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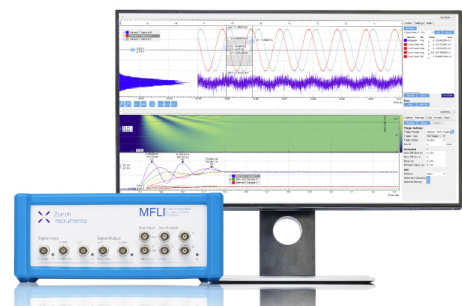
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ABSTRACT

Transition metal dichalcogenides such as molybdenum disulfide (MoS₂) may see service in the heart of next-generation nanoelectronic devices, where highly localized power dissipation can produce nontrivial temperature gradients over nanometer-scale distances. Here, we demonstrate that MoS₂ is a promising target for plasmon energy expansion thermometry (PEET), a high-spatial resolution temperature mapping technique employed in a scanning transmission electron microscope (STEM) equipped with electron energy loss spectroscopy (EELS). We first use a calibrated, commercial MEMS-style TEM sample heater chip to measure the temperature dependence of the MoS₂ bulk plasmon. We corroborate the chip's temperature calibration with Raman thermometry and determine the bulk thermal expansion coefficient (TEC) of MoS₂ in the temperature range of 300–1100 K. Applying this TEC value to PEET measurements on a suspended MoS₂ flake, we map 70–90 K/μm temperature gradients with a submicrometer spatial resolution.

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Transition metal dichalcogenides (TMDCs) draw attention because their weak out-of-plane bonding enables easy isolation of their atomically thin constituents. Their extraordinary electrical and optical properties and mechanical flexibility make TMDCs excellent candidates for novel nanoscale electronic and optoelectronic devices.¹ As device features approach or reach the atomic limit,² nanometer-scale thermometry techniques are required to better understand their thermal transport and heat management.

The currently available temperature measurement techniques with submicrometer resolution are generally optical,^{3,4} or scanning probe-based.^{5,6} Optical techniques (e.g., Raman, infrared detection) are noncontact, but diffraction limits their resolution to several hundred nanometers. Scanning probe techniques achieve ~10 nm resolution,⁷ but thermal contact requires perturbing the probed small volumes' temperature, introducing errors that are challenging to estimate and control.

Plasmon energy expansion thermometry (PEET) is a noncontact, nanoscale temperature mapping technique based on electron energy loss spectroscopy (EELS) in a scanning transmission electron microscope

(STEM).^{8,9} This approach exploits the temperature dependence of a material's bulk plasmon energy: thermal expansion or contraction changes the local electron density, which, in turn, shifts the bulk plasmon energy. In PEET, a STEM equipped with EELS rasters a focused probe across the sample and collects a spectrum at each beam position (the 3D data array is called a spectrum image). Analyzing a spectrum image produces a plasmon energy map, which is subsequently converted into a temperature map, using the material's known thermal expansion coefficient (TEC). For materials with sufficiently sharp plasmon resonances, PEET provides a method for obtaining temperature maps with a nanometer-scale resolution.

Other TEM-based temperature measurements use diffraction-based detection of thermal expansion,¹⁰ detailed balance using EELS,¹¹ or thermal population of Stokes/anti-Stokes excitations.¹² The phonon-based measurements are a novel and more fundamental means to measure temperature, but phonons are more delocalized than plasmons. So far, temperature gradients in the TEM have only been observed with PEET, although other techniques have such potential.

In the work presented here, we apply PEET to the prototypical member of the TMDC family, molybdenum disulfide (MoS_2). We use an FEI NanoExTM-i/v TEM microheater chip¹³ [Fig. 1(a)] with a vendor-supplied calibration of temperature-vs-microheater resistance. Depositing a MoS_2 flake, we corroborate the vendor calibration using both conventional (diffraction limited) Raman thermometry and PEET. Finally, we use a homemade microheater chip [Fig. 1(b)] to create switchable, opposing, nanoscale temperature gradients across a MoS_2 flake that we map using PEET.

