

Electronic Kapitza conductance at a diamond-Pb interface

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(Received 22 February 1994)

In recent experiments it was found that the Kapitza conductance between diamond and Pb at room temperature is as much as 100 times larger than predicted by the acoustic mismatch model. Because the diamond and Pb are in intimate contact, the phonon modes near the interface are joint vibrational modes, extending a phonon mean free path on each side of the interface. Since the phonon mean-free path at room temperature is of the order of 4000 Å in diamond but only of the order of 10 Å in Pb, the energy of a joint mode resides primarily in the diamond. Consequently, the thermal population of a joint mode is determined by the temperature of the diamond. It is shown here that the direct energy transfer between metallic electrons at the Pb temperature and joint vibrational modes at the diamond temperature explains the observed heat conduction across the interface.

I. INTRODUCTION

Kapitza¹ discovered that when heat flows from one material to another, there is a temperature jump ΔT at the interface, which is proportional to the heat flux j ,

$$j = \sigma_K \Delta T, \quad (1)$$

where σ_K is the Kapitza conductance.² In recent experiments,³ the heat transport across the interface between diamond and several metals (Ti, Al, Au, and Pb) was measured using picosecond optical techniques. For Ti and Al, the measured heat flow was in reasonable agreement with the predictions of the acoustic mismatch model,⁴ which assumes that the heat is carried across the boundary by thermal phonons. However, for Au and Pb, which are softer than Ti and Al and therefore have a greater mismatch with diamond, the measured heat flow was larger than expected from the acoustic mismatch model, being as much as 100 times larger in the case of Pb. Indeed, even if it is assumed that all the phonons in diamond whose frequencies are below the cutoff frequency of the phonons in Pb are transmitted into Pb with unit probability, the predicted value³ of the heat flow, known as the phonon radiation limit, is still an order of magnitude smaller than the observed value, suggesting the existence of another heat conduction mechanism.

In this research we investigate a mechanism which explains the observed heat conduction across the diamond-Pb interface. There are several key ideas to this proposed mechanism. The first is to recognize that, because diamond and Pb are in intimate contact, oscillations in one will induce oscillations in the other. Consequently, near the interface the appropriate phonon modes are the normal modes of the joint system, which are coherent across the interface. Since the coherence of a mode is destroyed by scattering processes, these joint vibrational modes extend a phonon mean free path on each side of the interface.

It is important to emphasize at the outset the difference between this viewpoint and that in the acoustic mismatch model. There the phonons are divided into two classes: those in diamond (at the temperature of diamond) and those in Pb (at the temperature of Pb). Near the interface, this distinction between phonon modes in diamond and Pb breaks down, since the atoms near the interface are coupled together into joint vibrational modes by Newton's laws of motion.

The second key idea is to realize that, because diamond and Pb are at different temperatures, the temperature difference being proportional to the thermal boundary resistance, the thermal population of these joint vibrational modes in general will not be at the temperature of either diamond or Pb, but at some weighted average. A heuristic approximation is to weight the temperatures of diamond and Pb by the fraction of the energy of the mode in each.

It is important again to contrast this viewpoint with that in the acoustic mismatch model. There the atoms in diamond and Pb are assigned different temperatures, the temperature changing abruptly at the interface. This abrupt change of temperature at the interface is unphysical, since the atoms near the interface are coupled together into joint vibrational modes, each mode having its own intermediate temperature.

Finally, since the effective temperature of the joint vibrational modes is different from the conduction-electron temperature in Pb, a net heat transfer will occur between the electrons and joint modes. (In contrast, in the acoustic mismatch model, which assumes that the electrons and phonons in Pb are both at the same temperature, there is no net heat transfer between these two systems.) Since part of the energy of each joint mode is located on the diamond side, this process transfers energy across the interface. A heuristic approximation for the fraction of the energy transferred between the electrons and a joint mode which is then transferred across the interface is to take this fraction equal to the fraction of the energy of

the joint mode which is on the diamond side of the interface. (Note that the reflection and transmission of a phonon at the interface are already included in the solution for the joint vibrational mode.)

The phonon mean free path in diamond is found from the measured thermal diffusivity. For isotopically enriched diamond which was used to measure the thermal boundary resistance between diamond and Pb,³ the measured thermal diffusivity at room temperature is $\alpha = 18.5 \text{ cm}^2/\text{s}$.⁵ For a dielectric solid, the thermal conductivity in the relaxation-time approximation is

$$\kappa = \frac{1}{3} C v^2 \tau, \quad (2)$$

where C is the heat capacity per unit volume, v is the phonon group velocity, and τ is the phonon relaxation time.⁶ The derivation of Eq. (2) treats all phonons as having the same average speed and relaxation time. Since the Debye temperature of diamond is $\Theta_D = 2240 \text{ K}$, the occupied phonon states at room temperature are near the Brillouin-zone center, and therefore the dispersion of the phonon frequency spectrum can be safely neglected. Since the phonon spectrum has two transverse branches and one longitudinal branch, the average phonon velocity can be simply approximated by averaging the transverse and longitudinal wave velocities (see Table I) in the ratio 2:1. Using the definition $\alpha \equiv \kappa/C$ for the thermal diffusivity and the relation $\Lambda = v\tau$ for the phonon mean free path, we find that the phonon mean free path in diamond at room temperature is of the order of 4000 \AA .

The phonon mean free path in Pb is found from the energy broadening of inelastically scattered neutrons. Since the Debye temperature in Pb is $\Theta_D = 102 \text{ K}$, the mean energy of a normal mode at room temperature is approximately the thermal energy $k_B T$. Since the phonon density of states (in the Debye model) is proportional to the square of the frequency, most of the heat energy in Pb at room temperature is carried by high-frequency phonons near the Brillouin-zone edge. In inelastic neutron-scattering experiments on crystalline Pb,⁷ it was found that the short-wavelength phonons near the Brillouin-zone edge are markedly energy broadened, their energy widths increasing with temperature somewhat faster than linearly. At 425 K , the widths of phonons of wave vector $\mathbf{q} = (2\pi/a)(1, 0, 0)$ were found to be $\Gamma = 2.3 \times 10^{-3} \text{ eV}$ for the longitudinal branch and $\Gamma = 1.7 \times 10^{-3} \text{ eV}$ for the transverse branch, where $a = 4.95 \text{ \AA}$ is the lattice constant of Pb. Similar results were found for the widths of

phonons at $\mathbf{q} = (2\pi/a)(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. If a linear temperature dependence is assumed, the energy widths of phonons at room temperature are reduced by a factor of 0.7 compared to their values at 425 K . The lifetime of a phonon is related to its energy width by $\tau = \hbar/\Gamma$,^{7,8} and the mean free path of a phonon is related to its lifetime by $\Lambda = v\tau$. Using the wave velocities in Table I, we find that $\Lambda = 9 \text{ \AA}$ in the longitudinal case and $\Lambda = 5 \text{ \AA}$ in the transverse case, so that the mean free paths of short-wavelength phonons in Pb at room temperature are of the order of 10 \AA .

