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# Tuning electrical and interfacial thermal properties of bilayer MoS<sub>2</sub> via electrochemical intercalation

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## Abstract

Layered two-dimensional (2D) materials such as MoS<sub>2</sub> have attracted much attention for nano- and opto-electronics. Recently, intercalation (e.g. of ions, atoms, or molecules) has emerged as an effective technique to modulate material properties of such layered 2D films reversibly. We probe both the electrical and thermal properties of Li-intercalated bilayer MoS<sub>2</sub> nanosheets by combining electrical measurements and Raman spectroscopy. We demonstrate reversible modulation of carrier density over more than two orders of magnitude (from  $0.8 \times 10^{12}$  to  $1.5 \times 10^{14}$  cm<sup>-2</sup>), and we simultaneously obtain the thermal boundary conductance between the bilayer and its supporting SiO<sub>2</sub> substrate for an intercalated system for the first time. This thermal coupling can be reversibly modulated by nearly a factor of eight, from  $14 \pm 4.0$  MW m<sup>-2</sup> K<sup>-1</sup> before intercalation to  $1.8 \pm 0.9$  MW m<sup>-2</sup> K<sup>-1</sup> when the MoS<sub>2</sub> is fully lithiated. These results reveal electrochemical intercalation as a reversible tool to modulate and control both electrical and thermal properties of 2D layers.

Supplementary material for this article is available [online](#)

Keywords: two-dimensional, intercalation, tunable, Raman, thermal, electrical

(Some figures may appear in colour only in the online journal)

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## 1. Introduction

Two-dimensional (2D) materials such as molybdenum disulfide (MoS<sub>2</sub>) are widely considered to be promising candidates for next-generation nanoelectronics due to their unique material

properties (electrical, thermal, optical, mechanical) and their sub-nanometer thickness without dangling bonds [1, 2]. Potential applications for 2D materials include transistors [3, 4], interconnects [5], transparent electrodes [6], sensors [7], solar cells [8], thermoelectrics [9], and energy storage devices [10]. This wide range of applications has many requirements; thus, it is desirable to modulate the properties of 2D materials to optimize their performance for specific applications. Researchers have been actively developing ways to engineer electron and phonon transport in 2D materials via chemical doping, strain, and nanostructuring [11–13]. However, a controllable, reversible, and scalable method to engineer 2D material properties is still lacking. For instance, while some doping strategies effectively achieve high carrier concentrations [14, 15], it remains challenging to precisely control or modulate the doping level in 2D materials.

Intercalation has recently emerged as a promising technique to engineer 2D material properties for tunable electronic applications [16–20]. In layered 2D crystals, the van der Waals spacing between layers provides perfect sites to accommodate guest species such as ions and molecules (intercalants) through a process known as intercalation. These intercalants modify interlayer interactions in 2D thin films and affect material properties through charge transfer, band gap engineering, and phonon scattering [21–25]. Earlier efforts studying intercalation in 2D materials often employed a chemical intercalation approach [18, 26], where 2D crystals were immersed in a chemical solution containing the intended intercalant species (e.g. n-butyl lithium for chemical intercalation of lithium) for an extended time (up to 72 h). While chemical intercalation is relatively easy to perform and provides a convenient route to demonstrate its usefulness in doping and phase engineering 2D materials, this process is irreversible and lacks control over the intercalant concentration.

In contrast, in an electrochemical intercalation process, the number of ions (e.g.  $\text{Li}^+$ ) intercalated between the 2D layers can be precisely and reversibly modulated by controlling the number of electrical charges (charging current  $\times$  time) transferred to the host materials [16, 17, 27, 28]. While electrochemical intercalation is extensively studied and forms the backbone of Li ion battery (LIB) industry, there has been limited work done leveraging this technique to engineer fundamental material properties in 2D materials. Electrically, electrochemical Li intercalation has demonstrated tunable electrical conductance over 2–3 orders of magnitude in graphitic thin films [17], few-layer  $\text{MoS}_2$  [16], as well as 2D heterostructures [29]. Thermally, Li intercalation has been employed to achieve  $\sim 8\text{--}10 \times$  modulation of cross-plane thermal conductance (combined volumetric contribution of  $\text{MoS}_2$ , and interfacial contributions of  $\text{Al}/\text{MoS}_2$  and  $\text{MoS}_2/\text{SiO}_2$ ) in  $\text{MoS}_2$  thin films [30] ( $\sim 10$  nm thick) and up to  $5 \times$  change in cross-plane thermal conductivity in bulk  $\text{MoS}_2$  samples [21] ( $\sim 10\text{--}20$   $\mu\text{m}$  thick). However, while these earlier studies provided valuable insights on how intercalation affects electrical and thermal transport in 2D materials, detailed studies on the charge transfer process in intercalated 2D films and the effect of intercalants on thermal boundary conductance (TBC) between 2D films and the substrate, a key parameter in determining energy dissipation in 2D electronics [31–33], are still lacking. The TBC

plays an important role in thermal transport across sub-100 nm thin films, even when quasi-ballistic effects are considered [34].

In this work, we investigate how the carrier concentration in  $\text{MoS}_2$  and the TBC at  $\text{MoS}_2/\text{SiO}_2$  interface vary as a function of Li concentration through electrical Hall measurements and Raman spectroscopy. In addition, prior intercalation studies often employ the well-characterized liquid electrolyte and Li metal as the reference electrode [16, 17], which are highly reactive and flammable and may not be ideal for electronics applications. We report a solid-state electrochemical intercalation platform using  $\text{LiClO}_4$  in polyethylene oxide (PEO) as a solid electrolyte and the air-stable  $\text{LiFePO}_4$  (LFP) as the reference electrode (Li reservoir). Our work addresses reversible doping and thermal dissipation that are two fundamental issues in 2D electronics, and provides a promising path towards tunable 2D electronics.

