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Acoustic Mismatch Model for Thermal Contact Conductance of van der Waals Contacts Under Static Force

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Abstract

Van der Waals interfaces play a major role in technology today. Thermal transport in material systems with van der Waals interfaces is mainly limited by the contact conductance. Although the effects of static force, such as pressure or the electrostatic part of hydrogen bonds, on the thermal contact conductance of van der Waals interfaces have been examined in a few studies, the focus was either on numerical simulation using techniques such as molecular dynamics or on experimental investigation. In this paper, an analytical model of thermal contact conductance that accounts for the effects of static force and adhesion energy is presented. The application of static forces is found to cause a decrease of the intermolecular distance, which leads to increased interatomic forces across the interfaces and thus higher thermal conductance. The model is in good agreement with experimental data on the effect of pressure on thermal conductance collected by Gotsmann and Lantz (Nature Materials, Vol. 12, 59, 2012).

Van der Waals interfaces play a significant role in modern science and technology [1]. Thermal transport across van der Waals interfaces has been investigated by many research groups [2,3] including studies on the effects of static forces such pressure [4-6] and the electrostatic part of hydrogen bonds [1]. Recently, Koblinski's group [4] modeled the effect of pressure using non-equilibrium molecular dynamics simulation. Zhang et al. [1] studied the effect of hydrogen bonds using molecular dynamics simulation and revealed collaborative effects of the electrostatic and van der Waals portions in the hydrogen bonds. They observed that stronger hydrogen bonds pull molecules closer to the interface. This shorter intermolecular distance leads to increased interatomic forces across the interfaces, which result in larger interfacial heat and higher thermal conductance. This effect is similar to that of pressure observed by other researchers [4-6]. The author previously developed an analytical model for the contact conductance of van der Waals interfaces without incorporating the effects of static force [2]. In this paper, this model [2] is extended to incorporate the effects of static force. The model successfully explains the pressure dependence of thermal conductance of flat silicon (Si) tips [6] in contact with tetrahedral amorphous carbon (taC) without evoking the concept of quantized thermal conductance. Pettes and Shi [7] had argued that the concept of quantized thermal conductance is not needed to explain their data for a similar setup in which a round tip was used instead of a flat one. Thus, the findings in this paper greatly help to resolve the confusion in the literature [6,7].

Apart from incorporating the static force term, the current model also differs from the previous model [2] in its approach for the calculation of surface traction. In the previous model, the spring constant was calculated between a pair of atoms, and the interfacial spring constant per unit area was calculated by multiplying the atomic pair spring constant with the number of atoms

per unit area on the surface of the contacting substrates. The calculation of the number of atoms per unit area is inherently problematic and requires certain assumptions. In the previous model, the number of surface atoms per unit area was assumed to be $1/a_p^2$, where a_p is the dimension of the primitive cell. In the current model, this problem is circumvented by calculating the surface traction between two macroscopic bodies with a flat surface by integrating the forces over all the atoms. This formulation automatically transforms the interplanar interactions between two bodies interacting via the Lennard–Jones potential between two atoms into force/area, i.e., the form of surface traction [8]. The surface traction (force/area) is given by [8]

$$T(z) = \frac{8\Delta\gamma}{3z_0} \left[\left(\frac{z_0}{z} \right)^3 - \left(\frac{z_0}{z} \right)^9 \right], \quad (1)$$

where T is the surface traction, z_0 is the equilibrium distance between the two surfaces, z is the distance between the surfaces, and $\Delta\gamma$ is the adhesion energy of the interface. Adhesion energy in Eq. (1) is directly related to the Lennard–Jones parameters. Detailed derivation of Eq. (1) from Lennard–Jones potential between two atoms and how adhesion energy is related to the Lennard–Jones parameters are given in reference [8]. Note for two dissimilar materials, z_0 is given by⁸ $(2/15)^{1/6} (r_1+r_2)/2$, where r_1 and r_2 are the nearest-neighbor atomic distances at equilibrium in material 1 and 2, respectively. From Eq. (1), the spring constant per unit area is given by [2]

$$\left(\frac{\partial T}{\partial z} \right)_{z=z_0} = 16\Delta\gamma / z_0^2. \quad (2)$$

Because the spring constant is inversely proportional to z_0^2 , the application of a static force will increase the spring constant (making the interface stiffer) because a static force will lead to a lower value of z_0 . Using molecular dynamics simulations, Zhang et al.[1] observed a similar effect for hydrogen bonds. They found that stronger hydrogen bonds pull molecules closer to the interface,

which results in a shorter intermolecular distance, leading to increased interatomic forces across the interfaces.

