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Kapitza conductance and heat flow between solids at temperatures from 50 to 300 K

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Measurements of the Kapitza conductance σ_K made using a picosecond optical technique at temperatures between 50 and 300 K are presented for interfaces between metals and dielectrics. The Debye temperatures Θ_D of the metals (Pb, Au, Al, and Ti) were in the range from 102 to 426 K, while those of the dielectrics (BaF_2, sapphire, and diamond) varied from 287 to 2200 K. Conductances measured between materials with Θ_D differing by less than about a factor of 5 were found to be in reasonable agreement with calculations based on a lattice dynamical theory. However, for more widely mismatched solids the measured conductances were found to be greatly in excess of the lattice dynamical calculations. In some of these cases the conductances exceeded even the phonon radiation limit, indicating that much of the heat flow between the solids was via an inelastic channel. It was demonstrated experimentally that the inelastic channel does not involve an interaction between the phonons in the dielectric and electrons in the metal. We show that the anomalously large conductance can be understood in terms of a model in which the anharmonicity of the metal plays an important role.

I. INTRODUCTION

A discontinuity in temperature arises across an interface between two materials through which a heat current flows.[1,2] This was first noticed by Kapitza for interfaces between copper and liquid helium.[3] The proportionality constant relating the temperature jump ΔT to the heat current density $\dot{Q}$ is known as the Kapitza conductance σ_K. Thus,

$$\dot{Q}=\sigma_K \Delta T \ . \tag{1}$$

The temperature jump arises because the probability $t_{\mathbf{k}j}$ that a phonon of wave vector **k** and polarization j will be transmitted across the interface is less than unity. This limits the heat current out of the hotter medium, and so the interface acts as a thermal barrier across which a temperature difference can be sustained.

The Kapitza conductance is the derivative of the net heat flux transmitted across an interface with respect to the temperature difference between the two materials. Therefore, if we consider an interface between materials A and B we can write σ_K as follows:

$$\sigma_K(T)=\frac{1}{V}\frac{\partial}{\partial T}\sum_{\mathbf{k}j}^{A}\hbar\omega_{\mathbf{k}j}n(\omega_{\mathbf{k}j},T)|v_{\mathbf{k}jz}|t_{\mathbf{k}j} \tag{2}$$

$$=\frac{1}{V}\frac{\partial}{\partial T}\sum_{\mathbf{k}j}^{B}\hbar\omega_{\mathbf{k}j}n(\omega_{\mathbf{k}j},T)|v_{\mathbf{k}jz}|t_{\mathbf{k}j} \ , \tag{3}$$

where the labels A and B on the sums indicate that all quantities in each sum correspond to phonons incident on the interface from A and B, respectively. In Eqs. (2) and (3), $\omega_{\mathbf{k}j}$ is the phonon frequency, $v_{\mathbf{k}jz}$ is the component of the phonon group velocity normal to the interface, $n(\omega_{\mathbf{k}j},T)$ is the Bose-Einstein distribution function, and V is the volume (assumed the same for materials A and B). The sums are over all phonons in the Brillouin zone for which $v_{\mathbf{k}j}$ is directed toward the interface.

One can anticipate that the dependence of σ_K on the properties of A and B will be influenced by the following factors governing the transmission coefficients.

(1) A phonon incident on the interface from A whose wave vector makes an angle θ_i with the surface normal is refracted in accordance with Snell's law into an angle $\theta_t=\sin^{-1}(v_B\sin\theta_i/v_A)$. If $v_A<v_B$, phonons whose angles of incidence exceed a critical value experience total internal reflection. This gives rise to a *critical cone* for phonons incident on A from B. Only phonons in A whose wave vectors lie inside this critical cone can have nonzero transmission coefficients.

(2) Even at normal incidence the transmission coefficients are affected by the discontinuity in sound velocity and density at the interface. At low temperatures, where the phonons may be treated as elastic waves, $t_{\mathbf{k}j}$ can be calculated from the laws of classical acoustics. For a wave in A normally incident on B the transmission coefficient is

$$t_{\mathbf{k}j}=\frac{4Z_AZ_B}{(Z_A+Z_B)^2} \ , \tag{4}$$

where Z_A and Z_B are the acoustic impedances given by the product of the mass density and sound velocity in A and B, respectively. Similar relations can be written which take into account the polarization and angle of incidence of the wave.[4,5]

(3) The discontinuity in the phonon density of states across the interface can also play an important role in determining the transmission coefficients. Assume that a phonon in A of frequency $\omega_{\mathbf{k}j}$ can couple only to phonons of the same frequency in B. If the number of states available in B with this frequency is small compared to the number in A, then the transmission coefficients for phonons incident from A onto B must also be small. In general, one expects that when the Debye temperatures

Θ_D for A and B are very different, there will be a large difference in the phonon density of states, and consequently a small value for the average phonon transmission coefficient.

A large number of experiments have been carried out for interfaces between solids at liquid-helium temperatures[2,6–10] and the agreement between experiment and the general physical picture we have described has been satisfactory. Substantially fewer measurements have been made at higher temperatures (i.e., at room temperature). This is because at high temperatures the small value of the Kapitza resistance makes it hard to detect the jump in temperature at the interface in the presence of the thermal gradients that always occur within the volumes of the bulk materials on either side of the interface. Recently several new techniques have been developed which overcome this problem. Martinon and Weis[11] used Joule heating to raise the temperature of thin metal films deposited on dielectric substrates. The substrate temperature was kept constant at 4.2 K and the temperature rise in the film was measured via the change in its resistivity. Measurements were made for a variety of interfaces with temperature rises up to about 100 K, and reasonable agreement was found with calculations based on a continuum elasticity model incorporating the general principles discussed above. More recently, Swartz and Pohl[12] developed a technique in which metal films were patterned lithographically into pairs of 10μ wires. One of the wires served both as a heat source and thermometer, and the second wire was used to measure the substrate temperature a short distance away. The Kapitza conductance was determined by fitting the temperatures measured at the two wires to a continuum heat flow model. By using samples of small dimensions this technique reduces the temperature drop between the metal wires to a point where it no longer totally dominates the Kapitza drop, even at moderately high temperatures. To determine Kapitza conductances from these experiments it is necessary to know the thermal conductivity of the substrate accurately. Measurements have been reported for a variety of interfaces using this technique,[13,14] and agreement between these measurements and calculations based on continuum elasticity models in which dispersion is accounted for in an approximate way is found to be remarkably good.

In this paper we report the results of experiments in which a picosecond optical technique was used to measure σ_K at temperatures up to 300 K.[15] Measurements have been made at interfaces between four metals (Al, Ti, Au, and Pb) with Debye temperatures ranging from 426 to 102 K and three dielectrics (diamond, sapphire, and BaF_2) whose Debye temperatures range from 2240 to 287 K.[16] We review our technique and present our experimental results in Sec. II. In Sec. III, we compare our results with theoretical calculations and show that for the interfaces between solids with greatly differing Θ_D's, an inelastic energy transfer mechanism is needed to explain the experimental results. We consider possible inelastic mechanisms and present model calculations which show that the anomalous results may be associated with anharmonicity in the metals.

II. EXPERIMENT

A. Samples

The samples consisted of thin metal films evaporated onto dielectric substrates. A summary of the properties[17–20] of the materials used in the experiments is given in Table. I. The method of substrate surface preparation depended on the material. Barium fluoride optical flats[21] were lightly repolished with 0.03μ alumina grit in deionized water. Several micrometers of material were removed in this way along with residual scratches, leaving surfaces on which no scratches were visible under a high-power optical interference microscope. The sapphire substrates were polished with Syton (colloidal SiO_2 in water) on a tin lap.[22] From reflection high-energy electron diffraction it was determined that the surfaces of these substrates were virtually atomically perfect. The diamond substrates were grown[23] in a high-pressure liquid metal cell from isotopically pure (99.93%) ^{12}C. Etch-pit studies of the surfaces revealed fewer than 10^6 defects per cm^2. Both as-grown and mechanically polished diamonds were used in our experiments with similar results. We used (100) surfaces for all diamond and BaF_2 substrates. The sapphire was cut perpendicular to the $[2\bar{1}\bar{1}0]$ direction.

Final cleaning of the substrates was carried out in cleanroom conditions. Residual grit was removed from the polished BaF_2 surfaces by further polishing with deionized water. The sapphire and diamond substrates were immersed in 1:1, H_2SO_4:H_2O at 85–100 °C for 3 min to remove organic surface contamination, then rinsed in deionized water and transferred to a 1:1:5 solution of H_2O_2:NH_4OH:H_2O at 75–85 °C for 5 min, followed by further rinsing in deionized water.

Metal films were evaporated from high-purity sources onto the surfaces of the substrates in a vacuum of less than 10^{-6} torr. As high a growth rate as practicable (≥ 10–20 Å/s) was used in order to achieve the best possible quality films. The electrical conductivities of the films were found to be consistent with published values for the bulk materials indicating both that they were continuous and of good quality.

B. Experimental technique

In our picosecond optical experiments a laser pulse was focused onto a small spot on the surface of the metal film.

TABLE I. Deybe temperature Θ_D, Debye velocity c_D, and mass density ρ for the materials studied. The data are taken from Ref. 17.

