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

Cite as: Appl. Phys. Lett. **120**, 262202 (2022); https://doi.org/10.1063/5.0092553 Submitted: 23 March 2022 • Accepted: 07 June 2022 • Published Online: 28 June 2022

🔟 Christian Mateo Frausto-Avila, 🔟 Victor M. Arellano-Arreola, 🔟 Jose Martin Yañez Limon, et al.





• **• • •** • •

1 qubit

Shorten Setup Time Auto-Calibration More Qubits





visit our website >

Appl. Phys. Lett. **120**, 262202 (2022); https://doi.org/10.1063/5.0092553 © 2022 Author(s). **120**, 262202

Export Citatio

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

Cite as: Appl. Phys. Lett. **120**, 262202 (2022); doi: 10.1063/5.0092553 Submitted: 23 March 2022 · Accepted: 7 June 2022 · Published Online: 28 June 2022

Christian Mateo Frausto-Avila,^{1,2} (b) Victor M. Arellano-Arreola,² (b) Jose Martin Yañez Limon,² (b) Andres De Luna-Bugallo,^{2,3,a)} (b) Séverine Gomès,¹ (b) and Pierre-Olivier Chapuis^{1,a)} (b)

AFFILIATIONS

¹Univ Lyon, CNRS, INSA-Lyon, Université Claude Bernard Lyon 1, CETHIL UMR5008, F-69621 Villeurbanne, France ²Cinvestav Unidad Querétaro, Querétaro, Qro. 76230, Mexico

³Departamento de Nanotecnología, Centro de Física Aplicada y Tecnología Avanzada, Universidad Nacional Autónoma de México, Querétaro, Qro. CP 76000, Mexico

^{a)}Authors to whom correspondence should be addressed: aluna@fata.unam.mx and olivier.chapuis@insa-lyon.fr

ABSTRACT

We characterize heat dissipation of supported molybdenum disulfide (MoS₂) monolayers grown by chemical vapor deposition by means of ambient-condition scanning thermal microscopy (SThM). We find that the thermal boundary conductance of the MoS₂ monolayers in contact with 300 nm of SiO₂ is around 4.6 ± 2 MW m⁻²K⁻¹. This value is in the low range of the values determined for exfoliated flakes with other techniques such as Raman thermometry, which span an order of magnitude (0.44–50 MW m⁻²K⁻¹), and underlines the dispersion of measurements. The sensitivity to the in-plane thermal conductivity of supported MoS₂ is very low, highlighting that the thermal boundary conductance is the key driver of heat dissipation for the MoS₂ monolayer when it is not suspended. In addition, this work also demonstrates that SThM calibration using different thicknesses of SiO₂, initially aimed at being used with bulk materials can be extended to 2D materials.

Published under an exclusive license by AIP Publishing. https://doi.org/10.1063/5.0092553

Over the last two decades, there has been a growing interest in 2D materials due to their low dimensionality, making them attractive for various fields such as electronics, condensed matter, photonics, catalysis, among others. After the popularization of graphene, different layered materials have been discovered, including borophene,¹ hexagonal boron nitride (hBN),² and transition metal dichalcogenides (TMDCs).³ Molybdenum disulfide (MoS₂), a member of the family of TMDCs, is a semiconductor whose synthesis has been quite well developed and established by different approaches.^{4–9} In the case of a single layer (thickness around 7 Å), MoS₂ exhibits a direct bandgap¹⁰ (\approx 1.82 eV), reasonable electrical conductivity, large spin–orbit coupling, and strong exciton binding, which makes it suitable for several optoelectronic applications.^{11,12}

Investigating the properties of 2D materials and implementing various characterization techniques are challenging in many cases, particularly for thermal studies, due to the complexity associated with their extremely low thickness. With the advent of atomically thin materials, different thermal characterization techniques have been extrapolated from bulk to nanostructured materials. Techniques such as the 3ω method,¹³ photothermal characterization,¹⁴ and Raman thermometry¹⁵ have efficiently been translated for thermal conductivity measurements of such materials. Some of the techniques require depositing metallic contacts onto the samples,¹⁶ which is unfeasible for certain configurations of the systems, or high-frequency equipment¹⁷ with assumptions on the (ideal) optical absorption.