## 2. Experiments

### 2.1. Device fabrication

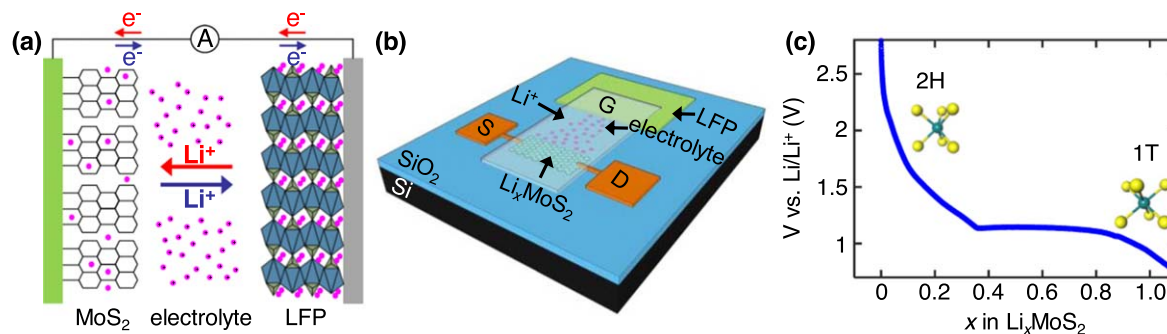
$\text{MoS}_2$  flakes were mechanically exfoliated onto a highly doped (p-type) Si substrate with 90 nm thick thermally grown  $\text{SiO}_2$ . The bilayer  $\text{MoS}_2$  flakes were pre-sorted with an optical microscope and later confirmed with Raman spectroscopy and atomic force microscopy (AFM). The  $\text{MoS}_2$  flakes were patterned into Hall structures with e-beam lithography and  $\text{XeF}_2$  etching. Electrode contacts (Ti/Au, 1/50 nm) were deposited with e-beam evaporation.

### 2.2. Intercalation

Our 2D electrochemical device is similar to a planar nano-battery with  $\text{MoS}_2$  and lithium iron phosphate  $\text{LiFePO}_4$  serving as the working and reference/counter electrodes, respectively. The solid electrolyte was prepared by dissolving  $\text{LiClO}_4$  (Sigma Aldrich) into PEO (Sigma Aldrich) matrix. PEO and  $\text{LiClO}_4$  powders (500 and 150 mg, respectively) were mixed with 7.5 ml of anhydrous methanol and stirred for 12 h at 50 °C. An electrochemical workstation (Biologic SP-150) was used to perform the Li intercalation and de-intercalation and measure  $\text{MoS}_2$  electrochemical potential.

### 2.3. Characterization and measurements

Raman spectroscopy was performed with a Horiba LabRam instrument with a 532 nm green laser and  $1800\text{ mm}^{-1}$  grating. A  $100\times$  long working distance objective (N.A. = 0.6) was used to accommodate the  $\text{MoS}_2$  electrochemical devices with PEO electrolyte. For Raman thermometry measurements, the temperature calibrations were carried out with a Linkam THMS600 stage. Part of the electrical transport measurements was carried out using a semiconductor parameter analyzer (Keithley 4200 SCS). The temperature-dependent Hall measurement was conducted in a Quantum Design PPMS-7 system with digital lock-in amplifiers (Stanford Research Systems SR830).



**Figure 1.** Electrochemical intercalation. (a) Schematics of electrochemical intercalation process with MoS<sub>2</sub> sheets as the working electrode and LiFePO<sub>4</sub> (LFP) as the reference electrode. The solid electrolyte is LiClO<sub>4</sub> in PEO. The red and blue arrows represent the direction of Li ions (in electrolyte) and electrons (in external circuit) movements during intercalation and de-intercalation, respectively. (b) Schematics of electrochemical intercalation device. (c) Galvanostatic discharge curve of MoS<sub>2</sub> with respect to LFP. The electrochemical potential (y-axis) is normalized to Li/Li<sup>+</sup> potential by adding 3.5 V, the potential of LFP with respect to Li/Li<sup>+</sup>. The 2H to 1T phase transition plateau for MoS<sub>2</sub> occurs at 1.1 V with respect to Li, similar to previous reports [16, 35, 36]. This indicates that LFP works well as an air-stable reference electrode to replace the highly reactive Li metal.

### 3. Results and discussion

#### 3.1. Electrochemical intercalation

Figure 1 shows the schematics of the electrochemical intercalation process, where Li ions move in and out of the interlayer spacing in 2D materials, depending on the external electrical current flow. Our intercalation platform consists of the 2D material (the working electrode), a Li reservoir (the reference electrode), and the electrolyte to allow ionic movement (no electron transport). The main advantage of electrochemical intercalation over the chemical intercalation process is that we can achieve reversible control over the intercalant concentration [16]. Governed by charge neutrality, for every negatively charged electron that flows from the reference electrode to the working electrode during intercalation, a positively charged Li<sup>+</sup> ion moves from the reference electrode to the working electrode, i.e. a Li<sup>+</sup> ion is intercalated in the 2D material. Thus, we can precisely modulate the Li concentration in the host 2D material by controlling the total amount of charge transferred from the reference electrode to the 2D material (current × time). To increase the Li content in 2D materials, i.e. intercalation, we will flow electrons from the reference electrode to the 2D material via an external circuit (indicated by the direction of red arrows in figure 1(a)); to decrease the Li content, i.e. de-intercalation, we will need to reverse the current flow (blue arrows in figure 1(a)). This allows us to engineer the electrical and thermal properties of our device by controlling the Li concentration in 2D materials.

