SUPPLEMENTARY MATERIAL TO

Thermal boundary conductance of CVD-grown MoS₂ monolayer-on-silica substrate determined by scanning thermal microscopy

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Contents

- 1. Procedure to determine if the sample is a MoS₂ monolayer
- 2. Procedure to determine the probe thermal conductance
- 3. Brief summary of the different steps for the data treatment
- 4. Thermal circuit associated to heat dissipation from the probe
- 5. 'Equivalent oxide thickness' procedure
- 6. Equivalent effective thermal conductivity
- 7. Determination of the thermal contact radius
- 8. Determination of the thermal boundary conductance

1. Procedure to determine if the sample is a MoS₂ monolayer

According to the methodology followed by Ganorkar *et al.*¹ for estimating the number of MoS₂ layers synthesized by Chemical Vapor Deposition (CVD), the difference between the Raman shifts of the E_{2g} and A_{1g} peaks is used to estimate the number of layers. The wavenumber difference is $\Delta \sigma = 21.5$ cm⁻¹ for a monolayer and $\Delta \sigma = 22.3$ cm⁻¹ for bilayers [1]. In this work we find a value of $\Delta \sigma = 21.6$ cm⁻¹, which can safely be considered as a MoS₂ monolayer crystal (see Suppl. Fig. 1).



Suppl. Fig. 1. MoS₂ Raman spectrum.

2. Procedure to determine the probe thermal conductance

The probe thermal conductance is defined as

$$G_{probe} = P/\theta, \tag{1}$$

where $P = R I^2$ is the Joule self-heating power inside the sensitive part (sensor) of the probe, R the electrical resistance of the sensitive part of the probe (sensor), I is the electrical current in the probe and θ its mean temperature rise with respect to ambient. The Wollaston probe resistance is inserted into a Wheatstone bridge (see Suppl. Fig. 2), and it is the bridge imbalance voltage ΔV that is provided in the thermal image. As a result, scans provide only temperature rises

$$\Delta \theta = \frac{\Delta V}{I} \cdot \frac{1}{\alpha R}$$
(2)

relative to an absolute reference temperature $\theta_{ref} + T_0$, which is selected by balancing the bridge ($\Delta V = 0$), and not directly the probe voltage V_p . It is customary to balance the bridge either far from contact ($\theta_{ref} = \overline{T} - T_0$, where \overline{T} is the average temperature in the sensor), or in contact at a given location on the sample ($\theta_{ref} = \overline{T} - T_{ref}$). Here, the second option is chosen.

Knowing the value of the electrical resistance in the bridge R_v , one can deduce the probe temperature $T_0 + \theta = T_0 + \theta_{ref} + \Delta\theta$. We use a symmetric bridge, so that $R_1 = R_2$ (taken as 50 Ω), and an input bridge current of $I_{in} = 80$ mA, i.e. I = 40 mA is supplied in the probe. The electrical resistance of the sensitive part of the Wollaston probe (Pt₉₀/Rh₁₀ filament) R_1 is computed by determining the geometrical parameters, noticing that the variable resistance, when the bridge is balanced, is equal to:

$$R_{\nu} = R_{wiring} + R_{Ag} + R \quad , \tag{3}$$

where the electrical resistance of the wiring is estimated to be $R_{wire} \approx 1 \Omega$, $R_{end} = R_0(1 + \alpha(\theta))$ and R_{Ag} is obtained by subtraction from room-temperature measurements ($\theta = 0$). R_{Ag} is the electrical resistance of the Wollaston wire (cantilever) assumed made of the silver shell.



The local probe thermal conductance variation is obtained as in Ref. [2] by differentiating logarithmically Eq. (1), which gives after straightforward algebra:

$$\Delta G_{probe} = G_{probe \ ref} - G_{probe \ ref} = G_{probe \ ref} \cdot \Delta \theta \cdot \left[\alpha(\theta) - \frac{1}{\theta} \right]$$
⁽⁴⁾

if the current variation in the probe ΔI is neglected (which we verified). $G_{probe\,ref}$ is the probe thermal conductance at absolute temperature $T_0 + \theta_{ref}$. Of course Eq. (4) is valid only provided the thermal conductance variations stay small. A direct calculation without linearization can be performed if this is not the case.