Using the solutions for the joint vibrational modes derived in the next section, we have evaluated the average energy density in Pb and diamond for each mode. Taking the phonon mean free paths in Pb and diamond, which determine the spatial extent of the joint modes on each side of the interface, equal to 10 and 4000 \AA , respectively, we have then evaluated the fraction f of the energy of each joint mode which is on the diamond side of the interface. Since the rate of energy transfer between the electrons and a joint mode is proportional to their temperature difference, which in turn is proportional to f , and since the fraction of the energy transferred between the electrons and a joint mode which is then transferred across the interface is also proportional to f , the rate of energy transfer across the interface is proportional to f^2 . Averaging f^2 over all the joint modes, we find that $\langle f^2 \rangle = 0.93$. It is therefore a good approximation to set $f = 1$, so that the temperature of the joint modes is equal to the diamond temperature and the energy transferred between the electrons and joint modes is entirely transferred across the interface.

It may seem puzzling that, even though the Pb atoms near the interface interact strongly with the Pb conduction electrons, nevertheless, these atoms are at the diamond temperature rather than the Pb temperature. The following analogy may help to explain this. Consider a spinning bicycle wheel with a streamer attached to the air valve. The spinning wheel with the streamer is analogous to a joint vibrational mode, the wheel being analogous to the diamond atoms on one side of the interface and the streamer, which interacts strongly with its environment, being analogous to the Pb atoms near the interface, which interact strongly with the Pb conduction electrons. Assigning different temperatures to the Pb atoms near the interface and the diamond atoms would be like asserting that the angular velocity of the streamer is different from the angular velocity of the wheel.

II. THEORY

The calculation of the heat transport by this mechanism is relatively straightforward. Diamond and Pb are treated as isotropic elastic continuous media, each characterized by a shear and bulk modulus. The normal modes of the joint system are found by matching the displacements and forces at the interface. The electrons in Pb are treated by a free-electron model. The interaction between electrons and joint modes is modeled by a deformation potential. (Since a deformation potential model only couples electrons to longitudinal phonons, thus

TABLE I. Longitudinal and transverse wave velocities, density, and elastic constants of Pb and diamond for an isotropic elastic model.

	Pb	Diamond
v_l (10^5 cm/s)	2.28	18.2
v_t (10^5 cm/s)	0.91	12.3
ρ (g/cm^3)	11.35	3.51
λ (10^{12} dyn/cm^2)	0.406	1.00
μ (10^{12} dyn/cm^2)	0.093	5.31

neglecting the interaction between electrons and transverse phonons, it underestimates the contribution of this mechanism to the heat transport across the interface.) The rate of energy transfer between the electrons and joint modes by emission and absorption of phonons is then found by Fermi's golden rule.

For an elastic continuous medium, the strain is related to the displacement by

$$u_{ik} = \frac{1}{2} \left[\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} \right]. \quad (3)$$

The general expression for the elastic energy density of an isotropic solid is

$$U = \frac{1}{2} \lambda u_{ii}^2 + \mu u_{ik}^2, \quad (4)$$

where u_{ii}^2 is the square of the sum of the diagonal components of the strain tensor, u_{ik}^2 is the sum of the squares of all the components of the strain tensor, and λ and μ are the elastic constants.⁹ (μ is the shear modulus, and $\lambda + \frac{2}{3}\mu$ is the bulk modulus.) Since $\sigma_{ik} = \partial U / \partial u_{ik}$, the stress is related to the strain by

$$\sigma_{ik} = \lambda u_{jj} \delta_{ik} + 2\mu u_{ik}. \quad (5)$$

The surface traction or force per unit area exerted across a surface is

$$f_i = \sigma_{ik} n_k, \quad (6)$$

where n_i is a unit vector along the outward normal to the surface. The equation of motion of an elastic medium is then

$$\rho \frac{\partial^2 u_i}{\partial t^2} = \frac{\partial \sigma_{ik}}{\partial x_k}, \quad (7)$$

where ρ is the mass density. Substituting (5) and (3) into (7) yields the wave equation, whose solutions are longitudinal plane waves,

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{x}} \exp[i(qx - \omega t)], \quad (8)$$

having the wave velocity

$$v_l = \left[\frac{\lambda + 2\mu}{\rho} \right]^{1/2}, \quad (9)$$

and transverse plane waves,

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{y}} \exp[i(qx - \omega t)], \quad (10)$$

having the wave velocity

$$v_t = \left[\frac{\mu}{\rho} \right]^{1/2}. \quad (11)$$

We determine the elastic moduli of the isotropic model as follows. Using the general expression for the elastic energy density of a cubic crystal⁹ and the experimentally measured elastic moduli of Pb (Ref. 10) and diamond,¹¹ we derive the longitudinal and transverse wave velocities in each of the high-symmetry directions [100], [110], and [111]. Since the unit sphere has 6 [100] directions, 12 [110] directions, and 8 [111] directions, we average these

velocities in the ratio 6:12:8 to find the longitudinal and transverse wave velocities of the isotropic model. Using these velocities and the known densities, we then find the elastic moduli of the isotropic model via Eqs. (9) and (11).

We adopt a Debye model for the phonons in Pb and diamond. For both Pb, which has a face-centered-cubic lattice with one atom per unit cell, and diamond, which has a face-centered-cubic lattice with two atoms per unit cell, the maximum phonon wave vector is given by

$$q_D^3 = 6\pi^2(4/a^3), \quad (12)$$

where the lattice constant a is 4.95 Å for Pb and 3.57 Å for diamond. For each branch of the phonon spectrum, having wave velocity v , the maximum angular frequency is then $\omega_D = vq_D$. Defining the Debye temperature by $\Theta_D \equiv \hbar\omega_D/k_B$, we find that the longitudinal and transverse Debye temperatures of Pb are 220 and 90 K, respectively, and the longitudinal and transverse Debye temperatures of diamond are 2400 and 1600 K, respectively.

