω , the angle θ between the phonon propagation direction and the normal to the interface, and the phonon mode *j*. With the assumption that the transmission probabilities are independent of the temperature on either side of the interface (and therefore are independent of the presence of other phonons), we close our eyes to the possibility of anharmonic interactions. The advantage of this last assumption was stated in the last section, namely, that the transmission probabilities from only one side of the interface need to be calculated. That is, the net heat flow from side 1 at temperature T_1 to side 2 at temperature T_2 is the difference between the gross heat flow from side 1 to side 2, when side 1 has temperature T_1 , and the gross heat flow from side 1 to side 2 when side 1 has temperature T_2 . Side 2 then need never be considered. For Kapitza boundaries, if we choose the helium to be on side 2, then the calculation looks much like that for solid-solid interfaces.

¹¹These assumptions are not essential, but a more general discussion in which the assumptions are relaxed would be unnecessarily complicated for this review.

The gross heat current density from side 1 to side 2, $(1/A)\dot{Q}_{1\rightarrow 2}^{\text{gross}}(T)$, is the sum over all frequencies and incident angles of the number of phonons with given frequency and incident angles θ and φ that are incident on a unit area A per unit time, times the phonon energy times the transmission probability $\hbar\omega = \hbar c_{1,j} k,$ $\alpha_{1 \rightarrow 2}(\theta, j, \omega)$. Here j is the phonon mode, k is the phonon wave vector, $c_{1,i}$ is the phonon propagation velocity in side 1 for phonons with mode *j*, and ω is the phonon frequency. The angles of incidence are φ (azimuthal) and θ (angle between the wave vector of the incident phonon and the normal to the interface). Let $c_{1,i}\cos\theta$ be the normal component of velocity, $d\Omega = d\varphi \sin\theta d\theta$, and $N_{1,i}(\omega, T)$ be the product of the density of phonon states with given angles φ and θ , times the Bose occupation factor. Then the number of phonons with given frequency and angles of incidence that are incident on the surface area A per unit time is

$$\frac{N_{1,j}(\omega,T)}{4\pi}d\Omega c_{1,j}\cos\theta$$

When this product is integrated over all angles, the result is $c_{1,j}N_{1,j}(\omega,T)$.

Therefore the gross heat current can be written as

$$\frac{\dot{Q} \,_{1 \to 2}^{\text{gross}}(T)}{A} = \frac{1}{2} \sum_{j} \int_{0}^{\pi/2} \int_{0}^{\omega_{1}^{\text{max}}} N_{1,j}(\omega, T) \hbar \omega c_{1,j}$$

$$\times \alpha_{1 \to 2}(\theta, j, \omega)$$

$$\times \cos\theta \sin\theta \, d\theta \, d\omega \,, \qquad (2.7)$$

(The integral over azimuthal angles contributed 2π .) Here, ω_1^{max} is the maximum phonon frequency on side 1. For small $(T_2 - T_1)/T_2$, the thermal boundary conductivity can be written as

$$h_{\rm Bd} = \frac{\dot{Q}_{1 \to 2}^{\rm gross}(T_2) - \dot{Q}_{1 \to 2}^{\rm gross}(T_1)}{A(T_2 - T_1)} \ . \tag{2.8}$$

The problem is in principle solved if the transmission probabilities are known.

C. The acoustic mismatch model

Calculating transmission probabilities between two solids is not a simple problem. In the acoustic mismatch model the only essential simplifying assumption is that the phonons are governed by continuum acoustics and the interface is treated as a plane. That is, phonons are treated as plane waves, and the materials in which the phonons propagate are treated as continua (no lattice). For phonons with wavelength much greater than typical interatomic spacings, this continuum approximation might be expected to be accurate. Given this (very strong) assumption, there are only a few results possible when a phonon is incident on the interface; the phonon can specularly reflect, reflect and mode convert, refract, or refract and mode convert. (That is, the final state is a superposition of these possibilities. The transmission probability is the total fraction of the energy transmitted across the interface.) Figure 12 shows these possibilities. The angles of reflection or refraction, with or without mode conversion, as well as the probabilities of each, are determined by the acoustic analog of Snell's law for electromagnetic waves. Consider first a liquid-helium-solid interface. Suppose a longitudinal phonon inside the helium is incident on the interface, at an angle θ_{in} from the normal to the interface, as in Fig. 13. If the result is a transmitted longitudinal phonon in the solid, then we can calculate the angle of transmission θ_{tran} using Snell's law:

$$\sin\theta_{\rm tran} = \frac{c_l^{\rm solid}}{c_l^{\rm helium}} \sin\theta_{\rm in} \; .$$

An analogous relationship holds if the transmitted phonon has a transverse polarization (i.e., the phonon is mode converted on transmission), with the longitudinal phonon velocity c_l^{solid} replaced by the transverse phonon velocity c_t^{solid} . The transmitted angle cannot exceed 90°. Thus, for incident angles greater than the angles where $\sin\theta_{\text{in}}$ equals $(c_l^{\text{helium}}/c_l^{\text{solid}})$ or $(c_l^{\text{helium}}/c_t^{\text{solid}})$, the probability of transmission to a longitudinal or transverse phonon vanishes, respectively. These are called the critical angles. The set of all angles less than the largest critical angle is called the critical cone. Only phonons incident from within this cone have a chance to be transmitted. On the side with greater phonon velocities, there is no critical cone; phonons with any incident angle have some



FIG. 12. Schematic of the many possibilities within the framework of the acoustic mismatch model for phonons incident on an interface. The picture simplifies if one of the sides is liquid helium; there are no transverse modes on that side.



FIG. 13. Incident and transmitted phonon angles, related according to Snell's law. If the velocities and angles of the incident and transmitted phonons are $c_{\rm in}$, $\theta_{\rm in}$, $c_{\rm tran}$, and $\theta_{\rm tran}$, respectively, then $\sin\theta_{\rm in}/c_{\rm in} = \sin\theta_{\rm tran}/c_{\rm tran}$.

chance for transmission. If a phonon with incident angle θ_{in} transmits to a phonon with transmitted angle θ_{tran} with probability α , then a phonon from the other side incident on the interface with incident angle θ_{tran} transmits with the same probability α back into a phonon with transmitted angle θ_{in} . This follows from Snell's law and is a consequence of the general principle of detailed balance. At a helium-solid interface, the critical angles (in the helium) are on the order of 5°. (See, for example, Haug and Weiss, 1972, or Peterson and Anderson, 1973.)

At solid-solid interfaces the critical angles are much larger and there are several additional critical angles corresponding to incident slow and fast transverse phonons transmitted with or without being mode converted. Apart from that, the calculation is the same.

Given the assumption that continuum acoustics governs the phonon system, the transmission probabilities are determined by the acoustic analog of the Fresnel equations. At a fundamental level, these are derivable (using continuum acoustics) from a set of boundary conditions. The normal component of displacement must be continuous at the interface; otherwise the interface would separate. The tangential component of displacement must also be continuous; otherwise the interface would slide. The "normal component" of the stress tensor (i.e., the contraction of the stress tensor with the unit vector normal to the surface) is a vector that is the force on the surface. (This force is not necessarily in the normal direction.) It is this force that is continuous across the interface. The proof is straightforward, given the formalisms of stress and strain tensors (see Auld, 1973). The intuitive explanation is that, if the force on the surface is not continuous, then the interface, having zero mass, would have infinite acceleration.¹²

These boundary conditions (which follow from continuum acoustics) have very important consequences; in particular, the possibility of scattering at the interface, whether elastic or inelastic, is implicitly ignored. Given the assumption that continuum acoustics is applicable, the transmission probabilities can (with effort) be calculated for any incident angle and mode. Once the transmission probabilities are known, the sum over phonon states must be taken to calculate the gross heat transport [Eq. (2.7)]. Transmission of phonons across the interfaces is much more complicated than transmission of signals in transmission lines because the phonon transmission probability depends on angles of incidence, because solids are anisotropic, because there are three phonon modes, etc. The simplest picture derivable from the acoustic mismatch model is that each material can be ascribed an acoustic impedance equal to the product $Z_i = \rho_i c_i$ of the mass density and the phonon velocity. The formula for the transmission probability from side i = 1 to side 2 can be made to look like the formula for transmission at a junction in a transmission line; for a phonon with normal incidence, for example, the energy transmission probability looks like

$$\alpha_{1 \to 2} = \frac{4Z_2 Z_1}{(Z_1 + Z_2)^2} \tag{2.9}$$

[see Little, 1959, Eq. (4), for example].

Several additional yet inessential assumptions are usually also made in order to simplify the calculations: Solids are assumed to be *isotropic Debye solids*, with the generalization that the longitudinal and transverse speeds of sound are different. Then, for frequencies below the Debye cutoff frequencies, ω_i^{Debye} ,

$$N_{1,j}^{\text{Debye}}(\omega,T)d\omega = \frac{\omega^2 d\omega}{2\pi^2 c_{1,j}^3 [\exp(\hbar\omega/k_B T) - 1]}$$

With

$$\Gamma_{1,j} = \int_0^{\pi/2} \alpha_{1 \to 2}(\theta, j) \cos\theta \sin\theta \, d\theta \,, \qquad (2.10)$$

Eqs. (2.7) and (2.8) can be rewritten as

$$h_{\rm Bd} = \frac{1}{2} \sum_{j} c_{1,j} \Gamma_{1,j} \int_{0}^{\omega_1^{\rm Debye}} \hbar \omega \frac{dN_{1,j}(\omega,T)}{dT} d\omega \ . \tag{2.11}$$

At low temperatures, the usual approximation of setting the upper limit in the frequency integral to infinity is made, and the integral can be done. [The integral turns out to be the same one as in the calculation of the Debye heat capacity (Debye, 1912).] The answer is

¹²The intuitive explanation of the continuity of the force on the interface was pointed out to us by one of the referees.

$$R_{Bd} = \left[\frac{\pi^2}{15} \frac{k_B^4}{\hbar^3} \left[\sum_{j} c_{1,j}^{-2} \Gamma_{1,j}\right]\right]^{-1} T^{-3}$$

= $\left[2.04 \times 10^{10} \left[\sum_{j} c_{1,j}^{-2} \Gamma_{1,j}\right]\right]^{-1}$
 $\times T^{-3} \left[\frac{\sec^2}{\operatorname{cm}^2} \mathrm{K}^3 \frac{\mathrm{K}}{\mathrm{W/cm}^2}\right].$ (2.12)

The Γ 's have been calculated numerically and are available in the form of tables (Cheeke, Ettinger, and Hebral, 1976) and in the form of a computer (FORTRAN) program (Peterson, 1973).

 $R_{\rm Bd}$ can be approximated as $(\frac{1}{4}Cc\alpha)^{-1}$, where C is the Debye specific heat, c is the Debye phonon velocity, and α is an appropriately averaged transmission probability. As a special case of Eq. (2.12), consider what happens if both sides of the interface are identical. Then the α 's are all unity and the Γ 's are all 0.5. Thus,

$$R_{Bd} = \left[1.02 \times 10^{10} \left[\sum_{j} c_{1,j}^{-2}\right]\right]^{-1}$$
$$\times T^{-3} \left[\frac{\sec^2}{\mathrm{cm}^2} \mathrm{K}^3\right] \frac{\mathrm{K}}{\mathrm{W/cm}^2} .$$

We see, as noted earlier, that the boundary resistance does not vanish at imagined interfaces.

Another result of the above analysis is that the transmission probabilities are independent of phonon frequency for phonons with frequency less than the Debye cutoff of the material with lower Debye cutoff; above the lower Debye cutoff frequency, the phonon transmission probabilities are zero. This analysis ignores the effects of elastic anisotropy and phonon dispersion. A more general treatment of phonon transport across solid-solid interfaces, including the effects of elastic anisotropy and phonon dispersion within the acoustic mismatch model, has been carried out by Weiss (1986). Young and Maris (1986, 1988) have calculated the thermal boundary resistance for a few specific solid-solid interfaces, using the measured (or calculated) phonon densities of states and phonon velocities.

1. A historical note

The approach taken in the above derivation is essentially that of Mazo¹³ (1955) for the Kapitza problem and of Little for the solid-solid problem. Khalatnikov (1965) performed the calculation from the helium side, which, if only the Kapitza problem is considered, is a clever shortcut: From the helium, there is only one mode, the longitudinal mode, so only one transmission probability must be calculated, ¹⁴ and from that, only one Γ must be calculated. In Khalatnikov's (1952) original work, he did the zeroth-order approximation to the above result. He calculated the normal component of vibration of a free solid surface in equilibrium at temperature T_1 , due to phonons' being reflected internally from that free surface. Then he calculated the amount of energy that these vibrations would couple into the helium. Since that energy is small, the error in initially assuming a free surface is small, and the answer is about the same as that of the above calculation.

2. A note on ³He

We have not concerned ourselves much with the complications introduced when calculating the thermal boundary resistance to 3 He. At temperatures above a few hundred mK, the thermal conductivity of ³He is quite low, and the thermal boundary resistance is thus difficult to measure. Below a few hundred mK, ³He is a Fermi liquid. Instead of phonons, the excitations in the ³He are quantum excitations of the Fermi liquid, including single-particle excitations called quasiparticles and collective excitations called zero sound. Zero-sound excitations act in some ways as phonons do (and can be of longitudinal or transverse nature). Semiclassical thermal boundary resistance calculations, in many ways analogous to acoustic mismatch model calculations, have been performed, for example, by Bekarevich and Khalatnikov (1960, 1961) and Gavoret (1965) and are based on Fermiliquid theory (Abrikosov and Khalatnikov, 1958). A fully quantum-mechanical calculation using a perturbationtheory approach has been carried out by Toombs, Sheard, and Rice (1980); it is based on Fermi-liquid theory and on the earlier perturbation-theory calculations of the ³He Kapitza resistance by Rice (1971), Sheard, Toombs, and Challis (1971), Rice, Sheard, and Toombs (1972), and Rice and Toombs (1972).

D. The diffuse mismatch model

1. Qualitative arguments

A crucial assumption made in the acoustic mismatch model is that no scattering occurs at the interface. However, reflection measurements of heat pulses from free solid surfaces have shown that they scatter highfrequency phonons ($\gtrsim 100$ GHz) unless they are cleaved *in situ* or laser annealed (Eisenmenger, 1986). The same scattering occurs at helium-solid interfaces and leads to a significant reduction of the thermal boundary resistance

¹³We should note that Mazo made a mistake in his original 1955 calculation, which resulted in a better agreement with the data (Mazo, 1988; see also Anderson and Johnson, 1972). The mistake did not qualitatively alter the general approach and does not detract from the significance of his efforts.

¹⁴This probability includes contributions from transmission into transverse phonons as well as the contribution from transmission into longitudinal phonons.

by opening up new channels for heat transport, which is discussed in Sec. III.B. The effect of phonon scattering on the thermal boundary resistance has been explored by Swartz (1987), who proposed what he termed the diffuse mismatch model, to be reviewed below.

In the diffuse mismatch model the assumption of complete specularity is replaced with the opposite extreme: *all* the phonons are diffusely scattered at the interface; this leads to an upper limit of the effect that diffuse scattering can have on the boundary resistance. In the diffuse mismatch model, acoustic correlations at interfaces are assumed to be completely destroyed by diffuse scattering, so that the only determinants of the transmission probability are densities of phonon states and the principle of detailed balance. The probability of transmission is nonetheless determined by a mismatch that between densities of states.

The effect on the thermal boundary resistance of diffuse scattering at the interface can be qualitatively understood with the following arguments. We shall assume that scattering destroys the correlation between the wave vectors of incoming and outgoing phonons. Simply put, we assume that a scattered phonon forgets where it came from. The probability that the phonon will scatter into a given side of the interface is then independent of where it came from. Instead, the probability of scattering into a given side is proportional to the density of phonon states on that side (see Fermi's "golden rule") and is also restricted by the principle of detailed balance.

Before presenting the quantitative analysis, let us see how much can be understood using only qualitative arguments. Consider what happens at low temperatures (~ 1 K) at a Kapitza boundary, say, between copper and helium, if we assume the diffuse mismatch model. When a phonon from the copper is incident on the interface it must decide whether to forward scatter or backscatter, and that decision is based only on the relative density of phonon states into which the phonon can scatter. Since there are vastly more phonon states of the initial phonon frequency in the helium,¹⁵ that phonon will almost certainly forward scatter. If we assume the acoustic mismatch model, that same phonon would almost certainly reflect due to the large acoustic mismatch between helium and copper. Thus the effect of the diffuse scattering event is predicted to be large at a Kapitza boundary (on the order of 2 orders of magnitude; see Fig. 1). At a boundary between two solids with *identical* acoustic properties, the transmission probability according to acoustic mismatch theory is unity. The transmission probability in the diffuse mismatch model is exactly 50%. In this case, the boundary resistance is *increased* by

diffuse scattering; specifically, the boundary resistance is doubled. At a more typical solid-solid boundary, the mismatch in the acoustic properties is small (but not negligible); roughly half of the phonons incident on the interface are transmitted according to the acoustic mismatch model. The mismatch in the density of phonon modes for most pairs of solids is also small, and therefore, again, roughly half of the scattered phonons at a solid-solid interface are transmitted. Thus the effect of diffuse scattering at a typical solid-solid boundary is small. For a pair of solids with relatively large differences in their acoustic properties, such as lead and diamond, the effect of diffuse scattering is to decrease the thermal boundary resistance just as it does for Kapitza boundaries (but to a much smaller extent). (A more quantitative analysis follows, in which it will be shown that diffuse scattering at typical solid-solid interfaces usually changes the thermal boundary resistance by less than $\pm 30\%$.) We can summarize these arguments by showing qualitatively what a plot of the effect of diffuse scattering on the thermal boundary resistance might look like. In Fig. 14 we plot the ratio of the (low-temperature) diffuse thermal boundary resistance R_{dm} to the acoustic thermal boundary resistance R_{am} against a unitless parameter which qualitatively represents the amount of dissimilarity between the two materials making up the interface. Think of this parameter as something like $(\rho c)_{max}$ $(\rho c)_{\min} - 1$, where $(\rho c)_{\max}$ is the product of the mass density and the Debye phonon velocity on the side where this product is the greater.¹⁶ Figure 14 shows that diffuse scattering increases the thermal boundary resistance at interfaces between very similar solids, decreases the thermal boundary resistance at interfaces between rather different solids, and greatly decreases the thermal boundary resistance at Kapitza interfaces, for which the dissimilarity is extremely large.

The pressure independence of the Kapitza resistance at temperatures above ~0.3 K can be qualitatively understood from the above simple arguments. For surfaces at which there is significant phonon scattering, the probability of transmission for a phonon incident from the solid is high because of the large density of phonon states in the helium. Although that phonon density of states is somewhat pressure dependent, changing the pressure does not change the fact that the phonon density of states in the helium is much larger than that in the solid. Therefore, independent of pressure, almost all phonons incident from the solid side are transmitted, and the pressure has only a small effect on the Kapitza resistance. We can also see from these arguments that the prediction

¹⁵The density of phonon states is proportional to ω^2/c^3 , where ω is the frequency and c the speed of sound. The speed of sound in helium is on the order of 20 times lower than in most solids. Thus the density of low-frequency phonon states in helium is on the order of 10⁴ as high as that in most solids.

¹⁶Unfortunately, there is no simple parameter that could be used quantitatively in such a plot, because the acoustic and diffuse mismatch models have different dependences on both the mass densities and the phonon velocities. The above formula for the parameter ignores the difference in the mass density dependences of the two models.



FIG. 14. Plot of the ratio of the diffuse mismatch model thermal boundary resistance to the acoustic mismatch model thermal boundary resistance vs the "amount of mismatch." The horizontal scale is qualitative and has arbitrary units; see text. The leftmost dotted line exemplifies a solid-solid boundary with relatively little dissimilarity, such as aluminum on quartz, the middle dotted line exemplifies a solid-solid boundary with large dissimilarity, such as platinum on quartz, and the rightmost dotted line marks the beginning of the region of extremely large dissimilarity, as found in the Kapitza (liquid helium-tosolid) case. The plot serves to summarize qualitatively the results of several calculations, including those in Table II. From Swartz (1987).

of the diffuse mismatch model is insensitive to the specific properties of liquid or solid helium. For that reason, it would not be very useful to refine the calculation of the diffuse mismatch model by using very precise estimates of the helium excitation spectrum. For example, the existence of a large roton density of states in the helium does not significantly change the value of the Kapitza resistance predicted using the diffuse mismatch model. Thus, for the purposes of the diffuse mismatch model, the excitation spectrum can be represented using just the longitudinal speed of sound.

2. Quantitative analysis

The only aspect of the calculation of a thermal boundary resistance using the acoustic mismatch model that must be modified for the diffuse mismatch model is the transmission probability. There are two inputs required to calculate the transmission probability for the diffuse mismatch model, namely, (1) the principle of detailed balance and (2) the definition of a diffuse scattering event at an interface.

The following definition of diffuse scattering will be used. A phonon with energy $\hbar\omega_i$, wave vector \mathbf{k}_i , and mode j_i is diffusely scattered if the resulting phonon's wave vector \mathbf{k}_f and mode j_f are completely independent of \mathbf{k}_i and j_i . Simply put, the definition is that, after a diffuse scattering event, a phonon loses the memory of where it came from and what mode it was. (We shall assume for simplicity that the scattering events are elastic, i.e., that $\hbar \omega_i = \hbar \omega_f$.) This, like most definitions of a diffuse scattering event, is quite restrictive. For example, all correlations (except energy) between incoming and outgoing phonons are ignored. All structure of the scatterers is ignored. Yet the definition is useful for two reasons: it allows a simple calculation, and it leads to interesting results.