Few layer MoS_2 flakes are dry-transferred via mechanical exfoliation (scotch tape method) to a commercial or homemade TEM chip (Fig. 1),¹⁴ avoiding some wet transfer contamination.¹⁵ The MoS_2 is exfoliated first from the bulk material (SPI Supplies, Inc.) onto a transparent polydimethylsiloxane (PDMS) substrate, and

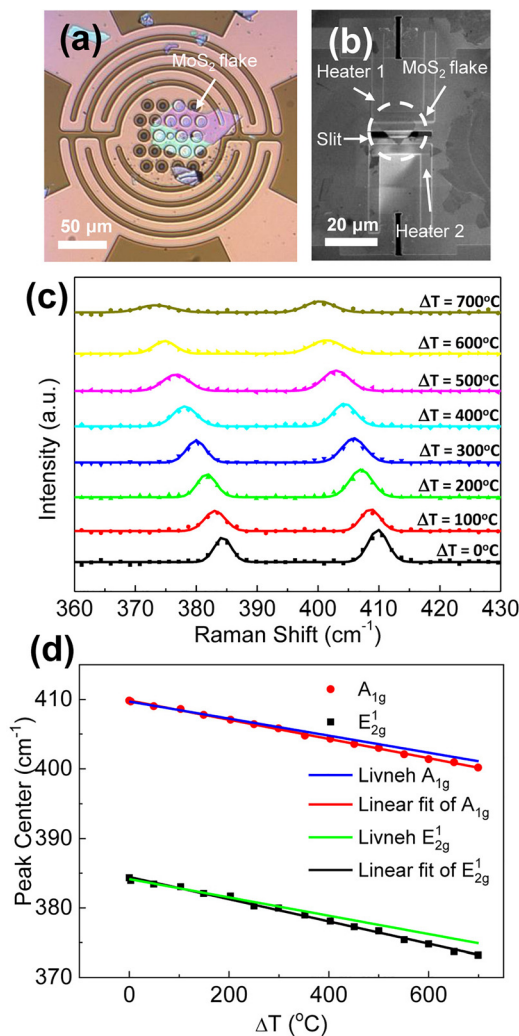


FIG. 1. Commercial [(a), optical image] and homemade [(b), SEM image] microheater chips with MoS_2 flakes deposited. (c) Raman spectra of MoS_2 at different temperatures. (d) Comparison of theoretical and experimental Raman peak positions as a function of temperature change (ΔT). In (d) the offsets were kept the same [from the value determined in (c)] and the different slopes are plotted.

then a thick flake (≥ 10 layers) is located using optical microscopy.¹⁶ The target flake is transferred to the TEM chip using a home-built contact aligner.¹⁷ The chips are wire bonded to PCB chip carriers customized for a TEM biasing holder (Hummingbird, Inc.), and electrical power is supplied by a Keithley 2400 Sourcemeter controlled with LabVIEW.

For Raman thermometry, the samples are placed in a cryostat (Cryo Industries, Inc.) with a base pressure of $<10^{-5}$ Torr. Raman spectra are collected using a Renishaw InVia confocal Raman microscope with 100 μW of a 532 nm laser excitation. TEM data are acquired in a JEOL JEM-2100F with a Gatan Quantum 963 GIF. Standard temperature mapping conditions used an 80 kV accelerating potential, a beam current of 0.3 nA, and a 12–14 mrad beam convergence semiangle (19–22 mrad spectrometer collection semiangle). PEET mapping conditions use a beam current of 0.3 nA, a 12–14 mrad beam convergence semiangle, a 19–22 mrad spectrometer collection semiangle, and a 0.025 eV/bin EELS dispersion. The calibrated heater experiment is performed with an accelerating potential of 200 kV and an EELS zero-loss peak (ZLP) full-width at half maximum (FWHM) of 0.95 eV, while the gradient experiment is performed with 80 kV and a ZLP FWHM of 0.65 eV.

Raman spectroscopy is a noncontact, nondestructive technique widely used to study mechanical and thermal properties of graphene and TMDCs. Raman peak positions shift and broaden with increasing temperature and have been measured for vapor-phase-grown, exfoliated, and hydrothermal-synthesized MoS_2 flakes. The in-plane mode E_{2g}^1 and out-of-plane mode A_{1g} frequencies show a linear variation with temperature.^{18,19} This temperature dependence can be mapped with $\sim 1 \mu\text{m}$ spatial resolution. Moreover, the temperature variation of these Raman frequencies is primarily due to thermal expansion.¹⁸ Thus, the shifting of Raman peaks and bulk plasmon peaks is similar, making Raman thermometry, which is comparatively well-established, an appropriate technique for calibrating temperature measurements in MoS_2 via PEET.