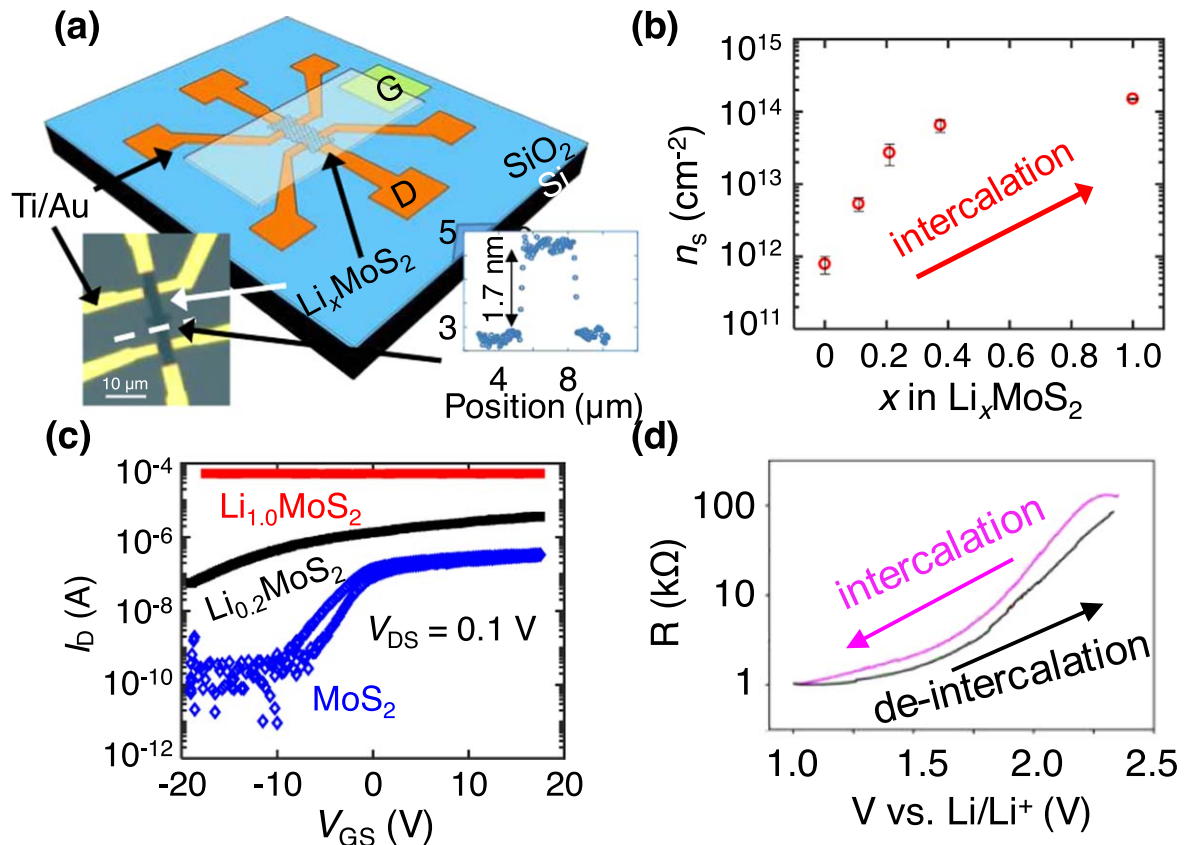
We adopt the air-stable LiFePO<sub>4</sub> (LFP) (instead of the highly reactive Li metal) as the reference electrode (figure 1(b)) in our device. LFP has a very stable electrochemical plateau (~3.4 V versus Li/Li<sup>+</sup>) when its lithiation content is between 10% and 90% [37, 38]. LFP also has a long lifetime and a high power density; therefore, it has been used as a cathode material in rechargeable LIB [37, 38]. We also utilize a solid electrolyte, LiClO<sub>4</sub>, in PEO for this work, instead of the liquid electrolyte (LiPF<sub>6</sub> in EC/DEC solution) that is prevalent in LIBs. Even though a liquid electrolyte

typically has better ionic mobility than the polymer electrolyte, it is prone to low-charge retention, leakage issues, and thermal instability at low and high operating temperatures [39]. This would severely limit the application of intercalation in tunable electronics. In contrast, the PEO electrolyte has better chemical and thermal stability, comparable ionic mobility, and higher energy density [40].

In principle, we could calculate the exact Li content in our 2D films, knowing the current flow and exact amount of charge flowing into the 2D nanosheet, as well as the exact molecular mass of the host 2D material. However, it is impractical to measure the weight of our MoS<sub>2</sub> nanosheet accurately. Thus, we first performed a galvanostatic discharge measurement (figure 1(c)) on bulk MoS<sub>2</sub> powder as a calibration, where we can measure the weight of the bulk MoS<sub>2</sub> accurately. Since the galvanostatic discharge measurement only depends on the chemical composition of the material, we expect the calibration result of the bulk powder to be transferable to MoS<sub>2</sub> thin films. From our calibration, we established a correlation between the electrochemical potential with respect to Li/Li<sup>+</sup> and Li concentration in intercalated Li<sub>x</sub>MoS<sub>2</sub> (figure 1(c)). The plateau at 1.1 V represents the 2H to 1T phase transition in MoS<sub>2</sub>, consistent with previously reported values [16, 35, 36]. With the calibration result, we can modulate the Li content in MoS<sub>2</sub> conveniently by varying the electrochemical potential of MoS<sub>2</sub> with respect to Li/Li<sup>+</sup> and read the Li concentration by comparing the potential in our calibrated sample. To prevent the irreversible conversion reaction that forms Li<sub>2</sub>S [35], we avoid lithiating MoS<sub>2</sub> below 0.9 V with respect to Li/Li<sup>+</sup> in this work.