Because of this definition of diffuse scattering, the transmission probability $\alpha_{i,j}(\omega, \mathbf{k})$ for phonons from side *i*, with mode *j* (longitudinal or transverse), and with a given energy $\hbar\omega$, is independent of its wave vector and mode:

$$\alpha_{i,j}(\omega, \mathbf{k}) = \alpha_i(\omega) . \qquad (2.13)$$

From our definition of diffuse scattering, it follows that¹⁷

$$\alpha_i(\omega) = 1 - \alpha_{3-i}(\omega) . \qquad (2.14)$$

This equation states that (since a phonon forgets where it came from) the probability of reflection from one side must equal the probability of transmission from the other.

The number of phonons of energy $\hbar \omega$ per unit area per unit time leaving side *i* is

$$\sum_{j} \int_{0}^{2\pi} \int_{0}^{\pi/2} d\theta \cos\theta \, d\phi \, c_{i,j} N_{i,j}(\omega, T) \alpha_{i}(\omega) \,. \tag{2.15}$$

Here, $N_{i,j}(\omega, T)$ is the density of phonons with energy $\hbar \omega$ on side *i* with mode *j* at temperature *T*. [See also the explanation of Eq. (2.7).] Because the transmission probabilities are independent of incident angle, the angular integrals can be done immediately; the result is

$$\frac{1}{4} \left[\sum_{j} c_{i,j} N_{i,j}(\omega, T) \right] \alpha_i(\omega) .$$
(2.16)

From detailed balance, this must equal the number of phonons of energy $\hbar\omega$ leaving side 3-*i* per unit area per unit time:

$$\sum_{j} c_{i,j} N_{i,j}(\omega, T) \alpha_i(\omega) = \sum_{j} c_{3-i,j} N_{3-i,j}(\omega, T) [1 - \alpha_i(\omega)] .$$
(2.17)

From this, we can solve for the transmission probabilities:

$$\alpha_{i}(\omega) = \frac{\sum_{j}^{j} c_{3-i,j} N_{3-i,j}(\omega, T)}{\sum_{i,j} c_{i,j} N_{i,j}(\omega, T)} .$$
(2.18)

¹⁷We use the subscript 3-*i* to denote the side opposite to side *i*; the side opposite side 1 is 2, and the side opposite to side 2 is 1.

We can now use this transmission probability to calculate the net heat flux, just as was done for the acoustic mismatch model. We shall again make the Debye approximation (though again with the generalization that the longitudinal and transverse phonon velocities are different) for the phonon velocities and phonon densities of states. Then we can write the transmission coefficients as

$$\alpha_{i}(\omega) = \frac{\sum_{j=1}^{j} c_{3-i,j}^{-2}}{\sum_{i,j=1}^{j} c_{i,j}^{-2}} .$$
(2.19)

To calculate the thermal boundary resistance, we can start at Eqs. (2.10) and (2.11), since these are not specific to the acoustic mismatch model. In the present case, we know the transmission coefficients and can calculate the averaged transmission coefficients (the $\Gamma_{i,j}$):

$$\Gamma_{i,j} = \int_{0}^{\pi/2} \frac{\sum_{j} c_{3-i,j}^{-2}}{\sum_{i,j} c_{i,j}^{-2}} \cos\theta \sin\theta \, d\theta$$
$$= \frac{1}{2} \frac{\sum_{j} c_{3-i,j}^{-2}}{\sum_{i,j} c_{i,j}^{-2}}.$$

Immediately from this,

$$\left[\sum_{j} c_{i,j}^{-2} \Gamma_{i,j}\right] = \frac{1}{2} \frac{\left[\sum_{j} c_{i,j}^{-2}\right] \left[\sum_{j} c_{j-1}^{-2}\right]}{\sum_{i,j} c_{i,j}^{-2}} . \quad (2.20)$$

In the low-temperature limit, the thermal boundary resistance in the limit of diffuse mismatch can therefore be written as

$$R_{dm} = \left[\frac{\pi^2}{15} \frac{k_B^4}{\hbar^3} \frac{1}{2} \frac{\left[\sum_{j} c_{i,j}^{-2}\right] \left[\sum_{j} c_{3-i,j}^{-2}\right]}{\sum_{i,j} c_{i,j}^{-2}} \right]^{-1} T^{-3}$$
$$= \left[1.02 \times 10^{10} \frac{\left[\sum_{j} c_{i,j}^{-2}\right] \left[\sum_{j} c_{3-i,j}^{-2}\right]}{\sum_{i,j} c_{i,j}^{-2}} \right]^{-1} \times T^{-3} \left[\frac{\mathrm{cm}^2}{\mathrm{sec}^2} \mathrm{K}^3 \frac{\mathrm{K}}{\mathrm{W/cm}^2} \right].$$
(2.21)

This is the direct analog of Eq. (2.12).

The ratio of the diffuse thermal boundary resistance R_{dm} to the acoustic thermal boundary resistance R_{am} is (in the low-temperature limit)

$$\frac{R_{dm}}{R_{am}} = 2 \frac{\left[\sum_{j} c_{i,j}^{-2} \Gamma_{i,j}\right] \left[\sum_{i,j} c_{i,j}^{-2}\right]}{\left[\sum_{j} c_{i,j}^{-2}\right] \left[\sum_{j} c_{3-i,j}^{-2}\right]} .$$
(2.22)

This relationship was qualitatively shown in Fig. 14. A

special case of Eq. (2.22) is the case of an imagined interface (both sides of the interface are identical). Then, as noted after Eq. (2.12), R_{am} simplifies, and $R_{dm}/R_{am}=2$. The only dependence on the mass densities in Eq. (2.22) lies in the average transmission probabilities calculated using the acoustic mismatch model (the $\Gamma_{i,i}$ above); there is no mass density dependence in the diffuse mismatch model. Because the acoustic mismatch model calculations (specifically the $\Gamma_{i,i}$) must be performed numerically, the best way to understand how the above ratio varies with mass densities and phonon velocities is to compare calculations of the boundary resistances for several interfaces. To do this, we have used the acoustic properties listed in Table I for several solids and for the helium liquids. Table II contains the calculated boundary resistances (in the low-temperature limit); for the acoustic mismatch model, we used the tables of Cheeke, Ettinger, and Hebral (1976), and for the diffuse mismatch model, we used Eq. (2.21).

At higher temperatures, where there is a non-negligible probability of excitation of phonons near the zone boundary in at least one side of the interface, the realistic phonon dispersion and density of states must be considered in the calculation; the Debye picture of the solid breaks down. One way to extend the model is to calculate for

TABLE I. Mass densities and speeds of sound of several materials. These are required for calculating the thermal boundary resistances in Table II. The properties of rhodium are from Walker *et al.* (1981). The properties of helium are from Folinsbee and Anderson (1974). The properties of all of the other materials can be found in Simmons and Wang (1971). *L*- and *S*denote liquid and solid, respectively, svp denotes saturated vapor pressure, and atm denotes atmospheres of pressure.

	Density	c_L	c_T
Material	(g/cm^3)	(10^5 cm/sec)	(10^5 cm/sec)
Aluminum	2.70	6.24	3.04
Chromium	7.19	6.98	4.10
Copper	8.96	4.91	2.50
Gold	19.3	3.39	1.29
Indium	7.47	2.699	.905
Magnesium	1.7752	5.940	3.298
Lead	11.59	2.35	.97
Nickel	8.81	5.63	2.96
Platinum	21.62	4.174	1.750
Rhodium	12.4	5.83	3.96
Silver	10.63	3.78	1.74
Tungsten	19.320	5.248	2.908
Sapphire	3.97	10.89	6.45
Quartz	2.66	6.09	4.10
Silicon	2.33	8.970	5.332
Diamond	3.512	17.50	12.80
Calcite	2.717	6.75	3.48
CaF ₂	3.217	6.92	3.69
$L-^{4}$ He (svp)	0.145	0.238	
S- ⁴ He (38 atm)	0.198	0.540	0.250
L^{-3} He (svp)	0.082	0.194	
L^{-3} He (27 atm)	0.114	0.390	
S^{-3} He (38 atm)	0.128	0.580	0.210

each phonon state the transmission probability using the exact phonon velocities and densities of states. Then the integral over frequencies is nontrivial and must be done numerically. To see the qualitative behavior of the boundary resistance at high temperatures, for an interface between solids in which one solid has much higher Debye temperature than the other (e.g., Rh:Fe on sapphire), we can assume the dispersion of a linear chain and the appropriate cutoff frequencies for the side with lower Debye temperature (e.g., Rh:Fe). In this case the calculation of the transmission probabilities and the numerical integration can be done relatively easily. The result of this calculation (Swartz, 1987) is used in this review as the prediction of the diffuse mismatch model in the plots of the thermal boundary resistance at the Rh:Fe-sapphire interfaces in Sec IV. The deviation of $R_{\rm Bd}$ from the T^{-3} behavior is seen at temperatures above about 60 K in Fig. 6 (see also Sec. IV). This is an effect of the phonon cutoff frequency in the Rh:Fe. There is also a slight decrease in $R_{\rm Bd}T^3$ at temperatures corresponding to the Rh:Fe zone boundary, due to the increased phonon density of states near the zone boundary. For a discussion of inelastic scattering in the diffuse mismatch model, see Sec. IV.D.

E. The phonon radiation limit

The Kapitza resistance has always been found to be lower than predicted by the acoustic mismatch model, often by orders of magnitude (see Fig. 1). This had led to the general question: What is the smallest thermal boundary resistance that can be realized, i.e., what is the maximum possible (phonon-dominated) thermal boundary conductivity for an interface? The answer first proposed was based on the so-called phonon radiation limit (Snyder, 1970). In this limit, all phonons from the side with the lower phonon density (the solid side at Kapitza boundaries) are assumed to be transmitted. From the side with higher phonon density (the helium side at Kapitza boundaries), precisely enough phonons are transmitted to satisfy the principle of detailed balance and the second law of thermodynamics. That this is the maximum transport which does not violate the principle of detailed balance is clear; no more than all the phonons from the solid side can be transmitted. This limit has sometimes been called the "perfect match model," a misnomer, because it is not a model, but simply a limit. There is no mention of the physics (or demon) which al-

TABLE II. Calculated low-temperature acoustic mismatch model thermal boundary resistances and low-temperature diffuse mismatch model thermal boundary resistances for several interfaces. The numbers reported are $R_{Bd}T^3$ with units $K^4/(W/cm^2)$. The materials are assumed to be isotropic Debye solids with the properties given in Table I. Note that the range of values is very limited, especially for a given substrate. Note also that, for solid-solid interfaces, there is very little difference between the predictions of the two models, and that the diffuse mismatch model prediction can lie either above or below the acoustic mismatch model prediction. The acoustic mismatch values were calculated using the tables of Cheeke, Ettinger, and Hebral (1976). The diffuse mismatch values were calculated using Eq. (2.21). The asterisks mark interfaces studied by Swartz and Pohl using techniques described in Sec. IV. B. 4. From Swartz and Pohl (1987). See Figs. 1 and 2 for the values of the Kapitza resistances to copper.

		Sapphire			Quartz		Sili	con
	AMM		DMM	AMM		DMM	AMM	DMM
Aluminum	21.0	*	21.4	6.50	*	10.8	11.8	15.9
Chromium	18.5		24.4	9.77		13.8	14.5	18.9
Copper	18.5		20.1	8.66		9.43	14.3	14.6
Gold	18.9	*	18.1	8.12		7.48	15.8	12.6
Indium	20.4		17.7	7.19		7.10	12.1	12.2
Lead	18.8		17.8	7.67		7.14	12.8	12.3
Nickel	19.7		21.1	9.32		10.5	15.5	15.6
Platinum	20.8		18.7	13.0		8.10	21.3	13.2
Rhodium	20.8	*	23.6	13.0	*	13.0	19.2	18.1
Silver	18.2		18.7	8.66		8.06	13.8	13.2
		Diamond	1		Calcite		Ca	\mathbf{F}_2
•	AMM		DMM	AMM		DMM	AMM	DMM
Aluminum	78.0		67.4	5.19		9.29	6.06	9.89
Chromium	60.0		70.4	8.33		12.3	8.24	12.9
Copper	61.3		66.1	6.77		7.95	6.94	8.56
Gold	60.3		64.1	7.98		6.00	7.70	6.60
Indium	88.0		63.7	5.51		5.62	6.26	6.22
Lead	75.0		63.8	5.85		5.67	6.34	6.27
Nickel	61.3		67.1	8.40		9.01	8.47	9.62
Platinum	60.5		64.7	9.31		6.62	9.39	7.22
Rhodium	62.0		69.6	10.9		11.5	10.2	12.1
Silver	61.5		64.7	6.51		6.58	6.71	7.19

lows precisely the correct number of phonons to transmit from the side with higher phonon density. The ratio R_{prl}/R_{am} can be interpreted as the fraction of the energy transported across the interface to that incident on the interface (from the side with lower phonon density). At boundaries to helium this is an averaged transmission probability for phonons incident from the solid side. Also, at boundaries to helium, $R_{prl} \simeq R_{dm}$, and thus R_{dm}/R_{am} can be interpreted in the same way.

In the previous section we discussed the diffuse mismatch model, which for Kapitza boundaries leads to a thermal boundary resistance nearly as low as that derived from the phonon radiation limit. Unlike the phonon radiation limit, the diffuse mismatch model provides physical justification for the high phonon transmission probability from a solid into helium and at the same time shows physically why the probability of phonon transmission from helium into a solid is such that the principle of detailed balance is satisfied. No demon is required. At the same time, the diffuse mismatch model can be used to predict that the phonon transmission probabilities at solid-solid interfaces are never vastly higher than those predicted using the acoustic mismatch model, but in some cases are slightly higher, and in other cases are slightly lower. Experimental tests of this prediction follow in Sec. IV.B.3.

In the following section, we shall review the studies of Kapitza resistance. In many of these studies, comparisons have been made with the phonon radiation limit. The diffuse mismatch model provides a justification for such comparisons, because of the proximity of the prediction of the diffuse mismatch model with this limit.

III. KAPITZA RESISTANCE

In this section, we shall first discuss the experimental techniques used to measure Kapitza resistance, i.e., the thermal boundary resistance between liquid helium and solids. We shall review the data, note some trends in the data, and discuss the mechanisms or "channels" for heat flow that have been proposed to explain the magnitude of the resistance and its dependence on temperature, pressure, and the physical condition of the solid surface. Some of the effects to be understood have been shown in the Introduction. For example, the Kapitza resistance fails to approach the acoustic mismatch value even at 0.1 K (see Fig. 1), and the disagreement increases as the temperature is raised toward 1 K. Moreover, the pressure dependence is qualitatively as expected from the acoustic mismatch model only well below 1 K (see Fig. 2); near 1 K, the Kapitza resistance is nearly pressure independent.

The condition of the surfaces has been seen to be very important, but from the Kapitza resistance measurements alone there was little chance of determining what aspects of the surface conditions were important. Phonon scattering experiments were designed to study the interaction of phonons at surfaces and interfaces. Although these experiments did not allow a direct measurement of the Kapitza resistance, they did allow systematic measurements on well-characterized surfaces and with phonons having specific propagation directions. The results of these experiments relate directly to the proposed phonon transmission mechanisms and in some cases suggest new mechanisms. Therefore experiments on phonon scattering at interfaces will also be reviewed here.

A. Techniques used

1. Steady-state measurements

In Fig. 15 is a diagram of a typical experimental arrangement of Kapitza resistance above 1 K, following Gittleman and Bozowski (1962). The helium is contained in a thin-wall stainless or glass tube, which has negligible



FIG. 15. Schematic diagram of a traditional Kapitza resistance experiment, adapted from Gittleman and Bozowski (1962). Electrical leads for the heaters and thermometers are fed through the vacuum chamber pump tube. The isolation tube is thin-walled stainless steel to minimize spurious heat flow. The solid that makes contact with the helium is sealed into the isolation tube in a superfluid leak-tight manner, usually by either soldering or epoxying. The helium bath is temperature controlled by controlling the pressure over the liquid. Thermometers are labeled T and the heater is labeled H.

thermal conductance. The interface is that between the helium in the tube and a material mounted into the tube, usually with solder or epoxy. The helium is supplied directly from a bath, which is temperature controlled by controlling the pressure above the bath. The tube is isolated from the bath by surrounding it with a vacuum space. Heat is applied at the end of the sample, and thermometers are mounted on the tube, on the sample, and in the helium. The temperatures are extrapolated to the boundary. Variations of this geometry have been used to allow careful preparation of the surface [see, for example, Snyder (1976)]. If the measured Kapitza resistance is to a pure metal like copper, and the metal is not in the superconducting state, then temperature gradients in the metal are usually small compared to the temperature discontinuity at the boundary. Temperature gradients in the helium are negligible if the helium is superfluid ⁴He. At temperatures above the lambda transition, temperature gradients in the helium are very large due to the low thermal conductivity of the normal fluid; great care must be taken to minimize the effects of these gradients on the measurement of the thermal boundary resistance.

For measurements using ³He, a different experimental geometry is used in order to reduce the volume of ³He required, to keep ³He from escaping, and to minimize the temperature extrapolation errors caused by the very low thermal conductivity of the liquid. In Fig. 16 is an example of a geometry for measuring the Kapitza resistance between copper and ³He or (with some modification) ⁴He from about 50 mK to nearly 1 K (Anderson and Johnson, 1972). The cell is designed with a very narrow space containing the helium to minimize both the thermal resistance and the heat capacity of the helium. The thermal resistance measured is the total of the resistance of the



FIG. 16. Improved geometry for measuring the Kapitza resistance to ³He or to ⁴He. The temperature difference between the closely spaced copper plates is measured, corresponding to the sum of two boundary resistances. The purpose of the guard ring is explained in the text. The gap between the guard ring and the lower plate is sealed with epoxy. Thermometers are labeled R and heaters H. Two fill ports are used to allow a check of the continuity of the cell and fill line. Adapted from Anderson and Johnson (1972).

metal-helium boundary, the resistance of the helium, and the helium-metal boundary, i.e., the thermal resistance between the upper plate and the lower plate in the figure. The upper plate mounts to a dilution refrigerator and the lower plate to a calibrated thermometer. The guard ring is kept at the temperature of the lower plate by supplying whatever heat is necessary. By controlling the temperature of such a guard ring to match that of the lower plate, heat flow from the lower plate to the guard ring is eliminated; therefore the only heat flow from the lower plate must be across the helium. The use of such a guard ring thus allows a mechanically robust cell to be used without the fear of extraneous heat flow around the edges. This design allows the measurement of the Kapitza resistance at helium pressures up to about 30 or 40 atm [see Folinsbee and Anderson (1974)].

2. Second-sound transmission measurements

The propagation of heat in helium by second sound allows an ac measurement of the Kapitza resistance. When a second-sound wave is incident on a thin foil with negligible heat capacity, that second-sound wave would transmit directly through the foil if the Kapitza resistance were zero. Because of the Kapitza resistance, though, most of the second-sound wave is reflected. The amplitude of the transmitted or reflected second-sound wave can be used to deduce a Kapitza resistance. This was first recognized by Osborne (1951), but the transmitted wave was too weak to be detected with the given sensitivity of his apparatus. An improvement in the apparatus was made by Brow and Osborne (1958), and preliminary measurements of the Kapitza resistance to copper were made. Further experimental refinements and a more complete series of experiments were made by Challis and Sherlock (1970). Their experimental apparatus is shown in Fig. 17. Two second-sound resonators are coupled through a thin foil. The drive cavity is driven at the first or second harmonic, usually a few hundred hertz. Resonance is indicated by a maximum in the signal from a thermometer at the opposite end of that cavity, near the foil. The second (detector) cavity is brought into resonance by adjusting the length of the cavity. The boundary resistance can then be deduced from the ratio of the temperature oscillations and the quality factor of the detector cavity.

Katerberg and Anderson (1981) made significant improvements on the second-sound technique for measuring Kapitza resistance and pointed out a serious flaw in all of the earlier second-sound Kapitza resistance measurements. They noticed systematic differences between the second-sound Kapitza resistance data and the dc Kapitza resistance data, namely, that the second-sound data had a stronger temperature dependence. This they attributed to a thermal communication between the two cavities via the bath. This communication is made possible because of the small holes connecting the cavities to the bath, as indicated in Fig. 17. By eliminating the thermal communication between the cavities and measuring the Kapitza resistance of a single sample using both techniques (dc and second sound) on the same sample, with the same experimental setup in the same cooldown, they were able to demonstrate that the second-sound technique was equivalent to the dc technique and that there is no frequency dependence of the Kapitza resistance at frequencies up to about 1 kHz. They were also able to reproduce the systematic difference in the measurements by reintroducing the connecting hole to the bath. Because of this investigation, the results of all prior second-sound Kapit-



FIG. 17. Experimental geometry for using the transmission of second sound to determine the Kapitza resistance across a superfluid/ metal-foil boundary. The second-sound heater is a planar-wound Constantan wire resistor, mounted directly above the base of the drive resonator. The thermometers are quartz plates painted with Aquadag colloidal graphite. Adapted from Challis and Sherlock (1970).

za resistance data must be viewed with the knowledge that there are likely to be systematic errors affecting the magnitude and temperature dependence of the results.

B. Experimental results and analyses

In conventional boundary resistance experiments, it is difficult to modify the surface *in situ*; it is even difficult to characterize the surface of the solid without that surface's changing in the time between characterization and measurement. For these and other reasons, few boundary resistance experiments have been performed during the past decade. Nevertheless, because so many measurements were done prior to that time at boundaries to many different metallic and dielectric solids, there is much that can be learned by a survey of those results. Several such surveys have been made; to avoid repetition, we shall refer to these surveys and cite many of the original studies only in the final list of references.