Material	Θ_D (K)	c_D (10^5cm/s)	ρ (g/cm^3)
Pb	102	0.92	11.34
Au	165	1.42	19.3
Al	423	3.62	2.70
Ti	426	3.73	4.50
Diamond	2240	13.43	3.51
Sapphire	1024	7.03	4.89
BaF_2	287	3.76	3.97

Partial absorption of this pulse (the pump) led to a small rise in the temperature of the metal film, which then cooled via conduction of heat into the substrate. The change $\Delta T(t)$ in the temperature of the metal film led to a small change in its optical reflectivity $\Delta R(t)$ which we measured using a second laser pulse (the probe).[24] This was time delayed relative to the pump pulse. Since the reflectivity of metals varies slowly with temperature[25] we can assume that $\Delta T(t) \propto \Delta R(t)$. The experimental arrangement is shown schematically in Fig. 1. From the measured cooling curve of the metal film the Kapitza conductance at the film-substrate interface could be determined as described below.

A room-temperature cooling curve for a Pb film deposited on diamond is shown in Fig. 2(a). The pump light pulse was absorbed by the electrons in the metal film with most of the energy being deposited within an absorption length ζ of the surface (for Pb, $\zeta \simeq 260$ Å when $\lambda = 6330$ Å). This produced a sudden rise in the local temperature of the electrons near the surface of the metal. In some metals this is evidenced by a sudden change in the optical reflectivity consisting of either a negative- or positive-going spike[26] very close to $t=0$. In Pb, this signature of electron heating is not seen, owing to the details of its electronic band structure; one sees instead a more gradual onset as the electrons give up their energy to the lattice over a period of several picoseconds.[27] The heated electrons diffused away from the surface of the film while cooling, depositing their energy by phonon emission throughout the film. The initial heat distribution set up in the film was therefore influenced by many factors, including the optical absorption length, the electronic mean free path, and the electron-phonon coupling strength. Intrinsic phonon lifetimes in solids are typically on the order of 10–50 ps (Ref. 28) so that the heat in the film was well described by a local temperature within about 100 ps, depending somewhat on the metal. For the film thicknesses used in our experiments, the film temperature could be considered to be uniform since the temperature variation across the film thickness was much smaller than the temperature jump that existed at the film-substrate interface. The initial drop in $\Delta R(t)$ seen in Fig. 2(a) just after $t=0$ corresponds to the rapid cooling of the region of the film near its front surface due to conduction into the interior of the film; the slower cooling that occurs at later times is due to energy transfer into the substrate. Heat flow in the film parallel to the interface is much slower than the heat flow across the interface and so can be ignored.[29,30]

The thicknesses of the metal films were determined as follows. The initial rise in the metal lattice temperature produced a thermal stress distribution in the film, which on relaxing launched a stress wave from the surface toward the interface. Because of the differing acoustic impedances of the metal and substrate this wave was partially reflected and some of it returned to the surface, causing a change in the optical reflectivity of the metal film. Repeated acoustic reflections within the film gave rise to features such as those seen in Fig. 2(b) at intervals corresponding to the round trip time $2d/v_s$ for longitudinal sound in the film (v_s is the longitudinal sound velocity). The detailed shape of these acoustic echoes is influenced by several factors involved with their generation and detection.[31–33] From the 84 ps round trip time,

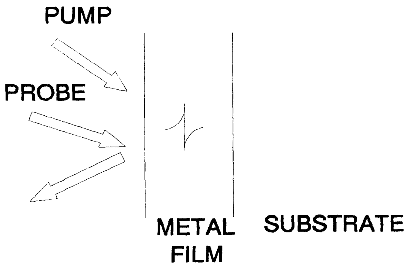

FIG. 1. Schematic diagram of the experiment. The metal film of thickness d is heated by the pump light pulse. The change in the optical reflectivity of the metal film due to its temperature change is measured by means of the time-delayed probe pulse. The sudden rise in temperature of the metal film also generates a stress wave which bounces back and forth inside the film.

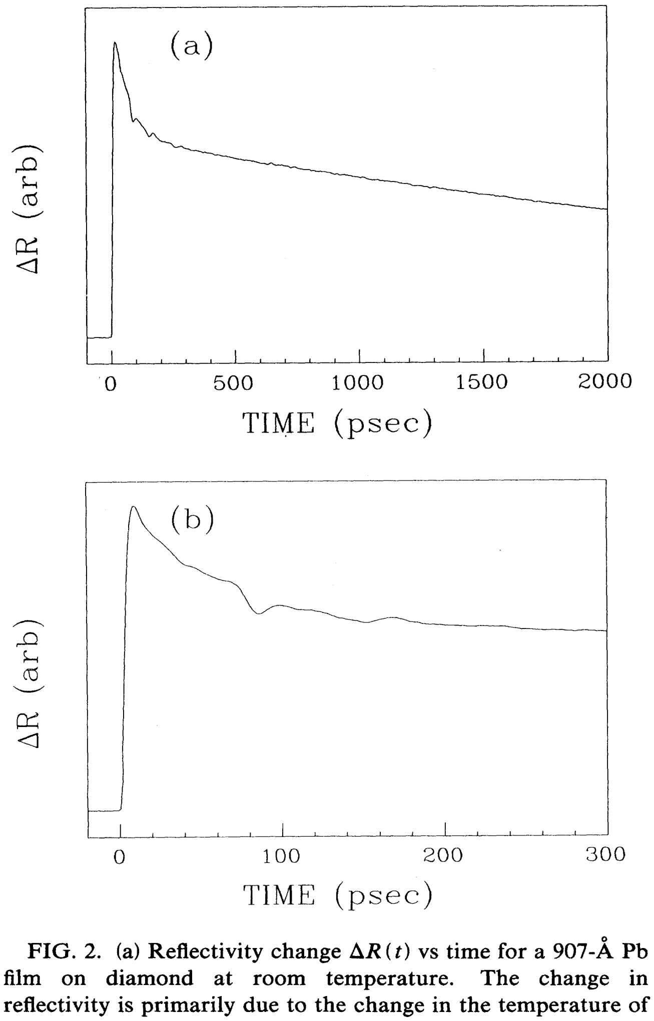

FIG. 2. (a) Reflectivity change $\Delta R(t)$ vs time for a 907-Å Pb film on diamond at room temperature. The change in reflectivity is primarily due to the change in the temperature of the film. (b) Data taken for the same sample but on a shorter time scale. The bumps in $\Delta R(T)$ at 84 and 168 ps are due to the stress wave bouncing back and forth inside the film.

and the sound velocity for Pb[34] which is 21.6 Å ps^{-1} (assuming our films to be polycrystalline of random orientation), we obtain a Pb film thickness of approximately 907 Å. The thickness of all of the films used in the experiments were found in a similar way.

Measurements were carried out in an optical cryostat at temperatures between 50 and 300 K. The samples were carefully thermally anchored to the cold finger of the cryostat in order to prevent a rise in the steady-state temperature of the substrate caused by successive laser pulses. For diamond and sapphire, whose thermal conductivities are high, we estimate that the steady-state temperature rise was negligibly small and so the substrate and cold finger temperatures were assumed equal. However, the thermal conductivity of $\mathrm{BaF_2}$ is much lower and the steady-state heat flow *through* the substrate could give rise to a significant difference in temperature between the cold finger and the film-substrate interface. Therefore with the $\mathrm{BaF_2}$ samples it was necessary to use less laser power than was used with the diamond and sapphire samples, and this led to a reduced signal-to-noise ratio. With the lower power we estimate the steady-state rise in temperature near the interface of $\mathrm{BaF_2}$ to have been about 8 K in the worst case.

The laser source used in the experiments was a hybrid mode-locked dye laser operated at a wavelength of 632 nm, having a repetition rate of 76 MHZ, and producing pulses of less than 150 fs FWHM.[35] The pump and probe beams were focused onto the sample through a single 5-cm focal length lens giving a spot of approximately 20 μm in diameter. From the heat capacity of a typical film, and a pump pulse energy of ≈ 0.1 nJ we estimate the initial temperature rise to have been typically on the order of 1 K. The corresponding change in the reflectivity of the metal films was on the order of 10^{-3} to 10^{-6}. To improve the signal-to-noise ratio, the pump beam was chopped at 1 MHz by an acousto-optic modulator. The reflected probe intensity was converted to an electrical signal in a photodiode, and the in-phase component at the chopping frequency was recorded by a computer at each delay time. The signal-to-noise ratio was improved by averaging over a number of traces, typically 10.

C. Analysis

The cooling of the metal film is governed by the following equations:

$$C_f d\frac{\partial T_f}{\partial t} = -\sigma_K[T_f - T_S(0)] , \tag{5}$$

$$C_S\frac{\partial T_S(z)}{\partial t} = \kappa_S\frac{\partial^2 T_S(z)}{\partial z^2} , \tag{6}$$

where T_f is the temperature of the film, $T_S(z)$ is the temperature of the substrate at a distance z from the interface, C_f and C_S are the specific heats of the film and substrate, and κ_S is the thermal conductivity of the substrate. Equation (5) relates the heat flux across the interface to the Kapitza conductance [see Eq. (1)], and Eq. (6) describes thermal diffusion in the substrate. Because of the large area of the film that is illuminated by the light pulses it is legitimate to neglect heat diffusion parallel to the interface.