Let us consider a bilayer consisting of Pb on the right ($0 < x < L$) and diamond on the left ($-L' < x < 0$). The joint vibrational modes of the bilayer are derived by matching the displacement and surface traction of elastic waves, having the same angular frequency ω , at the interface $x = 0$. The matching conditions at the interface require that the tangential component of the wave vector be the same for the elastic waves on both sides of the interface. For simplicity, we take this tangential component to be in the y direction, but the solutions are easily generalized to an arbitrary direction in the yz plane. Accordingly, on the Pb side, the general solution for a joint vibrational mode is a linear combination of the two longitudinal elastic waves

$$\mathbf{u}(\mathbf{r}, t) = (q_{lx}\hat{\mathbf{x}} + q_y\hat{\mathbf{y}}) \exp[i(q_{lx}x + q_yy - \omega t)], \quad (13a)$$

$$\mathbf{u}(\mathbf{r}, t) = (-q_{lx}\hat{\mathbf{x}} + q_y\hat{\mathbf{y}}) \exp[i(-q_{lx}x + q_yy - \omega t)] \quad (13b)$$

and the four transverse elastic waves

$$\mathbf{u}(\mathbf{r}, t) = (q_y\hat{\mathbf{x}} - q_{tx}\hat{\mathbf{y}}) \exp[i(q_{tx}x + q_yy - \omega t)], \quad (13c)$$

$$\mathbf{u}(\mathbf{r}, t) = (q_y\hat{\mathbf{x}} + q_{tx}\hat{\mathbf{y}}) \exp[i(-q_{tx}x + q_yy - \omega t)], \quad (13d)$$

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{z}} \exp[i(q_{tx}x + q_yy - \omega t)], \quad (13e)$$

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{z}} \exp[i(-q_{tx}x + q_yy - \omega t)]. \quad (13f)$$

Here

$$\omega^2 = v_l^2(q_{lx}^2 + q_y^2) = v_t^2(q_{tx}^2 + q_y^2), \quad (14)$$

where v_l and v_t are the longitudinal and transverse elastic wave velocities in Pb. Similarly, on the diamond side, the general solution for a joint vibrational mode is a linear combination of the two longitudinal elastic waves

$$\mathbf{u}(\mathbf{r}, t) = (q'_{lx}\hat{\mathbf{x}} + q_y\hat{\mathbf{y}}) \exp[i(q'_{lx}x + q_yy - \omega t)], \quad (15a)$$

$$\mathbf{u}(\mathbf{r}, t) = (-q'_{lx}\hat{\mathbf{x}} + q_y\hat{\mathbf{y}}) \exp[i(-q'_{lx}x + q_yy - \omega t)] \quad (15b)$$

and the four transverse elastic waves

$$\mathbf{u}(\mathbf{r}, t) = (q_y\hat{\mathbf{x}} - q'_{tx}\hat{\mathbf{y}}) \exp[i(q'_{tx}x + q_yy - \omega t)], \quad (15c)$$

$$\mathbf{u}(\mathbf{r}, t) = (q_y \hat{\mathbf{x}} + q'_{tx} \hat{\mathbf{y}}) \exp[i(q'_{tx} x + q_y y - \omega t)] , \quad (15d)$$

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{z}} \exp[i(q'_{tx} x + q_y y - \omega t)] , \quad (15e)$$

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{z}} \exp[i(-q'_{tx} x + q_y y - \omega t)] . \quad (15f)$$

Here

$$\omega^2 = v_l'^2(q'_{tx}{}^2 + q_y^2) = v_t'^2(q'_{tx}{}^2 + q_y^2) , \quad (16)$$

where v_l' and v_t' are the longitudinal and transverse elastic wave velocities in diamond. Since there are 6 independent elastic waves on each side, having 12 unknown coefficients, and since there are 3 matching conditions each for the displacement and surface traction, giving 6 equations for these coefficients, there are 6 linearly independent joint vibrational modes for a given ω and q_y .

Just as a clapper excites standing waves in a bell, so too the Pb conduction electrons excite standing waves in the bilayer. Because of the nature of the mechanism which is treated here, we elect to use a representation having standing waves in the direction normal to the interface and traveling waves in the directions parallel to the interface. Taking linear combinations of the above solutions gives, for the six independent elastic waves on the Pb side,

$$\mathbf{u}(\mathbf{r}, t) = (q_{lx} \hat{\mathbf{x}} \cos q_{lx} x + i q_y \hat{\mathbf{y}} \sin q_{lx} x) \exp[i(q_y y - \omega t)] , \quad (17a)$$

$$\mathbf{u}(\mathbf{r}, t) = (q_{lx} \hat{\mathbf{x}} \sin q_{lx} x - i q_y \hat{\mathbf{y}} \cos q_{lx} x) \exp[i(q_y y - \omega t)] , \quad (17b)$$

$$\mathbf{u}(\mathbf{r}, t) = (q_y \hat{\mathbf{x}} \cos q_{tx} x - i q_{tx} \hat{\mathbf{y}} \sin q_{tx} x) \exp[i(q_y y - \omega t)] , \quad (17c)$$

$$\mathbf{u}(\mathbf{r}, t) = (q_y \hat{\mathbf{x}} \sin q_{tx} x + i q_{tx} \hat{\mathbf{y}} \cos q_{tx} x) \exp[i(q_y y - \omega t)] , \quad (17d)$$

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{z}} \cos q_{tx} x \exp[i(q_y y - \omega t)] , \quad (17e)$$