1. Experimental results

The measured thermal boundary resistance between helium and a given solid at a given temperature (say, 1 K) can vary by an order of magnitude or more. We shall not present the specific data, as they have been collected in many earlier reviews. [The values for copper or lead have been reviewed by Pollack (1969) and Snyder (1970). See also the reviews by Cheeke (1970a, 1970b), Challis (1974), and Harrison (1974).] Instead of presenting these data directly, we shall discuss some of the trends. In many of the experiments, the effect of damaging or oxidizing the surface of the solid was ambiguous. It was even observed (Challis, 1968a, 1974) that the lowest Kapitza resistances corresponded to samples with the least damaged surfaces, contrary to the present understanding.

In spite of the confusion and experimental irreproducibility, there were definite trends in the data for different solids. For example, Challis (1968a) showed that the dependence of the boundary resistance on the Debye temperature of the solid (at 1.5 K) is approximately linear, not cubic, as predicted using the acoustic mismatch model. In 1974, Challis showed in another survey that the average phonon transmission probability (from the solid side into the helium at 1.5 K) is proportional to the Debye speed of sound in the solid (it should be inversely proportional, according to acoustic mismatch theory), and inversely proportional to the mass density of the solid.¹⁸ These trends imply that the resistance to dense, soft (low-Debyeboundary temperature) solids will be much closer to the prediction

¹⁸The linear dependence of the transmission probability on the sound velocity of the solid is nearly equivalent to a linear dependence of the boundary resistance on the Debye temperature.

of acoustic mismatch theory than will be the boundary resistance to light, hard (high-Debye-temperature) solids. This trend is independent of whether the solid is metallic or dielectric. Some noteworthy examples: the phonon transmission probability from LiF into helium (Johnson and Little, 1963) and Al_2O_3 (sapphire) into helium (Gittleman and Bozowski, 1962) were both measured to be about 70% at 1.5 K, according to the analysis in Sec. II.A (see also Cheeke, 1970b).

Harrison (1974) noted an interesting trend similar to the above observations made by Challis. He plotted the reduced boundary resistance P (reduced logarithmically so that a value of 1 corresponded to the acoustic mismatch model and a value of 0 corresponded to the phonon radiation limit) versus $T\Theta_D$, the product of the temperature and the Debye temperature of the solid. Harrison's plot is reproduced as Fig. 18. For a given solid (see also Figs. 1 and 2), $R_{Bd}T^3$ drops from near the acoustic mismatch prediction at low temperatures toward a very low value at higher temperatures. By plotting the reduced boundary resistances versus $T\Theta_D$, all the data follow approximately the same curve. This means that the boundary resistance to solids with high Debye temperatures, like sapphire or LiF, will drop relative to the acoustic mismatch model at lower temperatures than will the boundary resistance to softer solids, such as lead or mercury. These trends are very perplexing.



FIG. 18. Plot of the reduced thermal boundary resistance P as a function of $T\theta_D$. The stars indicate measurements between the solid and liquid ³He. From Harrison (1974). The data are from: CPA (Vilches and Wheatley, 1966), CMN (Harrison and Pendrys, 1973), Cu (Challis, Dransfeld, and Wilks, 1961; Johnson and Little, 1963, Anderson, Connolly, and Wheatley, 1964; Anderson and Johnson, 1972); Hg (Neeper, Pearce, and Wasilik, 1967), In and Al₂O₃ (Gittleman and Bozowski, 1962), Pb (Challis, 1962; Guan, 1962; Challis and Cheeke, 1965), Au (Johnson and Little, 1963; Whelan and Osborne, 1968), Ag (Clement and Frederking, 1966), Sn (Gittleman and Bozowski, 1962; Guan, 1962), Nb and Al (Mittag, 1973), Ni (Guan, 1962; Cheeke, Hebral, and Richard, 1973), Co and Fe (Cheeke, Hebral, and Richard, 1973), quartz (Challis, Dransfeld, and Wilks, 1961), W, Si, and LiF (Johnson and Little, 1963), KCl (Johnson, 1964).

2. Analyses

The analyses of many of the existing Kapitza resistance data often involve the proposal of a new transmission mechanism or "channel." No single channel can be responsible for all the observations; rather, for a given observation, several channels may contribute, and for different interfaces different channels may be important. We shall discuss the proposed channels separately and shall mention the experimental evidence that the channel does promote phonon transmission. A few of the calculational techniques will be discussed in simple terms, but for details we shall generally refer the reader to the original papers.

a. Boundary layers

At the interface between liquid helium and a solid, due to the van der Waals interaction between the helium and the surface of the solid, the first few monolayers of helium are under great pressure (tens of atmospheres). As a result, the first few monolayers are solid. In addition, the density near the wall falls off gradually to the bulk helium density in the next several monolayers. The total thickness of the denser layer of helium at the interface is on the order of 30 Å (Franchetti, 1956). One of the first attempts to explain the difference between the observed Kapitza resistance and that calculated using acoustic mismatch theory involved the idea that this layer would form effectively an antireflection coating (Challis, Dransfeld, and Wilks, 1961). For long-wavelength phonons (wavelength in the layer much longer than the thickness of the layer), the effect of the dense layer would be small, but for shorter-wavelength phonons (wavelength in the layer comparable to four times the layer thickness), the layer could greatly enhance transmission. Challis et al. calculated the effect of such a boundary layer. They found that, at low enough temperatures, the phonon wavelengths are long enough to be unaffected by the boundary layer, but as the temperature approaches about 0.5 K, the effect of the boundary layer becomes significant. If the thermal boundary resistance is plotted as $R_{\rm Bd}T^3$ versus temperature, then the effect of the boundary layer is seen as a drop in the boundary resistance at temperatures above a few tenths of a kelvin. Challis et al. also explained the pressure independence of the Kapitza resistance at temperatures above a few tenths of a kelvin by observing that the pressure in the interfacial layer (due to van der Waals attraction) is very large and is independent of the pressure in the bulk. At low temperatures, where the boundary layer is unimportant, the pressure dependence is unaffected by the layer, but at temperatures where the layer is important, the pressure dependence is reduced. Although these theoretical predictions agree qualitatively with the data, the predicted drop in $R_{Bd}T^3$ is much smaller than the measured drop in the boundary resistance.

Two conclusions follow from this work. First, any

theory of Kapitza resistance must account for the effects of the boundary layer. Further, there must be mechanisms in addition to acoustic mismatch and boundary layer effects, and these mechanisms further enhance the transport across interfaces. Many of the mechanisms discussed below specifically involve the boundary layer or excitations in it.

b. Attenuation

The effect of scattering in any system is to weaken conservation laws and boundary conditions. This general principle also applies to Kapitza boundaries. Consider what happens when a phonon from the helium (or in general from the side of the boundary with lower phonon velocities) is incident on an interface from outside the critical cone (see Fig. 13). The phonon will be totally internally reflected. In order to satisfy the boundary conditions, an evanescent wave is generated at the interface; the amplitude of the evanescent wave decays exponentially into the solid. If the evanescent wave scatters in the solid, then it can couple energy across the boundary. Thus a totally reflected phonon can end up being transmitted. Physically, this effect can be accounted for by including a classical attenuation of the phonons. That makes the wave vectors complex. In order to satisfy the boundary conditions, the resulting equations to be solved are complex, but can be solved on a computer. The effect of the attenuation is always to reduce the thermal boundary resistance and broaden the critical cone; the magnitude of the reduction of the thermal boundary resistance increases with the amount of attenuation. Since the critical cone lies on the side with the lower acoustic impedance, scattering only on the side with the higher acoustic impedance is important. (There are no evanescent waves on the same side as the critical cone.) This approach was taken independently by Peterson and Anderson (1972, 1973) and Haug and Weiss (1972). For an interface between liquid helium and a pure and wellannealed metal, let us assume that the attenuation in the metal is due to electron-phonon scattering. Then the attenuation length varies as T^{-1} , as does the dominant phonon wavelength. The enhancement of the transmission depends only on the ratio of the attenuation length to the dominant evanescent wave penetration. The dominant evanescent wave penetration is about the same length as the dominant phonon wavelength. Since both this length and the phonon attenuation length vary as T^{-1} , the boundary resistance still has a T^{-3} temperature dependence; the prefactor is lowered due to the attenuation. As much as a factor-of-3 reduction of the Kapitza resistance can reasonably be explained by considering the effects of attenuation. This argument has been used to explain the magnitude of the low-temperature asymptotic limit of the thermal boundary resistance relative to acoustic mismatch (see Figs. 1 and 2). Since this enhancement is classical and acoustic in nature, it can be considered to be part of a more general acoustic

mismatch model. In fact, the term *modified acoustic mismatch model* is often used to refer to this more general model. There is little controversy surrounding the validity of the effect of phonon attenuation described in the modified acoustic mismatch model.¹⁹ As with the effect of boundary layers, the effect of attenuation is to lower the predicted boundary resistance toward the measured boundary resistance. In this case the boundary resistance in the limit of low temperatures (below 0.1 K) can now be qualitatively understood, although the amount of phonon attenuation near the interface has not yet been experimentally measured independent of the boundary resistance.

If a measurement of the Kapitza resistance to a wellpolished dielectric crystal were performed at low enough temperatures (~ 0.1 K), then, because of the absence of electrons, the phonon attenuation should become negligible, and the phonon wavelength would be much larger than the solid helium layer thickness. Therefore the boundary resistance should approach the prediction of the acoustic mismatch model. No such experiment has been undertaken; performing such an experiment successfully (measuring the appropriate temperature distributions and avoiding stray resistances and conductances) would be quite challenging.

In a series of experiments to test the modified acoustic mismatch model, Folinsbee and Anderson (1974) were able to fit at low temperatures $(T \sim 0.03 - 0.3 \text{ K})$ the low-pressure Kapitza resistance (for both ³He and ⁴He) to a variety of metals, with the phonon attenuation in the metal as a parameter. Then, without changing that parameter, they were able to predict the Kapitza resistance to the same samples at higher pressures. Unfortunately, there was no experimental or theoretical method for verifying the value of the phonon attenuation determined by the fits to the low-pressure data. Nonetheless, the success of these experiments was strong evidence for the validity of the modified acoustic mismatch model. Rawlings and van der Sluijs (1979; see also van der Sluijs and Alnaimi, 1976a, 1976b, and Alnaimi and van der Sluijs, 1973, 1974, 1975) also supplied data in support of the effects of attenuation and scattering from dislocations in the metal, although quantitative analyses were not performed and the temperature range of measurement was very limited (1.2-2 K). At temperatures above 0.3 K, the pressure dependence of the thermal boundary resistance (see Fig. 2) was much weaker than expected. This indicates that the boundary conductivity is dominated by some other mechanism (such as the effect of the boundary layer; see above).

¹⁹Vuorio (1973) objected to the replacement of the phonon scattering with a classical attenuation length. However, the calculations of Peterson and Anderson (1973) have been consistent with a great many measurements made at Illinois and elsewhere, and little controversy remains about the usefulness of the modified acoustic mismatch model.

Opsal and Pollack (1974) calculated the combined effect of phonon attenuation and the helium boundary layer. Thus they could qualitatively explain the boundary resistance in the low-temperature limit, i.e., the drop in $R_{\rm Bd}T^3$ with increasing temperature, as well as the decrease in the pressure dependence above a few tenths of a kelvin. However, this is still insufficient to explain the measurements quantitatively.

c. Conduction electrons

In addition to the above effects of electrons in metals attenuating phonons and therefore broadening the critical cone, other effects of electrons on the thermal boundary resistance have been suggested. For example, an electron reflecting off the boundary between a metal and an insulator can cause the boundary to recoil, thus radiating phonons in the same manner as in the original Khalatnikov (1952) calculation. This channel was considered by Little (1961c, 1962), Andreev (1962a, 1962b), and Challis and Cheeke (1968), and was calculated to be a small effect, at most of the same order as the elastic channel at Kapitza boundaries, and negligible at solidsolid boundaries.

Experimentally, the role of electrons can be measured at a Kapitza interface to a superconductor. When the metal is in the superconducting state, the electrons play little or no role, as in insulators or semiconductors at sufficiently low temperatures. Application of a magnetic field to drive the superconductor normal allows the electrons to contribute to the thermal transport across the interface. The effect of the magnetic field can be significant [see the review by Cheeke (1970b) on the Kapitza resistance between lead and ⁴He]. However, the very low electronic thermal conductance in superconductors and the strong phonon scattering at defects near strained interfaces can make an accurate Kapitza resistance measurement very difficult; the resulting errors, which affect only the measurements of the Kapitza resistance to superconductors, are likely to have been much larger than the direct electronic contribution to the interfacial transport when the metal is driven normal. Thus these measurements were inconclusive.

Wagner, Kollarits, and Yaqub (1974) and Wagner and Yaqub (1975a) performed measurements of the boundary resistance between single-crystal gallium and superfluid helium in a magnetic field using the second-sound technique (see Sec. II.A.2). The effect of the magnetic field was quite complex; the boundary resistance initially dropped with increasing field at low fields and then increased, saturating at about four times its zero-field value at a field of about 4 kG. This indicated that in gallium electrons dominate the thermal transport across the interface. (It seems unlikely that the phonon transport could be significantly affected by the magnetic field.) Because the electronic properties are anisotropic and the electrons apparently affect the Kapitza resistance, one might expect an anisotropy in the Kapitza resistance. In a follow-up experiment, Wagner and Yaqub (1975b) observed no anisotropy in the magnitude of the Kapitza resistance to gallium, but they did observe an anisotropy in the temperature dependence of the Kapitza resistance to gallium. Wagner and Yaqub (1975a) also measured the magnetic field dependence of the Kapitza resistance to copper and found negligible transverse field dependence for fields up to 4 kG. We know of no other measurements of the magnetic field dependence of the Kapitza resistance. No specific mechanism was suggested that could cause the electrons in gallium to affect the Kapitza resistance so dramatically. We note that the secondsound technique used to measure these field dependences (see also Wagner, Kollarits, Wilkes, and Yaqub, 1975) was flawed as described in Sec. II.A.2 (see Katerberg and Anderson, 1981). We would expect the flaw in the measurement technique to affect the magnitude and temperature dependence systematically, but it might not affect relative measurements. It is therefore difficult to ascribe the dependence of the Kapitza resistance of gallium on magnetic field to systematic errors, especially as the Kapitza resistance to copper showed negligible magnetic field dependence. The strain dependences also described in the above papers (including strain in single-crystal metal foils *increases* the Kapitza resistance) are similarly difficult to interpret.

Halbritter (1979) suggested a relationship between the observed rf losses at nonideal metal surfaces and interfaces and the observed phonon scattering at such interfaces. The coupling arises because electrons can be localized in, for example, oxide layers at the metal surface. Phonons incident on these localized electrons cause them to vibrate and generate rf fields, which interact with other localized electrons. The result is strong diffuse scattering. (See also Halbritter, 1974, 1981.) Farmer, Rogers and Buhrman (1987) have studied the effects of individual and coupled electron traps in thin oxide layers and have proposed models for the microscopic interactions; this may also relate directly to diffuse phonon scattering. Singh, Lehndorff-Junges, and Dransfeld (1984) observed an anomalously high polarizability of thin adsorbed films of helium on sapphire. This polarizability must arise from excitations in the helium layer. In this case, there are probably no electrons localized in traps in the helium layer, but the nature of the excitations in the helium layer may be similar to that of the traps in the oxide layers.

d. Surface roughness

The acoustic mismatch theory assumes perfect, planar interfaces. Real interfaces are usually quite rough on the scale of hundreds of Å, i.e., the typical phonon wavelength in solids at 1 K. The effect of that roughness on the transmission of phonons at Kapitza boundaries has been studied theoretically by Lapin (1969), Sheard and Toombs (1972a, 1972b; see also the discussion below of the perturbation-theory approach to Kapitza resistance analysis), Khater (1978), Shiren (1981a, 1981b), and Nakayama (1985a, 1985b, 1986). One effect of the roughness is to couple bulk phonons (or other excitations) on either side of the interface with surface or Rayleigh waves. The phonons with wavelength comparable to the characteristic dimension of the roughness are most strongly scattered. The effect of surface roughness on the thermal boundary resistance can be understood as follows: A phonon incident on the interface can, among other things, be reflected, be transmitted, or excite a surface wave. The probability of transmission through the acoustic mismatch channel may be small. If the phonon excites a surface wave, then that wave will eventually scatter, coupling to an excitation in either the solid or the helium. The physics that governs the direction into which the Rayleigh wave scatters is no longer acoustic mismatch; the probability of transmission (relative to the initial incident phonon that coupled to a Rayleigh wave) may be much larger than the acoustic mismatch probability of direct transmission.

Using reasonable parameters describing the roughness of polished copper, Shiren (1981a, 1981b) was able to fit (from 0.2 to 1 K) the thermal boundary resistance between ³He and copper measured by Anderson, Connolly, and Wheatley (1964). (The data are shown in Fig. 2.) In his calculation, Shiren assumed the helium to be an ideal liquid and therefore neglected viscosity and other channels, e.g., boundary layer effects. Recently, Nakayama (1985a, 1985b, 1986) performed a similar calculation involving bulk phonons converting to surface phonons (Rayleigh waves) because of surface roughness. These Rayleigh waves then interacted with defects (e.g., tunneling states) in the adsorbed helium boundary layer (Nakayama, 1976a, 1976b, 1977, 1978), resulting in transmission of energy across the interface. The calculated transmission probabilities can be made to agree with the measured transmission probabilities both in magnitude and frequency dependence for temperatures between about 0.1 and 1 K, using reasonable parameters describing the surface roughness and the properties of the helium layer. These calculations account for many of the observations and effects that are generally accepted and that must be included in any realistic model: Real surfaces are rough, and the coupling to Rayleigh waves is certainly a significant factor in phonon transmission.

There is much work to be done before the thermal coupling across interfaces via Rayleigh waves is quantitatively understood; there may be several additional mechanisms coupling Rayleigh waves to phonons or other excitations in the solid, to excitations in the helium film, or to bulk phonons in the helium. There have been many theoretical studies of the coupling between Rayleigh waves and phonons or defects; see Maradudin and Mills (1968), King and Sheard (1969, 1970), Steg and Klemens (1970), Nakayama and Sakuma (1971, 1976a, 1976b), Sakuma (1972, 1973), Sakuma and Nakayama (1974), Maradudin, Mills and Wallis (1976), and Narita, Sakuma, and Nakayama (1978).

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e. Localized states at the interface

All nonacoustic channels invoked to couple phonons across a boundary require excitations at the interface which couple to phonons on both sides of the interface. In the case of surface roughness, the excitations are interface waves, which are always present at interfaces. Other models that follow very closely this general scheme start with the assumption that the surface contains a large density of defects and impurities. Vuorio (1972) modeled these low-energy vibrational modes at the interface by assuming a layer of loosely bound solid at the interface. Gel'fgat and Syrkin (1978) showed that a layer of loosely bound impurities on the surface could increase phonon transport across the interface by coupling through surface waves. Similarly, van der Sluijs and van der Sluijs (1981a, 1981b, 1981c; see also Jones and van der Sluijs, 1973; van der Sluijs, Jones, and Alnaimi 1974; van der Sluijs and Alnaimi, 1976a, 1976b) modeled the defects as localized masses and springs. These defects and impurities promote the coupling between bulk phonons and surface acoustic waves, which couple to the phonons in the helium. The model has not been worked out quantitatively in three dimensions, due to the complications introduced by the inclusion of phonon attenuation and boundary layers.

If resonant defect states are replaced with tunneling states, the result is a model proposed by Kinder (1981), and Kinder and Weiss (1986). Kinder ignores the effects of phonon attenuation and helium boundary layers and assumes the entire enhancement in phonon transmission probability to be caused by tunneling states at or near the interface. He calculates explicitly the transmission coefficients assuming a given set of tunneling densities and energies. To do this, Kinder calculates the effective classical (complex) impedance at the interface due to the tunneling states and does a classical transmission calculation. The model predicts a saturation of the enhanced phonon transmission probability at high phonon intensities. The model also predicts a lower transmission probability for normally incident transverse phonons relative to obliquely incident transverse phonons, due to the reduced normal stress at the interface. For supporting experimental results, see the phonon scattering experiments of Schubert, Leiderer, and Kinder (1980, 1982), Basso, Dietsche, and Kinder (1984, 1986), Kinder, De Ninno, et al. (1985, 1986), and Koester, et al. (1986), to be discussed in Sec. III.C, below. Although some of the predictions of the model have been observed experimentally, in order to fit the data it was necessary to assume an unrealistically high density of tunneling states (see, for example, the remarks by Nakayama, 1985b).

f. Excitations in the helium boundary layer

Another coupling mechanism, suggested by Johnson and Little (1963; see also Anderson and Johnson, 1972), is that phonons from the solid can directly desorb or excite helium atoms in the boundary layer. This mechanism cannot be effective without the influence of other mechanisms, because if it were, it would be effective at any Kapitza interface; however, at interfaces to in situ laser-annealed silicon surfaces (discussed later in this section), the transmission is not significantly enhanced (relative to the acoustic channel), even for phonons with frequencies of several hundred GHz. Thus there must be some unspecified defect at the surface (e.g., roughness), which either causes additional excitations in the boundary layer or causes the phonons to couple more effectively to the existing excitations in the boundary layer, or both. Desorption of helium atoms from the bound laver cannot explain the thermal boundary resistance between solids and solid helium, or the thermal boundary resistance between solids and solid adsorbed hydrogen, deuterium, neon, or argon (Taborek and Goodstein, 1981). Moreover, there is little or no change in the transmission across a helium-solid interface when the helium solidifies (Goodstein et al., 1981). Therefore the desorption or direct excitation of the helium atoms from the interface cannot be the sole cause, but is at most a contributing factor to, or an effect of, the enhanced phonon transmission mechanism.