#### 3.2. Electrical transport

We fabricated Hall bar structures on bilayer MoS<sub>2</sub> samples to investigate how much carrier modulation we can achieve with electrochemical intercalation. The schematics and optical image of our electrochemical devices are shown in figure 2(a). Figure 2(a) inset shows the MoS<sub>2</sub> thickness (*t*<sub>MoS<sub>2</sub></sub>) to be ~1.7 nm (bilayer) through AFM. More fabrication details can be found in the Methods and supporting information



**Figure 2.** Tuning MoS<sub>2</sub> carrier concentration via intercalation. (a) Schematics of Hall bar structure with electrochemical intercalation platform. Inset is an optical image of fabricated MoS<sub>2</sub> device with Hall bar structure. The inset shows the AFM measurement of the MoS<sub>2</sub> device, confirming bilayer thickness ( $\sim 1.7$  nm). (b) Carrier concentration in MoS<sub>2</sub> as a function of Li concentration, controlled by the potential difference of MoS<sub>2</sub> with respect to the reference electrode. (c) Transfer characteristics of pristine MoS<sub>2</sub> (blue), Li<sub>0.2</sub>MoS<sub>2</sub> (black), Li<sub>1.0</sub>MoS<sub>2</sub> (red). The channel conductance increases as carrier concentration increases due to Li intercalation. Li<sub>1.0</sub>MoS<sub>2</sub> shows metallic behavior (no  $V_{GS}$  dependence) because it is in the IT phase. (d) Reversible electrical properties as we lithiate and de-lithiate the MoS<sub>2</sub> thin film (a different bilayer sample than the one depicted in (a)–(c)) via changing its electrochemical potential.

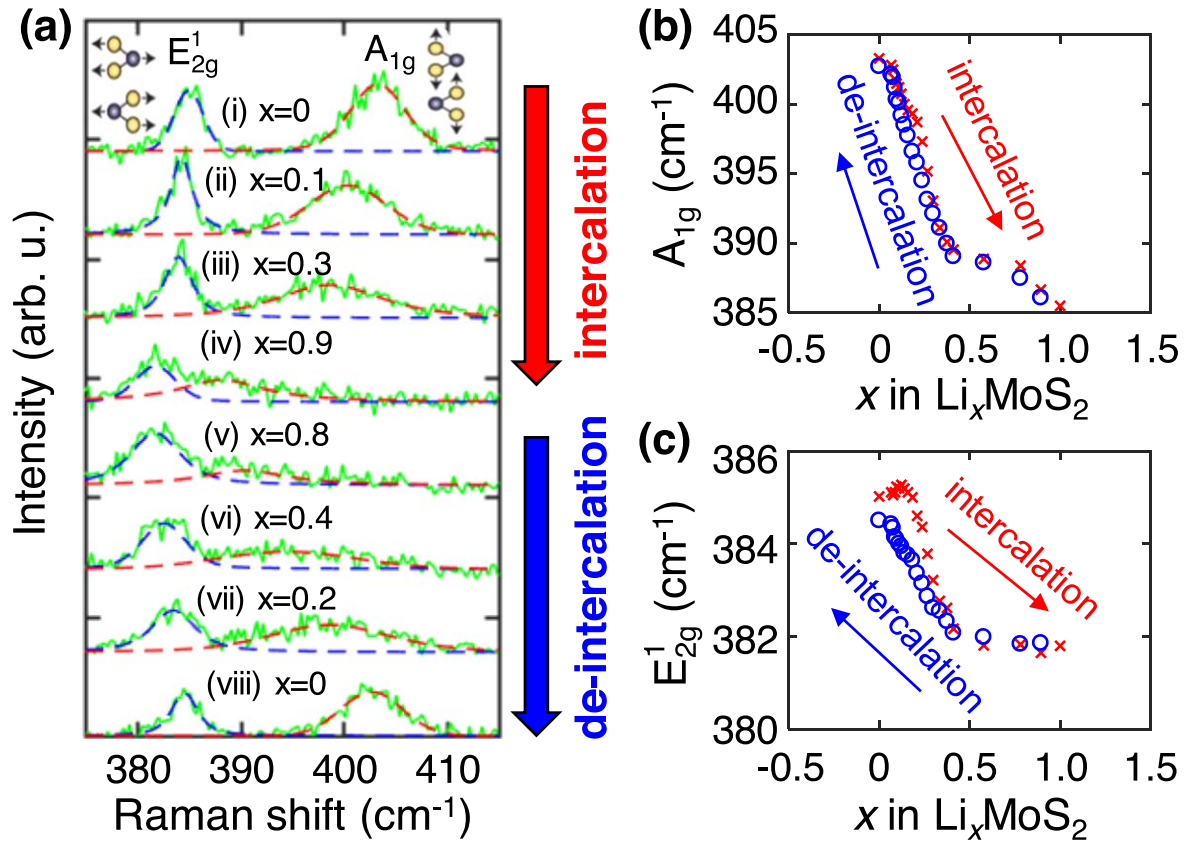
sections (available online at [stacks.iop.org/NANO/32/265202/mmedia](https://stacks.iop.org/NANO/32/265202/mmedia)).

Next, we applied the polymer electrolyte (LiClO<sub>4</sub> in PEO) to cover the entire MoS<sub>2</sub> channel and the reference electrode (LFP, see Methods for more details). This electrically-insulating solid polymer allows ionic exchange between LFP and the 2D material. Unlike an electric-double-layer transistor (EDLT), which does not have a reference electrode (only a metal contact) [41–44], we can controllably perform electrochemical intercalation in our device with a well-defined electrochemical reference potential (figure 1(a)). This provides additional control in electronics applications because the effect from electrochemical intercalation is long-term, while the ionic gating effect in EDLT is short-term (only persist with applied gate voltage). As we monitor the electrochemical potential of MoS<sub>2</sub> with respect to the reference electrode, we can precisely and reversibly tune the Li concentration in MoS<sub>2</sub> by adjusting the potential between MoS<sub>2</sub> and the reference electrode (figure 1(c)).

