Acoustic mismatch model for thermal contact resistance of van der Waals contacts

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Nanoparticles are typically in contact with another surface through weak van der Waals force. Thermal transport in these nanostructured systems is mainly limited by the contact resistance ($R_c$). $R_c$ of nanoparticles have been typically calculated using the traditional acoustic or diffuse mismatch models, which assume very strong bond at the interface. In this paper, an analytical model of $R_c$ that accounts for the strength of the interfacial bonding is presented. Conductance/area is proportional to the square of the adhesion energy of the interface for weak bonding and is the same as that given by traditional acoustic mismatch model for strong bonding. © 2009 American Institute of Physics.

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Thermal contact resistance ($R_c$) is very important for nanostructure-based technologies and metrologies.\textsuperscript{1-4} Typically, nanostructure/nanostructure or nanostructure/bulk substrate contacts are weak contacts due to weak van der Waals (vdW) forces.\textsuperscript{2,5} For example, thermal resistance of carbon nanotubes (CNTs) array-based thermal interface material is dominated by the large $R_c$ due to weak bonding between the CNTs and the substrate. $R_c$ between these types of interfaces\textsuperscript{5,6} in solids has been modeled using acoustic mismatch model (AMM) or diffuse mismatch model.\textsuperscript{6} These models assume perfectly welded contact (very strong bond) at the interface. Assumption of welded contact leads to continuity of stress and displacement at the interface. For weak bonding of atoms at the interface such as those due to vdW contact, assumption of welded contact is clearly not accurate. In this letter, we modify traditional welded AMM (w-AMM) to incorporate the vdW bonding parameters (v-AMM). Furthermore we relate $R_c$ to the adhesion energy of the interface.

We make a few simplifying assumptions in the development of the model. Mode conversion at the interface has been ignored, which is a good approximation.\textsuperscript{7} Velocities of the three polarizations are assumed to be the same, which is equal to the Debye velocity in the Debye model. In the case of phonon dispersion, it can be calculated using Chen’s approach. The results are applicable to nanoparticles if the size of the nanoparticles and the contacts are larger than the dominant phonon wavelength otherwise phonon confinement and diffraction effects need to be taken into account.\textsuperscript{2}

Due to the assumption of welded contact, the transmission coefficient ($\tau$) of phonons in w-AMM is given by\textsuperscript{6}

$$\tau_{w-AMM} = \frac{4z_1z_2 \cos \theta_1 \cos \theta_2}{(z_1 \cos \theta_1 + z_2 \cos \theta_2)^2},$$

(1)

where $z_1 = \rho_v v_1$ and $z_2 = \rho_v v_2$ are the acoustic impedances, $\rho$ is the density, $v$ is the velocity, and $\theta$ is the angle between the normal and the phonon propagation direction. $\theta_1$ and $\theta_2$ are related by Snell’s law as $\sin \theta_1 / v_1 = \sin \theta_2 / v_2$. Equation (1) does not include any parameter related to the strength of the bond between the atoms at the interface due to the welded interface assumption.

For v-AMM, we employ the widely used Lennard-Jones (LJ) potential to describe the vdW interaction between a pair of atoms.\textsuperscript{8} LJ potential is given as

$$\phi(r) = -4\varepsilon \left[\left(\frac{\sigma}{r}\right)^6 - \left(\frac{\sigma}{r}\right)^{12}\right],$$

(2)

where $\varepsilon$ is a parameter determining the depth of the potential well and $\sigma$ is a length scale parameter. Under the harmonic approximation, the spring constant ($K$) between a pair of atoms is given by\textsuperscript{9}

$$K = \left(\frac{\partial^2 \phi}{\partial r^2}\right)_{r=r_0} = \frac{72\varepsilon}{2^{1/3} \sigma^2},$$

(3)

where $r_0 = 2^{1/6} \sigma$ is the equilibrium nearest neighbor atomic separation. For the surface atoms, one can define spring constants between planes of atoms\textsuperscript{5} using nearest neighbor interactions if the details of the atomic arrangement of the surface atoms are known. For example, for fcc lattice the spring constant using LJ potential for the longitudinal and transverse modes between an atom and a plane of atoms\textsuperscript{9} are $2K$ and $K/2$, respectively, along the [111] direction. Therefore the average spring constant is $K$ (note that all the polarization is treated the same). Similarly along the [001] direction the spring constants\textsuperscript{1} are $2K$ and $K$ for longitudinal and transverse directions, respectively, i.e., the average spring constant is $1.33K$. If the exact arrangement of atoms at the interface is known, then one can easily calculate the $K$ between planes of atoms for each polarizations, however, this information in real cases is rarely available. Since the average $K$ between one atom and plane of atoms is approximately same as the $K$ between two atoms in the direction of the bond between the atoms, it can be assumed that $K$ between an atom and a plane of atoms is the same as that given in Eq. (3). Figure 1 schematically shows that the surface of two solids are connected by springs, where the $K$ is given in Eq. (3). Therefore the spring constant per unit area is given by $K_A = nK$, where $n$ is the number of surface atoms per unit area. The geometry shown in Fig. 1 is a very common geometry in understanding the impact of defects, voids, and cracks in the field of nondestructive evaluation of...
interfaces. Schoenberg calculated the τ of acoustic waves for the geometry and conditions shown in Fig. 1 as

\[
\tau_{v-AMM} = \frac{4z_1z_2 \cos \theta_1 \cos \theta_2}{(z_1 \cos \theta_1 + z_2 \cos \theta_2)^2 + \omega^2 (z_1 \cos \theta_1 \cos \theta_2)^2}.
\]

(4)

For the welded interface, i.e., \( K_A \rightarrow \infty \), Eq. (4) reduces to Eq. (1). Similarly for \( \omega \rightarrow 0 \), Eq. (4) reduces to Eq. (1), i.e., for very long wavelengths the acoustic waves do not see imperfections in the interface.