Cheeke, Hebral, Richard, and Turkington (1974) observed the temperature jump at an interface between helium and copper or between helium and lead to depend nonlinearly on the heat flux across the interface. This was later studied for interfaces between helium and copper or helium and silver by Rawlings and van der Sluijs (1977, 1978a, 1978b), van der Sluijs (1979), and Bishop and van der Sluijs (1980a, 1980b), who suggested that the nonlinearities (and time dependences) were caused in a layer of the superfluid near the surface.

There have been many theoretical studies relating to the effect of the van der Waals bonded surface helium layer and excitations in it. Challis and Toombs (1971; Toombs and Challis, 1971) investigated theoretically the interaction between continuum states of independent helium atoms and the van der Waals attraction at the solid surface. Toombs, Sheard, and Rice (1972) considered the effect of ³He adsorption and desorption on the energy transfer between a solid and 3 He. Long (1974) and Long, Sherlock, and Wyatt (1974) suggested that surface phonons (Rayleigh waves, which coupled to bulk phonons in the solid through an unspecified mechanism) were responsible for the coupling to the excitations of the helium atoms in the boundary layer. Cheeke and Ettinger (1976, 1979, 1980) and Guermeur and Jacolin (1977) investigated the effect of phonon attenuation in the bound helium layer on the phonon transmission into the layer. Maris (1979) investigated generally the effect of excitations in the helium (in the bulk as well as in the bound layer) on phonon transmission and derived a set of sum rules that govern the transmission probabilities under ideal circumstances. The sum rules of Maris are not valid once either the phonons or the excitations in the helium have a finite lifetime. Thus the effects of phonon scattering at the surface of the solid from defects at the surface of the solid (such as roughness, tunneling states, and resonant states) are excluded. Haug, Sigmund, and Weiss (1987) investigated the effect of the finite lifetime of the excitations in the helium boundary layer. They found that the shorter the excitation lifetime the greater is the effect on the phonon transmission probability. They investigated the effect of roughness on the atomic scale, but not the (much more important?) effect of roughness on greater length scales. Nakayama has investigated the effect on the Kapitza resistance of localized excitations (two-level systems) in the helium boundary layer coupled through Rayleigh waves, which couple through surface roughness to bulk phonons in the solid. (See the discussion of the effects of surface roughness.)

g. Perturbation-theory approach

The transmission due to the acoustic mismatch channel is small compared to the transmission caused by imperfections at the interface; these imperfections are thus hardly a perturbation. However, if the transmission is treated as a perturbation of the phonon reflection from a free solid surface (as is done in Khalatnikov's initial calculation in 1952), then both the acoustic channel and the other channel can be treated as perturbations and the procedure will have the potential to be at least qualitatively correct even at interfaces where, say, 30% of the incident phonons from the solid are transmitted. One way to treat the perturbation is to use a transfer Hamiltonian. This is the approach taken by Sheard and Toombs (1972a, 1972b), Sheard, Bowley, and Toombs (1973), and Sluckin, Sheard, Bowley, and Toombs (1975) to study the transmission from superfluid ⁴He to a solid. Their approach allows inclusion of the effects of any type of defect for which a transfer Hamiltonian can be defined and even appears to have the potential for studying the interaction of different transmission channels. The equivalence of the perturbation approach with the acoustic approach was implicitly shown by Khalatnikov (1952) and Mazo (1955), since their treatments gave essentially the same answer.

Sheard and Toombs (1972a), using their transfer Hamiltonian approach, first established the equivalence with the acoustic mismatch results and then calculated the effects on the transmission of both point-defect scattering and hard-sphere defect scattering at the interface. The idea was to model the effects of mass defect scattering and scattering due to surface roughness on scales larger than atomic scales, respectively. The result due only to the hard spheres was a boundary conductance $h_{\rm Bdy}^{\rm hs}$

$$h_{\rm Bd}^{\rm ns} \propto (c_D/c_h)(\rho_h/\rho)$$
.

For the total boundary conductance the classical result must be added. The mass densities of the solid and liquid are ρ and ρ_h , respectively, and c_D and c_h are the Debye phonon velocity in the solid and the longitudinal phonon velocity in the helium. Sheard and Toombs's calculated dependence of the phonon transmission on the phonon velocity in the solid and on the mass density of the solid was consistent with the dependences observed in the data (cf. Challis, 1974). Moreover, the dependence on the liquid density resulted in a pressure dependence closer to what is seen experimentally.

For short-wavelength (high-frequency) phonons Sluckin *et al.* (1975) calculated the effect of roton excitation, dispersion, and multiphonon, multiroton, and phononplus-roton excitation. They found very sharp transmission peaks near the phonon maximum and roton minimum of the superfluid dispersion curves (the transmission approaches unity in these peaks). The effects of multiple excitations were calculated to be small. Toombs and Bowley (1973) extended the above perturbation treatment to all orders, calculating the exact quantum-mechanical analog of classical acoustic mismatch theory; their results confirmed the classical treatment for long-wavelength phonons.

For interfaces to ³He, analogous perturbation treatments have been carried out (Toombs, Sheard, and Rice, 1980; see also the earlier work by Rice, 1971; Sheard, Toombs, and Challis, 1971; Rice, Sheard, and Toombs, 1972; Rice and Toombs, 1972). The transmissions due both to individual quasiparticle excitation and to zerosound excitation have been calculated and compared. Comparison with the semiclassical Boltzmann equation approach of Bekarevich and Khalatnikov (1960, 1961) was made by Sheard, Toombs, and Challis (1971); if Fermi-liquid effects are ignored in the perturbation approach, then the two approaches are equivalent, although Sheard, Toombs, and Challis point out that Fermi-liquid effects should not be ignored.

h. Quantum effects specific to the Kapitza problem

It has been noted that the large drop of $R_{Bd}T^3$ with increasing temperature in the range from 0.1 to 1 K has been observed only at interfaces to quantum materials, such as the helium liquids, and to solid hydrogen or neon (Buechner and Maris, 1975; Reynolds and Anderson, 1976, 1977). It certainly cannot be ruled out that the quantum aspect of helium plays some role in this behavior, but there remains little reason to believe that the quantum nature of helium is the dominant cause of enhanced phonon transmission at Kapitza boundaries.

One of the first attempts to explain the existence of a temperature drop at an interface between superfluid helium and a solid involved the two-fluid model of superfluid helium. The idea was that conversion from superfluid to normal fluid at the boundary could take place only at a finite rate (Landau, 1941). For more detailed discussions, see Kronig and Thellung (1950), Gorter, Taconis, and Beenakker (1951), Ginzburg and Sobaynin (1976), and Onuki (1984). The effect of such a finite conversion at the interface has recently been seen as a singularity in the Kapitza resistance near the superfluid transition temper-

ature (Duncan, Ahlers, and Steinberg, 1987). The singularity has a width of only a few mK, and at its peak contributes about 10% of the measured boundary resistance.

Saslow (1973) discussed the effects of shear-wave dissipation on the thermal boundary resistance to 4 He or 3 He. These effects, although not specifically quantum, depend on the fact that both are liquids. Saslow did not explicitly include the effects of defects at the interface; on defect-free surfaces, such as *in situ* laser-annealed silicon and *in situ* annealed NaF, the phonon transport across the liquid-solid interface is not significantly greater than that predicted by the acoustic mismatch model. Moreover, at temperatures above 1 K the phonon transport across interfaces between helium and solids does not significantly depend on whether the helium is liquid or solid. Therefore dissipation in the liquid cannot be critical.

i. Spin-spin and size effects at mK temperatures

In the ³He Kapitza problem, the fact that the ³He has a nuclear spin allows a transport channel not available in the ⁴He Kapitza problem. The boundary resistance between ³He and magnetic solids can be several orders of magnitude lower than that expected from acoustic mismatch alone, and the temperature dependence of the magnetic transport is much weaker than T^{-3} . In fine metal sinters used because of their large surface-tovolume ratio, the Kapitza resistance at millikelvin temperatures can also be unexpectedly low due to finite size effects and collective excitations of the sinter. These effects are dominantly responsible for allowing transfer of heat in the millikelvin regime. The physics of these phenomena is complex and exciting, but is outside the scope of this review. A review of the heat transfer between helium and solids below 100 mK is given by Harrison (1979).

C. Phonon scattering at surfaces and interfaces

From Kapitza resistance measurements, we have seen that low-frequency phonons ($f \ll 100 \text{ GHz}$), corresponding to temperatures much lower than 1 K, and dominant phonon wavelengths much greater than 500 Å in solids) appear to transmit across polished surfaces according to acoustic mismatch theory (modified to include the effects of attenuation by electrons in the metal), whereas highfrequency phonons ($T \gtrsim 1$ K) are transmitted more readily than allowed by this theory. Typical models of the Kapitza resistance at these elevated temperatures involve the scattering of a phonon at the interface due to an interaction with an excitation at the interface. The outcome of the scattering, particularly whether the scattering is forward or backward, determines the extent of its effect on the thermal boundary resistance. Therefore much can be learned from phonon scattering experiments on well-characterized surfaces.

The experiments reviewed here can be divided into two groups-phonon-pulse experiments, in which the pulse is detected after partially reflecting from a surface or partially transmitting across it, and thermal conductance measurements, in which the thermal conductance of a dielectric crystal is measured at temperatures low enough that the phonon scattering dominantly occurs at the surface (the Casimir regime); the measured thermal conductance can then be related to the probability of scattering at the crystal surfaces. With both types of experiments, well-characterized surfaces have been studied, and results analogous to ideal acoustic behavior have been observed. Also with both types of experiments, near-ideal surfaces have been modified by the deposition of films of different materials and of varying thickness in situ at low temperatures; the effect of the added films on the phonon scattering at the surface has been studied. Similarly, by comparing the phonon scattering at uncharacterized surfaces with that at well-characterized surfaces, it is possible to derive information about the nature of the uncharacterized surfaces.

We shall review the experimental techniques and technical considerations for these two types of experiments. Then experiments on near-ideal surfaces and the effects of added defects on the phonon scattering at these surfaces will be reviewed. We shall discuss the question: Which properties of the added films are important in causing phonon scattering, i.e., what are the relevant scattering mechanisms? In addition to the experiments with well-characterized surfaces, there have been many other studies of the interactions of heat pulses with incompletely characterized surfaces, which will subsequently be reviewed. These experiments were designed to study specific questions about the nature of phonon scattering and its relationship to the Kapitza resistance. For example: What is the angular dependence of the scattering probabilities? What is the angular dependence of the phonon transmission probability at a helium-solid interface? What is the dependence of the phonon reflection probability (from the solid side of a heliumsolid interface) on the amount and state of the helium on the solid's surface?

1. Phonon-pulse techniques

Figure 19 contains a schematic of an experimental geometry used in heat-pulse experiments (from Marx and Eisenmenger, 1982). A thin-film generator and a detector are deposited onto one side of a crystalline dielectric substrate, such as silicon, quartz, or sapphire. The generator may be a Joule-heated thin film or a Josephson junction, and the detector may be a fast bolometer, such as a superconducting thin film at its transition temperature, or another Josephson junction. The sample is cooled to a low temperature, usually near 1 K. The experiment is performed by pulsing the generator and detecting the phonons that have reflected from the prepared surface.

The signal is recorded as a function of time. If the



FIG. 19. Experimental geometry used for measuring reflection of phonons from a prepared crystal surface, adapted from Marx and Eisenmenger (1982). The reflected phonon spectrum can be measured before and after modifications are made to the surface.

crystal is isotropic, then the detected signal may be described as follows: The first phonons to reach the detector are the specularly reflected longitudinal phonons, with detected pulse width ideally about the same as the generator pulse width. The diffusely scattered longitudinal phonons reach the detector as a broad tail, starting with the specular peak. The fast and slow transverse phonon peaks occur during the diffuse longitudinal tail and are followed by their corresponding diffuse tails. Often the fast and slow transverse phonon peaks are not separately resolvable. Additional peaks correspond to phonons that were mode converted upon specular reflection. The individual mode-converted peaks (longitudinal to fast transverse versus longitudinal to slow transverse, for example) are not usually resolvable; they show up as a peak between the longitudinal peak and the transverse peak(s). The mode-converted peak is usually broader than the other peaks. The shape of the detected signal will be discussed in greater detail when we discuss the effects of phonon focusing.

The reflecting surface can be modified in several ways, for example, by coating it with helium, from fractions of a monolayer to bulk liquid. The reflecting surface can also be coated with other adsorbed gases or metals, it can be heated to remove the gases, or it can be laser annealed or cleaved *in situ* to remove defects as well as adsorbed gases.

The analysis of the signal depends on several factors like the properties of the generator, detector, and the crystal used and their geometries and orientation. Before discussing the data, we shall outline some of the factors that enter the analysis and that affect how the measurements are made.

a. Emitted phonon frequency spectrum from Joule-heated thin films

The phonon spectrum emitted by a Joule-heated thin metal film is, to first order, the blackbody spectrum at the temperature of the film times the transmission probability for phonons incident on the substrate from the film. According to acoustic mismatch theory, the transmission probability is independent of the phonon frequency; therefore the spectrum should not be distorted by transmission from the film to the substrate. We shall show in Sec. IV.C that at nonideal solid-solid interfaces the transmission probability for high-frequency phonons (> a few hundred GHz) is not correctly predicted using the acoustic mismatch model; the error can be as large as a few tens of percent (but not much more), depending on the materials on each side of the interface. Thus, if one uses the acoustic mismatch model to calculate the temperature of the film, the result will be in error by as much as 10% or 20%.

There is another problem, though, namely, that the temperature of the film is not well defined, i.e., the temperature of the electrons in the film and the temperature of the phonons in the film are not necessarily equal. Radiation of phonons by metallic films and the effect of inelastic scattering of electrons at impurities in the film are discussed by Maris (1979). The emitted phonon spectrum of a metal film during an intense heat pulse is calculated by Perrin and Budd (1972a, 1972b) and Perrin (1975, 1976, 1978). A special case, the steady-state solution for small power inputs ($\Delta T \ll T$), is explicitly solved by Swartz (1987). The general effect is that the emitted phonon distribution is distorted; the emitted phonon spectrum will contain more high-frequency phonons than does a blackbody distribution. There is significant uncertainty in the spectrum because of the unknown strength of the electron and phonon scattering mechanisms in the film. Murmann and Heber (1977) measured the phonon spectrum emitted from Joule-heated metal films and verified qualitatively the predictions of Perrin, and Perrin and Budd. If the metal film is deposited on a glassy substrate, or if there is significant frequency-dependent phonon scattering in the substrate near the surface, then this scattering can affect the phonon distribution during a heat pulse; see Bron and Grill (1977a, 1977b), Schaich (1978, 1984), Bron, Patel, and Schaich (1979), and Wilson and Schaich (1984). The steady-state solution is also discussed by Jäckle (1972) and by Matsumoto, Reynolds, and Anderson (1977).

Yet another effect that causes uncertainty in the emitted phonon distribution is that of the finite thickness of the film on the longer-wavelength phonons. When the thickness of the film approaches the dominant phonon wavelength at the temperature of the film, then the finite thickness of the film will influence the emitted phonon spectrum. This is discussed by Frick, Waldmann, and Eisenmenger (1975).

We estimate that the result of these complications (diffuse scattering, nonequilibrium between electrons and phonons, and film size effects) is that the emitted phonon frequency spectrum (amplitude, frequency dependence, and center frequency) is uncertain by at least a few tens of percent.

b. Phonon emission and detection using Josephson junctions

The use of superconductors as incoherent phonon generators and detectors was first demonstrated by Eisenmenger and Dayem (1967). Kinder (1972a, 1972b) extended the usefulness of the technique by demonstrating that the sharp edge of the phonon bremsstrahlung spectrum (the relaxation phonons discussed below) could be used to produce an emitted phonon spectrum that can be modulated in a narrow band around a single, tunable frequency. In Fig. 20 is an energy-level diagram of a biased superconductor-insulator-superconductor junction in which the two superconductors are made of the same metal. The structure is made using thin films deposited onto the substrate to be studied. When the applied bias energy eV exceeds the gap 2Δ , pairs can cross the barrier and split. There are two processes by which phonons can



FIG. 20. Josephson junction phonon generation by quasiparticle relaxation $(E_{\rm ph} \leq eV - 2\Delta)$ and recombination $(E_{\rm ph} \geq 2\Delta)$. The bias voltage must exceed the gap; $eV > 2\Delta$. The shaded area represents the electron density of states. Adapted from Eisenmenger (1976b).



FIG. 21. Josephson junction phonon detection of phonons with $E_{\rm ph} > 2\Delta$ by quasiparticle generation and subsequent tunneling. Here the gap exceeds the bias; eV < 2 Δ . Adapted from Eisenmenger (1976b).

be emitted: recombination and relaxation. Recombination phonons have energy no less than 2Δ and relaxation phonons have energy no greater than eV-2 Δ . The relaxation phonons are often called bremsstrahlung phonons. The energies of the recombination phonons are strongly peaked at 2Δ , and the energies of the relaxation phonons are peaked at eV-2 Δ . There are several reviews of the phonon generation processes in tunnel junctions, including those by Long (1973), Eisenmenger (1976a, 1976b, 1981), and Kinder (1985). The efficiency of the phonon generation process has been a topic of several studies (see Trumpp and Eisenmenger, 1977, and the above reviews). Figure 21 contains an energy-level diagram of the same Josephson junction as Fig. 20, this time biased below the gap 2 Δ . A phonon with energy at least as great as 2 Δ can break a pair, which can then contribute to the tunneling current across the junction. Such junctions thus serve as detectors of phonons with energy greater than the cutoff 2 Δ . The phonon detection sensitivity of tunnel junctions is discussed by Trumpp, Epperlein, and Lassmann (1972). Both the generation and the detection of phonons by tunnel junctions are affected by the uncertainty of the transmission probability across the interface between the junction and the substrate. Another phonon loss mechanism includes reabsorption of phonons in the film; see the above reviews. The frequencies corresponding to the energy gaps in the commonly used junctions are $2\Delta(\text{aluminum}) = 72.5 \text{ GHz}$, $2\Delta(\text{tin}) = 285.5 \text{ GHz}$, and $2\Delta(\text{lead}) = 653 \text{ GHz}$ (Eisenmenger, 1976a).

c. Effect of phonon focusing on the detected signal

In an *isotropic* medium with generator and detector deposited as in Fig. 19, the expected response of the detector to a pulse at the generator is relatively simple. The

specularly reflected phonons that reach the detector have the shortest path length and therefore reach the detector first. There will be several distinct peaks, corresponding to the longitudinal, fast transverse, slow transverse, and mode-converted phonons. Crystals are generally not isotropic, however, and that fact leads to several complications. The phonon wave vector and the group velocity (the direction of energy transport) are not parallel. There can be directions in which a disproportionate fraction of phonons are "focused," i.e., phonons within a large solid angle in k space have a group velocity direction lying in a small solid angle in group velocity space. For information on phonon focusing, see, for example, Taylor, Maris, and Elbaum (1971), Northrup and Wolfe (1979), Marx and Eisenmenger (1982), McCurdy (1982), and Maris (1986).Because of phonon focusing, a phonon generator-detector pair deposited on a real crystal will have a more complex response than described above. If the measurement is made with sufficient time resolution. there will be additional structure (Taborek and Goodstein, 1979, 1980a, 1980b, 1980c). The diffusely scattered phonons, which were expected to arrive at the detector as a broad background, arrive at the detector as a sharp structure; this structure was at first misinterpreted as being due to specular phonons. In addition, the peaks due to the specular phonons are modified due to phonon focusing. A Monte Carlo analysis of the detector signal must be made to determine the measured ratio of specular to diffuse reflection at the surface (see Basso, Dietsche, Kinder, and Leiderer, 1984; Basso, Dietsche, and Kinder, 1986; Eisenmenger, 1986, and references therein).

d. Effects of generator and detector efficiencies

The detected phonon signal in a real heat-pulse experiment is affected not only by the complexities of the phonon interactions at the surfaces and interfaces, but also by the complexities of the phonon generation and detection processes. It is difficult to calculate these efficiencies quantitatively and include them in the analysis; instead, a constant efficiency of the generation-detection process is assumed. This makes the absolute magnitude of the signal an unknown. Nevertheless, the shape of the detected signal contains the desired information. From the shape, i.e., the relative sizes of the peaks and the background, or through curve fitting using a Monte Carlo analysis, the ratio of specularly to diffusely scattered phonons is determined. This ratio should be independent of the magnitude of the detector and generator efficiencies, but will not necessarily be correctly calculated if the detector efficiency is frequency dependent or dependent on the amplitude of the pulse. For small enough pulse powers, the dependence of the detector signal on phonon-pulse amplitude should be negligible, but tunnel junction detectors, for example, are certainly not frequency independent, as discussed above. If the scattering at the surface is elastic, then the diffusely scattered phonons have the

same frequency dependence as the specularly reflected phonons, and the detector's frequency dependence is unimportant. If, on the other hand, there is any frequency downconversion (or upconversion) on scattering, then the diffusely scattered phonon frequency distribution may be shifted to a different region of the detector response function. In that case, the calculated ratio of diffuse to specular scattering will be in error (Taborek and Goodstein, 1981). If there are frequency shifts on reflection, different types of detectors which respond differently at different phonon frequencies, may report different diffuse scattering ratios. This effect affords a method for detecting the presence of inelastic scattering (Taborek and Goodstein, 1981). See Fig. 26 below and its discussion for an example of the use of detectors with different frequency response to observe inelastic scattering. (In that case, downconversion is detected in the transmitted phonons.)