By controlling the electrochemical potential of MoS<sub>2</sub> with respect to our reference LFP electrode, we can intercalate/de-intercalate Li ions and therefore modulate the carrier concentrations in MoS<sub>2</sub>, similar to a charging/discharging process

in a LIB. Initially, the pristine MoS<sub>2</sub> had an electrochemical potential of 2.5 V versus Li/Li<sup>+</sup>, i.e.  $-0.95$  V with respect to LFP, since partially lithiated LFP has a stable potential plateau of 3.45 with respect to Li/Li<sup>+</sup> (see Methods for more details) [37, 38]. The 2D carrier concentration  $n_s$  in this MoS<sub>2</sub> flake was  $8 \times 10^{11}$  cm<sup>-2</sup> before Li intercalation, as extracted from Hall measurement. As we intercalate more Li ions into MoS<sub>2</sub> by lowering the potential of MoS<sub>2</sub> with respect to the reference electrode (figure 1(c)), we observe a significant increase in carrier concentration (figure 2(b)), with  $n_s$  reaching  $1.5 \times 10^{14}$  cm<sup>-2</sup> when Li concentration approached the maximum intercalation capacity Li<sub>1.0</sub>MoS<sub>2</sub>, equivalent to one Li ion per unit cell. This large increase in carrier concentration is likely due to the combined results of charge transfer from the intercalated Li ions and the effect of ionic gating. We also measured the temperature dependence of the lithiated MoS<sub>2</sub> device's electrical resistance from room temperature down to 2 K (see figure S2 in the supporting information) but did not observe any super-conductive behavior in our device.

The Hall bar structure also behaves as a 2D transistor with a Si back gate below the 90 nm thick SiO<sub>2</sub>. Figure 2(c) shows the transfer curves of a typical device at room temperature with different Li concentrations under a source-drain



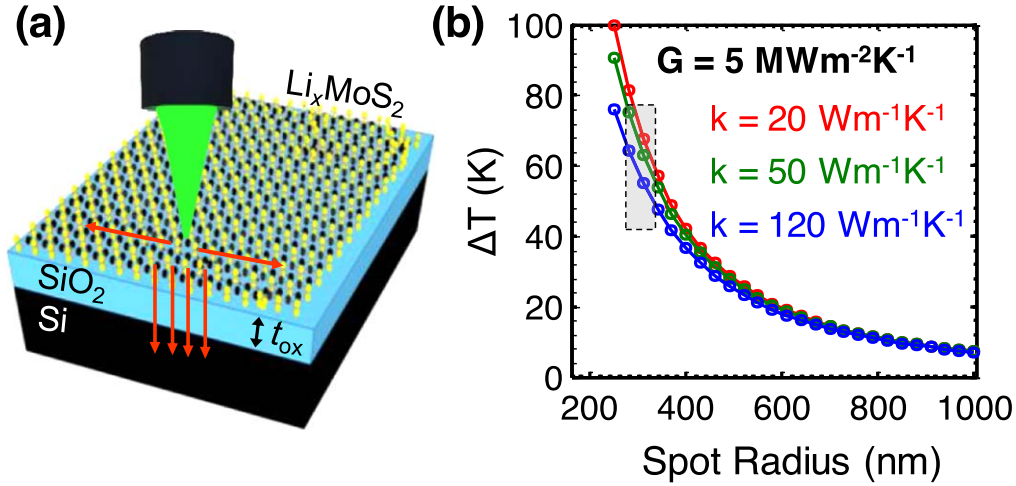
**Figure 3.** Raman spectroscopy of Li-intercalated MoS<sub>2</sub>. (a) Raman spectroscopy of bilayer MoS<sub>2</sub> thin film during the intercalation (i)–(iv) and de-intercalation (v)–(viii) steps. Green curves are the raw data; blue and red curves are the fitted results for the Raman modes,  $E_{2g}^1$  and  $A_{1g}$ , respectively. The insets show the corresponding vibrational modes. (b)–(c) The measured frequency shift of  $A_{1g}$  and  $E_{2g}^1$  modes as a function of Li content in  $\text{Li}_x\text{MoS}_2$ . Red crosses and blue circles represent the intercalation and the subsequent de-intercalation steps, respectively.

bias  $V_{DS} = 0.1$  V. Before Li intercalation, the exfoliated MoS<sub>2</sub> (blue curve in figure 2(c)) was in a semiconducting 2H phase and showed an on/off ratio of  $\sim 1000\times$  through back-gate control. Upon Li intercalation (black curve,  $\text{Li}_{0.2}\text{MoS}_2$ ), we noticed an increase in channel conductance due to doping and decreased on/off ratio ( $\sim 50\times$ ). When the MoS<sub>2</sub> flake was fully lithiated to  $\text{Li}_{1.0}\text{MoS}_2$  (red curve), the channel became metallic as MoS<sub>2</sub> underwent a 2H (semiconducting) to 1T (metallic) phase transformation upon lithiation as previously reported [16, 18, 35, 36]. We observed  $\sim 500\times$  improvement in drive current  $I_{on}$  upon Li intercalation ( $\text{Li}_{1.0}\text{MoS}_2$ ) likely because of the higher carrier concentration [29] ( $\sim 200\times$ ) upon intercalation as well as lower contact resistance due to the semiconducting-to-metallic phase transition [18, 45]. This 2H to 1T phase transition is likely triggered by the strain from the influx of Li ions [46]. Upon intercalation, Li atoms occupy octahedral sites between the 2H-MoS<sub>2</sub> layers, forming an unstable 2H- $\text{Li}_x\text{MoS}_2$  phase. As more Li atoms enter the MoS<sub>2</sub> structure [47], the strain causes the crystal structure to transform into the thermodynamically-stable 1T- $\text{Li}_x\text{MoS}_2$  phase with stacking sequence AA shown in the figure 1(c) inset. The associated ( $\sim 2\%$ – $5\%$ ) increase in interlayer spacing allows it to accommodate more Li atoms in the unit cell, as similarly reported in previous *in situ* XRD measurements [23] and first-principle

calculations [46]. The change in electrical conductance is reversible as we lithiate and de-lithiate the MoS<sub>2</sub> device (figure 2(d)), opening up opportunities for applications in memory and synaptic devices[48–51].

