Measurements of the Kapitza Conductance between Diamond and Several Metals

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The Kapitza conductance between isotopically enriched synthetic diamond and Ti, Al, Au, and Pb has been studied using picosecond optical techniques. For Ti and Al, which have Debye temperatures roughly $\frac{1}{5}$ of that of diamond, the measured heat flow is in reasonable agreement with calculations of the phonon heat transport based on a simple lattice dynamical model. However, for Au and Pb, which have Debye temperatures 0.07 and 0.04 times that of diamond, the measured Kapitza conductances are as much as 100 times larger than expected, suggesting the existence of a second conductance mechanism.

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Kapitza [1] discovered that when heat flowed between liquid helium and copper a temperature jump ΔT occurred at the interface. The Kapitza conductance σ_K is the ratio of the heat flow $\dot{Q}$ per unit area to ΔT. This conductance has since been studied at interfaces between liquid helium (^{3}He and ^{4}He) and a wide variety of materials, and also at the interface between two solids [2].

Most theories of the Kapitza resistance are based on the assumption (due to Khalatnikov [3]) that the heat is carried across the boundary by thermal phonons. Liquid helium has a sound velocity and density which are much smaller than those of most solids. Consequently, the probability that a phonon will be transmitted from a solid into helium is predicted to be very low, and so the Kapitza conductance should be small. However, the experimental values of σ_K are usually much larger (by a factor of 10 or 100) than estimated by this approach, and no satisfactory explanation of this discrepancy has been given [2]. For solid-solid interfaces there is much more limited experimental information [2,4–7], but what there is is in at least rough agreement with theoretical predictions. However, for the solid-solid interfaces investigated so far the difference in the elastic properties (e.g., density and sound velocity) between the two materials has been fairly small. To make a more rigorous test of the theoretical understanding of the Kapitza conductance at solid-solid interfaces it is of interest to make measurements for interfaces between materials that have widely differing elastic properties. This naturally suggests studies of σ_K at interfaces between diamond and other materials, since diamond has the highest sound velocity and Debye temperature Θ_D (2240 K) of any material. In this Letter we report measurements of σ_K as a function of temperature between diamond and four metals whose Debye temperatures range from 88 to 426 K.

The value of σ_K between diamond and other materials is also of technical interest. Isotopically enriched diamond [8] has the highest room-temperature thermal conductivity of any material (8 times that of copper), and therefore has potential application as a heat sink for high-power-density semiconductor devices. The Kapitza resistance at the interface between the diamond substrate and the device may be a limiting factor in these applications.

The samples were isotopically enriched (99.9% ^{12}C) low-defect density diamonds grown at General Electric [8]. Etch-pit studies of the surfaces of these crystals showed surface defect densities less than 10^6 cm^{-2}. We obtained similar results using both as-grown and polished surfaces. Metal films of thickness in the range 200 to 1000 Å were deposited onto (100) surfaces of the diamond by either electron beam or resistive evaporation at pressures less than 10^{-6} torr. The thermal conductivities of the films, as deduced from their measured electrical conductivities, were in agreement with accepted bulk values. In the experiment a hybrid mode-locked laser producing 200-fs pulses of 6320-Å light at a rate of 76 MHz was focused onto a region of the film approximately 20 μm in diameter. A fraction of the incident light was absorbed by electrons in the metal which rapidly came to equilibrium with the lattice [9]. The resulting phonon distribution in the metal film in our experiments is well-described by a temperature since phonon lifetimes in the temperature range of interest are typically less than 50 ps. Based on the heat capacity of the heated region and the absorbed pulse energy we estimate that the initial transient temperature rise in the heated spot was at most a few degrees. Owing to the very high thermal conductivity of the diamond substrates the average temperature of the spot differed by a negligible amount from that of the cold finger to which the samples were thermally anchored. The heating of the metal film caused a small change in its optical reflectivity (on the order of 10^{-5}) [10]. This change $\Delta R(t)$ was measured by means of a probe light pulse time delayed relative to the heating pulse by up to 2500 ps [11]. Since the temperature excursion is small, we assume that $\Delta R(t)$ is proportional to the temperature change $\Delta T(t)$, and so the measurement of $\Delta R(t)$ can be used to determine the cooling curve of the metal film.