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{z}} \sin q_{tx} x \exp[i(q_y y - \omega t)] , \quad (17f)$$

and for the six independent elastic waves on the diamond side,

$$\mathbf{u}(\mathbf{r}, t) = (q'_{lx} \hat{\mathbf{x}} \cos q'_{lx} x + i q_y \hat{\mathbf{y}} \sin q'_{lx} x) \exp[i(q_y y - \omega t)] , \quad (18a)$$

$$\mathbf{u}(\mathbf{r}, t) = (q'_{lx} \hat{\mathbf{x}} \sin q'_{lx} x - i q_y \hat{\mathbf{y}} \cos q'_{lx} x) \exp[i(q_y y - \omega t)] , \quad (18b)$$

$$\mathbf{u}(\mathbf{r}, t) = (q_y \hat{\mathbf{x}} \cos q'_{tx} x - i q'_{tx} \hat{\mathbf{y}} \sin q'_{tx} x) \exp[i(q_y y - \omega t)] , \quad (18c)$$

$$\mathbf{u}(\mathbf{r}, t) = (q_y \hat{\mathbf{x}} \sin q'_{tx} x + i q'_{tx} \hat{\mathbf{y}} \cos q'_{tx} x) \exp[i(q_y y - \omega t)] , \quad (18d)$$

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{z}} \cos q'_{tx} x \exp[i(q_y y - \omega t)] , \quad (18e)$$

$$\mathbf{u}(\mathbf{r}, t) = \hat{\mathbf{z}} \sin q'_{tx} x \exp[i(q_y y - \omega t)] . \quad (18f)$$

Since the energy of a joint mode resides primarily in the diamond, it is convenient to take the form of the solution for a joint mode on the diamond side to be the same as if the mode resided entirely in the diamond. Consequently, the solutions on the diamond side are chosen to be either pure transverse or pure longitudinal standing waves, i.e., the waves in Eq. (18). (The utility of this choice will become apparent later when we count and normalize the modes.) The solution on the diamond side is then extended to the Pb side by applying the matching conditions at the interface. When the solution on the diamond side is a transverse wave whose oscillations are parallel to the interface, i.e., the wave in Eq. (18e) or (18f), the nature of the wave is unchanged across the interface, the solution on the Pb side being also a transverse wave whose oscillations are parallel to the interface. Since the deformation potential model for the electron-phonon interaction which we employ here only couples electrons to longitudinal phonons, these joint modes do not interact with the Pb conduction electrons and therefore need not be considered further. When the solution on the diamond side is any of the other waves in Eqs. (18), the solution on the Pb side has a longitudinal component, so that these joint modes interact with the Pb conduction electrons. Accordingly, the joint vibrational modes

$$\mathbf{u}_{\lambda q'}(\mathbf{r}, t) = \mathbf{u}_{\lambda q'}(\mathbf{r}) e^{-i\omega_{\lambda q'} t} \quad (19)$$

that we consider here are

$$\mathbf{u}_{1q'}(\mathbf{r}) = \begin{cases} A_{1q'} \left\{ \left[1 + \frac{2(\mu' - \mu)q_y^2}{\mu q_t^2} \right] \frac{q'_x}{q_{lx}} (q_{lx} \hat{\mathbf{x}} \cos q_{lx} x + i q_y \hat{\mathbf{y}} \sin q_{lx} x) \right. \\ \left. - \frac{2(\mu' - \mu)q_y^2}{\mu q_t^2} \frac{q'_x}{q_y} (q_y \hat{\mathbf{x}} \cos q_{tx} x - i q_{tx} \hat{\mathbf{y}} \sin q_{tx} x) \right\} \exp(iq_y y), & x > 0 , \\ A_{1q'} (q'_x \hat{\mathbf{x}} \cos q'_x x + i q_y \hat{\mathbf{y}} \sin q'_x x) \exp(iq_y y), & x < 0 , \end{cases} \quad (20a)$$

$$\mathbf{u}_{2q}(\mathbf{r}) = \begin{cases} A_{2q} \left[\frac{[(\lambda' + 2\mu')q'^2 - 2(\mu' - \mu)q_y^2]}{(\lambda + 2\mu)q_l^2} (q_{lx} \hat{\mathbf{x}} \sin q_{lx} x - iq_y \hat{\mathbf{y}} \cos q_{lx} x) \right. \\ \left. + \left[\frac{[(\lambda' + 2\mu')q'^2 - 2(\mu' - \mu)q_y^2]}{(\lambda + 2\mu)q_l^2} - 1 \right] \frac{q_y}{q_{tx}} (q_y \hat{\mathbf{x}} \sin q_{tx} x + iq_{tx} \hat{\mathbf{y}} \cos q_{tx} x) \right] \exp(iq_y y), & x > 0, \\ A_{2q} (q'_x \hat{\mathbf{x}} \sin q'_x x - iq_y \hat{\mathbf{y}} \cos q'_x x) \exp(iq_y y), & x < 0, \end{cases} \quad (20b)$$

$$\mathbf{u}_{3q}(\mathbf{r}) = \begin{cases} A_{3q} \left[\left\{ 1 - \frac{[\mu'q'^2 - 2(\mu' - \mu)q_y^2]}{\mu q_l^2} \right\} \frac{q_y}{q_{lx}} (q_{lx} \hat{\mathbf{x}} \cos q_{lx} x + iq_y \hat{\mathbf{y}} \sin q_{lx} x) \right. \\ \left. + \frac{[\mu'q'^2 - 2(\mu' - \mu)q_y^2]}{\mu q_l^2} (q_y \hat{\mathbf{x}} \cos q_{tx} x - iq_{tx} \hat{\mathbf{y}} \sin q_{tx} x) \right] \exp(iq_y y), & x > 0, \\ A_{3q} (q_y \hat{\mathbf{x}} \cos q'_x x - iq'_x \hat{\mathbf{y}} \sin q'_x x) \exp(iq_y y), & x < 0, \end{cases} \quad (20c)$$

$$\mathbf{u}_{4q}(\mathbf{r}) = \begin{cases} A_{4q} \left[\frac{2(\mu' - \mu)q_y^2}{(\lambda + 2\mu)q_l^2} \frac{q'_x}{q_y} (q_{lx} \hat{\mathbf{x}} \sin q_{lx} x - iq_y \hat{\mathbf{y}} \cos q_{lx} x) \right. \\ \left. + \left[1 + \frac{2(\mu' - \mu)q_y^2}{(\lambda + 2\mu)q_l^2} \right] \frac{q'_x}{q_{tx}} (q_y \hat{\mathbf{x}} \sin q_{tx} x + iq_{tx} \hat{\mathbf{y}} \cos q_{tx} x) \right] \exp(iq_y y), & x > 0, \\ A_{4q} (q_y \hat{\mathbf{x}} \sin q'_x x + iq'_x \hat{\mathbf{y}} \cos q'_x x) \exp(iq_y y), & x < 0. \end{cases} \quad (20d)$$

The joint modes are labeled by q' , which is the wave vector of the joint mode on the diamond side, in addition to the index $\lambda = 1, \dots, 4$. The angular frequencies of the joint modes are $\omega_{\lambda q'} = v_l' q'$ for $\lambda = 1, 2$ and $\omega_{\lambda q'} = v_t' q'$ for $\lambda = 3, 4$, where v_l' and v_t' are the longitudinal and transverse wave velocities in diamond. ρ , λ , and μ are the density and elastic constants of Pb; ρ' , λ' , and μ' are the density and elastic constants of diamond. The constants $A_{\lambda q'}$ are normalization constants which will be determined later.