For diamond the thermal conductivity is so large[36] that the transient rise in T_S as the film cools is negligible. Consequently, from Eq. (5) one finds that the excess temperature of the film decays exponentially with a time constant τ which is equal to $C_f d/\sigma_K$. We obtained τ from the slope of a plot of $\ln\Delta R(t)$ as a function of t. The Kapitza conductance could then be found from τ by using the measured value of d and literature values of C_f.

For sapphire and $\mathrm{BaF_2}$ the transient rise in T_S near the interface as the film cooled, although small, could not be neglected and so the cooling curves were not exponential. We assumed trial values for σ_K and κ_S and solved Eqs. (5) and (6) numerically to obtain the cooling curve. Both σ_K and κ_S were then varied to obtain the best fit to the data. The quality of the fit depended strongly on σ_K but only weakly on κ_S. The best fit to the data was usually obtained for κ_S smaller than typical published bulk values by a factor of 2 to 5. This may indicate that a very thin damaged layer remained near the surface of these substrates after polishing. The experiment is sensitive to the thermal conductivity within a surface layer of thickness a few microns.

D. Results

The results are summarized in Figs. 3–5, together with theoretical curves which are discussed below. Room temperature results for $\hat{\sigma}_K$ are summarized in Table II. Errors arise from the uncertainty in the fitting procedure described above, as well as from the uncertainties in d and C_f. For a given sample the uncertainty in σ_K was typically ±10% for diamond and sapphire, and was ±20% for $\mathrm{BaF_2}$. For interfaces between metal films and the diamond and sapphire substrates we investigated the variation between different samples and found it to be ±10% for Al and Ti, and ±20% for Au and Pb. We were unable to get reliable results for Ti and $\mathrm{BaF_2}$. As already mentioned, for $\mathrm{BaF_2}$ it was necessary to lower the laser power to avoid steady-state heating, and at this lower power level the reflectance change of the Ti film was very small.

TABLE II. Values of Kapitza conductance in units of $\mathrm{W\,cm^{-2}\,K^{-1}}$ measured at room temperature for the combinations of metals and dielectrics studied. For each material the Debye temperature is indicated in parentheses.

	Pb (102K)	Au (165 K)	Al (423 K)	Ti (426 K)
$\mathrm{BaF_2}$ (287 K)	6 200	4 000	10 000	
Sapphire (1024 K)	5 500	4 500	10 500	11 200
Diamond (2240 K)	3 100	4 000	4 600	10 000

III. COMPARISON WITH THEORY

A. Comparison with lattice dynamical theory

A quantitative interpretation of our results must be made using a theory which can be applied at high temperatures. We have therefore compared our results with a lattice dynamical theory which takes into account in an approximate way the effects of phonon dispersion and the form of the density of states of the metals. The theory has already been described in detail;[37] here we present a summary of the major assumptions and the results of our calculations.

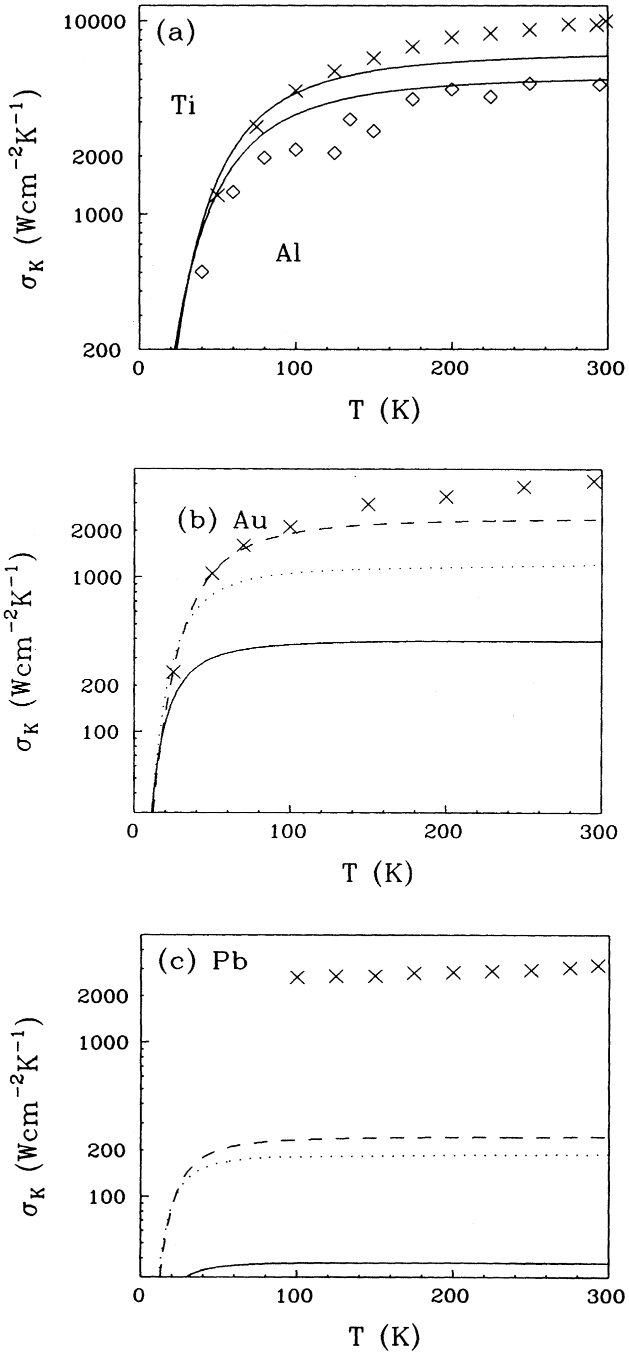

FIG. 3. The Kapitza conductance as a function of temperature between metals and diamond: (a) Ti (×) and Al (◇), (b) Au, and (c) Pb. The solid curves are the results of lattice dynamical calculations. The dotted curves in (b) and (c) show the diffuse mismatch limit, and the dashed curves indicate the radiation limit.

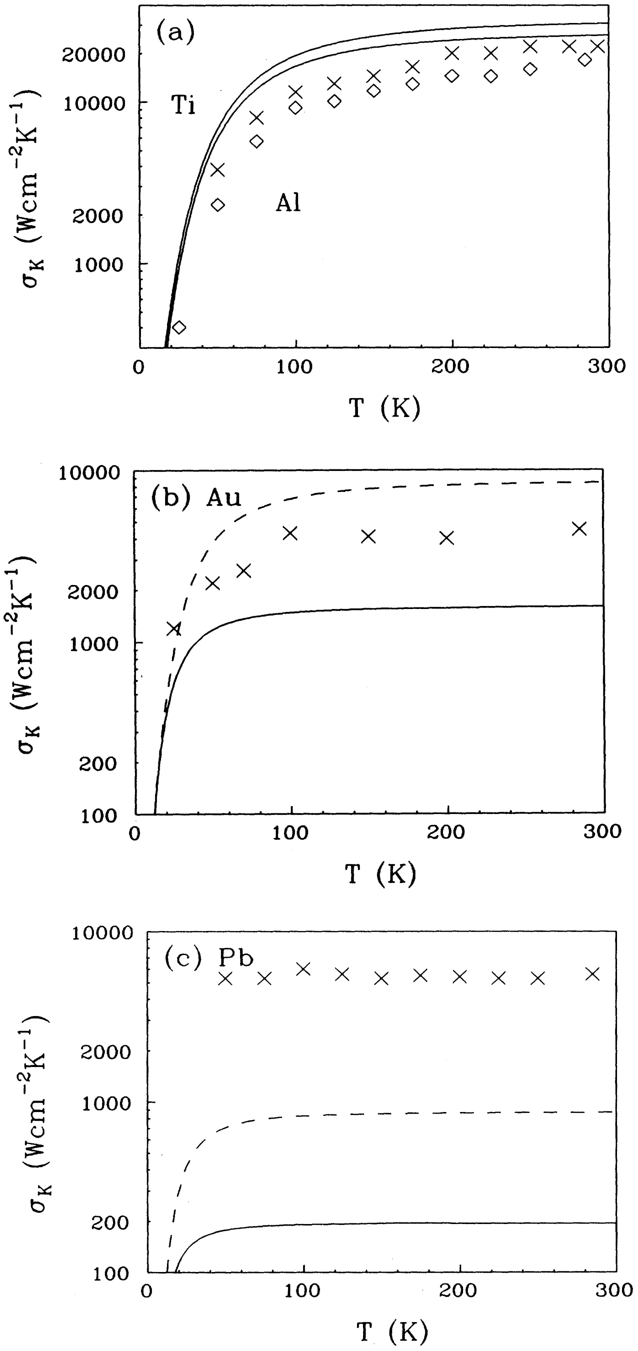

FIG. 4. The Kapitza conductance as a function of temperature between metals and sapphire: (a) Ti (×) and Al (◇), (b) Au, and (c) Pb. The solid curves are the results of lattice dynamical calculations. The dashed curves in (b) and (c) indicate the radiation limit.

In the theory both solids are replaced by monatomic fcc lattices with harmonic nearest-neighbor interactions. The lattices are assumed to both have the atomic spacing of the metal and to be in registry at the interface. The atomic masses and interatomic force constants in each lattice are chosen in such a way as to make the mass densities ρ and Debye velocities c_D of the model solids come out to be the same as those of the actual solids.