### 3.3. Raman spectroscopy

We also studied how intercalation affected the doping and strain in these 2D layers via Raman microscopy. Figure 3 illustrates our Raman measurements on a bilayer MoS<sub>2</sub> flake as we intercalate and de-intercalate the device. Green curves are the original Raman spectra; blue and red curves represent the fitted  $E_{2g}^1$  and  $A_{1g}$  modes, respectively. By intercalating  $\text{Li}^+$  ions into a bilayer MoS<sub>2</sub> film (series (i)–(iv) in figure 3(a)), we noticed a significant peak shift and broadening of the  $A_{1g}$  mode (figure 3(b)), which is attributed to doping in MoS<sub>2</sub> [16, 52, 53]. Similarly, the  $E_{2g}^1$  mode also decreased in frequency (figure 3(c)) and broadened upon intercalation, albeit to a much lesser extent. This likely indicates strain build-up in the lattice due to the influx of  $\text{Li}^+$  ions [16, 54]. These changes in Raman spectra were fully reversible as we moved  $\text{Li}^+$  ions out of MoS<sub>2</sub> during the de-intercalation process [series (v)–(viii) in figure 3(a)] and are consistent with our density functional theory calculations (see figure S7 in the supporting information). The  $A_{1g}$  mode shift



**Figure 4.** MoS<sub>2</sub>/SiO<sub>2</sub> thermal boundary conductance (TBC) before and after intercalation. (a) Schematic of Raman thermometry platform of the bilayer MoS<sub>2</sub> on the SiO<sub>2</sub>/Si substrate. (b) The simulated temperature rise of MoS<sub>2</sub> as a function of laser spot size at an absorbed power of 1.2 mW, when the MoS<sub>2</sub>/SiO<sub>2</sub> TBC = 5 MW m<sup>-2</sup> K<sup>-1</sup>, near the lower bound of the TBC after intercalation. Red, green and blue curves are for in-plane thermal conductivity of MoS<sub>2</sub>  $k = 20, 50,$  and  $120 \text{ W m}^{-1} \text{ K}^{-1}$ , respectively. This suggests that the temperature rise in MoS<sub>2</sub> is insensitive to its in-plane  $k$ , most of the heat flows through the SiO<sub>2</sub>/Si substrate, and we can fit the TBC of MoS<sub>2</sub>/SiO<sub>2</sub> by assuming a typical  $k \approx 50 \text{ W m}^{-1} \text{ K}^{-1}$ .

was as large as  $18 \text{ cm}^{-1}$  at Li<sub>1.0</sub>MoS<sub>2</sub> (figure 3(b)), confirming a heavy doping concentration ( $n_s = 1.5 \times 10^{14} \text{ cm}^{-2}$  from our Hall measurement in figure 2(c)) [25].

### 3.4. Raman thermometry

In addition to the use of Raman spectroscopy to probe the carrier concentration of intercalated Li<sub>*x*</sub>MoS<sub>2</sub>, we also employ a Raman thermometry technique (figure 4(a)) to characterize how the TBC between bilayer MoS<sub>2</sub> and its supporting substrate (SiO<sub>2</sub>/Si) changes as we intercalate and de-intercalate Li ions into the van der Waals gap between MoS<sub>2</sub> layers and between the bottom MoS<sub>2</sub> layer and the SiO<sub>2</sub> substrate. As illustrated in figure 4(a), TBCs between 2D thin films and their supporting substrate play significant roles in limiting the heat and energy dissipation of 2D devices [31–33, 55, 56]. Drive currents in 2D transistors are limited by their local temperatures, which TBCs largely determine [57]. However, measuring the TBC in ultrathin 2D electronics has been challenging due to uncertainties in measuring the heat flow and the local temperature [58, 59].

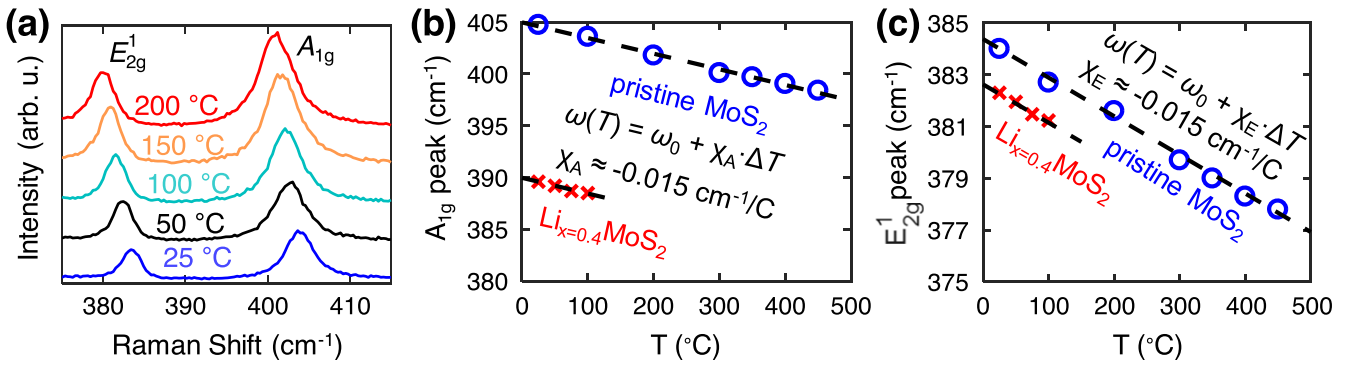
Recently, we reported the TBC of pristine monolayer MoS<sub>2</sub> on SiO<sub>2</sub> via Raman thermometry [31, 33]. In a Raman thermometry measurement, we characterize the temperature rise of 2D thin films optically (through peak shift of the Raman mode) as a function of the input power (either optically from the Raman laser or electrically by Joule heating). We then fit the temperature rise and the input power into a thermal model [60] to calculate the TBC and the in-plane thermal conductivity of the 2D film by solving the heat diffusion equation in our model [33].

