

# Phase Transition of MoTe<sub>2</sub> Controlled in van der Waals Heterostructure Nanoelectromechanical Systems

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This work reports experimental demonstrations of reversible crystalline phase transition in ultrathin molybdenum ditelluride (MoTe<sub>2</sub>) controlled by thermal and mechanical mechanisms on the van der Waals (vdW) nanoelectromechanical systems (NEMS) platform, with hexagonal boron nitride encapsulated MoTe<sub>2</sub> structure residing on top of graphene layer. Benefiting from very efficient electrothermal heating and straining effects in the suspended vdW heterostructures, MoTe<sub>2</sub> phase transition is triggered by rising temperature and strain level. Raman spectroscopy monitors the MoTe<sub>2</sub> crystalline phase signatures in situ and clearly records reversible phase transitions between hexagonal 2H (semiconducting) and monoclinic 1T' (metallic) phases. Combined with Raman thermometry, precisely measured nanomechanical resonances of the vdW devices enable the determination and monitoring of the strain variations as temperature is being regulated by electrothermal control. These results not only deepen the understanding of MoTe<sub>2</sub> phase transition, but also demonstrate a novel platform for engineering MoTe<sub>2</sub> phase transition and multiphysical devices.

CDs, DVDs, and Blu-ray discs.<sup>[8]</sup> Besides memory applications, the fast switching and high endurance also make phase transition devices important components in neuromorphic computing hardware such as artificial synapses<sup>[9]</sup> and artificial neurons.<sup>[10]</sup> For instance, chalcogenide-based phase transition devices, which undergo phase transitions at nanoseconds time-scale, have engendered integrate-and-fire functions as artificial neurons with both deterministic and stochastic dynamics.<sup>[10]</sup> These have played significant roles and witnessed promising advances in enabling state-of-the-art phase transition devices in modern and emerging technologies.

The advent of atomically thin layered crystals and 2D semiconductors (e.g., transition metal dichalcogenides, i.e., TMDCs)<sup>[11–13]</sup> offers exciting opportunities for novel nanoscale phase transition devices.

## 1. Introduction

Phase transition (or phase change) devices, which operate via exploiting salient differences in electronic and photonic properties between two phases, have been emerging as attractive and exquisite platforms for scalable and energy-efficient memory and information processing applications.<sup>[1–4]</sup> Compared with traditional flash-based memory, phase change memory exhibits remarkable metrics, including superior endurance,<sup>[5]</sup> excellent data retention,<sup>[6]</sup> ultrafast write, and read speed.<sup>[7]</sup> For example, Ge<sub>2</sub>Sb<sub>2</sub>Te<sub>5</sub> (GST) phase-change memory exhibits 10<sup>9</sup> cycles endurance,<sup>[5]</sup> ten-year data retention in 300 to 500 K temperature range,<sup>[6]</sup> and sub-ten nanosecond switching speed.<sup>[7]</sup> These encouraging attainments make GST widely used in

tion devices. Compared with traditional phase transition materials often having one amorphous phase and one crystalline phase, most 2D TMDC materials own two stable crystalline phases: 2H hexagonal phase and 1T' monoclinic phase. In general, 2H phase TMDCs are endowed with bandgaps in their band structures and thus have been widely employed as channel materials for field-effect transistors (FETs). In contrast, 1T' phase TMDCs exhibit metallic band structures without bandgap, thus offering low contact resistances with metals. Among the family of TMDCs, MoTe<sub>2</sub> has the lowest energy barrier between 2H phase and 1T' phase (0.04 eV per MoTe<sub>2</sub> unit)<sup>[14,15]</sup> and thus it is regarded as the most promising candidate for 2D phase transition devices. In experiments, MoTe<sub>2</sub> phase transition has been realized by global heating with local strain applied from atomic force microscope (AFM),<sup>[16]</sup> laser heating,<sup>[17,18]</sup> mechanical strain-induced from ferroelectric substrates,<sup>[19]</sup> and electrostatic doping.<sup>[20]</sup> However, several deficiencies exist in these approaches. For example, the laser irradiation method continues to “sputter” MoTe<sub>2</sub> layer by layer and the remaining MoTe<sub>2</sub> sample permanently stays at 1T' phase. Besides, the electrostatic doping approach inputs energy to 2H MoTe<sub>2</sub> with ionic liquids to realize phase transition, which makes it challenging to be integrated into CMOS for memory and computing applications. Therefore, novel reversible schemes and control of MoTe<sub>2</sub> phase transition on CMOS-friendly platforms are highly desirable and needed.