Another potential source of error in the determination of the diffuse scattering ratio is the angular dependence of the phonon generation and detection. Ideally, one assumes that the generator emits phonons with a cosine distribution (Lambert's law) and the detector absorbs phonons with a probability independent of the incident phonon angle. The generator and detector are thin films deposited onto the crystal; for a phonon to be generated or detected, it must cross the solid-solid interface between the film and the crystal. This process can be modeled using the acoustic mismatch model to determine the angular dependence of the generator and detector, assuming all phonons with a given frequency actually entering the detector contribute equally to the signal. Even this analysis has rather severe limitations, as the assumption of acoustic mismatch behavior at such a solidsolid interface is not necessarily a good assumption (see Secs. II and IV). Thus there is no reliable way to determine the angular dependence of the generation and detection processes, and this, too, adds to the uncertainty in the calculation of the diffuse scattering ratio in a heatpulse experiment.

Often in heat-pulse experiments, because of these uncertainties, the analysis is comparative; the conclusions are that a particular *in situ* modification of the surface or interface has decreased or increased the amount of diffuse scattering. Usually, the surface starts as having extremely low diffuse scattering, as with, for example, a laserannealed surface (Basso, Dietsche, and Kinder, 1984; Basso, Dietsche, Kinder, and Leiderer, 1984; Mok, *et al.*, 1986) or a surface cleaved in vacuum (Weber *et al.*, 1978a, 1978b). On these nearly perfect surfaces, the absence of diffuse scattering can be detected with a resolution of about a percent (see Eisenmenger, 1986, and references therein), essentially unaffected by the above considerations.

2. Thermal conductance measurements

In nearly perfect dielectric crystals at temperatures typically below 1 K, the dominant scattering of phonons



FIG. 22. Experimental geometry used to measure thermal conductance in the boundary scattering regime (from Klitsner, 1987). The two-heater method is drawn, although it is rigorously equivalent to the two-thermometer method (Klitsner *et al.*, 1988).

is at the boundaries of the crystal. In the experimental configuration shown in Fig. 22, the thermometers used to measure the thermal conductance of the sample are not infinitesimal. Were they, and were the scattering at the boundaries entirely specular, then the conductance would diverge. Instead, a finite ΔT is measured because of the phonon scattering at the thermometers.²⁰ If the scattering at the boundaries were entirely diffuse, then the conductance would approach the Casimir value, independent (almost) of the scattering at the thermometers, the sample geometry

 $^{^{20}}$ Because the conductance depends on the sample geometry, the state of the surfaces, and the positions and geometry of the thermometers, the thermal conductivity is not well defined; hence the term conductivity is avoided (Klitsner *et al.*, 1988).

dependences, and the effects of phonon focusing can (and must) be dealt with using a Monte Carlo calculation (Klitsner *et al.*, 1988); using the results of that calculation, we can calculate the probability for diffuse scattering at the boundaries from the finite thermal conductance measured in a real experiment.

Because the thermal conductance technique is a steady-state technique, the efficiencies of the generators and detectors (heaters and thermometers) do not affect the measurement. Moreover, the generated and detected phonons have a distribution perturbed only slightly from the equilibrium phonon distribution in the crystal, and thus whether or not the scattering at the surface is elastic should not be important. In determining the probability for diffuse scattering from these experiments, one assumes the scattering to be independent of the phonon's incident angle and the scattered phonon to be emitted with a cosine distribution. (These assumptions are equivalent, because of detailed balance.) The probability of diffuse scattering at the surface that best fits the data depends on this assumption, but the change in the fit of the diffuse scattering probability caused by assuming any other reasonable angular dependence (such as scattering of normally incident phonons with greater probability) is on the order of 10%. Any contribution from bulk phonon scattering (which should be small) so far has been included in the baseline diffuse scattering probability at the clean, unmodified surface; thus the measurements provide an upper bound for the diffuse scattering ratio at that surface. One advantage of the thermal conductance technique for studying phonon scattering at surfaces is its insensitivity to generator and detector efficiencies. Another advantage is its high sensitivity to diffuse scattering; an upper bound on the diffuse scattering probability at a polished silicon surface has been measured to be about 0.1% at 0.1 K. This resolution is obtainable because of the large number of reflections a phonon must make in traversing the length of the sample (Klitsner et al., 1988).

3. Phonon scattering results and interpretations

a. Thermal conductance

A number of experiments have been performed on clean surfaces which, at low enough temperatures, scatter phonons only very weakly, but which scatter phonons at higher temperatures; i.e., phonons with higher frequencies and shorter wavelengths. By starting with such well-characterized surfaces, one can learn a good deal by studying the effects of specific types of added defects on the phonon scattering at the surfaces. We shall discuss first a systematic study, using thermal conductance, of the effects of deposited thin films on the phonon scattering at silicon surfaces (Klitsner and Pohl, 1984, 1986, 1987; Klitsner, 1987; Klitsner *et al.*, 1988). From this study and an earlier study of the thermal conductance of polished sapphire (Pohl and Stritzker, 1982), it has been concluded that surface roughness even of very low amplitude causes strong geometric and Rayleigh scattering, suggesting that surface roughness could be a very important transmission channel in the Kapitza resistance.

The surfaces of a single-crystal silicon rod were Syton polished²¹ and then cleaned, thermometers, and heaters were mounted, and the assembly was mounted onto a dilution refrigerator. The thermal conductance of the sample (with thermometer clamps) was analyzed as discussed above with the following results: Phonons with frequencies well below 100 GHz diffusely scattered at the surfaces with a probability on the order of 0.1%. Phonons with frequencies approaching 100 GHz scattered with probability on the order of a few percent, and the scattering probability increased strongly with frequency above 100 GHz. In these experiments, the silicon surface was made to be nearly ideal for thermal phonons by lowering the temperature to well below 1 K, thus lowering the dominant phonon frequency to well below 100 GHz, and raising the dominant phonon wavelength in the silicon to well above 600 Å. At these temperatures, the dominant phonon wavelength greatly exceeded the characteristic size of any imperfections at the surfaces, whatever they were; the scattering from these relatively minute imperfections became almost undetectable.

The surfaces of the silicon were then modified by depositing in situ thin films of varying thicknesses of hydrogen, deuterium, or neon onto the silicon substrates. Again, by lowering the temperature to well below 1 K, Klitsner was able to observe the onset of diffuse scattering at the coated surfaces; at low enough temperatures (the minimum temperature of measurement was about 50 mK), the scattering from the films was small, but as the temperature was increased, the scattering from the films increased, often with a sharp onset. Depending on the film thickness and species, and on the deposition conditions, the films were made to possess islandlike structure or to be continuous. The dominant phonon wavelength at the onset temperature of diffuse scattering in all cases corresponded to the characteristic dimension of the film, whether it was the island diameter for discontinuous films or the thickness for the continuous films. The sharpness with which the scattering increased as the temperature increased correlated with the film structure; the onset of the scattering for the continuous, disordered films was broad, whereas the onset of the scattering for the islandlike films was sharp.

Other types of films studied included oxides, ionimplanted regions, and thin metal films, in addition to the adsorbed gas films. Surprisingly, the scattering rate

²¹Syton is a colloidal suspension of ~400-Å silica particles in a (slightly basic) chemical etch. Syton was developed for polishing silicon; it is surprisingly effective in polishing materials much harder than silica, for example, sapphire and boroncarbide (Fischer *et al.*, 1987).

reached a limit when the film thickness was no more than a few times the dominant phonon wavelength in the film (at a given temperature). Plotted in Fig. 23 are the effects of discontinuous 2- and 60-Å films of vapor-deposited gold, of 10- and 70-Å films of adsorbed neon that are believed to be discontinuous, and of a 10-Å film of adsorbed neon that is believed to be continuous, on the phonon scattering rate at a silicon surface. (The thicknesses



FIG. 23. Inverse phonon mean free path l^{-1} vs temperature for a series of deposited films, with average film thickness as indicated, on polished silicon. l^{-1} is defined through the gas kinetic equation $\Lambda = \frac{1}{3}Ccl$, where Λ is the measured thermal conductivity, C is the Debye specific heat, and c is the average speed of sound. The solid curves are for the clean surface. The gold film data use the left y axis, and the neon film data use the right yaxis, displaced for ease of comparison. The gold films were observed to be discontinuous using scanning electron microscopy. The continuous neon film (c) was deposited with the silicon held at 4 K and the discontinuous neon film (d) was deposited with the silicon held at 1 K. Note the abrupt onset of scattering in all of the discontinuous films (including both gold films) and the lack of a defined scattering onset temperature for what is believed to be a continuous film. The thick-layer scattering limit for the neon film at 1 K is about 0.6 cm⁻¹. If acoustic mismatch theory is used to determine which phonons enter the film, and all phonons scatter in the film, then the predicted thick-layer limit is about 0.5 cm $^{-1}$. Adapted from Klitsner and Pohl (1987).

given are averages.) Discontinuous gold films with thickness on the order of one monolayer caused the surface to become almost completely diffuse around 1 K, as indicated by the data approaching the line labeled Casimir. Even neon films only a few monolayers thick caused the silicon surface to become significantly diffuse. The sharpness in the onset temperature indicates that the film is indeed discontinuous. The sharpness of the scattering threshold for the discontinuous films also depends on the distribution of island sizes in the film. For a narrow distribution of island sizes, like that seen in the gold films, the scattering onset should be (and was) sharp. For continuous films, the onset temperature was more gradual; see Fig. 23.

The gold films and several other continuous and discontinuous metallic films were characterized using a scanning electron microscope in order to verify the correlation between the nature of the film and the nature of the scattering threshold. By comparing plots of the scattering rate versus temperature for many different films with the structures of the films, Klitsner showed that the scattering rate curve could be used qualitatively to determine the nature (continuous or islandlike), acoustic properties (large or small acoustic mismatch to silicon), and thickness (by the scattering threshold temperature if the film is continuous) of an unknown film, such as an adsorbed gas. This work showed that the phonon scattering from geometrical imperfections, such as roughness, adsorbates, or deposited films, is very large. Such strong sources of diffuse scattering at interfaces must be understood, particularly because diffuse scattering and enhanced phonon transmission are synonymous in the Kapitza problem.

The work of Klitsner and Pohl suggests that there should be correlations between phonon scattering at surfaces and the dominant longitudinal and transverse phonon wavelengths. The diffuse scattering above 1 K seen by Klitsner and Pohl has been seen universally; longwavelength phonons scatter with lower probability than short-wavelength phonons. (Historically, this observation has usually been stated in terms of the dominant phonon frequency.) Since the longitudinal phonon velocity is higher than the transverse phonon velocities, for a given frequency (i.e., for a given temperature in experiments using thermal phonons), longitudinal phonons have longer wavelengths than transverse phonons. In the temperature range of the onset of scattering, we would therefore expect longitudinal phonons to transmit across a helium-solid interface with smaller probability than do transverse phonons, although at higher temperatures we would not necessarily expect any polarization dependence because all phonons will be scattered. That is precisely what is seen: for example, from heat-pulse reflection experiments (Buechner and Maris, 1975, 1976) it was found that both longitudinal and transverse phonons have a smaller reflection probability at high temperatures than at low temperatures, but the onset of the reduced reflection probability was seen for the transverse

phonons at lower temperatures (frequencies) than it was for longitudinal phonons. This supports the idea that the scattering was more geometry dependent than frequency dependent. Similarly, Guo and Maris (1972, 1974) found that for phonons emitted from a source with temperature under 10 K (dominant phonon frequency about 900 GHz) the transverse phonons showed significantly greater enhanced transmission than did the longitudinal phonons. The same was seen by Kinder and Dietsche (1974) for phonons with frequency less than 870 GHz (corresponding to a temperature of about 10 K). On "dirty" silicon surfaces, Kinder et al. (1985, 1986) observed that the onset of enhanced transmission for longitudinal phonons occurred at a higher temperature than for transverse phonons; well above the longitudinal onset, both phonon modes transmitted with equal probability. The phonon temperatures were not precisely known, but the longitudinal transmission onset apparently occurred below 1 K. The observation that the transmission probabilities for longitudinal and transverse phonons are about equal at temperatures well above the phonon scattering onset has been made by several others as well: Swanenburg and Wolter (1973) and Horstman and Wolter (1977) studied the transmission of phonons from silicon to helium (via second sound) for heater temperatures between 10 and 20 K and found no polarization dependence. Taborek et al. (1981c) found that the transmission probabilities from sapphire into helium were polarization independent, regardless of whether the interface was to bulk liquid helium, through thin films of adsorbed helium into dense gaseous helium (which supported sound in the gas), or through thin films of adsorbed helium into lowpressure gaseous helium (in which the helium atoms travel ballistically). No estimate of the phonon frequencies or the dependence on phonon frequency was reported in this case, but we calculate that the power density in the film ($\sim 20 \text{ W/mm}^2$) should have heated the film to about 20 K. (The absolute transmission probabilities were not determined in these cases; only that the probabilities were independent of polarization was determined.)

Polished sapphire surfaces have also been studied using thermal conductivity techniques. Wybourne, Eddison, and Kelly (1984) and Eddison and Wybourne (1985) found that annealing the sapphire surface in a hydrogen atmosphere at 1200° C produced a surface that scattered only a small fraction of the incident phonons at temperatures near 1 K, but no quantitative analysis of the diffuse scattering fraction was performed. A 0.3-µm epitaxial layer of silicon deposited onto the sapphire was found to diffusely scatter almost all of the phonons that entered that layer, assuming the transmission into the silicon was governed by acoustic mismatch. Wigmore (1971), Wybourne, Eddison, and Kelly (1984), and Eddison and Wybourne (1985) suggested that vibrating dislocations were the cause of this surface (or slightly subsurface) scattering. They suggested that the phonons that entered the silicon layer were scattered by the large density of dislocations in the strained silicon layer. They compared this

scattering to the scattering from an uncoated but heavily damaged sapphire surface known to contain a large density of dislocations (as well as cracks, pits, surface roughness, etc.).

b. Heat-pulse reflection

Using phonon-pulse reflection experiments, researchers have been able to characterize several surfaces in terms of their tendency to scatter phonons. Two of the most striking surfaces characterized are in situ laser-annealed silicon (Basso, Dietsche, Kinder, and Leiderer, 1984; Mok et al., 1986) and in situ cleaved sodium fluoride (Weber et al., 1978a, 1978b). These surfaces showed almost no²² diffuse scattering of phonons at frequencies up to several hundred GHz. Moreover, the effect of covering these surfaces with helium was immeasurable, indicating that the transmission into the helium was not more than 1%. This upper bound in the transmission at several hundred GHz was nearly as low as the transmission predicted using the acoustic mismatch model (especially once the effects of the antireflection boundary layer and dispersion in the helium were considered). These experiments proved that nonacoustic phonon transmission is not observed at all surfaces, and therefore must be due to some sort of nonideal aspect of the surface.

Once the technique of producing nearly perfect surfaces by laser annealing had been proven, there were many opportunities for using in situ modifications of these surfaces to study the effects of specific defects, using phonons with frequencies in excess of 100 GHz. The effect of deposited layers of gold was studied by Basso, Dietsche, and Kinder (1984, 1986). The effect of adsorbed water was studied by Koester et al. (1986). As little as 0.2 monolayers of gold, and even less water, were enough to produce significant diffuse scattering at the surface. By adding helium to the chamber, experimenters were able to reduce the reflected signal by the amount transmitted into the helium. The reflection from the clean laser-annealed silicon surface was unaffected by the addition of helium to the surface, consistent with the behavior expected from the acoustic mismatch model. On the surfaces with the added layers of gold or water, adding the helium appeared to reduce the reflected signal by removing essentially all of the diffusely scattered phonons. Thus the diffuse scattering at the surface was again equated with phonon transmission into the helium.

In these experiments, the phonon generators and detectors were tunnel junctions. The signal could be measured as a function of phonon frequency by modulating the bias voltage, thus allowing a type of phonon spec-

²²Diffuse scattering of on the order of 1% of the incident phonons is resolvable with this technique. (see, for example, Basso, Dietsche, Kinder, and Leiderer, 1984; Basso, Dietsche, and Kinder, 1986; Eisenmenger, 1986; and references therein).

troscopy to be performed. Koester *et al.* (1986) found a scattering maximum at the as-received silicon surface at about 285 GHz. After the silicon surface was laser annealed, the feature at 285 GHz vanished. No features in the scattering as a function of frequency were seen when water was adsorbed onto this laser-annealed surface. Using a similar phonon spectroscopy technique, Koblinger *et al.* (1983, 1984) detected a strong threshold in the phonon transmission at a tin-⁴He interface at about 85 GHz and several transmission maxima at higher frequencies.

Burger, Eisenmenger, and Lassmann (1984) have characterized sapphire surfaces with various surface preparations using heat-pulse techniques. Sapphire surfaces polished with 500-Å alumina suspended in water or paraffin oil were found not to scatter phonons with frequencies up to 285 GHz. Syton-polished (see footnote 21) sapphire surfaces were found to scatter phonons of all frequencies above several tens of GHz, whether or not the surface had already been polished with alumina. Using the same techniques, Burger, Lassmann, and Eisenmenger (1985) found that Syton-polished silicon surfaces scattered few phonons with frequencies less than 85 GHz, but strongly scattered phonons with frequencies over 280 GHz. bin Rani et al. (1988) characterized sapphire surfaces using very fast (of ~ 12 nsec duration) heat pulses before and after ion bombardment (with aluminum ions having an energy of 40 keV and at doses of 1×10^{14} and $5 \times 10^{\overline{16}}$ per cm²). Their sensitivity to ion bombardment damage at the surface was comparable to or better than the sensitivity of Rutherford backscattering. They were able to observe a diminished specular reflection of each phonon polarization with increasing ion dose. Northrop and Wolfe (1984) showed for sapphire surfaces that the phonon image pattern from a reflected heat pulse could also be used to differentiate between specularly and diffusely reflected phonons (for each phonon polarization independently).

Heat-pulse experiments have shown that it is only the diffusely scattered phonons that are transmitted with probability enhanced relative to the acoustic mismatch model (see, for example, Taborek and Goodstein, 1981). This was seen by comparing the detected phonon signals from a pulse reflected from a free surface with the signal from a surface covered with helium. A careful analysis showed that the specular part of the signal is unaffected, whereas the diffuse part of the signal is almost entirely removed due to phonon transmission into the helium. The analysis is not trivial because phonon focusing causes the diffuse phonon peaks to be sharp and to be easily confused with the specular peaks.

c. Transmission into thin films of helium

In heat-pulse reflection experiments, it is usually seen that only a few monolayers of adsorbed helium are required to reduce the signal; more helium has little additional effect. The reflected signal as a function of helium coverage drops from its value at no helium coverage to its value at bulk helium coverage, all in the first three monolayers (see, for example, Guo and Maris, 1974; Kinder and Dietsche, 1974). This implies that the diffuse transmission process has nothing to do with the properties of bulk liquid (or solid) helium. In particular, it has nothing to do with phonons in the bulk liquid. If the surface is covered with bulk liquid, then it is likely that the phonons carry away the energy deposited into the helium side of the interface, but the coupling between phonons in the solid and phonons in the liquid is not direct.

If a surface is covered with three monolayers of helium, and enough power is deposited into that film for a long enough time (from an intense phonon pulse from the crystal), then the finite heat capacity of the film could eventually cause a saturation effect in the enhanced transmission. That is, the film would heat and the transmission would change. This question has been studied by Taborek *et al.* (1981a, 1981b). They used the power-sharing geometry (see Fig. 24) and found the usual three-layer effect when they used short pulses (150 nsec) with heater power density of 0.2 W/mm². At the same



FIG. 24. Power-sharing geometry. A voltage pulse causes the heater temperature to rise and emit phonons, detected by the detector underneath. Helium is added to the chamber above the heater and the experiment is repeated. The resulting difference in the detector signal is due to phonons being transmitted across the heater/helium interface. Adapted from Taborek *et al.* (1981a, 1981b).

heater power, using pulses lasting about 100 times as long (12 μ sec), the effect of three monolayers on the reflection was almost insignificant. The reason is that the helium film will heat to the temperature of the heater film in that time, and thus no significant further net transport can result. This experiment does not contradict the previous results. The reported existence and semiquantitative analysis of saturation effects can be used to determine whether an experiment will be affected by finite thickness of the helium layer. The phonons that enter the film need not desorb helium from the interface to cause saturation; they must merely heat the film. If, instead of a helium film, the solid is covered with bulk liquid, then the saturation effect is absent. This is because transport of heat from the helium side of the interface into bulk liquid is very efficient, especially if the helium is superfluid. Therefore the power density required in a given time period to cause saturation at an interface to bulk helium is much larger than the power density required in the same time period to cause saturation at an interface to a film of helium only a few monolayers thick.

Anderson and Sabisky (1970, 1971) observed transmission maxima for monochromatic phonons incident on an interface between a cleaved crystal and a helium film when the film thickness was an odd multiple of a quarter of the phonon wavelength. The phonon frequency for this experiment was between 18 and 58 GHz. To measure the transmission maxima, Anderson and Sabisky optically detected the spin temperature of divalent thulium dopants in CaF₂, SrF₂, and BaF₂. The spins generated and absorbed monochromatic phonons at an EPR frequency, tuned using a magnetic field. Anderson and Sabisky (1967, 1968, 1970, 1971) called this system a spinphonon spectrometer. Although a quantitative transmission coefficient could not easily be derived from such measurements, the observation indicated that at least some of the phonons were behaving according to classical acoustics. Wyatt, Lockerbie, Mills, and Sherlock (1972) calculated the phonon transmission from a crystal covered with a helium film; the results agreed with the experimental results of Anderson and Sabisky.