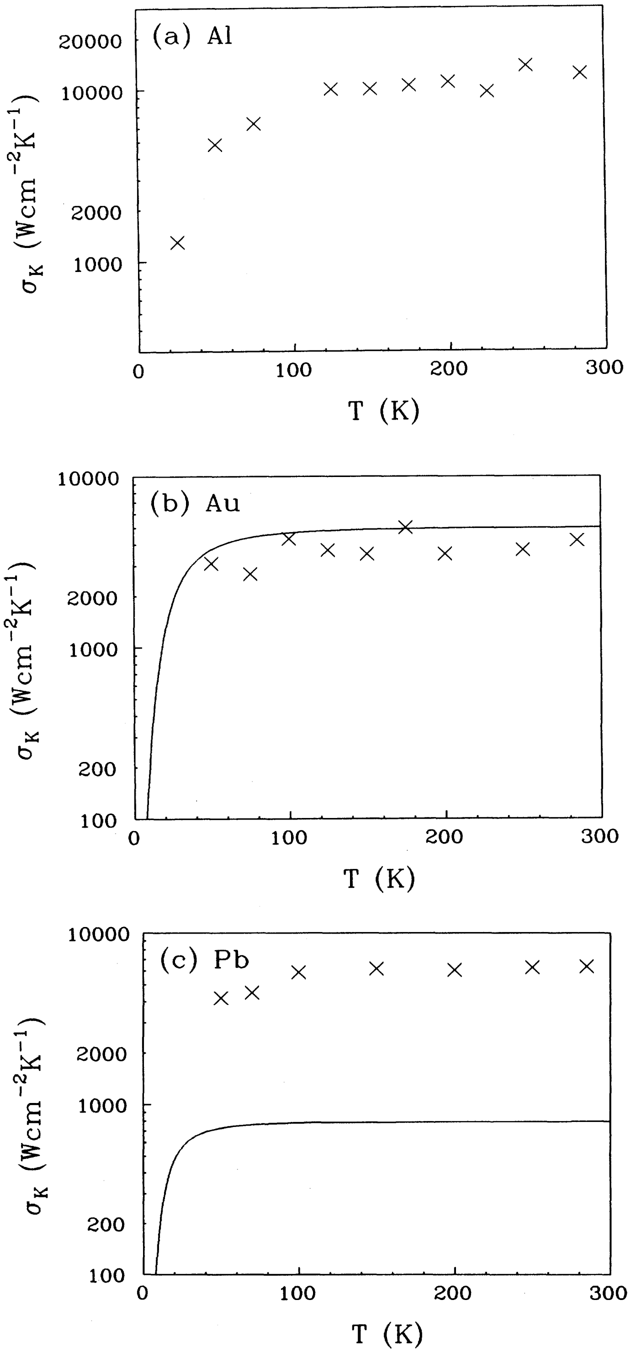

FIG. 5. The Kapitza conductance as a function of temperature between metals and BaF$_2$: (a) Al (◇), (b) Au, and (c) Pb. The solid curves in (b) and (c) are the results of lattice dynamical calculations.

The Kapitza conductance was calculated from Eq. (2) which for computational efficiency we divided into two parts as follows. We choose the metal to be side A and introduce the quantity $F_A(\omega)$ defined by

$$F_A(\omega)\equiv\frac{1}{V}\sum_{\mathbf{k}j}^{A}|v_{\mathbf{k}jz}|t_{\mathbf{k}j}\delta(\omega-\omega_{\mathbf{k}j}) \ . \tag{7}$$

$F_A(\omega)$ is a measure of the contribution to the flux arising from phonons at frequency ω. The Kapitza conductance is then

$$\sigma(T)=\int_0^\infty\frac{\partial n(\omega,T)}{\partial T}\hbar\omega F_A(\omega)d\omega \ . \tag{8}$$

The transmission coefficients $t_{\mathbf{k}j}$ are determined by finding the ratio of the transmitted to the incident energy flux. There are up to three phonons in the substrate of frequency $\omega_{\mathbf{k}j}$ to which the incident phonon can transfer energy. If we write the amplitudes of the incident phonon as A_0 and the amplitudes of the transmitted phonons as A_q ($q=4,5,6$) then

$$t_{\mathbf{k}j}=\frac{\rho_A\sum_{q=4}^{6}v_{qz}A_q^2}{\rho_B v_{0z}A_0^2} \ , \tag{9}$$

where ρ_A and ρ_B are the mass densities of the metal and the substrate, v_{0z} is the z component of the group velocity of the incident phonon, and v_{qz} ($q=4,5,6$) are the group velocities of the transmitted phonons. The amplitudes of the incident, transmitted, and the three reflected phonons are related by the following set of equations derived from the boundary conditions at the interface:

$$\sum_{q=1}^{6}M_{pq}A_q=-M_{p0}A_0 \ . \tag{10}$$

Expressions for the coefficients M_{qp} were misprinted in Ref. 37 and so we have included corrected expressions in the Appendix.

We obtained $F_A(\omega)$ in the following way. Approximately 30 000 wave vectors distributed randomly in the Brillouin zone of the metal having group velocities directed toward the interface were chosen. For each wave vector the three corresponding frequencies were determined. For each frequency the three corresponding phonons in each lattice propagating away from the interface (i.e., reflected into the metal, or transmitted into the dielectric), or which decayed exponentially with distance from the interface were identified. Transmission coefficients were found using Eq. (9) for each of the three incident phonons and together with the associated group velocities these were used to evaluate Eq. (7). The Kapitza conductance was then found from Eq. (8).[38]

The lattice models were also used to compute phonon densities of states $D(\omega)$ for the metals and substrates materials via the formula

$$D(\omega)=\frac{1}{V}\sum_{\mathbf{k}j}\delta(\omega-\omega_{\mathbf{k}j}) \ . \tag{11}$$

The results of these calculations are shown in Fig. 6. For

Pb, Au, and Al, which form fcc lattices, the calculated densities of states are in reasonable agreement with more elaborate models that are designed to reproduce the measured phonon dispersion curves. Titanium forms a hcp lattice, and so, although our model gives approximately the correct cutoff frequency, the density of calculated states is noticeably different from that calculated using a more realistic model. For the dielectrics the fcc models provide an adequate representation of the density of states at low frequencies, but fail completely to account for the part of the spectrum associated with optical phonons. However, this failure is not important for the Kapitza calculation if the Debye temperature of the metal is much less than that of the dielectric since then all of the phonons coming from the metal couple only to the acoustic phonon part of the spectrum in the dielectric. Consequently the calculations should be reasonably accurate for all of the metals on diamond, and for Pb and Au on sapphire, but much less reliable for Al and Ti on sapphire,[39] and for Pb and Au on BaF_2. We did not attempt calculations for Al and Ti on BaF_2.

Comparison of theory and experiment reveals the following. For Al and Ti on diamond and sapphire the calculated and measured σ_K's are in reasonable agreement over the entire range of temperatures [Figs. 3(a) and 4(a)]. Differences of the amounts seen in these cases could easily have been caused by the approximate nature of our lattice models. There is reasonable agreement for Au on BaF_2, but based on the discussion above this may be fortuitous. For Pb on BaF_2 [Fig. 5(c)] the theory underestimates the conductance by about a factor of 8. For Au and Pb on diamond and sapphire, the measured conductances are substantially larger than the calculated values [Figs. 3(b) and 3(c), and 4(b) and 4(c)]. The greatest discrepancy is for Pb on diamond, and is about a factor of 50. This is the combination of materials for which the lattice dynamics model should be most reliable.

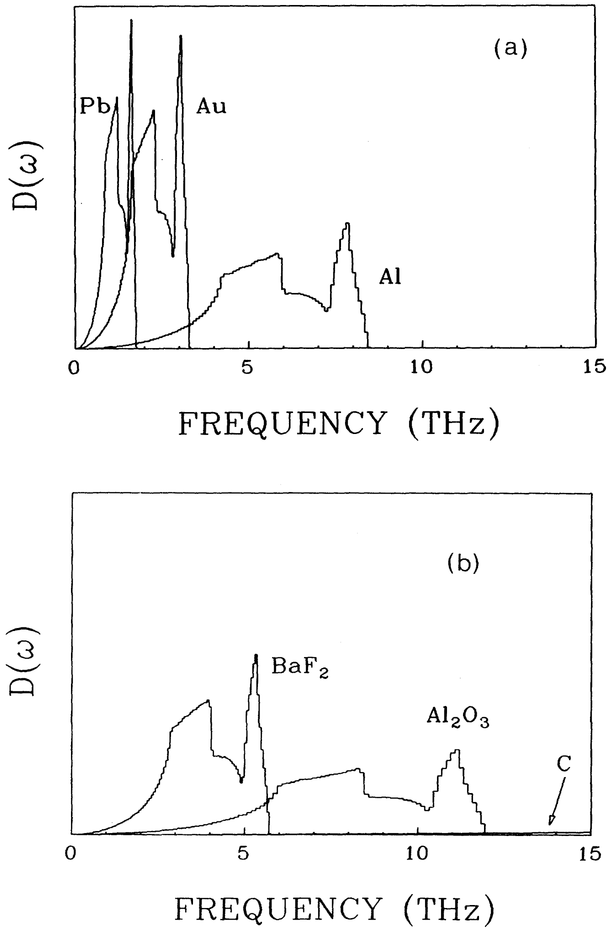

FIG. 6. Phonon densities of states calculated from fcc lattice models described in the text. (a) Results for Al, Au, and Pb. The model density of states of Ti (not shown) is quite similar to that of Al. (b) Results for diamond, sapphire, and BaF_2.