Figure 1 shows the NanoEx chip with a ~ 20 nm-thick MoS_2 flake and the shifts of the MoS_2 E_{2g}^1 and A_{1g} Raman modes at elevated temperatures. Temperatures are determined from the calibrated heater resistance. Linear fitting gives first order temperature coefficients of $\gamma(E_{2g}^1) = -1.60 \pm 0.03 \times 10^{-2} \text{ cm}^{-1}/\text{K}$ and $\gamma(A_{1g}) = -1.39 \pm 0.01 \times 10^{-2} \text{ cm}^{-1}/\text{K}$. We observe reasonable agreement ($\sim 10\%$) with the linear dependence reported by Livneh *et al.*, $\gamma(E_{2g}^1) = -1.47 \times 10^{-2} \text{ cm}^{-1}/\text{K}$ and $\gamma(A_{1g}) = -1.23 \times 10^{-2} \text{ cm}^{-1}/\text{K}$, shown as green (E_{2g}^1) and blue (A_{1g}) lines in Fig. 1(d).^{20,21}

After confirming the MoS_2 flake temperature via Raman thermometry, we switch to plasmon thermometry. EELS spectra (5 of 29) of suspended MoS_2 at different temperatures are shown in Fig. 2(a). The plasmon peak's maximum shift is equivalent to just seven spectrometer bins in the 2048-bin spectrum. Spectrum images are acquired at different temperatures, and the plasmon energy shifts are determined by fitting the ZLP and the MoS_2 plasmon peak with Gaussian and Lorentzian functions, respectively. The measured bulk plasmon energy of MoS_2 is defined as the difference between the ZLP's and MoS_2 plasmon peak's fitted centers, eliminating common-mode effects caused by the beam shifting relative to the spectrometer.

The curve fit spectrum images produce plasmon energy maps showing a clear systematic shift with temperature (supplementary material, Fig. 1). Each map produces a normally distributed set of plasmon energies

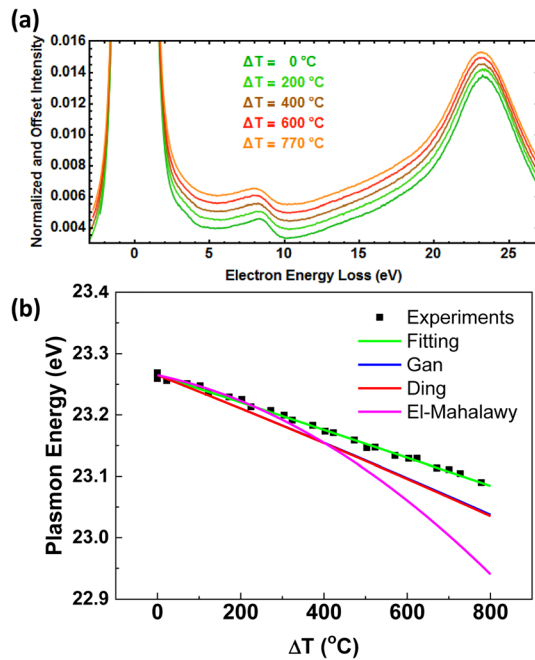


FIG. 2. (a) A sampling (5 of 29) of EELS spectra over a range of temperatures in the heating experiment. The spectra are offset vertically for clarity. (b) Comparison of theoretical and experimental plasmon energy shifts as a function of temperature change (ΔT).

per temperature (supplementary material, Fig. 1), from which a mean and standard deviation are extracted.