Thermal characterization aims mainly at obtaining parameters such as thermal conductivity and thermal boundary conductances (TBCs). Due to the quick preparation and crystal quality, most of the reports regarding the thermal properties of MoS₂ are normally performed using exfoliated samples (either supported by an arbitrary substrate or suspended in a micrometer-sized hole); however, MoS₂ crystals can also be grown by chemical vapor deposition (CVD), which appears more appropriate for device integration and scaling.¹⁸ For supported exfoliated monolayers (flakes), thermal conductivity values in the range 34.5–62.0 W m⁻¹ K⁻¹ are reported, while TBCs span 0.44–50 MW m⁻² K⁻¹.^{19–22} For the suspended configuration, thermal conductivity values between 23.2 and 84.0 W m⁻¹ K⁻¹ are reported.^{21,23,24} These values are more accurate when averaged over

large areas and were obtained by techniques with inherent limitations such as optical diffraction²⁵ in the best cases. Beyond such scales, scanning thermal microscopy (SThM), developed since the 1990s²⁵ on the atomic force microscopy (AFM) platform, is attractive since the spatial resolution can depend only on the radius of the thermal contact between the probe and the sample. Such radius can reach the sub-100 nm scale under certain operation conditions, making it an option for nanoscale thermal measurements, in particular thermometry.²⁴ Although SThM was already used on structures involving MoS₂ for thermometry in complex devices²⁶ and for an analysis of heat dissipation in samples where MoS₂ was coupled to graphene,²⁷ it has not been used for quantitative thermal-property determination of the TMDC yet. In the present work, we propose a methodology based on ambient-condition SThM to determine the TBC value for MoS2 monolayers grown by CVD on SiO2/Si substrates. It is demonstrated that (in-plane) thermal conductivity is not useful in practice for samples with several micrometers of lateral lengths, since heat dissipation takes place toward the substrate.

The MoS₂ crystals are grown by atmospheric CVD, and further details of this can be found in previous reports.²⁸ The studied systems are composed of a MoS₂ monolayer supported by a 300 nm-thick silica layer standing over a silicon wafer. Figure 1 shows an optical image of a typical MoS₂ monolayer, and the overall stack is reminded in the inset. The typical lateral size of the crystals is around 70–100 µm. As a large number of MoS₂ monolayer crystals (typical shapes as that of Fig. 1) can be present on the substrate, careful attention is paid to avoid thermal or optical crosstalk. Moreover, we use Raman spectroscopy to monitor the frequency difference between the E_{2g} and A_{1g} peaks⁷ to select only the single-layered MoS₂ crystals (see supplementary material Fig. 1).

Thermal scans are acquired by means of thermoresistive SThM, with two different thermal probes.²⁵ The data reported here are obtained using a Wollaston probe, whose sensor is a 5 μ m-in-diameter Pt₉₀/Rh₁₀ filament with a length of $\approx 200 \,\mu$ m. It is bent in a V shape with the tip contacting the sample, and anchored between unetched parts of the Wollaston wire, where the Pt₉₀/Rh₁₀ alloy is surrounded by a silver shell (\approx 75 μ m of diameter in total). This makes the sides of the filament less electrically resistive. As a consequence, the filament is self-heated when fed by an electrical current *I*. The SThM operation mode typically used in this work consists in bringing the heated probe into contact with the sample in order to heat it locally and scanning its



FIG. 1. (a) Optical microscopy image of a MoS_2 monolayer with a triangular shape. The light dot corresponds to a laser spot irradiating the surface. (b) Cross-section schematic of the analyzed system.