B. Interfacial scattering

Despite the care taken in preparing our samples it is likely that in all cases some amount of interfacial contamination or imperfection was present. It is therefore important to investigate the possible effect on σ_K of interfacial phonon scattering.

1. Diffuse mismatch limit

Swartz and Pohl[2] have considered how the Kapitza conductance is affected by strong scattering of phonons at the interface. They consider a model (the "diffuse mismatch limit") in which the scattering is sufficiently strong that a phonon impinging on the interface loses all memory of its former direction and polarization, as well as memory of which side of the interface it came from. We assume that the scattering is elastic, so that the frequency is unchanged. We write the probabilities that phonons coming from side A and side B are transmitted across the interface as $t_A(\omega)$ and $t_B(\omega)$, respectively, and the probabilities of reflection as $r_A(\omega)$ and $r_B(\omega)$. The model assumes that the probability $r_A(\omega)$ that a phonon of frequency ω coming from A is scattered back into A equals the probability $t_B(\omega)$ that a phonon of the same frequency is transmitted from B into A. In addition, we must have $t_A(\omega)+r_A(\omega)=1$ and $t_B(\omega)+r_B(\omega)=1$. It follows that

$$t_A(\omega)+t_B(\omega)=1 \ . \tag{12}$$

When the temperature is the same on both sides of the interface the fluxes of phonons at each frequency crossing the interface must be equal. Let $f_A(\omega)$ and $f_B(\omega)$ be the phonon fluxes incident on the interface from the two sides. Then

$$t_A(\omega)f_A(\omega)=t_B(\omega)f_B(\omega) \ . \tag{13}$$

It follows from Eqs. (12) and (13) that

$$t_A(\omega)=\left[1+\frac{f_A(\omega)}{f_B(\omega)}\right]^{-1} \ . \tag{14}$$

In terms of the lattice models of the last section, Eq. (14) becomes

$$t_A(\omega)=\left[1+\frac{\sum_{\mathbf{k}j}^{A}|v_{\mathbf{k}jz}|\delta(\omega-\omega_{\mathbf{k}j})}{\sum_{\mathbf{k}j}^{B}|v_{\mathbf{k}jz}|\delta(\omega-\omega_{\mathbf{k}j})}\right]^{-1} . \qquad (15)$$

Here, as in the preceding section the superscripts A and B indicate that each sum is over phonons traveling toward the interface from A and B, respectively.

The transmission coefficients given by Eq. (15) depend on $\mathbf{k}$ and j only through $\omega_{\mathbf{k}j}$. Thus, in this model phonons of the same frequency incident on the interface from all angles have equal transmission probabilities. In the calculations performed in the previous section, which assume a perfect interface and no scattering, phonons coming from a material of low sound velocity and incident on a material of higher sound velocity were reflected if their angles of incidence were larger than a critical value. In the diffuse mismatch limit all phonons, irrespective of their angles of incidence, have finite transmission probabilities, and so we expect σ_K to be increased relative to the perfect interface result.

We calculated σ_K in the diffuse mismatch limit for Au and Pb on diamond using the lattice models of the preceding section to find $t_A(\omega)$ from Eq. (15), and then substituting these in Eqs. (7) and (8) to obtain σ_K. The results are shown in Figs. 3(b) and 3(c) (dotted curves). The conductances are substantially increased relative to the values obtained above assuming no scattering. However, they are still much smaller than the measured values.

2. *Radiation limit*

The diffuse mismatch model just considered makes some implicit assumptions about the nature of the interfacial scattering. Now we investigate whether our experimental results can be explained by *any* model in which phonons are scattered *elastically* at the interface.

Regardless of the details of the scattering, one can establish an upper limit on the value of σ_K as follows.[40] Let us suppose that at a frequency ω the phonon flux $f_A(\omega)$ incident on the interface from A is greater than the flux $f_B(\omega)$ incident on the interface from B. The largest possible conductance occurs if all phonons coming from B have a probability of 1 of crossing the interface. For interfaces between any one of the metals and diamond or sapphire the phonon flux incident on the interface is always greatest from the metal (side A) for frequencies less than the maximum frequency $\omega_{\max}$ in the metal. At frequencies greater than $\omega_{\max}$ there can be no transport across the interface from either material provided that all scattering at the interface is elastic. Thus, from Eq. (3) the maximum possible value of the Kapitza conductance is given by

$$\sigma_K(T)=\frac{1}{V}\frac{\partial}{\partial T}\sum_{\mathbf{k}j,\,\omega\le\omega_{\max}}^{B}\hbar\omega_{\mathbf{k}j}n(\omega_{\mathbf{k}j},T)|v_{\mathbf{k}jz}| , \qquad (16)$$

where the superscript B indicates that all quantities in the sum refer to phonons incident on the interface from the B side (i.e., sapphire or diamond). Since the Debye temperatures of diamond and sapphire are much greater than those of Au and Pb, the only phonons involved in the transport in diamond and sapphire are long-wavelength acoustic phonons. Therefore, we can approximate both diamond and sapphire as elastic continua in order to evaluate $\sigma_K(T)$ from Eq. (16). Making the further assumption of elastic isotropy, we can derive the following formula for the radiation limit:

$$\sigma_K(T)=\frac{1}{16\pi^2}\left[\frac{1}{c_l^2}+\frac{2}{c_t^2}\right]\int_0^{\omega_{\max}}\frac{\partial n(\omega,T)}{\partial T}\hbar\omega^3 d\omega , \qquad (17)$$

where c_l and c_t are the isotropic longitudinal and transverse sound velocities. For diamond $c_l=1.81\times10^6$ cm sec^{-1} and $c_t=1.23\times10^6$ cm s^{-1}; for sapphire $c_l=1.08\times10^6$ cm s^{-1} and $c_t=0.64\times10^6$ cm s^{-1}.[34] To evaluate the integral as accurately as possible we have used the experimentally observed maximum frequencies[41,42] for Pb and Au, rather than the maximum frequencies from the fcc model. The results for the radiation limits for Au and Pb on diamond and sapphire are shown in Figs. 3(b) and 3(c), and Figs. 4(b) and 4(c) (dashed lines).

The experimental values exceed the radiation limits for Pb and Au on diamond and for Pb on sapphire. For Au on diamond and Pb on sapphire the difference between the experimental results and the radiation limit is not large, and might be explained by the approximations made in our calculations. For Pb on diamond, however, the experimental σ_K exceeds the theoretical value by more than 1 order of magnitude. This forces us to conclude that there must be a significant contribution to the heat flow from phonons that have a frequency above the frequency cutoff in the Pb. This means that in some way low-frequency phonons in the Pb transfer their energy to high-frequency phonons in the diamond substrates (and vice versa). Thus, it is essential to consider the role of *inelastic* processes, i.e., processes in which the phonon frequency changes.

C. Electronic contribution

We first consider the possible role of electrons in our experiments. Heat flow across interfaces between metals and liquid-helium resulting from coupling of electrons in the metal to phonons in the liquid has been considered theoretically by several authors,[43,44] and attempts have been made to detect this mode of energy transfer experimentally.[45,46] Phonons in helium have velocities which are small compared to those typical of phonons in metals, and so they can excite Stonely waves on metal surfaces if they are incident at angles near the critical angle. These surface waves have associated with them evanescent acoustic fields in the metal which, compared to the fields produced by phonons incident at larger angles, are very strong. The additional energy flux across the interface provided by the coupling of these fields to the electrons in the metal is found to be of the same order of magnitude as the amount of energy carried by phonons alone. Therefore, this channel leads to a significant increase in σ_K but still by much less than an order of magnitude. Irrespective of the strength of the electron-phonon interaction, this enhancement is limited by the fact that only a very small fraction of the phonons in the helium can couple strongly to the surface modes.[47]

For the combinations of materials that we have studied, the phonons which can excite surface modes are those coming from the metal side of the interface. Therefore, an interaction between these modes and the electrons cannot contribute to the energy flux across the interface. However, evanescent acoustic fields are produced in the metal by phonons coming from the dielectric which reflect from the interface because their frequencies exceed $\omega_{\max}$. As in the helium case, there can, in principle, be an energy transfer arising from a coupling of these fields to the electrons in the metal. We are not aware of any calculation of the expected rate of energy transfer due to this mechanism. Consequently, we have performed an experiment to look for an electronic contribution to σ_K directly.

