

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

Ravi Prasher^{a)}

Intel Corporation, CH5-157, 5000 W. Chandler Blvd., Chandler, Arizona 85226, USA

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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. [DOI: 10.1063/1.3075065]

Thermal contact resistance (R_c) is very important for nanostructure-based technologies and metrologies.¹⁻⁴ Typically, nanostructure/nanostructure or nanostructure/bulk substrate contacts are weak contacts due to weak van der Waals (vdW) forces.^{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^{3,5} in solids has been modeled using acoustic mismatch model (AMM) or diffuse mismatch model.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 adequate. 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.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's⁷ 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.²

Due to the assumption of welded contact, the transmission coefficient (τ) of phonons in w-AMM is given by⁶

$$\tau_{w-AMM} = \frac{4z_1z_2 \cos \theta_1 \cos \theta_2}{(z_1 \cos \theta_1 + z_2 \cos \theta_2)^2}, \quad (1)$$

where $z_1 = \rho_1 v_1$ and $z_2 = \rho_2 v_2$ are the acoustic impedances, ρ is the density, v is the velocity, and θ is the angle between the normal and the phonon propagation direction. θ_1 and θ_2 are related by Snell's law as $\sin \theta_1 / v_1 = \sin \theta_2 / v_2$. 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.⁸ LJ potential is given as

$$\phi(r) = -4\epsilon \left[\left(\frac{\sigma}{r} \right)^6 - \left(\frac{\sigma}{r} \right)^{12} \right], \quad (2)$$

where ϵ is a parameter determining the depth of the potential well and σ is a length scale parameter. Under the harmonic approximation, the spring constant (K) between a pair of atoms is given by⁹

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

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 atoms⁹ 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 atoms⁹ 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 constants⁹ 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

^{a)}Electronic mail: ravi.s.prasher@intel.com. Also at Arizona State University.

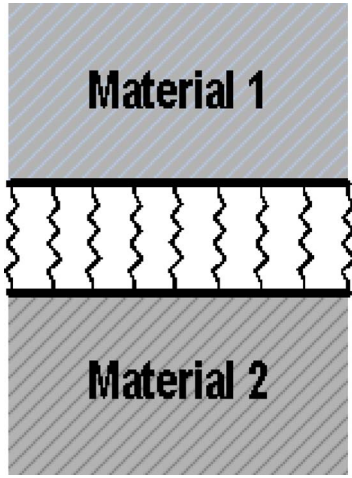


FIG. 1. (Color online) Schematic showing that two materials are connected by weak springs due to the vdW forces under the harmonic approximation.

interfaces.¹⁰ Schoenberg¹⁰ calculated the τ of acoustic waves for the geometry and conditions shown in Fig. 1 as

$$\tau_{v\text{-AMM}} = \frac{4z_1z_2 \cos \theta_1 \cos \theta_2}{(z_1 \cos \theta_1 + z_2 \cos \theta_2)^2 + \frac{\omega^2}{K_A^2}(z_1z_2 \cos \theta_1 \cos \theta_2)^2}. \quad (4)$$

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 as⁶

$$g = \frac{1}{2} \int_0^{\omega_m} \int_0^{\theta_c} c_\omega v_\omega \tau_\omega \sin \theta \cos \theta d\theta d\omega, \quad (5)$$

where ω_m is the maximum phonon frequency of side 1, c_ω is the frequency-dependent heat capacity per unit volume, and θ_c is the critical angle. We can define an average integrated transmissivity as

$$\Gamma(T) = \frac{\int_0^{\omega_m} \int_0^{\theta_c} c_\omega v_\omega \tau_\omega \sin \theta \cos \theta d\theta d\omega}{\int_0^{\omega_m} c_\omega v_\omega d\omega}, \quad (6)$$

where T is the temperature. Therefore, $g = \Gamma(T)/2 \int_0^{\omega_m} c_\omega v_\omega d\omega$. We present the results as the ratio of $\Gamma_{v\text{-AMM}}$ and $\Gamma_{w\text{-AMM}}$. ε in Eq. (2) is related to the Hamaker constant by¹¹ $H = 4\varepsilon \pi^2 N_1 N_2 \sigma^6$, where N_1 and N_2 are the number of atoms per unit volume in materials 1 and 2, respectively. H for various types of interfaces have been calculated¹¹ under vacuum or in the presence of liquids, however, H can vary dramatically from ideal values due to the presence of even one monolayer of adsorbed molecules.¹¹

Calculations are performed for Si/Si and Si/Pt contacts. For some 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.^{3,5} H for Si in vacuum¹¹ is 2.58×10^{-19} J and σ is 0.209 nm.⁸ To know n , exact information about the surface atoms is needed, which is rarely available.

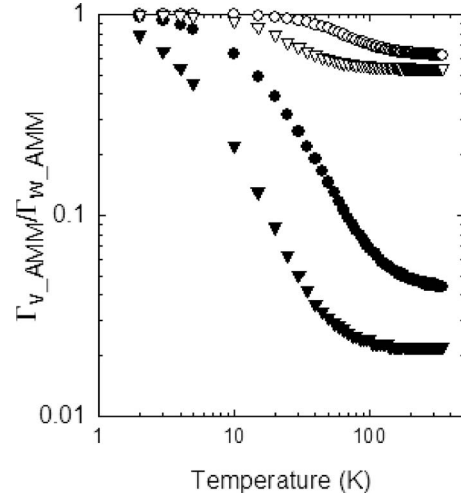


FIG. 2. Comparison between v-AMM and w-AMM for Si/Si and Si/Pt contacts. Circles represent Si/Si contacts and triangles represent Si/Pt contacts. Open symbols are for H calculated based on the literature values for ultraclean surfaces in vacuum. Filled symbols are the results if H is reduced by ten times thereby signifying a very weak interfacial bond.