surface at constant force as in AFM. Noticeably, the electrical resistance of the sensor *R* depends on the average temperature of the probe \overline{T} , so in addition to being a heat source, the probe is a thermometer

$$R = R_0 \cdot (1 + \alpha \theta), \tag{1}$$

where $\alpha = (1/R) \cdot dR/d\overline{T}$ is the temperature coefficient of the Pt₉₀/Rh₁₀ electrical resistance known to be $1.66 \times 10^{-3} \text{ K}^{-1}$, $R_0 = R(\overline{T}_0)$ and $\theta = \overline{T} - T_0$ is the temperature rise above ambient temperature T_0 (see supplementary material Sec. 2 for more details on SThM). The ratio between the heat input to the sensor $P = RI^2$ and the sensor average temperature rise θ provides a qualitative estimation of the sample ability to dissipate heat, which is known as the probe thermal conductance G_{probe} (see supplementary material Secs. 2–4) and its value is close to 95 μ W K⁻¹ in ambient condition.

In our setup, the current supplied to the probe is constant, and the voltage variation ΔV is monitored at the same time as the topography during the scan²⁵ of the sample surface. The voltage reference ($\Delta V = 0$) is taken at an arbitrary point on the surface. Note that thermal stabilization is reached by waiting around 45 min before scanning to minimize the impact of thermal drifts on the images. Figure 2 displays (a) the recorded AFM topography and (b) the raw thermal image (ΔV) of a MoS₂ monolayer. It is possible to directly correlate the crystal topography (here slightly different from the crystal of Fig. 1) with the thermal contrast. One can notice the strong difference between the thermal signal on the MoS₂ monolayer and that in the region around



FIG. 2. (a) Topography image obtained by atomic force microscopy with a Wollaston probe. A flat plane was subtracted from the raw image. (b) Raw thermal signal (ΔV) obtained during the same scan.

(SiO₂/Si substrate). Artifacts linked to scan direction are observed in the topography image and are also present in the raw thermal image.

The raw thermal image can be translated into a probe average temperature image with Eq. (1). One obtains the probe temperature variation $\Delta\theta$ as a function of location on the sample (with respect to some reference, here arbitrarily taken as the lowest value of the image). In order to smoothen the thermal signal fluctuations, we average the signal close to an edge as shown in Fig. 3 (the image is rotated with respect to that of Fig. 2). It is found that the probe temperature increases by approximately 0.1 K when it moves from SiO₂ to MoS₂, indicating that the MoS₂ layer induces an additional thermal resistance for the flux being dissipated into the sample. At first sight, this effect could be ascribed either to a worse contact between the SThM probe with MoS₂ than silica or to a weak thermal contact between MoS₂ and the silica. This is in striking contrast to supported graphene, which increases heat dissipation properties.^{29,30}

The temperature map is then translated into a map of the probe thermal conductance G_{probe} (again with respect to an arbitrary reference, see supplementary material Sec. 2). It is found that G_{probe} varies by $\Delta G_{\text{probe}} = 55 \times 10^{-9}$ W K⁻¹ close to the edge of the MoS₂ crystal. It is instructive to compare this value with that obtained when simply increasing the thickness of the silica layer (silica is a standard solidstate thermal insulator). In Ref. 31, some of us reported, with a similar Wollaston SThM probe, how G_{probe} varies with SiO₂ thickness (see supplementary material Sec. 5). Assuming similar thermal conductivity for the oxide in the SiO₂/Si substrate here and that of Ref. 31, we find that the decrease in probe thermal conductance when locating the probe on MoS₂ is the same as that while bringing it over an oxide layer thicker by 95 nm. This thickness is more than hundred times than that of MoS_2 , underlining the potential of the TMDC as thin but efficient heat barrier.