The surface traction under static traction (force/area) (p) is given by

$$T(z, p) = \frac{8\Delta\gamma}{3z_0} \left[\left(\frac{z_0}{z} \right)^3 - \left(\frac{z_0}{z} \right)^9 \right] + p. \quad (3)$$

The equilibrium separation distance under pressure (z_0') can be obtained by setting $T(z, p) = 0$, i.e.,

$$\frac{8\Delta\gamma}{3z_0} \left[\left(\frac{z_0}{z_0'} \right)^3 - \left(\frac{z_0}{z_0'} \right)^9 \right] + p = 0. \quad (4)$$

The modified interfacial spring constant (K') per unit area is given by $\left(\partial T / \partial z \right)_{z=z_0'}$:

$$K' = \frac{8\Delta\gamma}{3z_0} \left(\frac{-3z_0^3}{(z_0')^4} + \frac{9z_0^9}{(z_0')^{10}} \right). \quad (5)$$

Equation (5) shows that when $p = 0$, $K' = 16\Delta\gamma / z_0^2$ (for $p = 0$, $z_0' = z_0$), which is the spring constant for the unpressurized case. The transmissivity of phonons is given by [2]

$$\tau_{v-AMM} = \frac{4z_1 z_2 \cos \theta_1 \cos \theta_2}{(z_1 \cos \theta_1 + z_2 \cos \theta_2)^2 + \frac{\omega^2}{K'^2} (z_1 z_2 \cos \theta_1 \cos \theta_2)^2}, \quad (6)$$

where $z_1 = \rho_1 v_1$ and $z_2 = \rho_2 v_2$ are the acoustic impedances of material 1 and 2, respectively; ρ is the density; v is the velocity; and θ is the angle between the normal and phonon propagation direction. θ_1 and θ_2 are related by Snell's law as $\sin \theta_1 / v_1 = \sin \theta_2 / v_2$. The interface conductance per unit area (g) can be written as [2]

$$g = \frac{1}{2} \int_0^{\omega_{m1}} \int_0^{\theta_c} c_\omega v_\omega \tau_\omega \sin \theta \cos \theta d\theta d\omega. \quad (7)$$

In Eq. (7), it is assumed that side 1 has a smaller maximum phonon frequency than side 2. ω_{m1} is the maximum phonon frequency of side 1, v_ω is the group velocity, c_ω is the frequency-dependent heat capacity per unit volume, and θ_c is the critical angle.

As an example, calculations have been performed for Si and taC because of the availability of experimental data on a flat surface for these materials [6]. Data is also available on other material sets; however, the contact surface in those experiments was generally round,[7] adding the complexity of elastic deformation of a round tip [8]. Another reason for comparing the model with a Si/taC interface is that the conclusions of different groups appear to conflict [6,7]. For example, Gotsmann and Lantz [6] explained data on a Si/taC interface based on quantized thermal conductance, whereas Pettes and Shi [7] contradicted that conclusion but with data collected on a Si/Si interface for a round tip.

Table 1 lists the properties of taC and the sources from which they were obtained. Both the transverse and longitudinal phonon velocities were calculated using Young's modulus (E) and Poisson's ratio with the help of Lamé constants. For consistency, the density (ρ) of taC was also calculated using Young's modulus as they are related by [10] $\rho = 1.37 + E^{2/3}/44.65$. Another critical parameter for the calculation was the equilibrium nearest-neighbor atomic separation (r), which is also listed in Table 1 for taC. For Si, all these properties are given in Ref. 2. Finally, one of the most important interfacial parameters is the adhesion energy. For Si/taC, adhesion energies in the range of 0.1–0.35 J/m² have been measured experimentally [12]. Therefore, the calculations were been performed for both 0.1 and 0.35 J/m².

Figure 1 shows the effect of pressure on g . With increasing pressure, g approaches g_{amm} , where g_{amm} is the conductance given by the traditional acoustic mismatch model, which assumes an infinite interfacial spring constant [2]. All the calculations have been performed at 300 K. The reason g increases with increasing pressure is because applying pressure causes a decrease of the effective equilibrium separation distance between the two surfaces, thereby increasing the interfacial spring constant/stiffness. To the first degree, the spring constant is inversely proportional to the square of the equilibrium separation distance, as shown in Eq. (2). The exact value of K is given by Eq. (5). Figure 1 also shows that for a higher adhesion energy, g is higher for a given pressure because the interface is inherently stiffer. Initially, the pressure does not have much effect because the inherent stiffness due to the adhesion energy is much higher than the increase in stiffness due to pressure.