Two samples were prepared for this experiment. In one a 35-Å film of Bi was deposited on diamond under conditions similar to those described in Sec II.[48] On top of this Bi film we deposited 1000 Å of Pb.[49] The second sample was prepared under the same conditions, but had no Bi layer. The phonon spectra of Bi and Pb are similar, and so the interfacial Bi layer in the first sample was not expected to affect the flux of phonons impinging on the interface from the Pb. However, since Bi is a semimetal and has a much smaller electron density than Pb, Bi should act as a barrier that reduces the density of free electrons near to the surface of the dielectric. Thus, if the energy transfer from Pb to diamond is mainly via the electronic mechanism described above, the cooling rate measured for the sample with the Bi layer should be much less than for the sample without the layer. Cooling curves for the two samples measured at room temperature were very similar, and so we conclude that electrons do not contribute significantly to the heat flux across our interfaces.

D. Effect of interface quality

One can also consider the possibility that the anomalously large conductances observed for Au and Pb on diamond are not intrinsic, but are caused by some form of contamination at the interface. Phenomenologically, one could propose that phonons coming from the diamond with frequency above the cutoff in the metal are down converted in such a layer, and hence are able to enter the metal. To test this possibility experimentally we have conducted further experiments using more carefully prepared samples. Single-crystal Au films were grown on epitaxial-grade sapphire under UHV conditions. Three substrates with different crystal orientations (all perpendicular to [2110]) were used, and the substrates were carefully outgassed prior to growth of the Au films. The vacuum chamber in which the samples were grown was cryogenically pumped, and so was free of possible interfacial contaminants such as pump oil. Kapitza conductances for these samples were found to be larger by up to a factor of 3 than the conductances measured for Au on sapphire samples prepared under less ideal conditions, i.e., the data shown in Fig. 4. For some of the high-quality samples σ_K was found to be greater than the radiation limit. This result implies that the energy transfer across the interface is unlikely to be associated with contamination and defects, and is instead an intrinsic property of the metal-substrate interface.

E. Effect of anharmonicity

One can also consider the possibility that the excess energy transfer across the interface arises from anharmonicity. Even for a highly simplified model, it is much harder to develop an anharmonic theory. The key point is that in the harmonic lattice-dynamical theory discussed in Secs. III A and III B it was permissible to consider that the contributions from different phonon modes could simply be added together. The incident flux from a particular phonon mode was proportional to the phonon energy density (determined by the Bose-Einstein distribution function), the phonon group velocity, and the transmission coefficient which could be taken to be independent of temperature. When anharmonicity is included this division into separate contributions from individual modes is no longer possible. For a classical system the energy transfer can be calculated by molecular dynamics, but this approach is valid only for temperatures greater than the Debye temperature so that classical statistics apply.

For simplicity, in the following discussion we will focus on achieving an understanding of the Kapitza conductance at the Pb-diamond interface since it is for this interface that the measured values of σ_K show the largest disagreement compared to the harmonic lattice dynamical model. Fortunately, the characteristics of the materials that we are considering make it possible to allow for the effects of anharmonicity, at least in an approximate way. First, we note that the interatomic forces in diamond are much stronger than in Pb. The ratio of the bulk moduli, for example, is about 13. Thus, it makes sense to calculate the heat flow by the following scheme. As a *first* approximation we consider the surface of the diamond to act like a rigid wall. We then calculate, in a way described below, the fluctuating force exerted on this wall by the Pb atoms. Next we calculate, allowing for the finite stiffness of the diamond, the rate at which energy is radiated into the diamond as a result of these fluctuating surface forces. The second feature that we exploit is the low Deybe temperature of Pb. Provided that we restrict attention to temperatures comparable to or higher than Θ_D, we can calculate the forces exerted on the diamond wall by Pb by means of *classical* molecular dynamics.

A key parameter in the calculations is the strength of the interatomic potential acting between the Pb and diamond atoms. The subject of the interfacial bonding between the metals and dielectrics is complicated.[50] It is possible that because of the high concentration of dangling bonds on diamond surfaces dimers are formed with the Pb atoms,[51] and so the attraction between Pb and diamond might be strong. There may also be attractive image forces between charged defects near the surface of the dielectric and the metal.[52] We are not aware of published studies for Pb on diamond. For other systems [such as Pb on sapphire,[53] and Sb on Si (Ref. 54)] interfacial adhesion has been found to depend strongly on the preparation of the dielectric surface, strain in the film, the crystal orientation, surface reconstruction, and many other factors. Because of these uncertainties we have treated the strength of the Pb-diamond interaction $V_{\text{Pb-diam}}$ as an adjustable parameter.

We first calculated how anharmonicity would affect the Kapitza conductance if the Pb-diamond interaction had the same value as the Pb-Pb interaction. To do this we performed a classical molecular-dynamics simulation of a Pb lattice coupled to a rigid diamond wall. We calculated the magnitude and time dependence of the force exerted by Pb on diamond, both for a harmonic and an anharmonic Pb lattice. The harmonic Pb lattice was the same fcc model that we have considered above. To introduce anharmonicity we replaced the quadratic interatomic potential by a Lennard-Jones 6-12 potential, i.e., we used

$$V(r)=\epsilon_{\text{Pb-Pb}}\left[\left[\frac{a}{r}\right]^{12}-2\left[\frac{a}{r}\right]^{6}\right] . \tag{18}$$

The strength of the Lennard-Jones interaction was chosen so that for small interatomic displacements the effective spring strength between nearest neighbors was the same as in the harmonic model. The value of $\epsilon_{\text{Pb-Pb}}$ that was obtained in this way was 930 K. The results of this molecular-dynamics simulation showed that the Fourier spectrum of the force exerted by Pb on the diamond did indeed extend to frequencies higher than the highest frequency $\omega_{\max}$ in the Pb lattice. These higher frequencies can be considered to arise from two mechanisms. First, because of the anharmonicity the Fourier spectrum of the *displacements* of the Pb atoms will extend above $\omega_{\max}$. Second, because the relation between displacement and force is no longer linear, the force exerted on the diamond surface by the vibrating Pb atoms is not proportional to the displacements of the Pb atoms, but contains terms quadratic and higher in the displacements. The results of this simulation showed that although the Fourier spectrum did extend somewhat to frequencies in the range above $\omega_{\max}$, the strength of the fluctuations in this range was relatively weak. This indicates that the introduction of anharmonicity does not, by itself, provide an explanation of the large value of σ_K for the Pb-diamond interface. This result is consistent with earlier molecular-dynamics studies by Lumpkin and Saslow[55] and Ge and Chen[56] The models that they investigated were somewhat different from ours but they also found that σ_K was only slightly increased by the inclusion of anharmonicity.

We now investigate what happens when the interactions between Pb and diamond are assumed to be much stronger than the Pb-Pb interaction. The layer of Pb atoms next to diamond may then have a fundamental vibration frequency which is higher than the highest frequency in the Pb lattice. This leads to the following interesting situation. The Pb atoms in the bulk (i.e., those *not* in the layer next to the diamond) can transfer energy to the interfacial Pb atoms only via anharmonic interactions because the frequencies of vibrations of the bulk atoms are too low to excite the interfacial Pb atoms in a one-phonon process. The interfacial atoms, because of their high frequency and strong coupling to the diamond, can exert large forces on the diamond and hence act as efficient radiators. To calculate σ_K for this system it is therefore necessary to study two issues. The first is whether or not the radiation by the interfacial Pb atoms is sufficiently strong to explain the observed value of σ_K; the second issue is whether the anharmonicity of Pb is strong enough that the energy radiated into the diamond by the interfacial Pb atoms can be replaced at the same rate by an energy transfer from the bulk Pb. Thus, there are two steps to the heat flow as indicated schematically in Fig. 7.

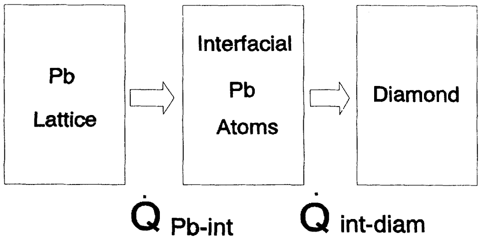

FIG. 7. Illustration of the path for the flow of heat from Pb to diamond. $\dot{Q}_{\text{Pb-int}}$ is the flux to the interfacial Pb atoms from the remainder of the Pb film, and $\dot{Q}_{\text{int-diam}}$ is the heat flux from the interfacial Pb atoms to the diamond substrate.

1. *Energy transfer from interfacial Pb to diamond*

We first consider the energy transfer rate $\dot{Q}_{\text{int-diam}}$ from the interfacial Pb atoms to diamond. Our calculations are based on a simple model in which a single Pb atom interacts with a rigid diamond substrate and a single stationary Pb atom via potentials $V_{\text{Pb-diam}}$ and $V_{\text{Pb-Pb}}$, respectively. The moving Pb atom vibrates along the z axis, i.e., along a line normal to the interface, the fixed Pb atom is at $z=-2a$, and the first atom of the diamond substrate is at $z=0$. Thus, the potential acting on the vibrating interfacial Pb atom is taken to be

$$V(z)=\epsilon_{\text{Pb-Pb}}\left[\left[\frac{a}{z+2a}\right]^{12}-2\left[\frac{a}{z+2a}\right]^{6}\right] +\epsilon_{\text{Pb-diam}}\left[\left[\frac{a}{z}\right]^{12}-2\left[\frac{a}{z}\right]^{6}\right] . \tag{19}$$

To calculate the properties of the fluctuating force we first select an energy E for the interfacial Pb atom. From the equation of motion of the interfacial Pb atom it is straightforward to calculate the power spectrum $|F(\omega)|^2$ of the fluctuating force exerted on the diamond. We repeat this procedure for a large number of other energies chosen at random using the canonical probability distribution $\exp(-E/k_BT)$, and obtain a temperature-dependent averaged power spectrum $\langle |F(\omega)|^2 \rangle$.