The joint vibrational modes which are considered here have two properties. First, on the diamond side of the interface, they are propagating (as opposed to evanescent). Because of the long phonon mean free path in diamond, the energy of such modes resides primarily in the diamond, and therefore they are at the diamond temperature. Second, on the Pb side of the interface, their longitudinal component is propagating. Because the deformation potential model couples electrons to longitudinal phonons, such modes interact strongly with the Pb conduction electrons. As a result of these requirements, the longitudinal Debye frequency of Pb, which is smaller than both the longitudinal and transverse Debye frequencies of diamond, is the maximum frequency of the joint vibrational modes which are considered here. (The higher-frequency vibrational modes in diamond, which have evanescent tails in Pb, also interact with the Pb conduction electrons. The contribution of these joint modes to the heat transport across the interface is not considered here.)

In the frequency range between the longitudinal and transverse Debye frequencies of Pb, the solution on the Pb side for a joint vibrational mode is a linear combination of a propagating longitudinal wave and an evanes-

cent transverse wave, whereas below this frequency range the solution on the Pb side for a joint vibrational mode is a linear combination of longitudinal and transverse waves, both of which are propagating. In the solutions adopted here, we approximate the solution on the Pb side in the entire frequency range as a linear combination of propagating longitudinal and transverse waves, as in Eq. (20). This is a reasonable approximation, since, if the transverse component on the Pb side is a small fraction of the total wave, the error committed thereby in the longitudinal component is small, whereas, if the transverse component on the Pb side is a large fraction of the total wave, the wave couples weakly to the Pb conduction electrons and therefore does not contribute significantly to the heat transport across the interface.

The directions of the waves on the Pb and diamond sides in a joint vibrational mode are related by Snell's law. For the joint vibrational modes considered here, the directions of the waves on the diamond side are unrestricted. However, because the elastic wave speeds are much slower in Pb than in diamond (see Table I), the directions of the waves on the Pb side are concentrated within a narrow critical cone about the interface normal. For the joint modes in which the solution on the diamond side is a pure longitudinal wave, the critical angles of the longitudinal and transverse waves on the Pb side are 7° and 3° , respectively; for the joint modes in which the solution on the diamond side is a pure transverse wave, the critical angles of the longitudinal and transverse waves on the Pb side are 11° and 4° , respectively. (If the angles of the waves on the Pb side are greater than the critical angles, the waves are totally reflected at the interface, the matching waves on the diamond side being evanescent rather than propagating. These joint modes,

which can transfer heat across the interface via anharmonic interactions with the diamond phonons, are not treated here.)

Since the joint modes reside primarily in the diamond, the number of joint modes is approximately the same as if the joint modes resided entirely in the diamond. Applying periodic boundary conditions in the diamond yields the allowed values of the wave vector $\mathbf{q}'=(q'_x, q'_y, q'_z)$ of the joint modes in the diamond,

$$q'_x = 2\pi n_x / L', \quad n_x = 0, 1, 2, \dots, \quad (21a)$$

$$q'_y = 2\pi n_y / L_y, \quad n_y = 0, \pm 1, \pm 2, \dots, \quad (21b)$$

$$q'_z = 2\pi n_z / L_z, \quad n_z = 0, \pm 1, \pm 2, \dots, \quad (21c)$$

where L' , L_y , and L_z are the thicknesses of the diamond in the directions perpendicular and parallel to the interface. (Because the representation we use has standing waves in the x direction, $q'_x \geq 0$). The density of states in q' space for each of the four joint vibrational modes in Eq. (20) is then

$$\rho(\mathbf{q}') = V' / (2\pi)^3, \quad (22)$$

where $V' = L' L_y L_z$ is the volume of the diamond.

Similarly, the normalization of the joint modes can be derived by considering only the part of the joint modes on the diamond side. We expand the displacement operator in the joint modes,

$$\mathbf{u}(\mathbf{r}) = \sum'_{\lambda, \mathbf{q}'} [a_{\lambda \mathbf{q}'} \mathbf{u}_{\lambda \mathbf{q}'}(\mathbf{r}) + a_{\lambda \mathbf{q}'}^\dagger \mathbf{u}_{\lambda \mathbf{q}'}^*(\mathbf{r})], \quad (23)$$

where $a_{\lambda \mathbf{q}'}^\dagger$ and $a_{\lambda \mathbf{q}'}$ are creation and annihilation operators for the joint mode $\mathbf{u}_{\lambda \mathbf{q}'}(\mathbf{r})$, and the prime on the summation sign indicates that the sum over \mathbf{q}' is restricted to values of \mathbf{q}' with $q'_x \geq 0$. [In principle, all the joint modes, not just the subset in Eq. (20), should be included in the expansion in Eq. (23), but this need not concern us here.] In terms of creation and annihilation operators, the Hamiltonian is

$$H = \sum'_{\lambda, \mathbf{q}'} \frac{1}{2} \hbar \omega_{\lambda \mathbf{q}'} [a_{\lambda \mathbf{q}'} a_{\lambda \mathbf{q}'}^\dagger + a_{\lambda \mathbf{q}'}^\dagger a_{\lambda \mathbf{q}'}]. \quad (24)$$

The energy density is the sum of the kinetic-energy density, given by $\frac{1}{2} \rho \dot{\mathbf{u}}_i^2$, and the elastic energy density, given by Eq. (4). Requiring that the total energy, given by the integral of the energy density over the volume of the diamond, reduces to Eq. (24), we find that the normalization constants, first introduced in Eq. (20), are

$$A_{\lambda \mathbf{q}'} = \frac{1}{q'} \left[\frac{\hbar}{\rho' \omega_{\lambda \mathbf{q}'} V'} \right]^{1/2}, \quad (25)$$

where ρ' and V' are the density and volume of the diamond, respectively. [Some readers may have expected an additional factor of $1/\sqrt{2}$ in Eq. (25); however, this factor is absent when a standing-wave representation is used instead of a traveling-wave representation.]