A similar experimental technique was used by Sabisky and Anderson (1975) to measure the phonon reflection coefficient from SrF_2 into liquid helium from 15 to 315 GHz. They observed a drop in reflectivity from 98% at 20 GHz to under 50% above 120 GHz. The decrease was enhanced and a resonance was observed at 85 GHz when the surface was exposed to pump oil vapors. Blackford (1972) observed phonon interference effects for helium films deposited on silicon monoxide films using Josephson junctions as phonon generators and detectors. The maximum phonon frequency used was 130 GHz.

d. Inelastic scattering results

The angular dependence of the transmission of phonons at a Kapitza boundary has been measured by Sherlock *et al.* (1972, 1975), Lockerbie (1978), and Wyatt and Page (1978). See also the review by Wyatt (1981). The experimental setup and some of the results are shown in Fig. 25 (Wyatt and Page, 1978). The critical cone on the helium side of the interface is evident in the angular



FIG. 25. Measured angular dependence of the transmission of phonons from NaF into helium and transmission of phonons from helium into NaF. The central peak corresponds to the classical (acoustic mismatch) channel, and the broad background that contains most of the phonon flux corresponds to the phonons in nonacoustically transmitted channels. The horizontal scale is not quite linear. A cartoon of the experimental geometry is in the corner of each graph. The solid black rectangles represent bolometers made with Aquadag colloidal graphite, and the outlined rectangle represents a gold thin-film heater (the phonon generator). The cubelike object is the NaF crystal, and the slide-shaped object is a Melenex strip (a glass substrate). In "from NaF to helium," the generated phonons travel across the NaF to the NaF-helium boundary. Those phonons transmitted to the helium are detected after traversing the helium to the Aquadag bolometer painted on the front surface of the glass substrate. In "from helium to NaF," the phonons generated in the thin gold film on the glass substrate first encounter a gold-helium Kapitza boundary, then traverse the helium and encounter the helium-NaF Kapitza boundary. Those which are transmitted into the NaF are detected using another Aquadag bolometer painted on the back surface of the NaF. Adapted from Wyatt and Page (1978).

dependence of both the emitted phonons from the solid (cleaved NaF) and the incoming phonons from the helium. The shape and width of the peak are consistent with the modified acoustic mismatch model (inclusion of phonon attenuation in the acoustic mismatch model, discussed earlier in this section); only a very small amount of phonon attenuation near the interface is needed to fit the data. The broad angular background in the phonons emitted from the solid dominates the energy transport; this is the channel not explainable using acoustic mismatch theory. Figure 26 compares the angular distribution as measured using a bolometer for detection with that using an Al tunnel junction for detection. The bolometer, which has phonon-frequency-independent sensitivity, measures a relatively higher background than the tunnel junction, which is sensitive only to phonons with frequency greater than about 90 GHz. This implies that the background phonons have lower frequency than those in the central peak. Therefore, in this experiment, the nonacoustically transmitted phonons, which are known to have been scattered, must have been inelastically scattered (downconverted) as they entered the bulk helium. This downconversion has been verified by Wyatt, Sherlock and Allum (1982), who measured the temperature of phonons emitted from a thin-film heater into liquid helium and compared them to the temperature of the heater. Figure 27 shows the results. The phonons emitted have a temperature of up to a factor of 3 lower than the heater; the temperature of the emitted phonons appears not to exceed about 0.7 K, independent of the temperature of the heater. The fact that the angular background of Fig. 25 for the phonons entering the



FIG. 26. Comparison of the angular distribution of phonons transmitted across a NaF/helium interface, measured using an aluminum tunnel junction, with that measured using a bolometer. The two curves have been normalized at the peak. The relatively small background measured with the tunnel junction (which is insensitive to phonons with frequency less than $4.2Kk_B/\hbar = \sim 90$ GHz) indicates that the phonons in the background are downconverted to below this frequency. Phonons in the peak are not downconverted. Adapted from Wyatt and Crisp (1978).



FIG. 27. The calculated temperature of a heater (upper curve) compared with the measured temperature of the phonons emitted from that heater into helium. The phonons have downconverted by the time they enter the bulk superfluid. The temperature of the phonon pulse was determined using a time-of-flight measurement, taking advantage of the frequency dependence of the phonon propagation velocity in superfluid helium at high pressures. Adapted from Wyatt, Sherlock, and Allum (1982).

crystal "from helium to NaF" is not as large as that for phonons emitted from the solid is further evidence for phonon downconversion; the phonons emitted from the heater on the glass substrate facing the crystal are downconverted as they enter the bulk helium, and therefore, when they are incident on the crystal, have a smaller chance of nonacoustic transmission. This downconversion process is not effective in reducing the background in the case of phonons going "from NaF to helium," presumably because the surface of the Aquadag bolometer is so poor that even the low-frequency phonons are transmitted and detected.²³

Upconversion and downconversion upon reflection have also been seen by Kinder and Dietsche (1974) and Dietsche and Kinder (1976a, 1976b). The frequency conversion process has also been observed in connection with time delays between generated and detected phonon pulses (Weber, Dietsche, and Kinder, 1977).

Challis, Ghazi, and Wybourne (1982), Challis, Kenmuir, Heraud, and Russell (1986; see also the reviews by Challis, 1983, 1986), and Kenmuir, Challis, Heraud, and Russell (1987) used a thermal conductivity technique to detect inelastic scattering at the surface of a doped sapphire bicrystal held in vacuum. The energy levels of the impurities (Fe in one end of the crystal and V in the other) could be tuned with a magnetic field. The impurities

²³The experimental asymmetry between "from helium to NaF" and "from NaF to helium" is subtle because, in both, the detected phonons have crossed from solid to liquid and then from liquid into a second solid (as well as crossing a solid-solid interface). The difference is the quality of the second Kapitza interface, where phonons are incident from the helium. In one case the second interface is a helium-NaF interface, which is relatively defect free. In the second case, the second interface is between helium and a painted-on Aquadag bolometer.

burned a hole in the phonon spectrum in the presence of a thermal current, and that hole was detected by monitoring the thermal conductivity while tuning a frequency of the second dopant to the hole frequency. When the frequencies matched (called a frequency crossing), the scattering due to the second dopant was minimized because phonons of that frequency were already depleted. If the phonons equilibrated (i.e., if there was inelastic scattering), then the effect of frequency crossing on the thermal conductivity was reduced. In this experiment, the inelastic scattering in the bulk was very small; thus thermalization of the phonon spectrum had to occur at the surfaces if at all. By using thermometers at several positions along the length of the bicrystal, Challis et al. could monitor the decay of the hole. It was found that the scattering from the (fine, ground, i.e., very rough and damaged) surfaces was predominantly inelastic, deduced from the fact that the inelastic decay length was equal to the diameter of the bicrystal.

D. Summary

Below 0.1 K, the Kapitza resistance to polished metal surfaces approaches the value predicted using the acoustic mismatch model as modified to account for the effects of phonon attenuation largely due to conduction electrons. It should be noted that measurements of the Kapitza resistance to single-crystal dielectrics at temperatures well below 1 K, where this kind of phonon attenuation does not exist, have not been performed. At temperatures above a few tenths of K, the effects of the helium boundary layer, and of imperfections in or near the interface, become important. The Kapitza resistance at 1 K is affected by many conduction channels and is typically about 2 orders of magnitude smaller than that predicted using the acoustic mismatch model. The keys to these mechanisms lie in understanding phonon scattering at surfaces and interfaces. Experiments designed specifically to study phonon scattering at surfaces have become very powerful tools for unraveling the mysteries of the Kapitza resistance.

IV. SOLID-SOLID THERMAL BOUNDARY RESISTANCE

The extension of the acoustic mismatch model to solid-solid interfaces by Little (1959) laid the foundation for the theory of solid-solid thermal boundary resistances and stimulated experimental studies. On the basis of Kapitza resistance studies, one might have expected similar surprises for solid-solid interfaces. Much of the early experimental work was done for practical reasons, for example, to find out the thermal resistance of a joint in the structure of a cryostat, and to determine how to control and minimize that thermal resistance.

A. Metallic contacts

Some of the early work was done on metal-metal interfaces, which have the advantage of good bonding. The interfaces produced by soldering metals together are mechanically sound; the two metals are in intimate contact. For contacts between normal metals, the electrons very effectively carry heat across the interface and short out the boundary resistance. However, when one or both of the metals is in the superconducting state, the transport by electrons is significantly reduced, leaving phonons as the dominant carriers across the interface. Moreover, an interface to a superconductor (such as a solder) can be manipulated with a magnetic field; the magnetic field drives the joint normal and thus allows the electronic contribution to the thermal transport. Unfortunately, the electronic transport is difficult to eliminate completely; alloying, the possibility of trapped magnetic flux, and the nonzero probability of electrons tunneling through the layer or breaking pairs in the layer can all cause the electrons to carry heat across the interface.

Measurements were made on copper-lead and coppertin interfaces between 1.3 and 2.0 K (Barnes and Dillinger, 1963, 1966), and copper-lead-copper sandwiches at temperatures between 1.3 and 4.0 K (Challis and Cheeke, 1963, 1964). At the lowest temperature of measurement (1.3 K), the boundary resistance at the copperlead interface was usually higher than the acoustic mismatch value. The discrepancy was attributed to stress and dislocations near the interface that caused additional phonon scattering. Measurements were not taken at low enough temperatures to see whether these effects would diminish as the thermal phonon wavelengths increase. The temperature dependence of the thermal boundary resistance (approximately T^{-4}) indicated that the contribution of the electrons to the transport became significant at the highest temperatures of measurement (~ 2 K); at 2 K the thermal boundary resistance was usually lower than the acoustic mismatch value. For the copper-lead-copper sandwiches, the boundary resistance depended on the thickness of the lead layer. For the thick layers ($\sim 200 \ \mu m$), the scattering in the stressed lead layer caused the total thermal resistance to exceed the thermal resistance predicted using acoustic mismatch theory by about a factor of 3 at 1.3 K. The temperature dependence was typically stronger than T^{-3} , so that at 4 K the total thermal resistance was comparable to or less than the prediction using acoustic mismatch theory. For thin lead layers (~ 40 μ m), the stresses in the lead were less evident and the thermal resistance was less than that predicted using acoustic mismatch theory by about a factor of 2, presumably due to the contribution of the electrons to the thermal transport. The boundary resistance of the copper-tin joints (Barnes and Dillinger, 1966) was measured to be lower than the acoustic mismatch value by a factor of about 2. Again, the difference was attributed to electron thermal transport across the interface. The

boundary resistance was found to be very sensitive to an alloyed layer near the interface; the originally reported boundary resistance (Barnes and Dillinger, 1963) was slightly higher than the acoustic mismatch value.

Alloying and phase separation have been observed at copper-solder-copper joints. These cause additional phonon scattering in the solder, which cannot be calculated from the bulk thermal properties of the solder (Steyert, 1967). Steyert measured the boundary resistance of copper-solder-copper sandwiches from 0.06 to 1.0 K using several types of solders. The results were generally within a factor of 4 of acoustic mismatch; the joints thought to be entirely superconducting had a boundary resistance higher than acoustic mismatch, and approached the acoustic mismatch results only at the lowest temperatures measured (about 60 mK). This suggested that the extra resistance was due to excess phonon scattering near the interface due to stresses and defects. These effects diminished with decreasing temperature.

To summarize, soldered interfaces and interfaces to superconductors provided a qualitative verification of acoustic mismatch theory and, in particular, provided the experimental parameters needed for the design of cryogenic equipment. Quantitative verification of acoustic mismatch theory at metal-superconductor interfaces, however, proved to be difficult due to heat transport by electrons, alloying, and phonon scattering by stresses inherent in interfaces between two bulk solids.

B. Metal-insulator contacts

1. Indium on sapphire

To eliminate the effects of electronic transport and interfacial alloying, interfaces between indium and sapphire were studied. Indium adheres relatively well to sapphire when ultrasonically soldered or vapor deposited. Neeper and Dillinger (1964), Wolfmeyer, Fox, and Dillinger (1970), Park and Narahara (1971a, 1971b), and Schmidt and Umlauf (1976) used the conventional geometry, which parallels the geometry for a thermal conductivity experiment (Fig. 3). Interfaces were prepared by ultrasonically soldering indium to sapphire and then vacuum casting an indium rod or another indium-coated sapphire rod to the result. The boundary resistance in these experiments ranged from 30% higher than that predicted using acoustic mismatch theory to four times the predicted value. In the latter case (Wolfmeyer, Fox, and Dillinger, 1970) the sapphire surface had a rough, ground finish. The measured thermal boundary resistance in these experiments typically had a temperature dependence of $\sim T^{-2.5}$. Schmidt and Umlauf argued that the poor reproducibility was caused by damage in the sapphire near the surface resulting from the ultrasonic soldering. The sapphire surface was severely damaged and the indium was driven into the resulting cracks. In order to avoid this damage, Schmidt and Umlauf (1976) made sapphire-indium-sapphire sandwiches by vapor depositing the indium onto the end faces of two indium rods and vacuum casting the rods together with a thin additional foil of indium. This had the advantage of eliminating the damage from the ultrasonic soldering. The measured thermal boundary resistance was typically 1.5-2 times the predicted acoustic mismatch value, again with a $T^{-\alpha}$ temperature dependence, with $\alpha < 3$, and with poor reproducibility (Fig. 4).

In the measurements of the thermal boundary resistance at superconductor-metal interfaces, driving the superconductor normal had a large effect on thermal transport because electrons were then allowed to participate (see, for example, Challis and Cheeke, 1964, and references therein). At metal-dielectric interfaces the contribution of electrons to thermal transport is much smaller and more subtle. Khalatnikov (1952), Little (1961c, 1962), Andreev (1962a, 1962b), and Challis and Cheeke (1968) have all calculated that the effect of electrons coupling energy across a metal-helium interface should be small. The effect of electrons coupling with phonons across a metal-dielectric solid-solid interface should be small for the same reasons. It was thought that this could be checked by observing the difference in the thermal boundary resistance at an indium-sapphire interface when the indium was driven normal by a magnetic field. The thermal boundary resistance was observed to decrease when the indium was driven normal; the effect ranged from a few percent for the ultrasonically soldered interfaces (Neeper and Dillinger, 1964; Park and Narahara, 1971a, 1971b; Schmidt and Umlauf, 1976) to 10-30 % for the deposited and cast samples (Wolfmeyer, Fox, and Dillinger, 1970; Schmidt and Umlauf, 1976). The largest effect was observed when the indium was deposited onto rough, i.e., not optically polished, surfaces. The fact that the effect of driving the indium normal was not reproducible indicates that the effect is probably not intrinsic. A simple explanation of the effect is that when the indium is superconducting, the electrons in the indium are not available to transport heat, and thus the thermal conductivity of the indium is affected. Although that can be accounted for, the additional scattering from stresses and defects in the indium near the interface cannot be. The effect of such scattering is greater for the superconducting indium than for the normal indium. It is reasonable that the rough interfaces would show the largest effect.

Thus the apparent failure to eliminate the effects of interfacial stresses and damage rendered these data largely ineffective in determining the magnitude of an ideal thermal boundary resistance.

2. Lead and aluminum on sapphire

Nitsche and Schumann (1980) measured the thermal boundary resistance at an interface between bulk lead and sapphire. The lead was vapor deposited and then cast onto the sapphire without breaking vacuum. The measurement was similar to that described in Fig. 9, except that differential thermocouples were used instead of resistance thermometers. The thermal boundary resistance could be reliably measured from 2 to 25 K, and below ~ 10 K the temperature dependence was very nearly T^{-3} . Above 25 K the determination of the thermal boundary resistance was unreliable due to the large temperature gradients in the lead. Because of the arrangement of the thermometers (see the discussion of Fig. 9), the prediction of the acoustic mismatch model for the thermal boundary resistance measured at this interface is about a factor of 2 lower than that predicted using the Little (1959) calculation. The calculation by Simons (1974) more closely models the geometry used by Nitsche and Schumann (see also Katerberg, Reynolds, and Anderson, 1977). The intuitive explanation is as follows: Since most of the phonons incident from the sapphire side are transmitted (and are replaced with lowertemperature phonons incident from the lead side), the thermometer on the sapphire side measures a temperature that is a weighted average of the temperature of the phonons incident on each side. (The weighting somewhat favors the temperature of the phonons incident from the sapphire side.) Hence the measured ΔT is lower than that predicted using the Little calculation of the acoustic mismatch model (by almost a factor of 2). For temperatures below 7 K the thermal boundary resistance measured by Nitsche and Schumann (1980) was within about 10% of the value predicted using the Little (1959) calculation of the acoustic mismatch model. This value is, however, nearly twice that predicted using the Simons (1974) calculation; hence the data appear to agree with the acoustic mismatch model more closely than they actually do (the measured resistance being larger than expected). One might suspect problems with differential thermal contraction causing stresses and causing the lead to pull away from the sapphire. Although the leadsapphire interface reportedly had small bubbles where the lead did not make contact (these were accounted for in the data), the lead was apparently well bonded and did not pull away. However, the surface of the sapphire had been polished with $3-\mu m$ diamond abrasive before the deposition and casting of the lead. We believe, on the basis of the phonon reflection studies of Burger, Eisenmenger, and Lassmann (1984) and of thermal boundary resistance measurements by Swartz and Pohl (1987; see also Sec. IV.C), that such diamond-polished sapphire surfaces are strong scatterers of phonons, and the subsurface region of the sapphire is badly damaged by such treatment. Interfaces where the sapphire was polished with even coarser diamond abrasive were observed by Nitsche and Schumann to have a higher thermal boundary resistance than that predicted using the acoustic mismatch model (Little version), by a factor of 3 when the sapphire was polished with 30- μ m diamond abrasive, and by a factor of about 6 when the sapphire was polished with 125- μ m diamond abrasive. The temperature dependence below 7 K was again nearly T^{-3} . This could not be due

to scattering at the interface (see Table II), but must have been due to phonon-frequency-independent scattering in the damaged sapphire subsurface. We suppose that the polishing caused the sapphire to have a large density of macroscopic cracks, which could cause frequencyindependent scattering. In none of the measurements did the state of the lead (superconducting or normal) have a significant effect on the measured thermal boundary resistance. This probably indicates that the lead was not highly stressed near the interface.

Thus the thermal boundary resistance between lead and sapphire had a T^{-3} temperature dependence and a magnitude that appeared to agree remarkably well with the acoustic mismatch model. However, we suspect that the analysis was flawed and the data were affected by the roughness of the interface.

Sahling et al. (1981) measured the thermal boundary resistance at an interface between bulk aluminum and sapphire at temperatures between 0.1 and 6 K. Single crystals of aluminum were grown onto sapphire surfaces in vacuum. The bonding between the aluminum and sapphire was apparently very good; attempts to separate them inevitably resulted in the destruction of the sapphire. The measurement technique was that of Fig. 9; the temperatures were detected using resistance thermometers. The reported thermal boundary resistance for the aluminum in the normal state was about half of the value predicted using the acoustic mismatch model (Little calculation). The temperature dependence at temperatures below 2 K was nearly T^{-3} , and between 2 and 6 K there was only a small rise in $R_{Bd}T^3$ with increasing temperature. However, as for the lead-sapphire data of Nitsche and Schumann (1980), the calculation of Simons (1974) more closely models the experimental geometry. If we use the same argument for the analysis of the temperature of the thermometer on the sapphire, we can conclude that the measured thermal boundary resistance very nearly agrees with the prediction of the acoustic mismatch model. The sapphire surface preparation was very similar to that of Nitsche and Schumann (1980). Even for the samples that used their smoothest sapphire, the roughness may well have played a role in phonon scattering at (and underneath) the surface of the sapphire. For the temperature range of the measurement (below 6 K), the roughness in their best samples probably caused the phonons to be scattered only once at the interface, and this should not have affected the thermal boundary resistance significantly (see the discussion of the diffuse mismatch model). The thermal boundary resistance was observed to be larger for interfaces where the sapphire was polished with coarser diamond abrasive, similar to the increases in the thermal boundary resistance at the lead-sapphire interface when the sapphire was roughened. The effect on the thermal boundary resistance of roughening the sapphire was smaller at interfaces to aluminum than it was at interfaces to lead.

To summarize, the thermal boundary resistance between normal aluminum and sapphire measured by Sahling et al. (1981) was seen to agree remarkably well with the acoustic mismatch model when the temperatures are interpreted correctly, and the observed temperature dependence was very close to T^{-3} at temperatures below 2 K. These measurements appeared to present a very convincing verification of the acoustic mismatch model at solid-solid interfaces, although we emphasize that the measurement is insensitive to any diffuse scattering that might have occurred at the interface.

Sahling et al. (1981) reported the interesting observation that the measured thermal boundary resistance at the aluminum-sapphire interfaces increased dramatically when the aluminum was allowed to superconduct. $R_{\rm Bd}T^3$ increased with decreasing temperature below 0.7 K and reached a maximum at about 0.13 K, where it was nearly 100 times as large as at 1 K. Below 0.13 K, $R_{\rm Bd}T^3$ decreased with decreasing temperature. This peak in the apparent thermal boundary resistance was explained as an effect of nonequilibrium between the unpaired electrons and the phonons in the aluminum near the interface, using the theory of Zelikman and Spivak (1979). The error in the extrapolation of the temperature to the interface is large because the temperature profile in the aluminum is very nonlinear near the interface. Yoo and Anderson (1986) explained the large observed peak in the thermal boundary resistance at such an interface with a model that also included the effect of scattering from lattice defects in the aluminum. They also observed and explained a similar large peak in the observed thermal boundary resistance at an aluminum-epoxyaluminum sandwich. The peak was absent at tin-epoxytin interfaces. Using surface superconductivity as a thermometer, Ridner, Martínez, and de la Cruz (1975; Ridner, de la Cruz, and Martínez, 1980) measured the difference between the extrapolated temperature (of the electrons) and the measured temperature at an interface between lead and sapphire (and at lead-helium interfaces). The error in the extrapolation should be much larger at an aluminum-sapphire interface; a similar measurement for that interface would be very helpful.

