Impact of roughness on heat conduction involving nanocontacts

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Impact of roughness on heat conduction involving nanocontacts

The impact of surface roughness on conductive heat transfer across nanoscale contacts is investigated by means of scanning thermal microscopy. Silicon surfaces with the out-of-plane rms roughness of ~0, 0.5, 4, 7, and 11 nm are scanned both under air and vacuum conditions. Three types of resistive SThM probes spanning curvature radii over orders of magnitude are used. A correlation between thermal conductance and adhesion force is highlighted. In comparison with a flat surface, the contact thermal conductance can decrease as much as 90% for a microprobe and by about 50% for probes with a curvature radius lower than 50 nm. The effects of multi-contact and ballistic heat transfer at the probe-sample contact are discussed. Limits of contact techniques for thermal conductivity characterization are also discussed.

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The samples consist of four silicon surfaces that have differing roughness parameters, prepared by anodic oxidation13 and characterized by atomic force microscopy (AFM) (Fig. 1) by means of their root mean square roughness $\sigma_{\text{Z}_{\text{RMS}}}$, transverse correlation length $l_c$, and mean peak-to-peak distance $L_{\text{RMS}}$. All these parameters allow each sample to be accurately characterized in both the perpendicular and parallel directions to the sample. One can note a correlation between the trends of $\sigma_{\text{Z}_{\text{RMS}}}$ and $l_c$. In addition, an untreated sample of smooth silicon substrate ($\sigma_{\text{Z}_{\text{RMS}}} < 1 \text{ nm}$) from the same batch is used as a reference.

To assess the impact of surface roughness, measurements based on (i) AFM vertical approach curves and (ii) images obtained by $xy$ scanning were both made in ambient air and in primary vacuum (pressure $P \sim 0.28 \text{ mbar}$), where the air contribution to the tip-sample transfer is eliminated. Results are provided as thermal conductances (see the supplementary material for details on protocols). Figure 2 reports on the decrease in thermal conductance ($\Delta G_{\text{global}}$) due to the global thermal transfer ($\Delta G_{\text{global}}^\text{global}$) and (b) due to the tip jump to contact ($\Delta G_{\text{jump}}^\text{global}$), for the three probes and both types of experimental conditions. Mechanical contacts form after the jump, possibly with the water meniscus. We note strong differences between the behaviors of the different probes. When sample roughness increases, $\Delta G_{\text{global}}^\text{global}$ decreases by 30% for the Wollaston probe [Fig. 2(a-A)] and by about 10% for the Pd probe [Fig. 2(a-B)]. For the DS probe, $\Delta G_{\text{global}}^\text{global}$ remains constant [Fig. 2(a-C)]. The observed conductance decreases are the signature of the decrease in heat conduction through the mechanical contact, as the air heat transfer taking place over a $\sim$ micrometric zone is not expected to vary much when roughening the surface. For the largest probe [Wollaston, Fig. 2(a)], it is found that the heat conduction by mechanical contact (solid–solid and water meniscus) on a flat surface represents 20% of the overall transfer. This thermal transfer can almost be suppressed by roughening the surface (decrease by 95%) [Fig. 2(b-A)]. The overall decrease can be larger than 20%, so heat transfer through air is also slightly reduced, probably due to an effective tip-sample distance larger in the rough case. For the roughest sample, the mechanical contact accounts for only 2% of the overall heat transfer. In contrast, thermal transfer across mechanical contact accounts for less than 1% of the overall transfer on a flat surface for the smallest tip apex [DS probe, Fig. 3(c)]. Although this transfer decreases with increased roughness, it has no visible effect on the measured overall transfer. Finally, the Pd probe, which has intermediate dimensions, exhibits an intermediate behavior [Figs. 3(a-B)–(b-B)].

On a flat surface, about 11% of the global transfer is made through mechanical contact. With the increase in roughness, this transfer decreases by up to 30%, resulting in a 5%–10% decrease on the overall signal measured. These results on thermal transfer across the first contact during an approach curve of the SThM tip on the sample show that surface roughness results in a decrease in the heat transfer across the contact, presuming a decrease in the probe-sample contact area. Analysis of thermal images of samples leads to the same conclusion (see the supplementary material for images).

Measurements of the adhesion forces14 performed with the three probes on the rough samples are consistent with this observation. Figure 3 shows the thermal conductance $\Delta G_{\text{jump}}^\text{global}$ as a function of the average value of the adhesion force ($F_{\text{ad}}$) measured for each SThM probe. When sample roughness increases, the adhesion force decreases by up to 97%, about 30% and 50%, respectively, for the Wollaston, Pd and DS probes. This underlines the correlation between the quality of the probe-sample contact and the heat transfer across it. Roughness significantly deteriorates the quality of the probe-sample contact and, thus, the thermal transfer associated with mechanical contact. The effect seems more pronounced for the Wollaston microphone than for the nanopores. This can be explained by the roughness of the metallic microfilament.15 The Wollaston probe-planar sample contact is made by a multitude of small contacts, and the number of which is decreased
when the surface becomes rough, resulting in a significant decrease in the total contact area. Measurements with nanoprobes are less impacted for the studied roughness range.