The conduction electrons in Pb are treated by a free-electron model. The electron wave functions are

$$\psi_{\mathbf{k}} = \frac{1}{\sqrt{V}} e^{i\mathbf{k} \cdot \mathbf{r}}, \quad (26)$$

having energy $E_{\mathbf{k}} = \hbar^2 k^2 / 2m$, where V is the volume of

Pb. Applying periodic boundary conditions in Pb yields the allowed values of the wave vector $\mathbf{k}=(k_x, k_y, k_z)$,

$$k_x = 2\pi n_x / L, \quad n_x = 0, \pm 1, \pm 2, \dots, \quad (27a)$$

$$k_y = 2\pi n_y / L_y, \quad n_y = 0, \pm 1, \pm 2, \dots, \quad (27b)$$

$$k_z = 2\pi n_z / L_z, \quad n_z = 0, \pm 1, \pm 2, \dots, \quad (27c)$$

where L , L_y , and L_z are the thicknesses of Pb in the directions perpendicular and parallel to the interface.

We model the electron-phonon interaction in Pb by a deformation potential

$$V(\mathbf{r}) = \frac{2}{3} C \nabla \cdot \mathbf{u}(\mathbf{r}), \quad (28)$$

where C is the coupling constant. (In a free-electron model, $C = E_F$.) An appealing aspect of the theory presented here is that, provided the electron-phonon interaction in Pb is strong enough that the phonon mean free path in Pb is short and therefore that the joint vibrational modes between Pb and diamond reside primarily in the diamond, the heat flow across the interface caused by the electron-phonon interaction is independent of the magnitude of the coupling constant C . This is because the effect of the coupling constant on the strength of interaction between electrons and joint modes is canceled by its effect on their volume of interaction.

The volume of interaction between the Pb conduction electrons and the joint vibrational modes is related to the strength of the electron-phonon coupling, because the joint vibrational modes are attenuated, the attenuation length of the energy density of a joint mode being equal to the coherence length of the joint mode. (As a result of this attenuation, an electron far away from the interface, i.e., farther than the coherence length of a joint mode, cannot emit or absorb a phonon of that joint mode.) In practice, the dominant phonon scattering process in Pb which destroys the coherence of a joint mode is the emission and absorption of phonons by the conduction electrons. Since the deformation potential only couples electrons with longitudinal phonons, we take the attenuation length of the energy density of a joint mode on the Pb side equal to the mean free path Λ of the longitudinal phonon on the Pb side which is part of that joint mode.¹² Since the energy density is proportional to the square of the displacement, the attenuation length of the displacement of a joint mode on the Pb side is equal to 2Λ . Since the wave vector of short-wavelength phonons in Pb, which carry most of the heat energy at room temperature, is approximately equal to the Debye wave vector $q_D = 1.25 \text{ \AA}^{-1}$ and since the phonon mean free path Λ of short-wavelength phonons in Pb is of the order of 10 \AA , q_D is much greater than $1/2\Lambda$, so that the matching conditions of a joint mode at the interface are not significantly modified by the attenuation of the joint mode on the Pb side. Multiplying the solutions on the Pb side in Eq. (20) by $\exp(-x/2\Lambda)$ and substituting Eqs. (20), (23), and (25) into (28), we obtain the interaction between conduction electrons and joint vibrational modes,

$$V(\mathbf{r}) = \sum'_{\lambda, \mathbf{q}'} V_{\lambda \mathbf{q}'}(\mathbf{r}), \quad (29)$$

where

$$V_{1q'}(\mathbf{r}) = -\frac{2}{3} C \left[\frac{\hbar}{\rho' \omega_{1q'} V'} \right]^{1/2} \left[1 + \frac{2(\mu' - \mu) q_y^2}{\mu q_i^2} \right] \frac{q'_x q'_i}{q_{lx} q'} (a_{1q'} e^{iq_y y} + a_{1q'}^\dagger e^{-iq_y y}) \sin q_{lx} x \exp(-x/2\Lambda), \quad (30a)$$

$$V_{2q'}(\mathbf{r}) = \frac{2}{3} C \left[\frac{\hbar}{\rho' \omega_{2q'} V'} \right]^{1/2} \frac{[(\lambda' + 2\mu') q'^2 - 2(\mu' - \mu) q_y^2]}{(\lambda + 2\mu) q_i^2} \frac{q_i^2}{q'} (a_{2q'} e^{iq_y y} + a_{2q'}^\dagger e^{-iq_y y}) \cos q_{lx} x \exp(-x/2\Lambda), \quad (30b)$$

$$V_{3q'}(\mathbf{r}) = -\frac{2}{3} C \left[\frac{\hbar}{\rho' \omega_{3q'} V'} \right]^{1/2} \left\{ 1 - \frac{[\mu' q'^2 - 2(\mu' - \mu) q_y^2]}{\mu q_i^2} \right\} \frac{q_y q'_i}{q_{lx} q'} (a_{3q'} e^{iq_y y} + a_{3q'}^\dagger e^{-iq_y y}) \sin q_{lx} x \exp(-x/2\Lambda), \quad (30c)$$

$$V_{4q'}(\mathbf{r}) = \frac{2}{3} C \left[\frac{\hbar}{\rho' \omega_{4q'} V'} \right]^{1/2} \frac{2(\mu' - \mu) q_y^2 q'_x q'_i}{(\lambda + 2\mu) q_i^2 q_y q'} (a_{4q'} e^{iq_y y} + a_{4q'}^\dagger e^{-iq_y y}) \cos q_{lx} x \exp(-x/2\Lambda). \quad (30d)$$