As noted above this calculation of the fluctuating force is based on the idea that as a first approximation the diamond can be treated as a rigid wall. To calculate the rate at which energy is transferred to the diamond it is necessary to allow for a finite compliance of the diamond.

This calculation is simplified by the fact that the sound velocity in the diamond is very large, approximately 15 times that of Pb. Consequently, even if the vibrations of the interfacial Pb atom are at a frequency several times higher than the Debye frequency of Pb the waves launched into the diamond will have wavelengths which are many tens of lattice parameters. Thus, it is a reasonable approximation to treat the diamond as an isotropic elastic continuum. An elastic continuum can be completely described by the Debye velocity c_D, the value of Poisson's ratio σ (or equivalently the ratio of the transverse to longitudinal sound velocity), and the density ρ. Since the diamond is treated as linear the energy transfer at a frequency ω must be proportional to $\langle |F(\omega)|^2 \rangle$. It then follows from dimensional analysis that the rate of energy transfer $\dot{Q}$ must be given by

$$\dot{Q}=\frac{Q_0(\sigma)}{\rho c_D^3}\int \langle |F(\omega)|^2 \rangle \omega^2 d\omega \ , \tag{20}$$

where $Q_0(\sigma)$ is a dimensionless constant that depends only on Poisson's ratio. Thus, the significant quantity is the parameter Γ which we define to be the ratio of the value of the integral

$$\int \langle |F(\omega)|^2 \rangle \omega^2 d\omega \tag{21}$$

for a given value of $\epsilon_{\text{Pb-diam}}$, to the same quantity evaluated with $\epsilon_{\text{Pb-diam}}=\epsilon_{\text{Pb-Pb}}$. In our calculations the range parameter a was assumed to be the same for both potentials and equal to the nearest-neighbor separation in Pb (3.5 Å). For $V_{\text{Pb-Pb}}$ we took $\epsilon_{\text{Pb-Pb}}=930$ K [see earlier discussion and Eq. (18)]. We investigated the variation of Γ with $\epsilon_{\text{Pb-diam}}$, and the results are shown by the solid curves in Fig. 8 for temperatures of 100 and 300 K.

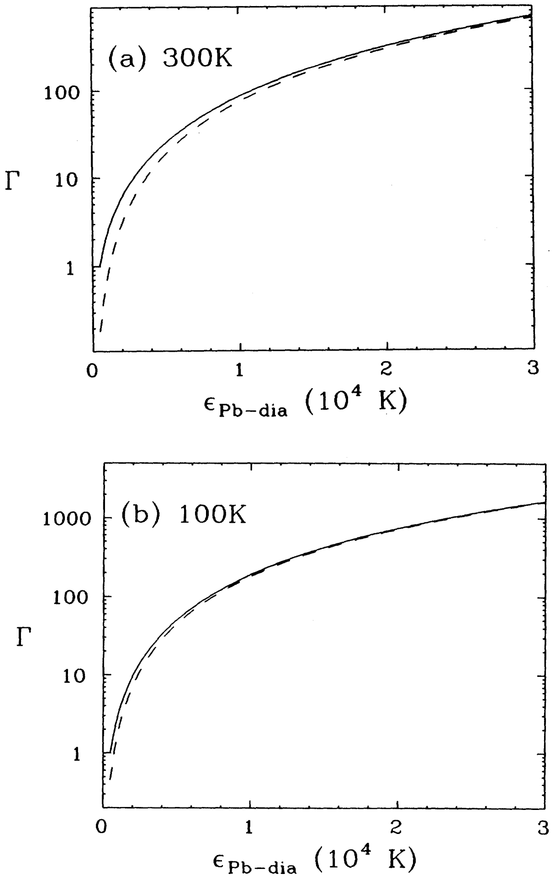

FIG. 8. Enhancement of the Kapitza conductance due to the strong bonding of Pb atoms to the diamond substrate. The quantity that is plotted is the ratio Γ of the calculated conductance for a given value of the Pb-diamond interaction $\epsilon_{\text{Pb-diam}}$ to the conductance for a model in which the Pb-diamond potential is the same as the Pb-Pb potential. Results are shown for temperatures of (a) 300 K and (b) 100 K. The solid curves are results for the Lennard-Jones potential, and the dashed curves are for a harmonic potential of the same effective spring constant.

One can see from these results that as the strength of the potential between the interfacial atom and the diamond is increased, the value of Γ goes up rapidly. To explain our experimental data it is necessary for the heat flow to be enhanced by a factor about 100 relative to the lattice dynamical model described in Sec. III A. This must be roughly equivalent to the requirement that Γ have a value of around 100. We see from Fig. 8 that this means that $\epsilon_{\text{Pb-diam}}$ must be greater than about 9000 K for this to happen.[57]

There are two important points to note regarding this calculation. The first is that the enhancement at large $\epsilon_{\text{Pb-diam}}$ is not a result of the *nonlinearity* of the Lennard-Jones potential. For each value of $\epsilon_{\text{Pb-diam}}$ we expanded the potential-energy function, Eq. (19), about the minimum at $z=-a$, and then repeated the calculation of Γ using just the quadratic term in $V(z)$. The results are shown by the dashed curves in Fig. 8. The effect of going to a quadratic potential is to decrease Γ by a factor which is at most a factor of 2 or 3.

The second point concerns the applicability of classical mechanics to the calculation of the rate of energy transfer. We have used the canonical ensemble together with classical mechanics to calculate the energy transfer. This should be valid provided that the frequency of vibration ω_{int} of the interfacial atom is less than or equal to $k_B T/\hbar$. In the harmonic approximation the vibrational frequency is

$$\omega_{\text{int}}=[72(\epsilon_{\text{Pb-diam}}+\epsilon_{\text{Pb-Pb}})/a^2 m]^{1/2} \ , \tag{22}$$

where m is the mass of the Pb atom. For $\epsilon_{\text{Pb-diam}}=9000$ K one obtains $\hbar\omega_{\text{int}}/k_B=116$ K, and so at least down to $T=100$ K it is reasonable to use classical statistical mechanics.

2. *Energy transfer from Pb lattice to interface atoms*

The calculations just described show that provided $V_{\text{Pb-diam}}$ is sufficiently strong the rate of energy transfer from the interfacial atoms to the diamond can be consistent with the measured Pb-diamond Kapitza conductance. However, it is also necessary for the energy lost by the interfacial atoms to be replaced at an equal rate by an energy transfer from the bulk of the Pb film. Otherwise the interfacial atoms will have a temperature that is closer to the temperature of the diamond than to the temperature of the Pb film, and the flow of heat will be reduced.

It is convenient to discuss the heat transfer in terms of characteristic times. If the number of interfacial Pb atoms per unit area is taken to be $1/a^2$ with $a = 3.5$ Å, and each atom is assumed to have the classical specific heat of $3k_B$, then the specific heat of the surface layer is 3.4×10^{-8} J cm^{-2} K^{-1}. The measured Kapitza conductance at 300 K for the Pb-diamond interface is about 3000 W cm^{-2} K^{-1}. Thus, if the energy lost by the interfacial layer to the diamond were not replaced by an energy flow from the remainder of the Pb film, the interfacial Pb layer would cool at a rate governed by a time constant τ of $3.4 \times 10^{-8}/3000 = 1.1 \times 10^{-11}$ s. We therefore want to compare this cooling time for energy loss into diamond with the time it takes heat to flow into the layer from the Pb film (Table II).

To study this heat transfer we used a molecular-dynamics simulation. To reduce the computer time needed for the calculations we used the two-dimensional lattice model shown in Fig. 9. In the figure, Pb atoms (solid circles) are joined to one another by the weak potential $V_{\text{Pb-Pb}}$ (light lines), and to diamond (open circles) by the stronger potential $V_{\text{Pb-diam}}$ (heavier lines). We continue to consider diamond as a rigid body so that open circles may be regarded simply as anchor points for the interfacial Pb atoms. We used the Lennard-Jones potential described above for $V_{\text{Pb-Pb}}$ and $V_{\text{Pb-diam}}$. For $V_{\text{Pb-diam}}$ we used $\epsilon_{\text{Pb-diam}} = 9000$ K. Simulations were carried out using a lattice consisting of 20 rows of Pb atoms with 20 atoms per row. Periodic boundary conditions were used at the edges perpendicular to the interface, and the surface of Pb away from the interface was free. The Pb atoms in the interfacial layer were initially placed at rest at their equilibrium positions. The remaining Pb atoms were also placed at their equilibrium positions, and were given a random kinetic energy E taken from a classical ensemble corresponding to the temperature T. The motions of the atoms were then evolved in time steps corresponding to less than one-tenth of the shortest phonon period in the Pb lattice. At each time step the total energy E_{int} of the atoms in the interfacial Pb row was calculated. Since the number of atoms in the Pb lattice is much larger than the number of interfacial atoms, the temperature of the Pb lattice remains approximately constant as the interfacial atoms gain energy.