From the parameters experimentally determined, $G_{probe\ 0} \approx 95 \,\mu\text{W}$. K⁻¹ and $\theta_{ffc} \approx 156$ K far from contact. When the probe contacts the sample, the temperature rise θ decreases by about 10% for materials of moderate thermal conductivities, so $\theta_{ref} \approx 140$ K. In principle $G_{probe\ ref}$ and $G_{probe\ 0}$ are different, but in the following Eq. (4) is used with the assumption $G_{probe\ ref} \approx G_{probe\ 0}$, which induces an uncertainty propagation in ΔG_{probe} . Note that maps of ΔG_{probe} or G_{probe} with respect to an arbitrary reference provide similar information.



3. Brief summary of the different steps for the data treatment

The raw SThM image allows only acquiring qualitative analysis of heat dissipation at the sample surface and a significant part of the work is therefore to deduce quantitative data from these images. We summarize the different steps (see Suppl. Fig. 3) mentioned in the main manuscript here, and more details are provided in the Sections below.

- First (1), the local probe thermal conductance G_{probe} is obtained from the electrical data (see above).
- Then (2), the calibration from Guen *et al.* [3] allows obtaining an oxide thickness that impacts the probe thermal conductance equivalently as MoS₂. This step is interesting for qualitative reasoning but not decisive for the following.
- More importantly (3), the same paper [3] allows determining the two effective bulk thermal conductivities k_{eff} that provide the same probe thermal conductance as that of the MoS₂/SiO₂/Si and the SiO₂/Si samples, respectively. Noticeably, all the previous steps do not require simulations. But they do not allow to determine the thermal contact radius *b*.
- The simulation steps (4) involve Finite Element (FE) modelling. We proceed in two steps: (4a) we first determine the thermal contact radius b, and then (4b) we use it to determine the MoS₂ thermal properties. The thermal radius is obtained by equating the thermal conductances of the effective bulk geometry (known to be $G_{sample} = 4k_{eff}b$) and of the exact SiO₂/Si geometry. Then for such radius a FE simulation of the MoS₂/SiO₂/Si stack is performed. The thermal conductivity of the monolayer k and the thermal boundary conductance between the monolayer and the supporting material G_{TBC} are varied in order to match the stack effective thermal conductivity determined in (3).



Suppl. Fig. 3. Schematic of the different steps of our approach for quantitative measurement.

4. Thermal circuit associated to heat dissipation from the probe

We provide a schematic clarifying the different thermal conductances involved in our SThM experiments. G_{probe} is in principle indeed the sum of the three channels allowing heat to dissipate from the probe, where only one is useful for the experiment. However $G_{environement} \approx 0$ when the probe is in contact³. G_{tip} includes the thermal contact conductance associated with the transport of heat from the tip into the sample, and is usually difficult to determine precisely. The method described below (Suppl. Sec. 5) avoids addressing this issue fully.



Suppl. Fig. 4. Suppl. Fig. 3. Thermal circuit associated with heat dissipation in the probe.

5. 'Equivalent oxide thickness' procedure (Step (2) of Suppl. Fig. 3)

To find the equivalent thickness of SiO_2 that induces a similar thermal resistance in the sample as that of the monolayer of MoS_2 , we use a calibration sample³ made of a mosaic of silicon oxide layers with different thicknesses coating a silicon wafer. It happens that the substrate below MoS_2 is similar, with a SiO_2 layer on top of the silicon wafer. Since the two samples were not prepared at the same time and with the same goal, some uncertainty is introduced by comparing the data, which propagates until the determination of the thermal boundary conductance. The calibration sample is shown in Suppl. Fig. 5 and detailed in the previous publication³ (beware that notations are not the same and that the probe thermal conductance defined here is based on the probe average temperature, not the probe apex one).