The results taken at room temperature for the cooling of an Al film are shown in Fig. 1. The cooling curve

should be given by

$$\Delta T(t) = \Delta T(0)\exp(-t/\tau), \quad (1)$$

where the cooling time $\tau = Cd/\sigma_K$, with C the specific heat of the film per unit volume and d the film thickness. The measured cooling curves can be fitted very well by an exponential (see Fig. 1), and from these fits τ could be determined. For C the bulk value of the specific heat for the metal film was assumed. To find the film thickness the reflectivity change was also measured on a much shorter time scale (inset in Fig. 1). It was then possible to resolve an oscillation in $\Delta R(t)$, originating from acoustic vibrations of the film excited by the pump pulse. From the period of these oscillations and the sound velocity of the metal the film thickness d could be determined. The results for σ_K obtained in this way are plotted as functions of temperature in Fig. 2, along with theoretical curves which we will discuss. The error bars reflect the uncertainty in determining τ for a particular sample; variations from sample to sample were typically 20% or less.

The *general trend* of the results for the different metals can be understood by comparing the density of states (DOS) [12] of the metals to that of diamond (Fig. 3). It is clear from this figure that diamond has a markedly different phonon spectrum from most other materials. The effect of this difference on the Kapitza conductance can be understood as follows. If one considers a measurement of σ_K for a Pb/diamond interface at 300 K, for instance, one would expect to be exciting in the Pb modes at all frequencies up to the allowed maximum ($\omega \sim 1.4\times10^{13}$ s^{-1}). The number of states in the diamond in this frequency range is extremely small, and so the phonons in the Pb film are expected to couple to the diamond lattice very weakly. Thus, very little thermal energy should be carried across the interface, and the Kapitza conductance is predicted to be small. On the basis of this sort of reasoning it would be expected that the Kapitza conductance should be greatest for the metal having the largest Debye temperature.

To make a qualitative calculation of σ_K one has to calculate the flux of phonons incident on the interface and the probability that a phonon of wave vector **k** and polarization j incident from the metal side will be transmitted into the diamond. To do this one has to set up a lattice dynamics model to represent the phonons in the metals and in the diamond and take account of the boundary conditions at the interface. We have used a simple model developed earlier [13]. Nearest-neighbor central forces are assumed, both in the diamond and in the metals. The strength of this interaction is chosen so that the Debye velocity comes out correctly. The interface is chosen to lie parallel to a (001) plane. While the model does not

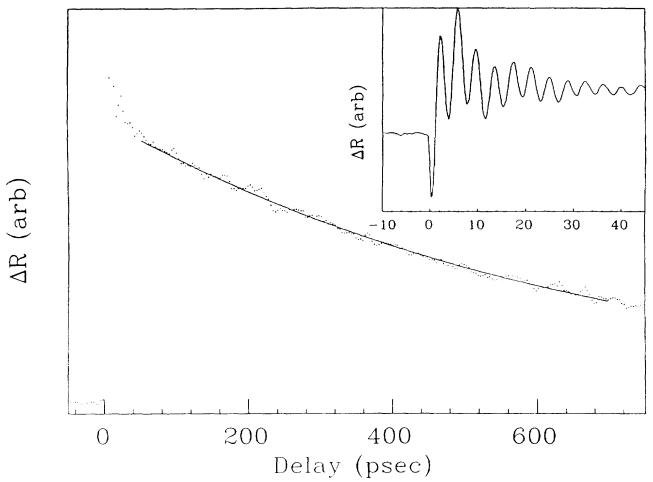

FIG. 1. Photoinduced change $\Delta R(t)$ of a 126-Å film of Al deposited on a diamond substrate. $\Delta R(t)$ is proportional to the change in film temperature. The solid line shows a fit to the data based on Eq. (1). Inset: $\Delta R(t)$ on a short time scale. The oscillations are due to acoustic vibrations of the film, and can be used to determine the film thickness.