In the following, we aim at obtaining quantitative thermal data for the MoS₂ monolayer (see supplementary material Sec. 3 for a graphical summary of the procedure). To determine these, one needs first to find an estimate of the thermal contact radius b, i.e., the size over which the SThM probe heats the sample. It is obtained by first comparing the probe thermal conductance with that obtained as a function of the silica thickness in Ref. 31. The effective thermal conductivity (that of a bulk leading to the same G_{probe}) determined for a layer of 300 nm of SiO_2 over Si is around $\lambda_{e\!f\!f}\approx 2\,W\,m^{-2}\,K^{-1}$ (see supplementary material Sec. 6). The radius can then be obtained from a finite element (FE) simulation solving the steady-state heat equation, in the sample only. Indeed, the sample thermal conductance (conductance associated with heat dissipation in the sample from a hot isothermal disk on the sample surface) is $4\lambda_{eff}b$ and equal to that of the exact geometry (300 nm SiO₂/Si) for an identical thermal contact radius. The radius determined from the FE simulation is around $4 \,\mu m$ (see supplementary material Sec. 7 for more details). This value underlines the well-known fact that heat spreads from the probe to the sample in the air, leading to a transfer over a much larger area than that of the mechanical contact.²⁵ Since heat is transferred mostly through air to the sample, the thermal boundary conductance at the mechanical contact is not a matter of concern. Note that the impact of the thermal contact conductance between the tip and the sample depends only on the effective (bulk) thermal conductivity felt by the probe for heat transfer through air.³² This is in stark contrast to many works where



FIG. 3. (Top) Probe temperature rise with respect to an arbitrary reference between the supported MoS₂ and the silica-over-silicon wafer. (Bottom) Probe thermal conductance deduced from the temperature measurements (arbitrary reference). Left panels show rotated images with respect to Fig. 2 and right ones vertical averages in the square for each (horizontal) position.

scitation.org/journal/apl

heat transfer inside the whole system made of the SThM sensor and at the probe-sample contact is also required to be modeled. This simplification is possible because the current work builds on the previous calibration in Ref. 31.

The final step is also performed with a FE simulation. The actual geometry, i.e., the stack shown in the inset of Fig. 1, is considered, with known thermal conductivity values for silicon and silica, and again with a disk of homogeneous temperature as heat input on the top (see supplementary material Sec. 8). The unknowns are the MoS₂ thermal conductivity, supposed isotropic in the 0.7 nm thickness, and the TBC between MoS_2 and silica. These two quantities are adjusted in the 2D cylindrical FE simulation to dissipate a power equivalent to that of a bulk with the effective thermal conductivity mentioned above (i.e., the bulk and MoS₂/SiO₂/Si sample thermal conductances are equal). It is found that the value of the thermal conductivity of MoS₂ impacts very weakly the temperature distribution, which is driven only by the TBC. The temperature profile in the center of the structure is provided in Fig. 4 as a function of depth. Note that we verified that the MoS₂ lateral size and shape do not matter provided that the size is larger than the thermal contact radius. The temperature profile is mostly flat in the thin TMDC layer (see supplementary material Fig. 8 for 3D temperature distribution), as a result of the insensitivity to thermal conductivity. Most importantly, there is a strong temperature discontinuity associated with the MoS₂/SiO₂ interface. Finally, one obtains a value of $4.6 \pm 2 \text{ MW m}^{-2} \text{ K}^{-1}$ for the thermal boundary conductance, which is close to the values found experimentally for flakes by Raman thermometry^{15,20} and of similar order of magnitude to a molecular dynamic study.³³ The value is intrinsically low as van der Waals bonding provides a much weaker connection between the monolayer and its support. It seems therefore that the quality of the material, being it an exfoliated flake or a CVD-grown crystal, is not key for heat conduction when it is supported.

In summary, this work has shown that, with a proper calibration technique, SThM allows for quantitative determination of key parameters associated with heat dissipation in supported 2D materials. Thermal conductivity may not be the relevant parameter, while van der Waals bonding leads to weak thermal coupling with substrates.



FIG. 4. Temperature profile in logarithmic scale as a function of depth below the heat source (FEM simulation). The upper region overlaid in green corresponds to the MoS₂ monolayer, the lower blue region corresponds to SiO₂, and, finally, the gray region corresponds to the contribution of the Si substrate.

In the near future, it will be useful to analyze TMDCs with a better spatial resolution, either by studying the jump at contact in probe approach curves or by implementing vacuum conditions. Analyzing heat dissipation in TMDCs as a function of temperature may also enable us to discriminate between the effect of thermal conductivity and thermal boundary conductance.²⁷

See the supplementary material for details on material, the temperature-probe thermal conductance connection, varying-thickness oxide calibration samples, and simulations.