Therefore n is approximated as $1/a_p^2$, where a_p is the dimension of the primitive cell. a_p for Si is calculated by assuming Si to have a fcc lattice structure. For simplicity we have assumed the Debye model for c_ω , however phonon dispersion can be incorporated using Chen's⁷ approach. Figure 2 shows that at room T $\Gamma_{v\text{-AMM}}/\Gamma_{w\text{-AMM}} \approx 0.6$ and $\Gamma_{v\text{-AMM}}/\Gamma_{w\text{-AMM}} \approx 1$ at lower T . $\Gamma_{v\text{-AMM}}/\Gamma_{w\text{-AMM}} \approx 1$ at lower T because at low T the ω of thermally excited phonons is small and in accordance with Eq. (4) v-AMM reduces to w-AMM. Figure 2 also shows that if H is reduced by a factor of 10 then $\Gamma_{v\text{-AMM}}/\Gamma_{w\text{-AMM}} \ll 1$ at room T . Figure 2 also shows the results for Si/Pt interface. Si/Pt interface has been chosen due to the two following reasons: (1) Pt has been used consistently as heaters and sensors in various metrologies to measure the thermal conductivity of nanowires and nanotubes⁴ and (2) $z_{\text{Pt}}/z_{\text{Si}}$ is very large (~ 3.1). Therefore, by calculating R_c for Si/Si and Si/Pt interfaces, we have covered a wide range of acoustic mismatch. For dissimilar materials, σ is given by⁸ $(\sigma_1 + \sigma_2)/2$ and the number of surface atoms per unit area is approximated by $(n_1 + n_2)/2$. H for metals in vacuum ranges¹¹ from 3×10^{-19} to 5×10^{-19} J. We could not find the H for Pt. Therefore for calculations, we assumed H of Pt as 3×10^{-19} . H for the Si/Pt interface is given by¹¹ $\sqrt{H_{\text{Si/Si}} H_{\text{Pt/Pt}}}$. Figure 2 also shows that if H is reduced by a factor of 10 then $\Gamma_{v\text{-AMM}}/\Gamma_{w\text{-AMM}} \ll 1$ 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 (γ) of the interface as it is easily measurable and technologically relevant quantity.¹² H can be expressed as⁸ $H = 16\pi z_0^2 \gamma$, where z_0 is the equilibrium distance between the two surface planes under zero applied load. z_0 is given by⁸ $z_0 = (2/15)^{1/6} \sigma$. γ is a strong function of surface conditions.¹³ The highest value of γ is the same as the fracture energy for monolithic material such as single crystal Si which is ~ 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 γ . Depending on the surface treatment and annealing temperature, the range of γ of Si (Ref. 13) is 2–2000 mJ m⁻². Figure 3 shows that $\Gamma_{v\text{-AMM}}$ shows

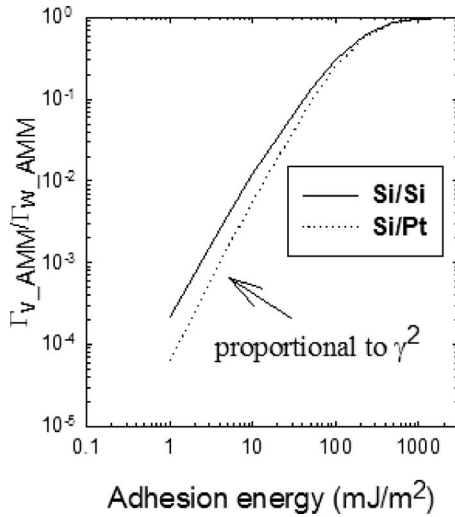


FIG. 3. Comparison between v-AMM and w-AMM for Si/Si and Si/Pt contacts at room temperature as a function of adhesion energy.

significant deviation from Γ_{w-AMM} for γ below 100 mJ/m^2 . It also shows that $\Gamma_{v-AMM}/\Gamma_{w-AMM}=1$ for larger values of γ . At very low γ , it can be shown that $\Gamma_{v-AMM}/\Gamma_{w-AMM} \propto \gamma^2$, i.e., $g_{v-AMM} \propto \gamma^2$. At very low γ , K_A in Eq. (4) is very small. Note that $K_A \propto \gamma$. Therefore from Eq. (4), it can be seen that $\tau_{v-AMM} \propto \gamma^2$. Figure 3 also shows the results for Si/Pt interface for the different γ . Although z_{Pt}/z_{Si} is large, Γ_{v-AMM} shows significant deviations from Γ_{w-AMM} for γ below 100 mJ/m^2 , which is similar to that for Si/Si interface.

Tong *et al.*² measured the R_c of the chemical-vapor deposition (CVD) grown CNT array on Si substrate and the R_c of the array in vdW contact with a glass substrate. They found that the R_c of the vdW contact was approximately one order of magnitude larger than the CVD surface. In a separate measurement Tong *et al.*¹² measured γ of the CNT/glass in the vdW contact. They found the γ was ~ 36 mJ/m^2 . For covalent bonding the γ is larger than 1000 mJ/m^2 whereas for the vdW contact it is less than 100 mJ/m^2 . Therefore for the CVD side, the interface condition is close to the welded contact. Therefore looking at Fig. 3, it can be seen that R_c of

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.*² 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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