The interface conductance per unit area can be written as

\[
g = \frac{1}{2} \int_0^{\omega_m} \int_0^{\theta_c} c_{w\nu} \sin \theta \cos \theta \, d\theta \, d\omega,
\]

where \( \omega_m \) is the maximum phonon frequency of side 1, \( c_{w\nu} \) is the frequency-dependent heat capacity per unit volume, and \( \theta_c \) is the critical angle. We can define an average integrated transmissivity as

\[
\Gamma(T) = \frac{\int_0^{\omega_m} \int_0^{\theta_c} c_{w\nu} \sin \theta \cos \theta \, d\theta \, d\omega}{\int_0^{\omega_m} c_{w\nu} d\omega},
\]

where \( T \) is the temperature. Therefore, \( g = \Gamma(T) / 2 \int_0^{\omega_m} c_{w\nu} d\omega \). We present the results as the ratio of \( \Gamma_{v-AMM} \) and \( \Gamma_{w-AMM} \).

Calculations are performed for Si/Si and Si/Pt contacts. For same materials on either side, Eq. (5) for w-AMM is only valid if the size of the contact is smaller than the mean free path of phonons. \( H \) for Si in vacuum is \( 2.58 \times 10^{-19} \) J and \( \sigma \) is 0.209 nm. To know \( n \), exact information about the surface atoms is needed, which is rarely available.

Therefore \( n \) is approximated as \( 1/a_0^2 \), where \( a_0 \) is the dimension of the primitive cell. \( a_0 \) for Si is calculated by assuming Si to have a fcc lattice structure. For simplicity we have assumed the Debye model for \( c_{w\nu} \), however phonon dispersion can be incorporated using Chen’s approach. Figure 2 also shows that if \( H \) is reduced by a factor of 10 then \( \Gamma_{v-AMM} / \Gamma_{w-AMM} \approx 1 \) at room \( T \). Figure 2 also shows the results for Si/Pt interface. Si/Pt interface has been chosen due to the following two reasons: (1) Pt has been used consistently as heaters and sensors in various metrologies to measure the thermal conductivity of nanowires and nanotubes and (2) \( \varepsilon_{Pt}/\varepsilon_{Si} \) is very large (~3.1). Therefore, by calculating \( \Gamma \) for Si/Si and Si/Pt interfaces, we have covered a wide range of acoustic mismatch. For dissimilar materials, \( \sigma \) is given by \( \sigma_1 + \sigma_2 \) and the number of surface atoms per unit area is approximated by \( (n_1 + n_2) / 2 \). \( H \) for metals in vacuum range from \( 3 \times 10^{-19} \) to \( 5 \times 10^{-19} \) J. We could not find the \( H \) for Pt. Therefore for calculations, we assumed \( H \) for \( Pt \) as \( 3 \times 10^{-19} \). J for the Si/Pt interface is given by \( \gamma H_{Si/Si}/H_{Pt/Pt} \). Figure 2 also shows that if \( H \) is reduced by a factor of 10 then \( \Gamma_{v-AMM} / \Gamma_{w-AMM} \approx 1 \) at room \( T \). Figure 2 shows that results for Si/Pt interface are similar to that of Si/Si interface.

Finally we express the results in terms of the adhesion energy/area (\( \gamma \)) of the interface as it is easily measurable and technologically relevant quantity. \( H \) can be expressed as

\[ H = 16\pi z_0^2 \gamma, \]

where \( z_0 \) is the equilibrium distance between the two surface planes under zero applied load. \( z_0 \) is given by

\[ z_0 = (2/15)^{1/6} \sigma. \]

\( \gamma \) is a strong function of surface conditions.

The highest value of \( \gamma \) is the same as the fracture energy for monolithic material such as single crystal Si which is \( \sim 2000 \) mJ m\(^{-2}\). This would correspond to very strong bonding and it is expected that v-AMM should reduce to w-AMM for this value of \( \gamma \). Depending on the surface treatment and annealing temperature, the range of \( \gamma \) of Si (Ref. 13) is 2–2000 mJ m\(^{-2}\). Figure 3 shows that \( \Gamma_{v-AMM} \) shows...
the vdW contact is expected to be approximately one order of magnitude larger than the welded contact. By thermally welding the glass/CNT interface with indium, Tong et al. saw an order of magnitude decrease in the \( R_c \), which is again consistent with the results shown in Fig. 3.

In conclusion, an analytical model for thermal contact resistance of the vdW contact was developed. The model captures the effects of appropriate interface bonding parameters. The model was expressed in terms of the adhesion energy, which is easily measurable and is a technologically relevant parameter.

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\[ R_c \approx \gamma \]

**FIG. 3.** Comparison between v-AMM and w-AMM for Si/Si and Si/Pt contacts at room temperature as a function of adhesion energy.