The rate of energy transfer between the conduction electrons (at the Pb temperature) and the joint vibrational modes (at the diamond temperature) is derived by Fermi's golden rule. The energy change of the electrons per unit time is the sum of the changes due to phonon emission and absorption,

$$\frac{dE}{dt} = \left(\frac{dE}{dt} \right)_{\text{em}} + \left(\frac{dE}{dt} \right)_{\text{abs}}, \quad (31)$$

where

$$\left(\frac{dE}{dt} \right)_{\text{em}} = 2 \sum_{\mathbf{k}, \mathbf{k}', \lambda, q'} \frac{2\pi}{\hbar} |\langle \mathbf{k}' | V_{\lambda q'} | \mathbf{k} \rangle|_{\text{em}}^2 \delta(E_{\mathbf{k}'} + \hbar\omega_{\lambda q'} - E_{\mathbf{k}}) f(\mathbf{k}) [1 - f(\mathbf{k}')] (-\hbar\omega_{\lambda q'}), \quad (32a)$$

and

$$\left(\frac{dE}{dt} \right)_{\text{abs}} = 2 \sum_{\mathbf{k}, \mathbf{k}', \lambda, q'} \frac{2\pi}{\hbar} |\langle \mathbf{k}' | V_{\lambda q'} | \mathbf{k} \rangle|_{\text{abs}}^2 \delta(E_{\mathbf{k}'} - \hbar\omega_{\lambda q'} - E_{\mathbf{k}}) f(\mathbf{k}) [1 - f(\mathbf{k}')] (\hbar\omega_{\lambda q'}). \quad (32b)$$

Here $\langle \mathbf{k}' | V_{\lambda q'} | \mathbf{k} \rangle_{\text{em}}$ and $\langle \mathbf{k}' | V_{\lambda q'} | \mathbf{k} \rangle_{\text{abs}}$ are the matrix elements for phonon emission and absorption, and $f(\mathbf{k})$ is the Fermi-Dirac distribution function for the electrons at the temperature of Pb. The matrix elements for phonon emission and absorption, $\langle \mathbf{k}' | V_{\lambda q'} | \mathbf{k} \rangle_{\text{em}}$ and $\langle \mathbf{k}' | V_{\lambda q'} | \mathbf{k} \rangle_{\text{abs}}$, are proportional to $\sqrt{n_{\lambda q'} + 1}$ and $\sqrt{n_{\lambda q'}}$, respectively, where $n_{\lambda q'}$ is the Bose-Einstein distribution function for the phonons of the joint modes at the temperature of diamond. Because the tangential components of the wave vector of a joint mode are conserved across the interface, the allowed values of q_y and q_z for the longitudinal part of a joint mode on the Pb side are the same as the allowed values of k_y and k_z for the electron wave function; compare Eqs. (21b) and (21c), and Eqs. (27b) and (27c). On the other hand, the allowed values of q_x for the longitudinal part of a joint mode on the Pb side are different than the allowed values of k_x for the electron wave function. At first sight it thus may seem that the usual requirement that the total wave vector be conserved in a transition can be satisfied for the y and z components, but not for the x component. However, because of the attenuation of a joint mode in the normal direction, the Fourier decomposition (with respect to x) of the longitudinal part of a joint mode on the Pb side does not consist of a single wave vector, but has a distribution of wave vectors, spread by an amount of the order of $1/2\Lambda$ about a central value. The effect of this is to relax the requirement of exact wave vector conservation for the x component (by an amount of the order of $1/2\Lambda$), so

that transitions in which the electrons emit and absorb phonons can occur.

Since all of the energy transferred between the electrons and joint modes is transferred across the interface, the Kapitza conductance is equal to the rate of energy transfer per unit area and per unit temperature difference. Making use of the condition that $2q_D \Lambda \gg 1$, we find that

$$\sigma_K = INk_B v_l f_D(\Theta_{Dl}/T), \quad (33)$$

where N is the atomic density of Pb, v_l is the longitudinal wave velocity in Pb, and Θ_{Dl} is the Debye temperature of longitudinal phonons in Pb. $f_D(y)$ is the Debye function¹³ defined by

$$f_D(y) = \frac{3}{y^3} \int_0^y \frac{e^x}{(e^x - 1)^2} x^4 dx. \quad (34)$$

For $y \rightarrow 0$, $f_D(y) \rightarrow 1$, whereas, for $y \rightarrow \infty$, $f_D(y) \rightarrow 4\pi^4/5y^3$. The constant I is

$$I = \sum_{\lambda=1}^4 I_\lambda, \quad (35)$$

where

$$I_1 = \frac{1}{2} \frac{\rho}{\rho'} \left\{ (\sin\theta_l - \theta_l \cos\theta_l) + 2 \left[2 \frac{(\mu' - \mu)}{\mu'} \frac{\rho'}{\rho} \frac{\mu'}{(\lambda' + 2\mu')} \right] \frac{(\sin\theta_l - \theta_l \cos\theta_l - \frac{1}{3}\sin^3\theta_l)}{\sin^2\theta_l} \right. \\ \left. + \left[2 \frac{(\mu' - \mu)}{\mu'} \frac{\rho'}{\rho} \frac{\mu'}{(\lambda' + 2\mu')} \right]^2 \frac{(\sin\theta_l - \theta_l \cos\theta_l - \frac{1}{3}\sin^3\theta_l - \frac{2}{15}\sin^5\theta_l)}{\sin^4\theta_l} \right\}, \quad (36a)$$

$$I_2 = \frac{1}{2} \frac{\rho'}{\rho} \sin\theta_l \left\{ 1 - \frac{4}{3} \left[2 \frac{(\mu' - \mu)}{\mu'} \frac{\mu'}{(\lambda' + 2\mu')} \right] + \frac{8}{15} \left[2 \frac{(\mu' - \mu)}{\mu'} \frac{\mu'}{(\lambda' + 2\mu')} \right]^2 \right\}, \quad (36b)$$

$$I_3 = \frac{1}{2} \frac{\rho}{\rho'} \left\{ \left[1 - \frac{\rho'}{\rho} \right]^2 (\theta_l \sec\theta_l - \sin\theta_l) + 2 \left[1 - \frac{\rho'}{\rho} \right] \left[2 \frac{(\mu' - \mu)}{\mu'} \frac{\rho'}{\rho} \right] \frac{(\theta_l \sec\theta_l - \sin\theta_l - \frac{2}{3}\sin^3\theta_l)}{\sin^2\theta_l} \right. \\ \left. + \left[2 \frac{(\mu' - \mu)}{\mu'} \frac{\rho'}{\rho} \right]^2 \frac{(\theta_l \sec\theta_l - \sin\theta_l - \frac{2}{3}\sin^3\theta_l - \frac{8}{15}\sin^5\theta_l)}{\sin^4\theta_l} \right\}, \quad (36c)$$

$$I_4 = \frac{4}{15} \left[\frac{\mu' - \mu}{\mu'} \right]^2 \frac{\rho'}{\rho} \sin\theta_l. \quad (36d)$$

Here $\theta_l = \sin^{-1}(v_l/v_l')$ is the maximum angle of the longitudinal component of the wave on the Pb side when the wave on the diamond side is longitudinal, and $\theta_t = \sin^{-1}(v_t/v_t')$ is the maximum angle of the longitudinal component of the wave on the Pb side when the wave on the diamond side is transverse.