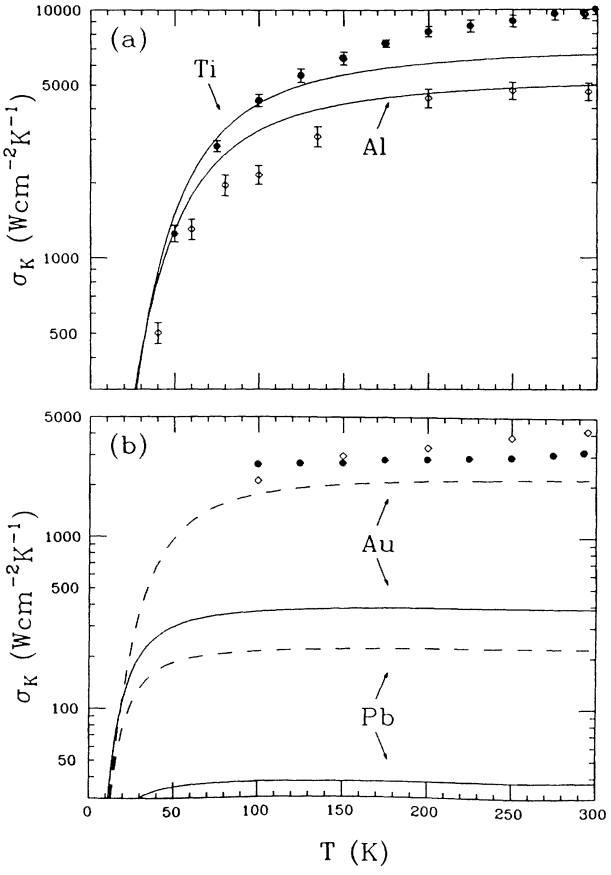

FIG. 2. Measured values of the Kapitza conductance σ_K at the interface between diamond and (a) Al (◇) and Ti (●) and (b) Au (◇) and Pb (●). For Au and Pb the error bars are about the size of the symbols used. The curves are the results of calculations based on the theoretical models discussed in the text and Ref. [16]. The dashed curves are the radiation limit σ_{max} discussed in the text.

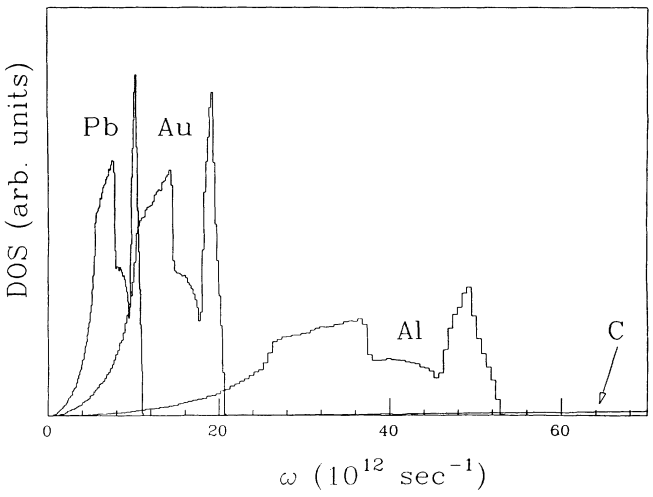

FIG. 3. The density of phonon states for diamond and the four metals studied.

include all the intricacies of the interatomic forces or the arrangement of the atoms at the interface, it does take into account, in an approximate way, Snell's law (an important effect because of the large velocity difference between diamond and the metals) and phonon dispersion in the metals (important because the temperature is comparable to Θ_D for the metals).

The results of calculations based on this model are included in Fig. 2. For Al and Ti the theoretical results are in reasonable agreement with experiment, both in magnitude and in temperature dependence. For Au and Pb, however, the experimental σ_K are much larger than the theory at all temperatures. For Pb the discrepancy is about a factor of 100 at room temperature [14]. These discrepancies are very unlikely to be due to the limitations of the lattice dynamical model we have used. The data suggest that there is some additional way in which energy is transferred across the interface which continues to be important even when the vibrational spectra of the two materials are very different.