3. Epoxied contacts

The total thermal resistance of a copper-varnishdielectric joint typical of that used to adjust sample-tobath thermal relaxation times for specific-heat measurements of dielectric crystals was measured by Harrison (1968) to be $\sim 1000T^{-3} \text{ K}^4/(\text{W/cm}^2)$ between 0.05 and 2 K. This value was surprisingly reproducible and has been used successfully for many years at Cornell to estimate this kind of thermal anchoring. The thermal boundary resistance, calculated using the acoustic mismatch model for this arrangement, is about 2 orders of magnitude smaller than the empirical result. This demonstrates that thermal contact at solid-solid interfaces can suffer considerably due to imperfect contact unless very special care is taken. Anderson, Salinger, and Wheatley (1961) succeeded in making a copper-(N- grease)-(chromium potassium alum) joint with total thermal resistance of $\sim 30T^{-3}$ K⁴/(W/cm²), which is much closer to the value predicted for such a joint using the acoustic mismatch model. The temperature range of the measurement was from 0.03 to 0.15 K. The reason for their success may have been lower differential thermal contraction at the joint, which resulted in lower stress and thus less damage, possibly a magnetic channel for thermal contact, or greater care in the preparation of the interfaces.

Epoxies can be used to bond two materials thermally as well as mechanically. Often, though, the thermal conductivity of the epoxy limits the thermal contact. At temperatures well above a few K, the thermal conductivity of epoxies can be improved by adding a filler of material with high thermal conductivity, such as a fine powder of copper, silver, or sapphire. At temperatures near or below a few K, the thermal boundary resistance between the particles and the surrounding epoxy matrix eliminates the thermal benefit of adding the filler; the filled epoxy can then have a lower thermal conductivity than the unfilled epoxy because heat is transported less easily across the interfaces and through the particles than it is around the particle (Anderson and Rauch, 1970). From the thermal conductivity of the filled epoxy, the thermal boundary resistance between the particle and the epoxy can be derived (Anderson and Rauch, 1970; Schmidt, 1975). This deduced thermal boundary resistance is consistent with direct measurements in the temperature range of 1-4 K (Schmidt, 1974; see also below). See also the work by de Araujo and Rosenberg (1976a, 1976b) and Garrett and Rosenberg (1974).

Several direct measurements of the boundary resistance across individual metal-epoxy interfaces were performed by Peterson and Anderson (1972), Schmidt (1974, 1977), Reynolds and Anderson (1975), and Matsumoto, Reynolds, and Anderson (1977). They used variations of the experimental arrangement used for the indium-onsapphire interfaces (shown schematically as the inset in Fig. 28). The sapphire rods were replaced with rods of high-purity copper (or some other metal), and the indium was replaced by a layer of epoxy with well-defined thickness. Thermometers were placed on the copper rods as close to the interface as possible, and the sum of the thermal boundary resistances of the copper-epoxy and the epoxy-copper interfaces was measured. The copperepoxy-copper geometry has some advantages over the sapphire-indium geometry. The epoxy layer can have a well-defined thickness and still be very thin, $\sim 10 \ \mu m$. The copper has a very high thermal conductivity, making bulk temperature drops in the copper unimportant, and the electrons, which dominate the heat transport in the copper, thermalize the phonons so that the thermometers measure the appropriate temperature distributions. Moreover, the quality of the interface between the epoxy and the copper is apparently quite good; the results were reproducible. Using this geometry, Matsumoto et al. (1977) found that the acoustic mismatch model agreed



FIG. 28. Measured total thermal resistance R of two copperepoxy-copper sandwiches, multiplied by the cube of temperature, plotted log-log vs temperature: \blacksquare , for a "thick" epoxy layer (about 80 μ m); \bigcirc , for a "thin" epoxy layer (about 16 μ m); dashed line, the sum of the acoustic mismatch thermal boundary resistances and the thermal resistance of the thin epoxy layer per unit area (the low-temperature limits agree with the acoustic mismatch prediction); solid lines, a calculation in which the total thermal resistance is calculated more realistically; see text. A schematic of the experimental geometry is shown in the inset. Adapted from Matsumoto, Reynolds, and Anderson (1977).

with the low-temperature limit (near 0.1 K) of the measured thermal resistance for copper-epoxy interfaces (see Fig. 28). However, above 0.05 K, the measured thermal resistance rose dramatically relative to the acoustic mismatch model; at 0.1 K the measured thermal resistance was more than a factor of 2 too high, and by 1 K the measured thermal resistance was more than 100 times as large as acoustic mismatch theory predicts for the interfaces alone. Much of the added thermal resistance could be accounted for by simply adding the bulk thermal resistance of the epoxy, but, as discussed in Sec. II, thermal resistances do not simply add. Therefore a more careful calculation had to be done (Matsumoto, Reynolds, and Anderson, 1977). In this calculation, the phonon transmission probability through the sandwich was calculated by modeling the thermal resistance at the interfaces with a single mean free path and adding this scattering rate to the known scattering rate in the epoxy. Due to its amorphous structure, the epoxy acts as a low

pass filter, allowing only the lowest-frequency phonons $(f \leq 25 \text{ GHz})$ to pass easily through the sandwich; the interfaces contribute significantly to the total thermal resistance only for these low-frequency phonons. The result of their calculation was consistent with the data (see the solid lines in Fig. 28).

Using a Boltzmann equation approach, Jäckle (1972) calculated the phonon transport across an interface between a metallic film and an amorphous solid (with large, frequency-dependent phonon scattering rates), including the effect of a small inelastic scattering rate in the glass. He found that the spectral redistribution of the phonons in the glass caused a significant contribution to the effective boundary resistance, in agreement with the observations and calculations of Matsumoto, Reynolds, and Anderson (1977).

In spite of their success in modeling the thermal behavior of the metal-epoxy-metal sandwiches, only below 0.05 K could Matsumoto, Reynolds, and Anderson accurately deduce from the total temperature drop the thermal boundary resistance at the interface. These measurements are the first quantitative verification of the acoustic mismatch model, although only for a limited temperature range. Another advantage of this experiment was that the analysis of the thermometry was unambiguous because the thermometers were attached to normal metals, where the electrons thermalized the phonons and also minimized thermal gradients (see Sec. II.A).

4. Metal films on dielectrics

In the early 1960s there was significant interest in the thermal time constants of structures made from thin films because of their application to superconducting devices, specifically cryotrons. In a typical geometry, a superconducting bolometer film (usually tin, indium, or lead) was deposited onto and covered by a SiO insulating film. In some cases another superconducting film, oxide layer, and heater were deposited on top. The thermal conductance across one or more of the bolometer-oxide interfaces was then deduced from the measured thermal decay time for the layered structure and the calculated heat capacity of the bolometer (Jones and Pennebaker, 1963), or from a dc temperature difference between two bolometers (Dorey, 1965; Griffiths and Watton, 1966). In all cases, the measured thermal boundary resistances were about a factor of 2 or 3 higher than predicted using acoustic mismatch theory. The temperature range of the experiments was typically 1.5-4 K; in this interval, the temperature dependence of the measured thermal boundary resistance was weaker than T^{-3} . In all these experiments, in series with the boundaries there was a relatively thick (several thousand Å thick) amorphous oxide film that is now known to introduce a significant thermal resistance, even greater than that calculated using the low thermal conductivity of the bulk oxide (Matsumoto, Reynolds, and Anderson, 1977; Swartz, 1987; Swartz and Pohl, 1987). In the pulsed experiments, the necessity of calculating the heat capacity of the thin film introduced an additional uncertainty. As a result, these measurements were inconclusive.

The use of thin films to measure thermal boundary resistance has many advantages, such as reliability and quality of the interface. The first direct measurements of the thermal boundary resistance between a single thin metal film and a single-crystal dielectric were performed by Holt (1966). The temperature range of the experiment was from 1.5 to 4 K. The temperature of the Jouleheated thin film was measured by epoxying a small carbon thermometer to its free surface. The thermal boundary resistances between films of tin and quartz or sapphire substrates were more than factor of 3 higher than the prediction of acoustic mismatch theory at 1.5 K, and the temperature dependence²⁴ was $T^{-2.0} - T^{-2.5}$. Better agreement with acoustic mismatch theory at 1.5 K was obtained for gold films on sapphire when the film was fired²⁵ onto the substrate, but the temperature dependence was still $T^{-2.5}$. We suggest as an additional difficulty the use of an epoxied-on thermometer, which can induce stresses in the film under the thermometer because of differential thermal contraction. This can affect the measurement, and can even lead to poor thermal and mechanical contact between the thermometer and the thin film.

von Gutfeld, Nethercot, and Armstrong (1966) deposited a thin-film metal heater/bolometer (pure indium or a lead-bismuth alloy) onto single-crystal sapphire or quartz, and measured the thermal relaxation time of the film. The measurement could be performed at only one temperature, the critical temperature of the bolometer film. To deduce the thermal boundary resistance from the measured thermal time constant ($\tau = RC$ is the product of the heat capacity of the thin film and the thermal boundary resistance between the film and the substrate) required a calculation of the heat capacity of the thin film. At ~ 8 K, the measured thermal boundary resistance between a Pb_{98.5}Bi_{1.5} thin film and sapphire fell 30% above the prediction of acoustic mismatch theory. The boundary resistance between the same film and quartz was a factor of more than 2 above the prediction of acoustic mismatch theory. These results were attributed to graininess in the film causing reduced contact area. The measured thermal boundary resistance at about 3.8

K between an $In_{94}Sn_6$ thin film and quartz matched the prediction of acoustic mismatch theory within the experimental uncertainty. The same film on sapphire had a thermal boundary resistance about 30% lower than the prediction of acoustic mismatch theory.

If, instead of measuring the relaxation time, one measured the temperature of the film during the pulse (but only after the film reached a steady-state temperature), then the thermal boundary resistance could be determined directly without a calculation of the film's heat capacity. If, in addition, the bolometer were replaced with a thin-film thermometer that was sensitive over a wide temperature range, then the thermal boundary resistance could be directly measured over a correspondingly wide temperature range. These considerations were the motivation for the experiments we describe next.

Wigmore (1972a, 1972b) evaporated a thin-film (Constantan) heater onto a polished and cleaned MgO substrate. The MgO was doped with Fe²⁺ resonant scatterers. The temperature of the MgO was maintained at about 2 K, and the heater was Joule heated with a known power input, varying from 2.5 to 150 W/cm², for a fraction of a microsecond. Phonons were detected by measuring the ESR signal of the Fe^{2+} (tuning the resonant frequency of the Fe^{2+} with the magnetic field) to determine the distribution of phonons emitted by the Constantan film, and therefore the film temperature. During the short pulse the MgO did not have time to heat; its temperature remained constant, and therefore the distribution of phonons incident on the Constantan film from the MgO had a temperature equal to the initial temperature of the system. This statement can be made more quantitative: since phonons traveled ballistically in the MgO for significant distances, they had insufficient time during the duration of the pulse to propagate in the MgO to a scattering site, scatter, and propagate back to the film.²⁶ Therefore the distribution of phonons incident on the film from the MgO during the pulse was unaffected by the pulse. The temperature rise during the pulse could be calculated by assuming that acoustic mismatch determined phonon emission from the Constantan film into the MgO substrate. The measured phonon distribution agreed with the calculation to within the experimental scatter, estimated to be about 5% (see Fig. 29). These were among the first experimental results at any temperature above 1 K that were clearly consistent with the acoustic mismatch model.

In the technique introduced by Weis (1969), the temperature of a thin film deposited onto a dielectric sub-

²⁴This temperature dependence, also seen in the indium/sapphire data, is consistent with the idea that the longer-wavelength (lower-temperature) phonons are less sensitive to the imperfections at or near the interface than are the shorter-wavelength phonons. See also the discussion by Little (1959, and see footnote 27) concerning the wavelength-dependent effects of imperfect (spatially intermittent) mechanical contact.

 $^{^{25}}$ "Firing" probably refers to some high-temperature heat treatment of the sample after the film deposition.

²⁶The possible exception would be if some scattering centers were concentrated very near the interface, as would be the case if the substrate surface were rough or strained. At low temperatures ($\lesssim 1$ K), the dominant phonon wavelength may be long enough to ignore such damage, but at sufficiently high temperatures we expect to see effects of this scattering on the temperature of the pulse-heated film. This is discussed below.



FIG. 29. The measured temperature of a Constantan film on MgO vs power per unit area in the film. The solid line is the calculated film temperature assuming the acoustic mismatch model. Adapted from Wigmore (1972a, 1972b).

strate is measured during a heat pulse by measuring the resistance of the film itself, using the fact that the heater film terminates the transmission line (coaxial cable) used to carry the heater current to the film. (Because the pulse is so short, transmission-line effects must be considered, including the effect of electrical impedance mismatch, which causes reflection of the pulse. The physics is analogous to the reflection of a phonon at an interface with an acoustic impedance mismatch.) The reflection of the heat pulse from the terminating resistance is a function of the resistance R of the heater and of the impedance Z of the coax (typically 50 Ω):

$$\frac{\text{reflected voltage amplitude}}{\text{incident voltage amplitude}} = \frac{R - Z}{R + Z} .$$
(4.1)

The film is a pure metal; therefore its resistance is temperature dependent, but only above $T \sim 15$ K, and therefore only above 15 K can the reflection of the pulse be used to determine the temperature. The experiment is not done with a small ΔT , but usually in the limit where the substrate temperature is small compared to the temperature of the film. The film temperature is measured as a function of the heater power per unit area. One point that is worth repeating is that, even though this heatpulse method uses short pulses to heat the sample, it is still essentially a dc technique, as is Wigmore's technique. During the pulse, the film does reach a steady temperature. The pulse must be short only in order that the extremely higher powers needed to produce the temperature rise in the film do not integrate to a large energy input into the crystal. By keeping the substrate at 4 K or less, one ensures that the energy is ballistically radiated away from the interface. In short, the problem of the high power requirement is solved by performing the entire experiment in < 100 nsec, and the problem of determining temperature gradients in the substrate is solved by keeping the substrate at 4 K, so that temperature gra-

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dients are negligible.

With this technique, phonon transport across interfaces at temperatures above 15 K (although not below because of insensitivity of the thermometers) could be studied. On some interfaces, experiment and theory agreed at temperatures up to about 100 K (for example, Herth and Weis, 1970, shown earlier in Fig. 5). This technique allowed the acoustic mismatch model to be checked on a broad range of interfaces at high temperatures. For details and for comparisons with theory, see Herth and Weis (1969, 1970); Weis (1969, 1972); Cheeke and Martinon (1972), Cheeke, Hebral, and Martinon (1972, 1973), Kappus and Weis (1973), Rösch and Weis (1977, 1978), and Martinon and Weis (1979).

The remarkable success of the acoustic mismatch model in predicting these results indicates that high-quality interfaces can be made by evaporation onto clean, polished crystal surfaces. The failures of many previous researchers to obtain agreement with the acoustic mismatch model, which was reviewed above, now appear to have been caused by imperfect interfaces or damage in the near-surface regions. However, the agreement between the acoustic mismatch model and the heat-pulse data of Weis et al. and of Wigmore seemed to be too good, given the evidence from Kapitza resistance data and phonon reflection experiments that even microscopic imperfections strongly affect heat transport. Thus the apparent quantitative success of the acoustic mismatch model in these heat-pulse experiments had to be viewed as a mystery. By using the diffuse mismatch model, it can now be understood why the effect of diffuse scattering at a solid-solid interface should be very small. In fact, evidence for the influence of diffuse scattering can indeed be seen in the data of Weis and co-workers. In Fig. 30, we show the measured radiation temperature for copper, gold, and lead on diamond. The dashed curves indicate the predicted temperatures based on the acoustic mismatch model. For copper, the radiation temperature exceeds the prediction, indicating a larger boundary resistance; for gold, the agreement is satisfactory, and for lead the boundary resistance is smaller than predicted. This behavior agrees at least qualitatively with the prediction of the diffuse mismatch model (see Table II): Diffuse scattering should raise the boundary resistance for copper on diamond, should have no effect for gold on diamond, and should lower the boundary resistance for lead on diamond.

Before the smallness of the effect of diffuse scattering at solid-solid interfaces was recognized, the apparent agreement between the heat-pulse data and the acoustic mismatch model prediction at temperatures above 10 K triggered more detailed calculations of the acoustic mismatch model solid-solid thermal boundary resistance, which included the effects of phonon focusing and more realistic density of phonon states at high phonon frequency (see, for example, Weis, 1986). However, if the phonons are scattering at the interface, the assumptions of these calculations are not valid. We hope that the machinery used in the calculations will soon be used to predict accurately the thermal boundary resistance for the diffuse mismatch regime, or better yet, for even more realistic models.

One drawback of the heat-pulse technique introduced by Weis was that measurements at temperatures below 15 K were inaccessible to his thin-film thermometers, and thus no connection to work at lower temperatures could be made. Another problem was that the thermal boundary resistance was obtained by taking the derivative of the measured temperature rise versus the power input, which led to a very large uncertainty, given the large scatter in the data. Furthermore, the assumption that the substrate temperature is unaffected by the heat pulse may not be valid at the highest heater powers, due to the possibility of phonons scattering back into the heater from imperfections in the substrate. Another simple way



FIG. 30. The measured temperature of different metal films on diamond, vs power per unit area in the film. The data are shown as shaded regions; the widths of the regions indicate the extent of scatter. The dashed lines represent the prediction of the acoustic mismatch model including the estimated (and small) effect of dispersion (using dispersion of a linear chain). Adapted from Kappus and Weis (1973).

to understand the backscattering effect qualitatively is to add to the thermal boundary resistance the thermal resistance caused by scattering in a thin, damaged region near the surface of the crystal (see Cheeke and Martinon, 1972; Martinon and Weis, 1979). As an example, Fig. 31 contains the thermal boundary resistance data for nickel on sapphire (Herth and Weis, 1970); above about 50 K, the temperature of the film greatly exceeds the predicted temperature assuming the acoustic mismatch model, also shown in Fig. 31. The prediction of the diffuse mismatch model is only slightly different from that of the acoustic mismatch model for nickel on sapphire (see Table II); therefore diffuse scattering *at* the interface cannot be used to explain these data.

Further evidence for strong phonon scattering in near-surface regions was presented by Wybourne, Eddison, and Wigmore (1985) and Wybourne and Wigmore (1986). They observed time delays in very short (~ 1 nsec) phonon pulses due to phonon scattering in a thin region between a metal heater and a polished sapphire substrate. The time constant for the heating of the film was much longer than the pulse duration; this was attributed to the very strong phonon scattering in the layer just underneath the film. Bron and Grill (1977a, 1977b) and Bron, Patel, and Schaich (1979), using longer (25-20 nsec) and very intense (up to several kW/mm²) pulses in a thin-film heater, observed the effects of strong frequency-dependent scattering at frequencies above 1 The effects of frequency-dependent phonon THz. scattering and of inelastic phonon scattering in the substrate on the time evolution of the phonon distribution in the metal film are discussed theoretically by Schaich (1978, 1984) and Wilson and Schaich (1984).

In order to resolve the transition at solid-solid interfaces from acoustic mismatch behavior expected at low temperatures to diffuse mismatch behavior expected at high temperatures, a new technique was required with which boundary resistance between a thin metal film and



FIG. 31. The temperature of three nickel films on sapphire vs power per unit area in the film. The temperature of the sapphire was 4.2 K. The dashed lines represent the prediction of the acoustic mismatch model, including the estimated (and small) effect of dispersion (using dispersion of a linear chain to approximate the dispersion in the nickel). Adapted from Herth and Weis (1970).

a crystalline dielectric substrate could be measured over the temperature range from below 1 K to over 100 K. Such a technique has recently been developed (Swartz, 1987) and is described in Sec. II (see Fig. 11). The thermal boundary resistance between Rh:Fe and polished sapphire, measured with this technique in the temperature range 0.6-200 K, is shown in Fig. 6 (Swartz and Pohl, 1987). The excellent fit with the solid curve, calculated using the acoustic mismatch model (containing no adjustable parameters), again seems to suggest that it does correctly predict transport across this solid-solid interface below about 40 K. Other metal-substrate interfaces (for example, Rh:Fe on quartz, Al on quartz and sapphire, and Pt on sapphire) have also been studied with this same technique; the data agree with the acoustic mismatch model in a similar way for all the samples (Swartz, 1987). The asterisks in Table II indicate interfaces that have been studied with this technique.

C. Evidence for diffuse scattering at solid-solid interfaces

1. T < 30 K: Specular versus diffuse

Using the diffuse mismatch model as an upper limit for the effect of diffuse scattering at solid-solid interfaces, we have shown (see Table II) that diffuse scattering does not have nearly as striking an effect on the solid-solid thermal boundary resistance as it does on the Kapitza resistance. Nevertheless, diffuse scattering at solid-solid interfaces should be observable, if sufficient care is taken in the measurement. On the basis of thermal conductivity and phonon reflection studies, we expect that at low enough temperatures (≤ 1 K), corresponding to low dominant phonon frequencies ($\lesssim 100$ GHz) and long dominant phonon wavelengths (several hundred Å in crystalline dielectrics), diffuse scattering at a well-prepared, smooth interface should be rare; at high enough temperatures (>3 K; >300 GHz), we expect that specular scattering should be rare. Therefore, even without a firm prediction of the magnitude of the effect of diffuse scattering, a transition in the thermal boundary resistance from the value predicted using the acoustic mismatch model to another value should mark the onset of diffuse scattering. The only measurements with the accuracy, temperature range, and resolution required to look for such a transition are those using deposited thin films of Rh:Fe on sapphire or quartz (Swartz and Pohl, 1986, 1987; Swartz, 1987).