For bilayer MoS<sub>2</sub> thin films on SiO<sub>2</sub>/Si substrates and with a spot size  $r_0$  between 310 and 370 nm (without and with electrolyte through a 100× objective, see figure S3 in the supporting information for more details), we expect most of the heat in MoS<sub>2</sub> will be dissipated through the substrate and rendering the system to be insensitive to the in-plane thermal conductivity

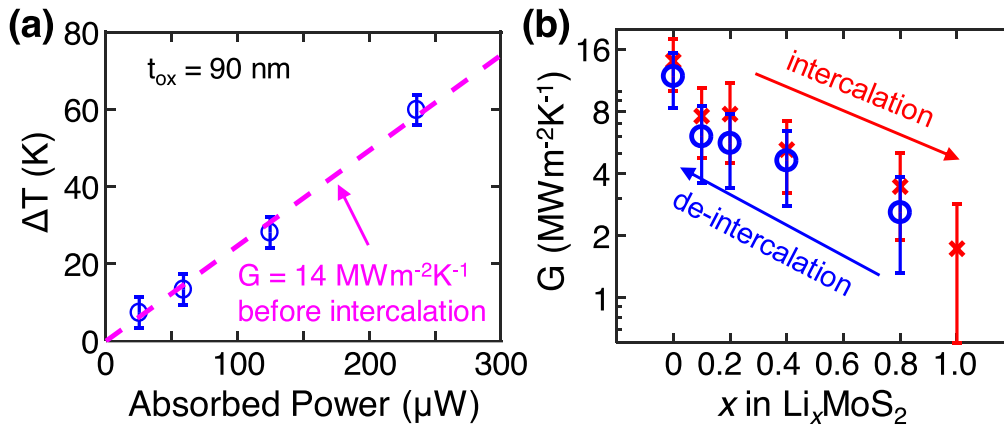
of MoS<sub>2</sub>, as shown in figure 4(b) (see figure S4 in the supporting information for more analysis). This allows us to only use a supported structure (figure 4(a)) to calculate our single fitting parameter, the TBC at the MoS<sub>2</sub>/SiO<sub>2</sub> interface significantly simplifies our measurement and analysis.

A key parameter in Raman thermometry is the power absorbed ( $P_{\text{abs}}$ ) by bilayer MoS<sub>2</sub>. A significant portion of the uncertainty in optically-heated Raman thermometry measurements stems from  $P_{\text{abs}}$  estimations because it is challenging to directly measure the absorbed power of a 2D thin film supported on a substrate where multiple optical reflections interfere. We employed the following approach to minimize those uncertainties. The power absorbed by such a 2D film is calculated as follows:  $P_{\text{abs}} = P_{\text{in}} \alpha_f E$ , where  $P_{\text{in}}$  is the incident laser power,  $\alpha_f$  is the absorption coefficient of free-standing MoS<sub>2</sub>.  $E$  is a wavelength-dependent enhancement factor, the intensity of the electric field (of the electromagnetic wave of the laser) at the top surface of SiO<sub>2</sub>/Si substrate relative to the intensity of the incident electric field, accounting for the multiple reflections within the SiO<sub>2</sub>/Si substrate. We characterized the incident laser power and the spot radius  $r_0$  during our calibration measurements (see figure S3 in the supporting information for more details).

We measured the absorption coefficient of free-standing monolayer MoS<sub>2</sub> over a range of temperatures (see figure S6 in the supporting information). This temperature-dependence of  $\alpha_f$  is important as we observed a 30% increase in absorption (at a laser wavelength of 532 nm) at 250 °C compared to that at room temperature. Neglecting this temperature dependence (and therefore underestimating  $P_{\text{abs}}$ ) will underestimate the TBC in the Raman thermometry analysis. We expect the optical dielectric functions of monolayer and bilayer MoS<sub>2</sub> to be very similar away from the peak energies of those transitions, such as at a wavelength of 532 nm. Thus we assume that the absorption of free-standing



**Figure 5.** Temperature dependence of Raman peaks. (a) Raman spectra of pristine bilayer MoS<sub>2</sub> as a function of temperature. (b)–(c) Temperature calibration by substrate heating for A<sub>1g</sub> and E<sub>2g</sub><sup>1</sup> modes, respectively. Blue and red circles represent calibration for pristine MoS<sub>2</sub> and intercalated Li<sub>0.4</sub>MoS<sub>2</sub>, respectively. The slopes of the linear fits are the temperature coefficients  $\chi_A$  and  $\chi_E$  for A<sub>1g</sub> and E<sub>2g</sub><sup>1</sup> modes, respectively.



**Figure 6.** Thermal boundary conductance (TBC) between MoS<sub>2</sub> and SiO<sub>2</sub>. (a) The measured temperature rise of a pristine bilayer MoS<sub>2</sub> device (no intercalation) versus absorbed laser power. The dashed line represents a thermal resistance with TBC =  $14 \pm 4$  MW m<sup>-2</sup> K<sup>-1</sup>. (b) TBC as a function of Li concentration in MoS<sub>2</sub>. As we intercalate (red crosses) more Li ions into MoS<sub>2</sub>, the TBC decreases to  $\sim 1.8$  MW m<sup>-2</sup> K<sup>-1</sup> at Li<sub>1.0</sub>MoS<sub>2</sub>. As we de-intercalate (blue circles) Li ions out of the system, the TBC almost recovers to its original value ( $\sim 11.8$  MW m<sup>-2</sup> K<sup>-1</sup>).

bilayer MoS<sub>2</sub> at 532 nm to be twice that of the monolayer MoS<sub>2</sub> at 14% at 300 K, with similar temperature dependence as monolayer MoS<sub>2</sub> (see more discussion in supporting information section S6) [33]. We measured the absorption coefficient of lithiated MoS<sub>2</sub> in an earlier study [16], where we observed a 60% decrease in absorption (8.4% for bilayer Li<sub>1.0</sub>MoS<sub>2</sub>) at 532 nm due to the 2H to 1T phase transformation and the associated elimination of a band gap (see figure S6 in the supporting information). We also calculated the enhancement factor ( $\sim 1.46$  at 532 nm) based on the thickness, interface, and refractive index of the materials in our stack (PEO/MoS<sub>2</sub>/SiO<sub>2</sub>/Si). More details on the estimation of the absorbed power can be found in the supporting information section S6 and our prior analysis [33].