According to the electron gas model, the bulk plasmon energy $E_p = \hbar\omega_p = \hbar\sqrt{4\pi ne^2/m}$. At elevated temperatures, the valence electron number density n in the MoS₂ decreases due to thermal expansion according to the relation $n(T) \approx n(T_0)[1 - f(T)]$, where $f(T) = \int_{T_0}^T \alpha_V(T')dT' \approx \alpha_1\Delta T + \alpha_2\Delta T^2$, and α_1 and α_2 are the first and second order volume (not to be confused with linear) TECs, respectively.^{22–24} Temperature changes $\Delta T = T - T_0$ are measured relative to a reference temperature, here taken to be room temperature $\equiv 300$ K. Combining the equations for E_p and $n(T)$ gives the bulk plasmon energy as a function of temperature, $E_p(T) \simeq E_p(T_0)[1 - \frac{1}{2}f(T)] = E_p(T_0)[1 - \frac{1}{2}(\alpha_1\Delta T + \alpha_2\Delta T^2)]$. Figure 2(b) shows, as a function of temperature change ΔT , the measured bulk plasmon energies, along with predictions of E_p and reported α_V values^{25,26} for comparison. Fitting the experimental values to a second order polynomial [green line in Fig. 2(b)] gives the estimates $\alpha_1 = 1.9 \pm 0.1 \times 10^{-5} \text{ K}^{-1}$ and $\alpha_2 = 5 \pm 8 \times 10^{-10} \text{ K}^{-2}$ in the range of 300–1100 K.

The literature values for the range of the TECs of MoS₂ are various and confusing. In the 1970s, El-Malahaway and Evans,²⁶ and Murray and the same Evans,²⁷ measured MoS₂'s lattice parameters in the temperature ranges of 20–850 °C and 10–320 K, respectively, using X-ray diffraction. Unfortunately, while the data are consistent in the overlap region, the lattice-constant fit parameters (from which the TECs are extracted) given by Murray and Evans, do not match their own data. Moreover, the low-temperature TECs in Ref. 27 are more than twice than the high-temperature TECs in Ref. 26, which is unphysical since TECs are expected to approach zero in the low-temperature limit. A more recent

theory by Ding and Xiao²⁵ and Gan and Liu²⁸ matches this expectation, indicating that the MoS₂ TEC increases rapidly from zero below room temperature and then more slowly above room temperature. In quoting the X-ray data for comparisons, however, these^{25,28} and other authors²⁹ note neither Murray and Evan's error nor that the high-temperature TECs²⁶ are referenced to 0 °C and the low-temperature TECs²⁷ to 0 K, where, by all accounts, the TEC is very different. Huang and Zeng³⁰ and Hu *et al.*²⁹ have studied the in-plane thermal expansion of single and few-layer MoS₂. Hu *et al.* used shifts in the plasmon energy to measure in-plane thermal expansion coefficient values, but unfortunately do not give bulk volume TEC values (no out-of-plane component is given); so, a comparison is not possible.

Given this state of affairs, we ignore the TEC data of Murray and Evans entirely. Table I lists the other values available. The values of El-Mahalawy and Evans are outliers and thus seem implausible, not only because of the small value of α_1 but also because their (linearized) volume TEC increases by a factor of $2\alpha_2\Delta T/\alpha_1 \sim 4$ over their measured temperature range of 20–850 °C. For the other three entries (two of them theoretical), the corresponding increase is $\leq 15\%$. The value on the last line is the result of a quadratic fit of $E_p(T)$ to the data [Fig. 2(b)], where the errors are statistical and reflect the scatter in the data plotted.

To determine Raman-based temperatures in MoS₂ flakes, we invert the equations describing the peak shift's temperature dependence. We have $\Delta T_{\text{Raman}} = [\omega(T) - \omega(T_0)]/\gamma$, where ω is the frequency, and γ is the first order temperature coefficient, of the Raman mode.¹⁸ In the case of PEET, we solve $E_p(T)$ for ΔT , finding

$$\Delta T_{\text{PEET}} = \frac{\alpha_1}{2\alpha_2} \left(\sqrt{1 - \frac{8R\alpha_2}{\alpha_1^2}} - 1 \right) \simeq -\frac{2R}{\alpha_1} = -\frac{2}{\alpha_1} \frac{\omega_p(T) - \omega_p(T_0)}{\omega_p(T_0)}, \quad (1)$$

where the normalized change in the plasmon energy $R \equiv [E_p(T) - E_p(T_0)]/E_p(T_0)$. Here, we show the linear approximation to emphasize the similarity (the origin of the “2” is the square-root in the plasmon energy) with the Raman case. For the Raman-based determination of the MoS₂ flake's temperature, only two Raman spectra are acquired: room temperature and an elevated temperature. In contrast, for the PEET-based temperature determination, R is obtained at each point from two spectrum images. Raman thermometry provides point measurements on the μm -scale, while PEET can provide nm-scale maps.