Figure 2 compares the model with the data collected by Gotsmann and Lantz [6], where m is given by g/p and p is the pressure in MPa. Within experimental error, the model matches well with the data both in magnitude and trend. Gotsmann and Lantz [6] explained their data by invoking quantized conductance of a one-dimensional body, whereas Pettes and Shi [7] did not find any evidence of quantized conductance for a Si/Si interface. The setup of Pettes and Shi [7] was more complicated because the tip was round, which leads to elastic deformation of the tip due to pressure and interfacial adhesion [8] that can lead to more uncertainty in the data analysis. Gotsmann and Lantz [6] used a flat tip that significantly simplified the modeling and data analysis. Based on the pressure-dependent model proposed in this paper, it appears that the data from Gotsmann and Lantz [6] can be explained based on the traditional 3D theory of phonon transport.

It is worth mentioning that in the current model, pressure can affect the stiffness of interatomic bonds in the materials themselves [4]; however, the pressure required is very high

because the interatomic bonds within the materials are very strong. Therefore, strictly speaking, this model assumes that both the materials are effectively rigid and pressure does not affect the inherent phonon properties of the materials. This is a very good approximation as long as the interfacial bonds are much weaker than the interatomic bonds within the adjoining materials. If the pressure approaches within an order of magnitude of elastic constants of the bulk materials then phonon transmissivity can change [13].

Although the model introduced in this paper is for flat surfaces, it can be extended for curved surfaces by allowing for elastic deformation of curve surfaces [14]. Once the constriction radius of the deformed surfaces is determined, the transmissivity of phonons calculated from the pressure dependent model given in this paper can be used to calculate the conductance of the deformed surface.

In summary, we have introduced a static force-dependent acoustic mismatch model for van der Waals interfaces. The model explains data reported in the literature and provides much needed clarity.

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Table 1: Physical properties of taC used for calculations

Young's modulus [9]	759 GPa
Poisson's ratio [9]	0.17
Longitudinal phonon velocity (Calculated using Young's modulus and Poisson's ratio using Lamé constants)	15884 m/s
Transverse phonon velocity (Calculated using Young's modulus and Poisson's ratio using Lamé constants)	10015 m/s
Debye velocity	11022. m/s
Nearest neighbor [11]	0.153 nm
Density [10] (Calculated using Young's modulus, see text for details)	3233 kg/m ³