The surface of a sapphire substrate was polished with a series of suspensions of alumina powders; the alumina particle size of the last step was 500 Å. Then the sample was Syton polished (see footnote 21) for 2 h. Afterwards, the sapphire was annealed in a hydrogen atmosphere at 1470 K for 30 min. According to Eddison and Wybourne (1985, and Wybourne, 1986), this procedure should produce an almost atomically smooth sapphire surface. A Rh:Fe film was then sputter deposited at a

rate of about 40 Å per second onto the sapphire substrate, which was kept at 1100 K during deposition to anneal the film during its growth, thereby reducing stresses. The two narrow, parallel strips were then lithographically patterned on the film, and the boundary resistance between the film and the substrate was measured as described in Sec. II. The results are plotted in Fig. 32 as $R_{\rm Bd}T^3$ in order to emphasize any deviations from a T^{-3} behavior. The solid curve is the acoustic mismatch model prediction, and the dashed curve the diffuse mismatch model prediction. The calculations are described in Secs. II.C and II.D.2.

We would expect the acoustic mismatch model to hold at sufficiently low temperatures, and the effects of diffuse scattering at the interface to start to become visible at some higher temperature. From Fig. 32, we see just that; the onset of diffuse scattering is seen at about 7 K. The *quantitative* agreement between the diffuse mismatch model and the data between 10 and 25 K is probably fortuitous, given the simplicity of the model.



FIG. 32. Thermal boundary resistance between Rh:Fe and hydrogen chemically polished sapphire: solid curve, prediction of the acoustic mismatch model; dashed curve, prediction of the diffuse mismatch model. The measured boundary resistance for this and the following three figures depends linearly on the measured widths of the Rh:Fe strips. We estimate the uncertainty of the widths at about 5%. In these plots of the boundary resistance between Rh:Fe and sapphire or quartz (except the log-log plot of Fig. 6), the thermal resistance resulting from the finite thermal transport between the electrons and the phonons in the Rh:Fe film has been fit and subtracted from the data. The analysis is discussed by Swartz (1987). The expected temperature dependence $(R_{ep}T^{-4})$ of the electron-phonon thermal resistance was clearly seen in the data for all the samples, but the prefactor R_{ep} was reproducible only to within a factor of 2 or 3 from sample to sample. R_{ep} was on the same order as that for a thin film of copper, measured by Roukes (1985); see also Roukes et al. (1985).

In order to show that for temperatures less than about 7 K phonons interact specularly at the interface between Rh:Fe and hydrogen-polished sapphire, Swartz and Pohl (1986, 1987) deposited Rh:Fe onto sapphire treated with each of the following polishing techniques: alumina polishing, Syton polishing, and hydrogen polishing. In addition, a diamond-polished (0.2 μ m) sapphire surface was tested. The Syton- and alumina-polished samples are compared in Fig. 33. As suggested by Eisenmenger (1986), we see that the Syton-polished surface is a strong diffuse scatterer of phonons even at 1 K (90 GHz), whereas the alumina-polished surface does not diffusely scatter phonons any more strongly than the hydrogentreated surface, (see Fig. 32). The boundary resistance to the diamond-polished surface is shown in Fig. 34. In this case, as expected, there is strong diffuse scattering down to 1 K; the thermal boundary resistance never does approach the acoustic mismatch value, but instead remains at the diffuse mismatch prediction to the lowest temperatures measured.

There is, however, one observation that does not fit into the picture presented here: the specular-to-diffuse transition seen in the hydrogen-polished sample should not be present in the Syton-polished or diamond-polished samples, if the analysis is competely accurate; yet in both cases there is an indication of the same feature. This phenomenon remains unexplained.

While for the Rh:Fe-sapphire interfaces diffuse scattering leads to a small increase in the thermal boundary resistance, in agreement with the theory, for Rh:Fe-



FIG. 34. Thermal boundary resistance between Rh:Fe and diamond-polished sapphire. The effects of the electron-phonon thermal resistance have been subtracted. Solid curve, prediction of the acoustic mismatch model; dashed curve, prediction of the diffuse mismatch model. From Swartz (1987).

quartz interfaces, the effect of diffuse scattering should be vanishingly small (see Table II). Figure 35 contains the measured thermal boundary resistance for that interface. The quartz surface was Syton polished. There is no hint



FIG. 33. Thermal boundary resistance between Rh:Fe and Syton-polished sapphire, and between Rh:Fe and aluminapolished sapphire. The effects of the electron-phonon thermal resistance have been subtracted. Solid curve, prediction of the acoustic mismatch model; dashed curve, prediction of the diffuse mismatch model. From Swartz (1987).



FIG. 35. Thermal boundary resistance between Rh:Fe and quartz. The solid curve represents both the diffuse mismatch model prediction and the acoustic mismatch model prediction; they coincide in this case. Note the quantitative agreement with either prediction at temperatures up to over 15 K, indicating that diffuse scattering, which must be present at some temperature well below 15 K, has no effect at this interface. From Swartz (1987).

of any effect of diffuse scattering; the absolute magnitude of the thermal boundary resistance lies right on the solid curve at temperatures up to 15 K. The solid curve is the prediction of the acoustic mismatch model (which is the same as the prediction of the diffuse mismatch model at this interface). This absence of any indication of the effect of diffuse scattering is perhaps the most compelling evidence for the validity of the diffuse mismatch model, since it is quite unlikely that the Rh:Fe-quartz interface could be smooth even for 20-K phonons. As predicted, diffuse scattering simply does not affect the boundary resistance at this interface.

2. T > 30 K: Substrate damage

Let us now turn to temperatures greater than 30 K. Here we see the strong increase in $R_{Bd}T^3$ with increasing temperature (see Figs. 6 and 32-35). We believe that this increase is not intrinsic to the boundary resistance, since it is far greater than can be explained by any improved calculation of the thermal boundary resistance. Our arguments are as follows. There is little difference between the predicted thermal boundary resistances of the acoustic and diffuse mismatch models; therefore, elastic scattering produces little uncertainty in the calculation. Conceivably, other effects could significantly affect the theory, such as discreteness of the lattice (Steinbrüchel, 1976; Lumpkin, Saslow, and Visscher, 1978) and phonon dispersion (Weis, 1979, and references therein). These effects decrease the transport across the interface at temperatures above 30 K, but not enough to explain the observations. Even though elastic anisotropy has a strong effect on heat-pulse reflection data (Taborek and Goodstein, 1979, 1980a), it has only a small quantitative effect on the thermal boundary resistance. Poor interface quality, such as incomplete contact, decreases the transport across interfaces, but this effect should lead to an increase in the thermal boundary resistance by a constant factor over the entire temperature range.²⁷ Phonon attenuation near the interface (the modified acoustic mismatch model, discussed in Secs. II and III) always increases transmission across an interface. In addition, that increase is small at solid-solid interfaces and competes with scattering at the interface. Koos et al. (1983) used a phonon imaging technique to observe an unusual angledependent phonon transmission at a copper-sapphire interface. They concluded that, because the copper film was weakly bonded to the sapphire, there existed a mechanism that coupled longitudinal phonons in the copper near the mode-conversion critical cone to transverse phonons in the sapphire via evanescent longitudinal phonons. This mechanism also increased transmission and hence could not be responsible for the rise of the boundary resistance at temperatures above $30 \text{ K}.^{28}$

Given no reasonable intrinsic mechanism, we concluded (Swartz and Pohl, 1986, 1987; Swartz, 1987) that the explanation of the strong increase in $R_{\rm Bd}T^3$ above 30 K must involve phonon scattering caused by damage in the substrate near the interface. The thermal resistance caused by this scattering is not experimentally separable from the thermal boundary resistance.

This increase in $R_{\rm Bd}T^3$ above 30 K was largest for the diamond-polished sapphire; in this sample we concluded that the final polishing step caused extensive damage both to the surface of the sapphire and to the region underneath the surface of the sapphire. The samples with the smallest upturn in $R_{Bd}T^3$ were the Syton-polished and the hydrogen-polished samples. These two samples apparently had very different surfaces (the Syton-polished surface scattered low-frequency phonons much more), but in neither sample is the region underneath the surface likely to have been damaged as severely by the final polishing treatment as by the diamond polishing. (Both hydrogen and silica are softer than sapphire.) The alumina-polished sample seemed to mirror the Sytonpolished sample; the surface of the alumina-polished sample was apparently smoother, according to the lowfrequency phonons, but the region underneath the surface of the alumina-polished sample was apparently more damaged, judging by the stronger upturn in $R_{Bd}T^3$ (see Fig. 33).

Understanding and controlling subsurface damage, such as cracks, dislocations, and grain boundaries, is critical to the understanding of transport in films and small structures. Because such damage has a large effect on the measurement of transport across interfaces, we can use this transport to study the damage. It seems reasonable to model the damaged layer as a highly disordered region of the substrate with thermal properties like those of amorphous solids. We deposited amorphous silicon dioxide layers of thicknesses varying from 70 to 1150 Å onto Syton-polished sapphire substrates using plasmaenhanced chemical vapor deposition. Afterwards, Rh:Fe films were deposited onto those layers and the effective²⁹ thermal boundary resistance of the sandwich structure was measured; see Fig. 36. Also shown in the figure are the data for the diamond-polished sample and for a sapphire sample that was bombarded with 500-V Ar⁺ ions before and during deposition of an aluminum film. The solid curve is the diffuse mismatch prediction for a single interface between Rh:Fe and sapphire (which is about the

 $^{^{27}}$ However, at temperatures below a few K, the effect of imperfect contact diminishes as the phonon wavelength becomes much larger than the spacing between points of contact between the solids (Little, 1959).

²⁸We suspect that the coupling mechanism is strongly related to the mechanism of the modified acoustic mismatch model (phonon attenuation near the interface) because that also leads to strong transmission near the critical cone.

²⁹The term "effective thermal boundary resistance" is used because the measured values include the thermal resistance of the silica films.



FIG. 36. The measured total effective thermal boundary resistance for a series of interfaces with oxide interlayers of varying thickness: \Diamond , thermal boundary resistance between Rh:Fe and diamond-polished sapphire; \bigcirc , thermal boundary resistance between aluminum and sapphire when the surface of the sapphire is damaged by Ar⁺ ion bombardment; solid curve, thermal boundary resistance at a single Rh:Fe/sapphire interface predicted using the acoustic mismatch model. The boundary resistance between aluminum and sapphire is about the same as between Rh:Fe and sapphire (see Table II); the solid curve thus also represents the predicted boundary resistance at a single aluminum/sapphire interface. From Swartz (1987).

same for aluminum on sapphire; see Table II). If we use the diffuse mismatch model and assume that phonons are not scattered in the silica layer, then the thermal resistance of the two interfaces is about 1.5 times that of a single interface. Even the thinnest silica layer caused significantly more phonon scattering than the damage from the diamond polishing. From Fig. 36 one sees that the effect of the diamond polishing is equivalent to a glassy layer of the order of about 30 Å thick. We expect that the damage in the sapphire from the diamond polishing extends significantly deeper than this. Thus, while the thermal resistance caused by the imperfect surface treatment may well swamp the intrinsic thermal boundary resistance, this added thermal resistance is small compared to the thermal resistance caused by a < 100 Å glassy interlayer.

A common method of promoting film adhesion is to etch $(Ar^+$ ion bombardment) the substrate by sputtering



FIG. 37. Thermal conductivity of thin amorphous SiO_2 layers derived from measurements shown in Fig. 36. The solid curve is the thermal conductivity of bulk amorphous SiO_2 (Cahill and Pohl, 1987). Note the drop in thermal conductivity with increasing film thickness, indicating an increase of disorder with increasing film thickness (Swartz and Pohl, 1987).

immediately prior to the film deposition (Maissel and Glang, 1970, pp. 6-41). Two methods of producing glassy excitations in quartz are neutron bombardment (see, for example, Gardner and Anderson, 1981; Laermans, 1985; de Goër, 1986) and electron bombardment (Vanelstraete and Laermans, 1986). We might expect Ar⁺ ion bombardment to have a similar effect on the outer layer of the bombardment substrate. Then, there is no longer a well-defined interface; there is instead a layer of bombarded sapphire between the metal and the sapphire. The thermal resistance of the aluminum-(bombarded sapphire)-sapphire sandwich has nearly the same magnitude as the thermal resistance of the Rh:Fesilica-sapphire with the thinnest silica layer (69 Å). We expect that the damage from the bombardment extends on the order of 50-100 Å into the sapphire.³⁰ Thus the damage in the sapphire layer must be very severe; it scatters phonons as much as if it were amorphous. These

³⁰The effects of bombarding substrates before and during deposition are quite complex. See, for example, Dautremont-Smith and Feldman (1985) or Rossnagel and Cuomo (1987) and references therein.

findings suggest that the price of improving mechanical contact by sputter etching is a weakening of the thermal contact.

From the data shown in Fig. 36 the thermal resistance of the silica layers was determined; from that, their thermal conductivity was determined by subtracting the thermal resistances of the interfaces; see Fig. 37. While this procedure is somewhat uncertain, since these thermal resistances do not simply add (see Sec. II, footnote 7), this procedure should lead to negligible errors for the thickest films, since they almost completely dominate the measured total thermal resistance above approximately 20 K. Yet the thermal conductivity obtained for the 1150-Å thick film differs from the known values for bulk silica (solid curve in Fig. 37) by the largest amount. We believe that the only way in which the thermal conductivity of an amorphous solid can be lowered below its bulk value in this temperature range is through microscopic flaws like cracks or voids, which significantly increase the distance heat must travel. We conclude that these measurements indicate the presence of such defects in our silica films. See Cahill et al. (1988) for further discussion of the thermal conductivity of thin films. In the very-large-scale integrated circuit industry, one of the problems associated with miniaturization is heat removal from the devices and the interconnecting thin films. The glassy layer often used between successive layers in the circuit is expected to create a thermal bottleneck. The effect of interfaces and in particular the effect of the unexpectedly low thermal conductivity of the glassy interlayers on heat removal from integrated circuits is discussed by Swartz (1987) and Swartz and Pohl (1987).

D. Inelastic scattering

If the temperature is much lower than the Debye temperatures of both solids, then the thermal boundary resistance at a solid-solid interface is not sensitive to whether a scattering event is elastic or inelastic. The reason is that the phonon density of states on both sides of the interface has the same frequency dependence $(\sim \omega^2)$ for low frequencies (much smaller than the Debye cutoff frequencies). Therefore the probability of scattering forward or back is independent of frequency.³¹ At higher temperatures, however, where the density of states no longer varies as ω^2 , at least on one side of the interface, inelastic scattering can have a large effect. For example, at an interface between lead and diamond at temperatures above 80 K, a majority of phonon modes excited in the diamond have frequencies above the maximum fre-

quency in the lead. All of these phonons must be reflected at the interface if inelastic processes are not allowed. If, on the other hand, they can be downconverted, the resulting phonons will be predominantly transmitted. Thus one of the ways that inelastic scattering at solid-solid interfaces can be observed is through a decrease of the thermal boundary resistance below the values predicted by either the diffuse or the acoustic mismatch model, when the temperature is comparable to the Debye temperature of the softer solid. The effect would be pronounced if the Debye temperatures of the two solids are very different and if one of the Debye temperatures is very low. It can be argued that the theoretical predictions of both the acoustic mismatch model and the diffuse mismatch model are too uncertain near room temperature to make such an observation meaningful. However, the magnitude and, in particular, the sign of the uncertainty are such that any observation of enhanced transport (lower boundary resistance than predicted using either model) is likely to be evidence for inelastic scattering. We cannot think of any mechanism other than inelastic scattering which would significantly decrease the boundary resistance at temperatures near the Debye temperature. See the above discussion of substrate damage at temperatures above 30 K for examples of ways that the boundary resistance might be increased in this temperature range.

In the previous subsection, we saw that for most thinfilm/substrate pairs at temperatures above 40 K, independent of measurement method, the heat transport across the interface is less than that expected from either boundary resistance model (acoustic or diffuse mismatch). However, evidence for inelastic scattering may have been seen in heat-pulse experiments on gold/diamond and lead/diamond interfaces (see Fig. 30) in which the heat transport exceeded the expected values. We suggest that some of the phonons in the diamond which would be reflected in the absence of inelastic scattering are being transmitted after having been scattered inelastically.

There are two more cases in which heat transfer greater than that predicted has been observed: Young et al. (1986) measured the thermal boundary resistance between gold and vitreous silica at room temperature using picosecond pulsed-laser techniques. They concluded that their measured value was a factor of 2 below their prediction based on acoustic mismatch. In the second, evidence was found in the thermal boundary resistance between a deposited film of gold and sapphire (Swartz and Pohl, 1986, 1987) from 10 K to about 300 K. At low temperatures (below 50 K) the measured thermal boundary resistance was a factor of 3 higher than that predicted using either diffuse or acoustic mismatch, presumably due to a small effective area of contact (thin films of gold adhere very poorly to most substrates). In spite of this enhancement of the thermal boundary resistance (which was not seen for any of the other film-substrate pairs), the measured boundary resistance at room temperature was

³¹This does not mean that inelastic scattering at solid-solid interfaces cannot be probed at low temperatures. See, for example, Challis, Ghazi, and Wybourne (1982), Challis (1983, 1986), and Challis, Kenmuir, Heraud, and Russell (1986) for a experimental arrangement that is sensitive to inelastic scattering at solid surfaces at temperatures low compared to the Debye temperature of the solid. This experiment is discussed in Sec. III.C.3.d.

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only about 40% higher than the theoretical prediction. If the area of contact is normalized so that the low-temperature boundary resistance matches the acoustic mismatch model prediction, then at high temperatures the boundary resistance is about a factor of 2 lower than expected, in general agreement with the results of both types of pulsed measurements described above.

E. Epitaxial films

In the work reviewed above, none of the solid-solid interfaces studied are ideal on the atomic scale. Certainly, characterization of intentionally added defects and other sources of scattering would be easier for a nearly atomically perfect interface, such as an interface grown using molecular-beam epitaxy. Only certain combinations of solids can form an epitaxial interface, the most common combination being GaAs/AlGaAs used in superlattices. Narayanamurti et al. (1979) studied the phonon transmission in a direction normal to the layers through a GaAs/AlGaAs superlattice with 100 interfaces. The phonon frequencies used ranged from 75 GHz (the detector was an Al tunnel junction) to 285 GHz (the generator was a Sn tunnel junction). They found selective transmission consistent with classical Bragg scattering, or stopbands. Although precise calculation of the average transmission coefficient of the phonons at a single interface is difficult from the data, the diffuse scattering at each interface must have been negligible in order to have observed the stopband.³²

Using phonon imaging techniques (see, for example, Wolfe, 1980, or Northrop and Wolfe, 1980), Hurley *et al.* (1987) and Tamura, Hurley, and Wolfe (1988) observed the phonon transmission through a 40-period (80interface) $In_{0.15}Ga_{0.85}As/AlAs$ epitaxial superlattice for arbitrary phonon propagation and polarization, for phonon frequencies in the range of 700–900 GHz. The fact that such high-frequency phonons would travel through so many interfaces with negligible diffuse scattering indicates that these interfaces are also nearly ideal. Hurley *et al.* (1988) used the same technique and frequency range to observe phonon stopbands in an epitaxial Fibonacci superlattice with over 750 interfaces.

Data on phonon interaction with epitaxial interfaces are essential to the understanding of phonon scattering, yet it is apparently limited to the above studies. For example, as far as we know, there are no studies of the interactions of phonons at a single epitaxial interface and no studies of interactions at epitaxial interfaces with intentionally added defects. Such studies would certainly be valuable.

F. Summary

Below approximately 30 K, the solid-solid thermal boundary resistance can be well described with either the acoustic mismatch or the diffusive mismatch model. The reason why either model works well is that the effect of diffuse scattering at the interface is small, in contrast to the liquid-helium-solid interface. This good agreement between theory and experiment is dependent on good contact between the two solids, resulting, for example, from the evaporation of one substance onto the other; early reports of evidence for disagreement between theory and experiment are explained through imperfect contacts. The difference between acoustic and diffuse transmission has been observed in one case, namely, for the interface between iron-doped rhodium (Rh:Fe) and sapphire.

Above 30 K, the observed thermal boundary resistance can be as much as an order of magnitude larger than that predicted by either theoretical model and also depends on the surface treatment of the substrate. Evidence has been presented that this additional resistance cannot be associated with the interface itself, but must be caused by disorder in the subsurface region. This region scatters high-frequency phonons as effectively as an amorphous layer approximately 50 Å thick. However, the dominant carriers of heat below 30 K, which have frequencies less than \sim 3 THz, are little affected by this disorder. Independent evidence for this disorder is lacking at this time. In particular, the nature and extent of the damage are unknown.

V. CONCLUSIONS

Almost five decades after its discovery, the Kapitza resistance, i.e., the thermal boundary resistance between a solid and liquid helium, is still not fully understood. The acoustic mismatch model suggested by Khalatnikov and independently by Mazo provides a fundamental, qualitative explanation. However, it provides only an upper limit to the resistance. There are many different mechanisms that can lead to a reduction of the Kapitza resistance, and in most real cases the resistance results from several mechanisms acting simultaneously. A lower limit of the Kapitza resistance is given by the diffuse mismatch model, which assumes that all phonons are scattered at the interface. It leads to a thermal boundary resistance between typical solids and liquid helium that is several orders of magnitude smaller than that predicted using the acoustic mismatch model. The observed Kapitza resistances always lie between these two limits.

For solid-solid interfaces, the two theoretical limits of the thermal boundary resistance, based on the acoustic mismatch and the diffuse mismatch model, differ typically only by a small fraction (<30%). Therefore the experimentally observed thermal boundary resistances agree well with either model. Significant deviations are probably the result either of partial contact or of bulk

 $^{^{32}}$ In a similar experiment, Koblinger *et al.* (1986) observed phonon stopbands for normal incidence in amorphous (not epit-axial) SiO₂/Si superlattices.

disorder in the near-surface region. Independent evidence for this disorder is not available at this time. Considering its importance for heat transfer across interfaces near and above room temperature, an understanding of this disorder is highly desirable.

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