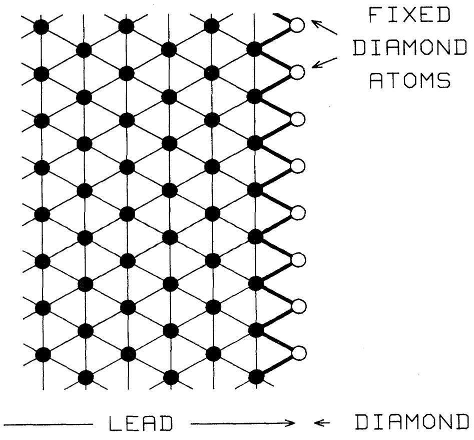

FIG. 9. Two-dimensional model of the Pb-diamond interface used in molecular-dynamical simulations. The Pb and diamond atoms are shown as solid and open circles, respectively. The thick lines indicate the strong potential coupling the interfacial Pb and diamond atoms.

We carried out these simulations at 100 and 300 K, averaging the results over 20 different initial velocity distributions at each temperature. The characteristic times for the energy of the interfacial layer to rise to its equilibrium value were found to be 1.0×10^{-11} s at 300 K, and 1.8×10^{-11} s at 100 K. Thus, to within the accuracy of the calculations, the rate of energy transfer at 300 K is limited equally by the difficulty of heat transfer between Pb and the interfacial atoms, and between the interfacial atoms and diamond. At 100 K it appears that the process of heat transfer from the Pb film to the interfacial atoms provides the main barrier to the overall heat flow.

IV. SUMMARY AND CONCLUSIONS

We have measured the Kapitza conductance for interfaces between solids for which the ratio R of the Debye temperatures ranged from 0.05 (Pb on diamond) to 2 (Ti on BaF$_2$). For R greater than about 0.2 the results could be understood quantitatively if we viewed the heat as being carried across the interface by phonons whose transmission probabilities were computed using a lattice dynamical theory. For smaller R the experimental conductances were generally larger than predicted by the theory. The disparity between the theoretical conductances and the experimental values was greatest for Pb on diamond where $R \simeq 0.05$. Smaller discrepancies were found for larger R's. The anomalous conductances could not be explained by elastic phonon scattering by interfacial contaminants or defects. In three cases, Pb and Au on diamond and Pb on sapphire, the measured conductances were shown to exceed the phonon radiation limit. This forced us to conclude that inelastic processes played a major role in transporting energy across these interfaces. We determined experimentally that this mechanism did not involve electrons in the metal. An inelastic channel was then proposed in which the metal atoms were required to be bound to the substrate by a potential whose strength was much greater than the strength of the potential binding the metal atoms together elsewhere in the lattice. The metal lattice was also required to be anharmonic. It was not necessary to assume that the *interfacial* potential was anharmonic.[58] We used molecular-dynamics simulations to show that our anomalous Pb-diamond measurements could be accounted for by the proposed mechanism.[59]

We have concentrated on the development of a theoretical explanation of the Kapitza conductance between Pb and diamond. This strategy is chosen because the disagreement between the lattice dynamical theory and experiment is the largest for this combination of materials. Anharmonicity must also modify the Kapitza conductance for the other combinations of materials. However, the relative importance is presumably much less because the harmonic channel is large and because anharmonic effects are weaker in the metals other than Pb.

As just mentioned, our results show that the Kapitza conductance between Pb and Au and diamond, and between Pb and sapphire cannot be explained by the effect of elastic scattering at the interface. However, it is important to note that this does *not* mean that elastic scattering is not taking place. As far as we can see the experimental results do not give a clear indication on this issue. Elastic scattering (and anharmonicity) may contribute to some extent to the discrepancy between experiment and the harmonic lattice dynamical theory for all of the interfaces studied.

Finally, it may be interesting to attempt to use our ideas to understand the anomalous Kapitza conductances measured for interfaces between solids and liquid helium. For liquid helium both the sound velocity and the density are much less than for ordinary solids. Consequently, the transmission coefficients of phonons at a helium-solid interface are predicted to be very small, and the Kapitza conductance is expected to be low.[1,4] It is found that at temperatures below about 0.1 K measured values of σ_K are in reasonable agreement with theory.[60] However, at higher temperatures the phonon transmission and the conductance are larger than predicted by a factor of the order of 100 and this is referred to as the anomalous Kapitza conductance.[61] It is interesting to note that our results for the Pb-diamond interface give a comparable enhancement. It appears from recent experiments[62] that the extra conduction mechanism for a helium-solid interface is related to contamination of the metal surface.[63] In a sense the problem is the reverse of the solid-solid problem we have considered; the phonon flux from the metal is limited not by the smaller density of states of the substrate (helium) but by its own small density of states. Therefore, to have a larger σ_K it is necessary to have additional modes at the surface of the metal of *low* frequency. In analogy with the *tightly* bound interfacial Pb atoms in our molecular-dynamical model the needed low-frequency modes could be associated with *weakly* bound contaminants at the helium-solid interface.[63] These modes would then serve to channel energy from phonons of high energy impinging on the interface from the metal into the large number of available states in the helium, thus giving an increase in σ_K. It is also apparent that the additional channel must "freeze out" if the proposed scheme is to be consistent with experimental observations, which show that σ_K is well described by the general physical picture described in Sec. I for temperatures below about 100 mK. Thus, there must be a minimum allowable binding strength for the interfacial contaminants. In principle, calculations similar to the ones we have carried out in this paper could be used to test these ideas.

ACKNOWLEDGMENTS

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APPENDIX: COUPLING MATRIX FOR LATTICE DYNAMICAL THEORY

For the incident wave ($q=0$) and the three reflected waves ($q=1,2,3$)

$$\begin{aligned}
M_{1q} &= (K-K'')e_x^{(q)} \\
&\quad - K[\cos(X_q)e_x^{(q)} + i\sin(X_q)e_z^{(q)}]\exp(iZ_q)\ , \\
M_{2q} &= (K-K'')e_y^{(q)} \\
&\quad - K[\cos(Y_q)e_y^{(q)} + i\sin(Y_q)e_z^{(q)}]\exp(iZ_q)\ , \\
M_{3q} &= -iK[\sin(X_q)e_x^{(q)} + \sin(Y_q)e_y^{(q)}]\exp(iZ_q) \\
&\quad + \{2(K-K'') \\
&\qquad - K[\cos(X_q)+\cos(Y_q)]\exp(iZ_q)\}e_z^{(q)}\ , \\
M_{4q} &= K''[\cos(X_q)e_x^{(q)} - i\sin(X_q)e_z^{(q)}]\ , \\
M_{5q} &= K''[\cos(Y_q)e_y^{(q)} - i\sin(Y_q)e_z^{(q)}]\ , \\
M_{6q} &= -iK''\sin(X_q)e_x^{(q)} - iK''\sin(Y_q)e_y^{(q)} \\
&\quad + K''[\cos(X_q)+\cos(Y_q)]e_z^{(q)}\ .
\end{aligned}$$

For the three transmitted waves ($q=4,5,6$)

$$\begin{aligned}
M_{1q} &= K''[\cos(X_q)e_x^{(q)} + i\sin(X_q)e_z(q)]\exp(iZ_q)\ , \\
M_{2q} &= K''[\cos(Y_q)e_y^{(q)} + i\sin(Y_q)e_z(q)]\exp(iZ_q)\ , \\
M_{3q} &= iK''[\sin(X_q)e_x^{(q)} + \sin(Y_q)e_y^{(q)}]\exp(iZ_q) \\
&\quad + K''[\cos(X_q)+\cos(Y_q)]e_z^{(q)}\exp(iZ_q)\ , \\
M_{4q} &= (K'-K'')e_x^{(q)}\exp(iZ_q) \\
&\quad - K'[\cos(X_q)e_x^{(q)} - i\sin(X_q)e_z^{(q)}]\ , \\
M_{5q} &= (K'-K'')e_y^{(q)}\exp(iZ_q) \\
&\quad - K'[\cos(Y_q)e_y^{(q)} - i\sin(Y_q)e_z^{(q)}]\ , \\
M_{6q} &= iK'[\sin(X_q)e_x^{(q)} + \sin(Y_q)e_y^{(q)}] \\
&\quad + 2(K'-K'')\exp(iZ_q) \\
&\quad - K'[\cos(X_q)+\cos(Y_q)]e_z^{(q)}\ ,
\end{aligned}$$

where, for example, $X_q = ak_x^{(q)}$. $a\sqrt{2}$ is the nearest-neighbor separation. These coefficients are written under the assumption that phonons are incident from a lattice with springs of strength K connecting nearest neighbors onto an interface with a lattice in which atoms are connected with springs of strength K'. The strength of the springs connecting the atoms across the interface is K''.

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[59] To verify that we have not used an unrealistic amount of anharmonicity we evaluated the coefficient of thermal expansion of our two-dimensional lattice. This was done by determining the change in the mean area of a 50-atom by 50-atom Pb lattice with $\epsilon_{\text{Pb-Pb}}=930$ K when the temperature was varied by a small amount near 300 K. We estimated a thermal expansion coefficient of 3.5×10^{-5} K^{-1} which compares favorably with the experimentally determined value (Ref. 18) for Pb of 2.8×10^{-5} K^{-1}.

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