Suppl. Fig. 5. (Left) SThM scan of the mosaic sample made of 9 different thicknesses of silica. The heights of the silicon oxide steps are indicated on the images. (Right) Probe thermal conductance for the different SiO₂ thicknesses. The probe thermal conductance reference ($G_{probe} = 0$) is the far-from-contact position.

More precisely, G_{probe} varies of $\Delta G_{probe} = 55 \times 10^{-9} \text{ W.K}^{-1}$ when the probe moves from the MoS₂ crystal to the oxide surface (see Fig. 3 in the main paper). Note that $\Delta G_{probe} = G_{probe}(\text{MoS}_2) - G_{probe}(\text{SiO}_2 300 \text{ nm})$. From Suppl. Fig. 5, we find that $G_{probe}(\text{SiO}_2 300 \text{ nm}) + \Delta G_{probe} = G_{probe}(\text{SiO}_2 395 \text{ nm})$ in the calibration sample.

6. Equivalent effective thermal conductivity (Step (3) of Suppl. Fig. 3)

The effective thermal conductance for the {oxide on silicon} sample is obtained from the calibration curve in Ref. [3], as shown in Suppl. Fig. 6. The advantage of this method is that it provides a quantity that depends only on the sample and does not require the knowledge of the thermal conductance corresponding to the heat transfer between the probe and the sample included in G_{tip} . For the value of G_{probe} found in Suppl. Fig. 5, we find $k_{eff} \approx 2.1 \text{ W.m}^{-1}$.K⁻¹ in Suppl. Fig. 5.



Suppl. Fig. 6. Variation of the probe thermal conductance (reference far from contact) as a function of the effective thermal conductivity.

7. Determination of the thermal contact radius

The thermal contact radius, i.e. the size of the hot zone on the sample surface (assumed to be a disc), is required for the final step. One can consider that there is a single thermal contact radius for each effective thermal conductivity, i.e. the radius does not depend on the exact configuration within the sample but only on the thermal conductance G_{sample}^4 . This conductance is not known initially, and we are required to perform simulations in order determine the thermal contact radius for the {silica on silicon} sample.

With finite-element (FE) simulations, we compute the thermal conductance of a medium consisting of a silica layer (300 nm) over a silicon wafer, for a given radius (see Suppl. Fig. 7). This can be done for an arbitrary temperature on the top T_{top} of the simulated sample provided the thermal conductivities are considered temperature-independent. The lateral and bottom sides of the domain are considered at a fixed ambient temperature $T_{ambient}$. The sample thermal conductance within such geometry $G_{sample} = Q/(T_{top} - T_{ambient})$ is computed and compared to the conductance associated with the effective thermal conductivity, known analytically to be $G_{sample} = 4k_{eff}b$ (defined as that of an equivalent-bulk thermal conductance for the same radius), determined from Suppl. Fig. 6. When the two thermal conductances are equal, this process provides the thermal contact radius *b*. We find $b \approx 4 \mu m$, which confirms that air heat transfer predominates.

This work can also be performed for a 395-nm silicon oxide layer, which provides the equivalent sample thermal conductance as that of the $MoS_2/SiO_2/Si$ system. Note that we do not consider here possible partially-ballistic dissipation in contrast to Ref. [4].



Suppl. Fig. 7. Temperature distribution on the equivalent (395 nm SiO₂ film on Si substrate) sample surface.

8. Determination of the thermal boundary conductance

In the final step, we perform simulations with the thermal radius previously determined by varying the thermal conductivity of $MoS_2 k$ and the thermal boundary conductances G_{TBC} at its boundary with SiO₂. While the values of thermal conductivity k do not impact much on the total sample thermal conductance (which is known to be $4k_{eff}b$, where b is the thermal radius), one value of the boundary conductance provides the correct sample thermal conductance. The cross section temperature field is shown in Suppl. Fig. 8, and as a function of depth z on the revolution axis in the core paper.



Suppl. Fig. 8. Temperature field in a cross section of the MoS2/SiO2/Si sample.

References

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