In this work, we describe the first experimental demonstration of controllable MoTe<sub>2</sub> phase transition directly on free-standing device platforms of van der Waals (vdW) heterostructure

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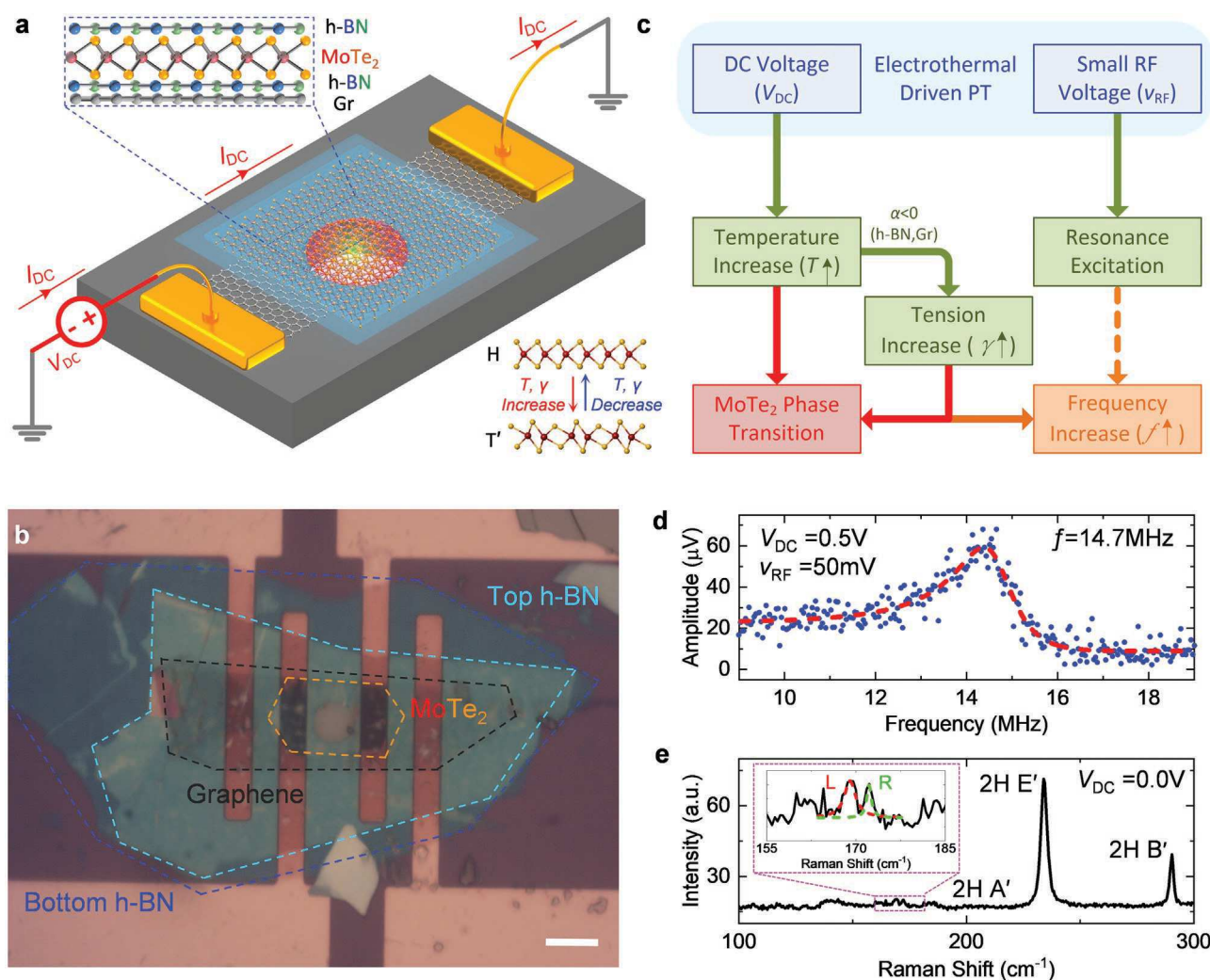
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nanoelectromechanical systems (NEMS). We fabricate suspended graphene/h-BN/MoTe<sub>2</sub>/h-BN (Gr/h-BN/MoTe<sub>2</sub>/h-BN) vdW heterostructures and investigate the phase transition behavior of MoTe<sub>2</sub>. We conveniently use the DC bias voltage ( $V_{DC}$ ) to electrothermally heat up the suspended graphene that serves as a nanoscale hotplate. As the temperature of graphene increases, the heat propagates to the upper layers and the temperature of the MoTe<sub>2</sub> increases. As the temperature rises, the tension level of the whole suspended vdW heterostructure membrane increases due to the negative thermal expansion coefficients (TECs) from both hexagonal boron nitride (h-BN) and graphene. As the applied voltage  $V_{DC}$  gradually increases, we observe clear, controllable, and reversible phase transitions between 2H phase and 1T' phase. After the MoTe<sub>2</sub> fully converts to 1T' phase, we gradually decrease the applied voltage and the MoTe<sub>2</sub> changes back to 2H phase. With solid experiments and comprehensive clarifying results, this work demonstrates a novel

platform for building MoTe<sub>2</sub> phase transition nanoelectronic devices, toward future integration of phase-change random access memory and artificial neurons with tunable NEMS platforms.