In the theoretical model phonons crossing the interface are refracted according to Snell's law. For the interfaces we have considered phonons incident on the interface from the metal side undergo total internal reflection unless they lie within a critical cone whose semiangle is $\sin^{-1}(v_{\text{met}}/v_{\text{diam}})$, where v_{met} and v_{diam} are the phonon velocities in the metal and in the diamond. The solid angle formed by this cone, and hence σ_K, is thus smallest when the velocity mismatch is largest. This effect together with the density-of-states effect already described mainly determine σ_K. To explain the experimental results it is necessary to find a mechanism which either allows phonons from outside the critical cone to carry energy across the interface or extends the range of available frequencies in the metal.

There are several mechanisms to consider, most of which have already been discussed as possible explanations of the anomalously large value of σ_K between helium and solids [2]. Phonons within a narrow range of angles outside the critical cone can excite surface waves on the diamond. These surface waves can in turn decay or scatter into bulk phonons in the diamond that would carry the heat away from the interface. However, as in the helium case [3] one can show that the maximum enhancement of σ_K that can result from this mechanism is a factor of 3 or less. A second effect comes from the evanescent waves excited in the diamond by *all* phonons coming from the metal side which are outside the critical cone [15]. If these evanescent waves are attenuated or scattered in some way, the energy will remain in the diamond thereby giving an energy transfer across the interface. The strength of this process is critically dependent on the rate of dissipation about which essentially nothing is known. Since we obtain the same results (to within $\pm 20\%$) for σ_K at the Pb/diamond interface for several different samples, it appears that for this process to explain the experimental results the dissipation mechanism probably must be intrinsic to diamond, i.e., not associated with defects near the interface.

The above discussion focuses on the relation between σ_K and the transmission coefficient $t_{\text{met-diam}}$ of phonons coming to the interface from the metal side. One can equally discuss σ_K in terms of the transmission coefficient $t_{\text{diam-met}}$ for phonons going from the diamond to the metal. The average of $t_{\text{diam-met}}$ must be much larger than the average of $t_{\text{met-diam}}$, since the flux of phonons incident (in the range of phonon frequencies of the metal) from the diamond side is much smaller than that from the metal side. We have calculated the value ($\sigma_{\max}$) that σ_K would have if $t_{\text{diam-met}}$ were equal to 1 for all phonons coming from the diamond side with a frequency in the range of allowed phonon frequencies (0 to $\omega_{\max}$) in the metal. (In the Kapitza literature $\sigma_{\max}$ is referred to as the radiation limit.) To make this calculation as accurate as possible we have used the experimental value [16] of $\omega_{\max}$ for Pb (1.4×10^{13} s^{-1}). The flux coming from the diamond is calculated using an elastic continuum model for diamond with the known elastic constants. The result is included in Fig. 2. One finds that for Pb $\sigma_{\max}$ is *less* than the experimental result. This is remarkable since it is extremely unlikely that $t_{\text{diam-met}}$ can really be close to 1 for all of these phonons. It implies that phonons above the highest frequency in the metal *must* contribute substantially to the energy transfer. This is evidence that inelastic processes play a major role at Kapitza boundaries [2], although, by what mechanism is not clear. The additional energy flow could arise via either electron-phonon or phonon-phonon coupling in the metal near the interface. If the Pb or Au were replaced by a material with a similarly low Θ_D, but a much lower concentration of free electrons such as Bi, σ_K should be substantially reduced if electron-phonon coupling is important. We have found this to be difficult to test experimentally because of the

very low thermal conductivity of Bi.

In conclusion, we have measured Kapitza conductances for interfaces between Al, Ti, Au, and Pb and isotopically enriched diamond. The measured values can be explained at temperatures from 40 to 300 K for both Al and Ti by a lattice dynamical theory which takes account of the phonon dispersion in an approximate way. In the cases of Au and Pb the measured conductances are more than an order of magnitude larger than predicted by the same theory, suggesting that there is an additional path for heat transfer across the interface. The existence and origin of such a path is of potential importance to the application of isotopically enriched diamond as a heat sink, as well as to the general understanding of the Kapitza problem.

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