We thank J.-M. Bluet for helping with the Raman characterization and A. Alkurdi for discussions. C.M.F.-A. acknowledges a CONACYT scholarship funding. S.G. and P.-O.C. acknowledge support of project TIPTOP (No. ANR-16-CE09-0023), and A.D.L.-B. acknowledges the support of project No. UNAM-PAPIIT-A101822.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflict of interest to disclose.

Author Contributions

Christian Mateo Frausto-Avila: Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Visualization (equal); Writing – original draft (equal). Victor M. Arellano-Arreola: Investigation (equal). Jose Martin Yañez Limon: Conceptualization (equal); Resources (equal); Supervision (equal); Writing – review and editing (equal). Andres De Luna-Bugallo: Conceptualization (equal); Funding acquisition (equal); Project administration (equal); Resources (equal); Supervision (equal); Writing – review and editing (equal). Séverine Gomès: Conceptualization (equal); Methodology (equal); Project administration (equal); Resources (equal); Supervision (equal); Visualization (equal); Writing – review and editing (equal). Visualization (equal); Writing – review and editing (equal). Supervision (equal); Visualization (equal); Methodology (equal); Supervision (equal); Visualization (equal); Writing – original draft (equal); Writing – review and editing (equal).

DATA AVAILABILITY

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

REFERENCES

- ¹R. Peköz, M. Konuk, M. E. Kilic, and E. Durgun, ACS Omega 3, 1815 (2018).
- ²T. Knobloch, Y. Y. Illarionov, F. Ducry, C. Schleich, S. Wachter, K. Watanabe, T. Taniguchi, T. Mueller, M. Waltl, and M. Lanza, Nat. Electron. 4, 98 (2021).
- ³L. M. Xie, Nanoscale 7, 18392 (2015).
- ⁴I. D. Marion, D. Čapeta, B. Pielić, F. Faraguna, A. Gallardo, P. Pou, B. Biel, N. Vujičić, and M. Kralj, Nanotechnology **29**, 305703 (2018).
- ⁵Y. Lee, X. Zhang, W. Zhang, M. Chang, C. Lin, K. Chang, Y. Yu, J. T. Wang, C. Chang, and L. Li, Adv. Mater. **24**, 2320 (2012).
- ⁶P. Yang, S. Zhang, S. Pan, B. Tang, Y. Liang, X. Zhao, Z. Zhang, J. Shi, Y. Huan, and Y. Shi, ACS Nano 14, 5036 (2020).
- ⁷S. Ganorkar, J. Kim, Y.-H. Kim, and S.-I. Kim, J. Phys. Chem. Solids 87, 32 (2015).
- ⁸K. M. McCreary, E. D. Cobas, A. T. Hanbicki, M. R. Rosenberger, H.-J. Chuang, S. V. Sivaram, V. P. Oleshko, and B. T. Jonker, ACS Appl. Mater. Interfaces 12, 9580 (2020).
- ⁹G. Deokar, D. Vignaud, R. Arenal, P. Louette, and J.-F. Colomer, Nanotechnology 27, 075604 (2016).