When normalized for the integration time, the statistical precision of Raman thermometry and PEET are comparable in MoS₂. Raman spectra, acquired in 30 seconds, have a statistical uncertainty of

TABLE I. Comparison of the first and second order volume thermal expansion coefficients of MoS₂.

Author (T_0)	α_1 (10^{-5} K^{-1})	α_2 (10^{-10} K^{-2})
El-Mahalawy and Evans ²⁶ (273 K)	1.25	280
Ding and Xiao ²⁵ (300 K)	2.3	21
Gan and Liu ²⁸ (300 K)	2.3	18
Present work, EELS (300 K)	1.9 ± 0.1	5 ± 8

about 0.4 K (2 K/√Hz). PEET spectra, acquired in 0.05 s, have a statistical uncertainty of about 12 K (3 K/√Hz, similar to aluminum in Ref. 8).

Plotting ΔT_{PEET} and ΔT_{Raman} vs the vendor-calibrated chip temperature ΔT_{holder} (Fig. 3) shows that the Raman E_{2g}^1 and A_{1g} peaks give values that are systematically +9% and +13% high, respectively. PEET values, calculated using the TEC values of Gan and Liu for illustration purposes, are systematically low by 22%. (The values in the last line of Table I would give perfect agreement by construction.) Clearly systematic errors dominate statistical errors in both Raman and PEET thermometry (their scatter is small). One possibility is that the chip calibration, specified to be good at 5%, is incorrect. The chip's heater/thermometer could have been miscalibrated or altered, either during flake deposition or use. However, a different chip calibration would not make the Raman data agree with the PEET data. Thus, it appears that the temperature coefficients of the Raman modes, the available MoS₂ TEC values, the chip calibration, or some combination of these three are incorrect. Clearly, more work is required to demonstrate consistent and reproducible accuracy with these three thermometric techniques. On a more promising note, the lack of scatter in the data indicates that relative thermometry with precision at the few percent level is already possible. Because the Raman and the chip thermometries agree better than the other two pairs, we elect to take those values as more reliable and thus the value for MoS₂'s volume TEC given in the last line of Table I as the best available.

We now apply PEET to mapping a nanoscale temperature gradient in a suspended 30 nm-thick MoS₂ flake. To create temperature gradients, we use a homemade chip [Figs. 1(b) and 4(a)], applying heater power alternately on one side or the other (the heater on the opposing side is grounded). Spectrum images are acquired with neither heater powered (reference temperature T_0) and one heater powered (unknown temperature T), aligned to remove spatial drifts, and fit to give the plasmon energies. With the aligned maps of plasmon energies, the normalized change R is calculated for each pixel and then converted to a temperature map.

Applying 100 mW to the two heaters alternately gives two temperature maps [Fig. 4(b)]. Averaging the mapped region along the

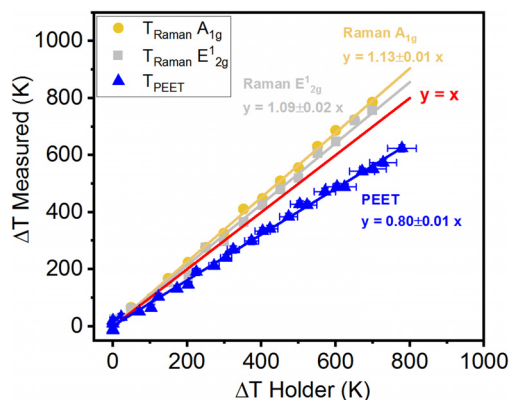


FIG. 3. Measured temperature changes (ΔT measured) obtained from PEET and Raman measurements plotted as a function of chip temperature (ΔT holder), with corresponding fits. The x-error bars are derived from the commercial MEMS chip vendor's specification of the accuracy of the heater/thermometer calibration (5%). The y-error bars (partially hidden by the plot markers) are statistically based on a reduced χ^2 analysis (~ 12 K).

direction transverse to the gradient gives a 1D plot of temperature as a function of the position along the flake [Fig. 4(c)]. Fitting these temperature profiles reveals that the (switchable) temperature gradients are 70–90 K/μm (−90 K/μm for left heating, blue line, and 70 K/μm for right heating, red line). The error in temperature, determined from the normally distributed temperatures in the isothermal columns in Fig. 4(b)'s temperature maps, is 20 K.