Another important parameter in Raman thermometry is the temperature rise  $\Delta T = T - T_0$ , where  $T$  and  $T_0 = 300$  K are the sample and background temperature, respectively. As shown in figure 5(a), we first performed the temperature calibrations using a Linkam heating stage to measure the Raman peak position (A<sub>1g</sub> and E<sub>2g</sub><sup>1</sup> modes for MoS<sub>2</sub>) as a function of the sample temperature (through stage heating). This allows us to fit a linear relationship between the Raman peak position  $\omega$  and the sample temperature  $T$ :  $\omega(T) = \omega_0 + \chi(T - T_0)$ , where  $\omega_0$  is the

peak position at room temperature  $T_0$  and  $\chi$  is the temperature dependence coefficient for the particular Raman mode. For E<sub>2g</sub><sup>1</sup> and A<sub>1g</sub> modes in pristine bilayer MoS<sub>2</sub>,  $\chi_E$  and  $\chi_A$  were fitted to be  $-0.015 \pm 0.002$  cm<sup>-1</sup> °C<sup>-1</sup> and  $-0.015 \pm 0.002$  cm<sup>-1</sup> °C<sup>-1</sup>, as shown in the blue dashed lines in figures 5(b) and (c).

For intercalated Li<sub>x</sub>MoS<sub>2</sub>, the initial peak position and temperature dependence coefficient can differ from that of pristine MoS<sub>2</sub>, as illustrated in figure 3. Thus, we performed a series of temperature calibrations at different Li concentrations,  $x = 0, 0.2, 0.4, 0.8,$  and  $1.0$ , for our Li<sub>x</sub>MoS<sub>2</sub> samples. The calibration and fitted results for Li<sub>0.4</sub>MoS<sub>2</sub> are shown in figures 5(b) and (c), where the temperature dependence coefficients are found to be similar for Li<sub>x</sub>MoS<sub>2</sub> samples (more details on the calibrations can be found in figure S5 in the supporting information). We kept the laser power low (and therefore Li<sub>x</sub>MoS<sub>2</sub> temperature  $< 100$  °C) during our calibration and actual thermometry measurements to avoid unintentional phase transformation and minimize ionic diffusion (and therefore a change in carrier concentration) that has been observed in high-temperature annealing ( $> 300$  °C) of chemically lithiated MoS<sub>2</sub> thin films [18, 61].

As shown in figure 6(a), the TBC at the bilayer MoS<sub>2</sub>/SiO<sub>2</sub> interface before intercalation is  $G = 14 \pm 4.0$  MW m<sup>-2</sup> K<sup>-1</sup>,



consistent with previous reports ( $\sim 15 \text{ MW m}^{-2} \text{ K}^{-1}$ ) for monolayer  $\text{MoS}_2$  with electrical [31] and optical [33] heating. Upon Li intercalation (figure 6(b)), we observe a significant decrease in TBC at the  $\text{MoS}_2/\text{SiO}_2$  interface, to  $G = 1.8 \pm 0.9 \text{ MW m}^{-2} \text{ K}^{-1}$  for  $\text{Li}_{1.0}\text{MoS}_2$ . This large decrease in TBC is likely due to a combination of effects, including an increase in the van der Waals spacing at the  $\text{MoS}_2/\text{SiO}_2$  interface and changes in  $\text{MoS}_2/\text{SiO}_2$  bonding. Upon de-intercalation, the TBC can be mostly recovered to its pre-lithiation value when we removed Li ions from the system ( $G = 11.8 \pm 3.5 \text{ MW m}^{-2} \text{ K}^{-1}$ ). This large and reversible TBC modulation opens interesting opportunities to engineer the heat dissipation for 2D applications such as nanoscale thermal transistors [30] and thermoelectric devices.

#### 4. Conclusion

In summary, we developed a solid-state Li intercalation platform for 2D materials and achieved reversible control of their electrical and thermal properties via electrochemical intercalation. Through Hall measurements, we demonstrated  $\sim 200\times$  tuning of carrier concentration in Li-intercalated bilayer  $\text{MoS}_2$ , ranging from  $0.8 \times 10^{12}$  to  $1.5 \times 10^{14} \text{ cm}^{-2}$ . Combining with an improved contact resistance, almost three orders of magnitude increase ( $500\times$ ) in the drive current of such bilayer  $\text{MoS}_2$  transistors. Raman spectroscopy confirmed the reversible doping effect, recording peak shifts as large as  $18 \text{ cm}^{-1}$  and  $3 \text{ cm}^{-1}$  for  $A_{1g}$  and  $E_{2g}^1$  modes of  $\text{MoS}_2$ , respectively. Using Raman thermometry, we extracted the TBC between  $\text{MoS}_2$  and  $\text{SiO}_2$  to be  $14 \pm 4.0 \text{ MW m}^{-2} \text{ K}^{-1}$  before intercalation, and as low as  $1.8 \pm 0.9 \text{ MW m}^{-2} \text{ K}^{-1}$  after intercalation, due to weakened bonding and phonon scattering. Thus our study suggests that electrochemical intercalation is a powerful technique to engineer and reversibly tune 2D material electrical and thermal properties. Our findings also provide a convenient method to measure the carrier concentration and TBC of 2D materials using Raman spectroscopy, crucial for 2D electronic applications.

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#### Data availability statement

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

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