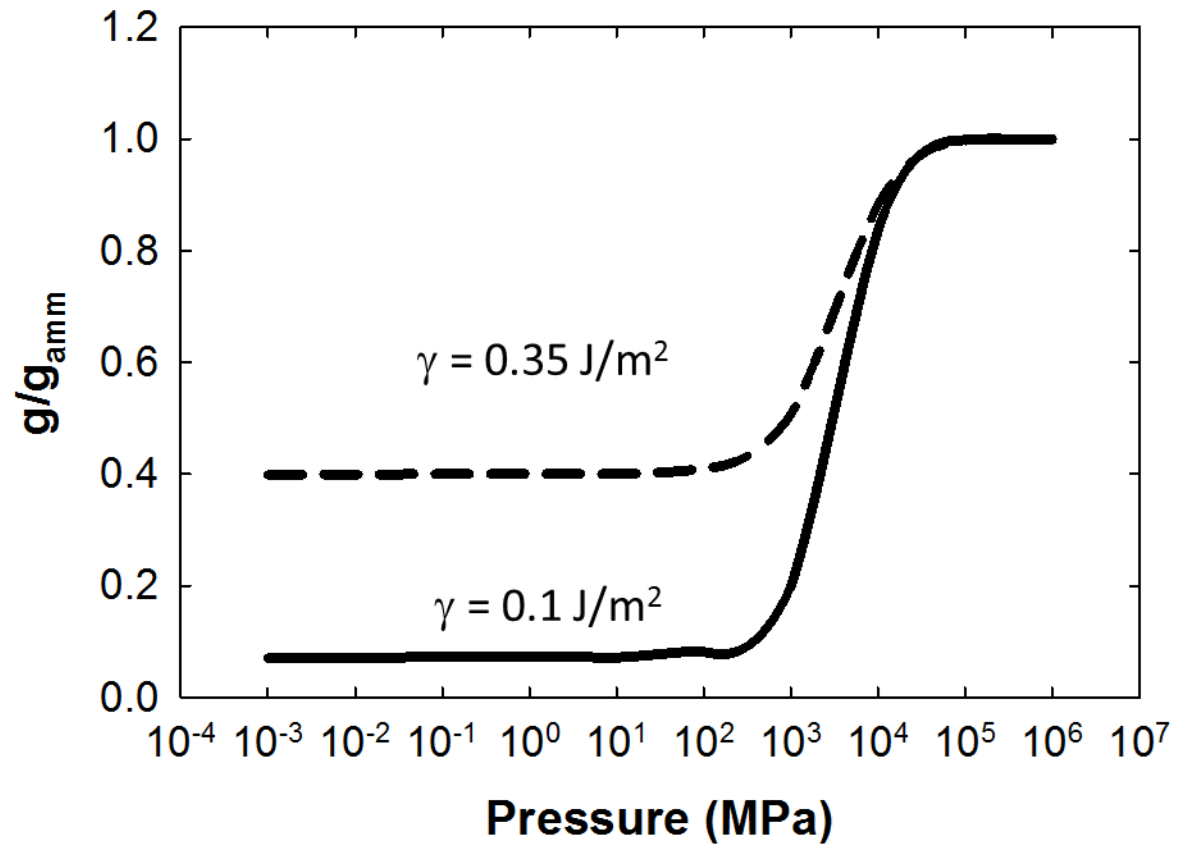


Figure 1: Ratio of thermal conductance based on pressure-dependent van der Waals model and traditional acoustic mismatch model

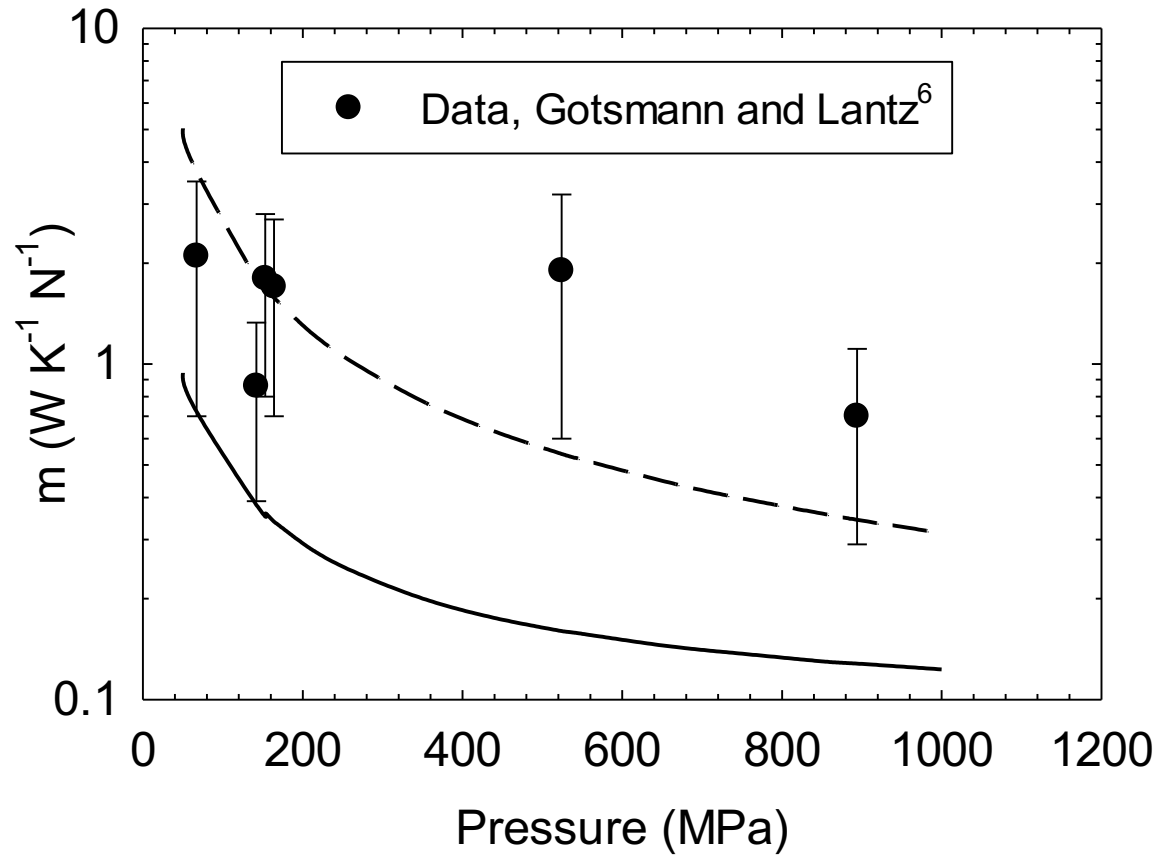


Figure 2: Comparison of proposed model with data collected by Gotsmann and Lantz⁶. m is the ratio of thermal contact conductance and pressure.