- ¹⁰D. P. Rai, T. V. Vu, A. Laref, M. P. Ghimire, P. K. Patra, and S. Srivastava, Nano-Struct. Nano-Objects 21, 100404 (2020).
- ⁿP. Ni, A. De Luna Bugallo, V. M. Arellano Arreola, M. F. Salazar, E. Strupiechonski, V. Brandli, R. Sawant, B. Alloing, and P. Genevet, ACS Photonics 6, 1594 (2019).
- ¹²F. Xia, H. Wang, D. Xiao, M. Dubey, and A. Ramasubramaniam, Nat. Photonics 8, 899 (2014).
- ¹³D. G. Cahill, Rev. Sci. Instrum. **61**, 802 (1990).
- ¹⁴A. Abareshi, M. Arshadi Pirlar, and M. Houshiar, Mater. Res. Express 6, 105050 (2019).
- ¹⁵E. Yalon, Ö. B. Aslan, K. K. H. Smithe, C. J. McClellan, S. V. Suryavanshi, F. Xiong, A. Sood, C. M. Neumann, X. Xu, K. E. Goodson, T. F. Heinz, and E. Pop, ACS Appl. Mater. Interfaces 9, 43013 (2017).
- ¹⁶T. Tong and A. Majumdar, Rev. Sci. Instrum. 77, 104902 (2006).
- ¹⁷P. Jiang, X. Qian, X. Gu, and R. Yang, Adv. Mater. **29**, 1701068 (2017).
- ¹⁸S. Shree, A. George, T. Lehnert, C. Neumann, M. Benelajla, C. Robert, X. Marie, K. Watanabe, T. Taniguchi, U. Kaiser, B. Urbaszek, and A. Turchanin, 2D Materials 7, 015011 (2019).
- ¹⁹S. Sahoo, A. P. S. Gaur, M. Ahmadi, M. J.-F. Guinel, and R. S. Katiyar, J. Phys. Chem. C 117, 9042 (2013).
- ²⁰A. Taube, J. Judek, A. Łapińska, and M. Zdrojek, ACS Appl. Mater. Interfaces 7, 5061 (2015).
- ²¹X. Zhang, D. Sun, Y. Li, G.-H. Lee, X. Cui, D. Chenet, Y. You, T. F. Heinz, and J. C. Hone, ACS Appl. Mater. Interfaces 7, 25923 (2015).
- ²²J. Judek, A. P. Gertych, M. Świniarski, A. Łapińska, A. Dużyńska, and M. Zdrojek, Sci. Rep. 5, 1 (2015).

- ²³A. J. Gabourie, S. V. Suryavanshi, A. B. Farimani, and E. Pop, 2D Materials 8, 011001 (2021).
- ²⁴R. Yan, J. R. Simpson, S. Bertolazzi, J. Brivio, M. Watson, X. Wu, A. Kis, T. Luo, A. R. Hight Walker, and H. G. Xing, ACS Nano 8, 986 (2014).
- ²⁵S. Gomès, A. Assy, and P. O. Chapuis, Phys. Status Solidi A **212**, 477 (2015).
- ²⁶S. Vaziri, E. Yalon, M. M. Rojo, S. V. Suryavanshi, H. Zhang, C. J. McClellan, C. S. Bailey, K. K. H. Smithe, A. J. Gabourie, V. Chen, S. Deshmukh, L. Bendersky, A. V. Davydov, and E. Pop, Sci. Adv. 5, 1 (2019).
- ²⁷C. Evangeli, J. Spiece, S. Sangtarash, A. J. Molina-Mendoza, M. Mucientes, T. Mueller, C. Lambert, H. Sadeghi, and O. Kolosov, Adv. Electron. Mater. 5, 1900331 (2019).
- ²⁸V. M. A. Arreola, M. F. Salazar, T. Zhang, K. Wang, A. H. B. Aguilar, K. C. S. Reddy, E. Strupiechonski, M. Terrones, and A. D. L. Bugallo, 2D Materials 8, 025033 (2021).
- ²⁹F. Menges, H. Riel, A. Stemmer, C. Dimitrakopoulos, and B. Gotsmann, Phys. Rev. Lett. **111**, 205901 (2013).
- ³⁰M. E. Pumarol, M. C. Rosamond, P. Tovee, M. C. Petty, D. A. Zeze, V. Falko, and O. V. Kolosov, Nano Lett. **12**, 2906 (2012).
- ³¹E. Guen, P.-O. Chapuis, R. Rajkumar, P. S. Dobson, J. M. R. Weaver, and S. Gomes, J. Appl. Phys. **128**, 235301 (2020).
- ³²A. M. Massoud, J.-M. Bluet, V. Lacatena, M. Haras, J.-F. Robillard, and P.-O. Chapuis, Appl. Phys. Lett. 111, 063106 (2017).
- ³³Z.-Y. Ong, Y. Cai, G. Zhang, and Y.-W. Zhang, Nanotechnology **32**, 135402 (2021).