In deriving Eq. (33), we substituted, for the mean free path of a longitudinal phonon in Pb due to electron-phonon scattering,

$$\Lambda = \frac{9\pi\hbar^3 \rho v_l}{2m^2 C^2 q}. \quad (37)$$

This result applies when the product of the electron mean free path l and the phonon wave vector q is greater than unity.¹⁴ The electrical resistivity of Pb at room temperature is $\rho_e = 21.0 \mu\Omega \text{ cm}$. Since $\rho_e = m/ne^2\tau$ and $l = v_F\tau$, the electron mean free path in Pb at room temperature is $l = 23 \text{ \AA}$. Since the Debye wave vector in Pb is $q_D = 1.25 \text{ \AA}^{-1}$, the condition $ql > 1$ is easily satisfied at room temperature.

For $T = 295 \text{ K}$ and $\Theta_{Dl} = 220 \text{ K}$, $f_D(\Theta_{Dl}/T) = 0.97$. Evaluating Eq. (33), we find that the Kapitza conductance between diamond and Pb at room temperature is

$$\sigma_K = 3.1 \times 10^3 \text{ W cm}^{-2} \text{ K}^{-1}, \quad (38)$$

which is the same as the experimentally measured value.³ (This agreement is so much better than can reasonably be expected from this theory that it is almost embarrassing.) From 300 to 100 K, the measured value of the Kapitza conductance between diamond and Pb decreases smoothly, falling by about 15%. Over the same temperature range, the theoretical value¹⁵ predicted by Eq. (33) also decreases smoothly, falling by about 20%, in substantial agreement with experiment. (The faster falloff predicted by theory is likely an artifact of our using the Debye model, which neglects the leveling off of the phonon spectrum at the zone boundary.)

III. DISCUSSION

The primary difference between the mechanism presented here and the acoustic mismatch theory is that in the acoustic mismatch model there is no transfer of energy between the conduction electrons and the phonons in Pb, since in that model they are both at the same temperature. The crucial ingredient of the mechanism presented here is that the Pb atoms within a phonon mean free path of the interface are at the same temperature as the diamond atoms, since these Pb and diamond atoms are strongly coupled together into joint vibrational modes. Because the Pb atoms near the interface are at a different temperature than the Pb conduction electrons, energy is transferred between them.

In an attempt to determine whether the conduction electrons in Pb are involved in the heat transport across the interface with diamond, the following experiment was performed.³ Two samples were prepared. In one, a 35- \AA layer of Bi was deposited on the diamond substrate before depositing a 1000- \AA layer of Pb. In the second, the Pb layer was deposited directly on the diamond substrate. Because Bi and Pb, which are next to each other in the Periodic Table, have similar Debye temperatures, the heat transport across the interface by phonons is not expected to be significantly affected by the Bi interlayer. On the other hand, because Bi is a semimetal, having an electron density much less than Pb, the heat transport across the interface by the interaction of diamond phonons with conduction electrons might be expected to be severely reduced by the presence of the Bi interlayer. What was found experimentally was that the cooling rate of the Pb layer was substantially the same with or without the Bi interlayer.

Our interpretation of this experiment is as follows. The dominant cause of phonon scattering in a metal is the electron-phonon interaction. Since Bi has a much smaller electron density than Pb, phonons in Bi are ex-

pected to be weakly attenuated compared to those in Pb. Consequently, in the sample with the Bi interlayer, a joint vibrational mode maintains its coherence through the thin Bi layer, extending about 10 Å into the Pb, just as in the sample without the Bi interlayer. Thus, in both samples, the Pb conduction electrons interact directly with the joint vibrational modes, and therefore the cooling rates of the Pb layers in the two samples are similar.

The reader is cautioned not to apply indiscriminately the simple formula in Eq. (33) to other metals besides Pb, since this formula assumes that the energy of a joint vibrational mode resides primarily in diamond, and this assumption will surely fail in metals having a weaker electron-phonon interaction and therefore a longer phonon mean free path. For example, if one naively applies the formula in Eq. (33) to calculate the Kapitza conductance at room temperature between diamond and Au or between diamond and Al, the predicted values are, respectively, 2.5 times and 20 times larger than the observed values.³ (In the acoustic mismatch model, the predicted values³ are, respectively, 10 times smaller than and equal to the observed values.) Since the room-temperature electrical resistivity due to electron-phonon scattering is 2.20 $\mu\Omega$ cm in Au and 2.74 $\mu\Omega$ cm in Al,

compared to 21.0 $\mu\Omega$ cm in Pb, it is expected that the electron-phonon interaction is weaker in Au and Al than in Pb, and therefore that the phonon mean free path is longer in Au and Al than in Pb. Since the transfer of energy across the interface by the interaction between the conduction electrons and a joint vibrational mode is reduced when the energy of the joint mode is distributed between the metal and diamond rather than being primarily in diamond, this may explain why the formula in Eq. (33), which assumes that all the energy of a joint vibrational mode resides in the diamond, overestimates the Kapitza conductances of Au and Al with diamond, compared to the observed values.

Finally, since the caveat which applies to Au and Al does not apply to Pb, we believe that the observed anomaly in the Kapitza conductance between diamond and Pb is explained by the theory presented here.

ACKNOWLEDGMENTS

One of us (M.L.H.) thanks L. C. Davis, K. C. Hass, G. Mozurkewich, and D. A. Young for helpful discussions. The other (A.W.O.) is grateful to the National Science Foundation for partial support.

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