From the temperature gradient of 80 K/μm and our 20 K temperature uncertainty, we can estimate our spatial resolution σ_R to be 250 nm (slope $\sim \sigma_T/\sigma_R$). This resolution is not limited by the thermometry technique's spatial resolution *per se*, which is determined by (see Ref. [8]) the image pixel size (6 nm) and the plasmon delocalization length (~ 2 nm, which is similar to our probe size of 1–2 nm), but by its precision and the temperature gradient's size. Our gradient of 80 K/μm = 8×10^5 K/cm is already large by macroscopic standards, but the ideal target for a resolution test would feature a step change in the temperature. Such a step is impossible to arrange and difficult to even approximate, since the thermal conductivities of real materials span only a few orders of magnitude (compare, e.g., electrical conductivities, which span many).

In summary, we apply Raman thermometry and PEET to a MoS₂ flake on a commercial, calibrated MEMS-style TEM heating chip. Ramping the chip temperature from room temperature to 1100 K (according to the chip calibration) gives corresponding Raman and PEET temperature curves that are internally consistent at the few percent level, but that gives values 10% high and 20% low relative to

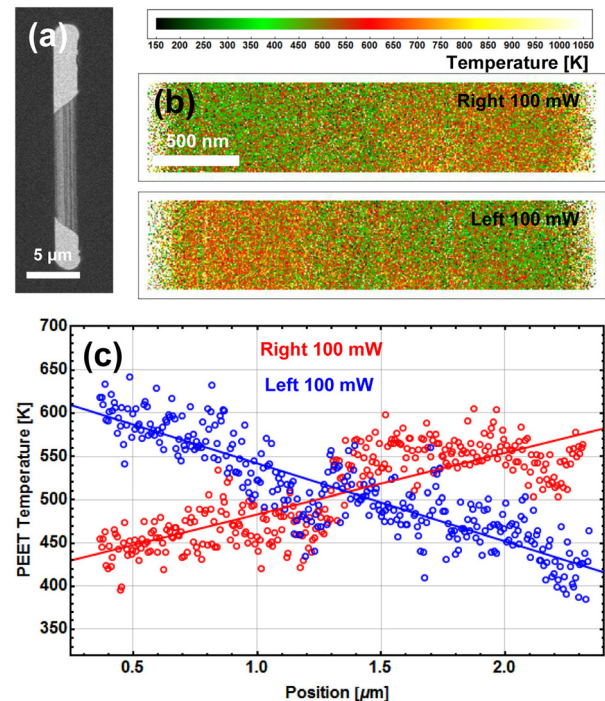


FIG. 4. (a) Higher-magnification STEM image of the device of Fig. 1(b), rotated 90°, showing the MoS₂ flake spanning the slit in the chip. PEET-derived 2D temperature maps (b) and 1D temperature profiles (c) plotted along the suspended length of a MoS₂ flake. A rectangular ROI is used (long direction across the slit), and the results are averaged in the short direction (~ 80 points).

the chip temperature, respectively. These systematic discrepancies highlight the need for more careful studies of the TECs and Raman-peak-frequency temperature coefficients in MoS₂ and other TMDCs. Interpreting the STEM EELS experiment as a measurement of the MoS₂ volume TEC gives $\alpha_1 = (1.9 \pm 0.1) \times 10^{-5} \text{ K}^{-1}$ and $\alpha_2 = (5 \pm 8) \times 10^{-10} \text{ K}^{-2}$ in the measured temperature range of 300–1100 K. We then used this TEC value to map 70–90 K/ μm temperature gradients in a suspended flake of MoS₂ with a submicrometer spatial resolution. The basic procedure reported here, which determines the TEC if unknown and then maps the plasmon energy to determine the temperature, is general and can be most profitably applied in nanostructures made of materials (e.g., many semiconductors) with sharp plasmon resonances and sizable TECs.

See the [supplementary material](#) for two figures describing the curve fitting used to determine the plasmon energy, plasmon energy maps at different temperatures, a schematic of the experiment, and images of the connections to the MoS₂ flake. The first supplementary figure shows the plasmon energy maps and characteristics of the fitting, and the second shows an overview of how the Joule heaters are connected to the device.

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