An upper bound for the curvature radius $R$ at the contact can be obtained by neglecting the influence of the water meniscus on adhesion (note that we do not expect the water meniscus to be predominant for heat transfer\cite{9-15}). $R$ is determined from the adhesion force, measured when retracting the probe from the sample, by using the mechanical model of Rabinovich et al.\cite{16,17}. This model considers a rough surface with periodic peak-to-peak distance $\delta L_{\text{RMS}}$ and mean

![Graphs showing thermal conductance measurements](image)

**FIG. 2.** Measured global (a) and mechanical contact-related (b) thermal conductances according to the Si roughness, for the Wollaston (A), Pd (B), and doped Si (C) probes. (a) refer only to air measurements. Inset schematics represent the percentage of the mechanical contact heat transfer in the global heat transfer between probe and sample. For vacuum measurements $\Delta G_{\text{global}} = \Delta G_{\text{mech}}$. Error bars represent dispersion of the measurements.

![Graphs showing adhesion force vs. contact radius](image)

**FIG. 3.** Thermal conductance due to mechanical contact as a function of the adhesion force ($F_{\text{ad}}$) and corresponding contact radii for experiments performed in air conditions on the rough samples with (a) the Wollaston probe, (b) the Pd probe and (c) the DS probe.
out-of-plane deviation $\delta Z_{\text{RMS}}$ associated with hemispherical asperities of
curve radius $r = f(\delta Z_{\text{RMS}}, L_{\text{RMS}})$,

$$F_{\text{id}} = \frac{A_H R}{6 R^2} \left( \frac{1}{1 + 58.14 \frac{R \delta Z_{\text{RMS}}}{L_{\text{RMS}}}} \right) \left( 1 + 1.817 \frac{\delta Z_{\text{RMS}}}{H_0} \right)^2, \quad (1)$$

where $A_H \approx 2.65 \times 10^{-19}$ J is the Hamaker constant and $H_0 = 0.3$ nm is the minimum separation distance between the tip apex and the asperity. Using this expression, one finds $R = 9 \pm 2$ nm for the DS probe in accordance with previous estimate, $R = 6.4 \pm 0.5$ nm for the Pd probe, which is ten to five times lower than the values announced by the provider. This difference could be understood as a contact considered to be made through an asperity at the apex of the tip. For the Wollaston probe that is the largest and roughest probe, $R$ values are very dispersed and the mean is around 450 nm as found in Ref. 15. Equation (1), which assumes that the surface is rougher than the probe ($R > L_{\text{RMS}}, \delta Z_{\text{RMS}}$), could, therefore, be applicable for the Wollaston probe but is only approximate for the two other probes. Adding the Derjaguin–Müller–Toporov (DMT) model for the sphere-plane configuration, we can determine, for each surface, a lower bound for the contact radius $b_0$ when zero force is applied,

$$b_0 = \sqrt[3]{\frac{1}{E'} \frac{R r}{R + r} F_{\text{id}}}, \quad (2)$$

where $r$ is the curvature radius of rough surface asperities and $E'$ is the generalized Young modulus. Figure 3 provides this quantity for the various probes. For the Wollaston probe, $\Delta G_{\text{mech}}$ is found proportional to $b_0^3$, for the Pd nanoprobe to $b_0^2$ and for the DS probe to $b_0$. It is known that when $\Delta G_{\text{mech}} \propto b_0$ heat transfer is diffusive and that $\Delta G_{\text{mech}} \propto b_0^2$ for a ballistic or thermal-boundary limited transfer. The oxide layer that is present on the surface of samples and probably on the resistive elements of the probes can be involved in this interfacial thermal resistance. For the three probes, the exponent is larger than 1, which suggests that thermal transfer is not only diffusive. Let us note, however, that $b_0$ values are well below the phonon average mean free path of Si (around 250 nm) so that an exponent closer to 2 would be expected. The role of water meniscus on adhesion, which is here neglected, could explain the difference with such value. A generalization of Eq. (1) to arbitrary values of $R$, which would include contact of the probes’ sides with the samples, would be useful to clarify these observations.

It is interesting to analyze the heat transfer reduction in light of the usual SThM measurement process, which involves first a calibration with samples of well-known thermal conductivity surfaces as flat as possible. Figure 4 provides such a calibration curve, which underlines the lack of sensitivity for large thermal conductivity. Most importantly, it highlights that using the average value of $\Delta G_{\text{mech}}$ for a rough sample of unknown thermal conductivity would lead to determining a thermal conductivity much lower than the correct value (see red arrow), as if an insulating body was present below the surface. Using the upper values of the conductance range may be better (possibly also for the calibration curve) but induces also uncertainty. As a consequence, a detailed analysis of roughness is essential prior to SThM thermal-conductivity determination from the contact.

In conclusion, we have measured thermal conductances across micro- to nanocontacts by means of SThM probes on Si samples. For roughness $\delta Z_{\text{RMS}}$ close to 10 nm, the decrease in contact thermal conductance can reach as much as 90% at a microcontact and about 50% at nanocontacts. In all cases, surface roughness strongly alters the mechanical contact, resulting in most cases in multi-contacts reducing the apparent contact radius. It is found that heat transfer is not only diffusive, but that ballistic or boundary-limited heat conduction can also be involved. Finally, we demonstrate that sample roughness can completely distort the analysis of SThM measurements when estimating thermal conductivity of materials. It will be needed to study the effect of roughness on materials covering the whole thermal conductivity range in order to be able to analyze correctly the thermal data. Another pending issue is that current mechanical models consider only the mechanical properties of solid materials, so the water meniscus and its contact radius deserve to be further investigated.

See the supplementary material for further details on SThM probe and sample characterizations, SThM images, and protocols.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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