## 2. Results and Discussions

Figure 1a illustrates the scheme of Gr/h-BN/MoTe<sub>2</sub>/h-BN (counting from the bottom to top according to stacking during fabrication) vdW heterostructure nanoelectromechanical resonators with electrothermal excitation and tuning. The Gr/h-BN/MoTe<sub>2</sub>/h-BN vdW heterostructure device is fabricated using the all-dry transfer method.<sup>[21]</sup> The graphene flake is first exfoliated on PDMS stamp and transferred onto pre-patterned substrates, followed by the transferring and stacking of h-BN, MoTe<sub>2</sub>, and h-BN, consecutively. The optical microscopy image of the Gr/h-BN/MoTe<sub>2</sub>/h-BN vdW heterostructure resonator is shown in



**Figure 1.** Graphene/h-BN/MoTe<sub>2</sub>/h-BN van der Waals (vdW) heterostructure nanoelectromechanical resonators. a) Illustration of a circular drumhead graphene/h-BN/MoTe<sub>2</sub>/h-BN resonator on a pre-patterned substrate. (upper left inset: cross-section view of the vdW heterostructure; bottom right inset: crystal structure of MoTe<sub>2</sub>). b) Optical microscopy image of a graphene/h-BN/MoTe<sub>2</sub>/h-BN vdW heterostructure nanoelectromechanical resonator. c) Analysis of coupling effects and signal transduction mechanisms of the vdW heterostructure resonators under Joule heating, for controlling the phase transition (PT). d) Fundamental-mode resonance of the vdW heterostructure in (b). e) MoTe<sub>2</sub> Raman signatures measured from the suspended region in (b) (inset: zoom-in figure of 2H peak A'). Scale bar: 10  $\mu\text{m}$ .

Figure 1b. The few-layer graphene flake (black dashed lines) is first transferred onto the substrate, followed by another few-layer h-BN flake (outer blue dashed lines) stacking on top of the graphene. The bottom h-BN layer on top of graphene is used to provide electrical isolation for MoTe<sub>2</sub> from graphene. A few-layer MoTe<sub>2</sub> flake (orange dashed lines) is then transferred on top of the bottom h-BN. Finally, another few-layer h-BN (inner blue dashed lines) is stacked to cover the MoTe<sub>2</sub> to avoid environmental degradation and MoTe<sub>2</sub> sublimation at high temperatures.<sup>[16]</sup> All the devices are fabricated on a pre-patterned substrate with a circular drumhead micro-cavity (diameter ≈74 μm and a trench depth 2200 nm). After the device fabrication, the suspended vdW heterostructure is electrothermally heated up by a DC current from drain to source. In the meantime, a small RF voltage ( $V_{RF}$ ) is applied to excite the resonance. The small RF voltage gives rise to a small temperature vibration periodically, which generates a periodical thermal force on the vdW heterostructure. As the DC voltage ( $V_{DC}$ ) increases, several effects are coupled together to trigger the phase transition (Figure 1c). First, due to the electrothermal effect, the heat propagates to the upper layers and thus the temperature of MoTe<sub>2</sub> increases. Second, due to the negative TECs of graphene and h-BN, the tension level of the whole suspended structure rises. To probe the temperature and strain level changes, Raman shifts and mechanical resonances are measured using the combined Raman and laser interferometry system (Supporting Information S1). Such a vdW heterostructure device shows clear resonances in moderate vacuum, with a fundamental-mode frequency  $f \approx 14.7$  MHz (Figure 1d), demonstrating the heterostructure membrane is clearly suspended and vibrating. We also perform Raman measurements to characterize the device. From the Raman measurement results, characteristic peaks of MoTe<sub>2</sub> (Figure 1e),<sup>[22]</sup> graphene,<sup>[23]</sup> and h-BN<sup>[24]</sup> (Supporting Information S2) are observed. In particular, the phase of the MoTe<sub>2</sub> is confirmed as 2H after fabrication. It should be noted that the 2H A' peak splits into two subpeaks, which is a unique Raman characteristic for few-layer 2H MoTe<sub>2</sub> samples.<sup>[22]</sup>

We then investigate the phase transition of sandwiched MoTe<sub>2</sub> sample by gradually increasing  $V_{DC}$ . Figure 2 shows the Raman evolution of an 8-layer MoTe<sub>2</sub>-based vdW device with voltage sweeping. For pristine MoTe<sub>2</sub> sample before Joule heating ( $V_{DC} = 0.0$  V, Figure 2a), 2H E' peak and 2H B' peak are dominant, while in contrast, the 2H A' peak is almost invisible. When the  $V_{DC}$  increases to 5.0 V, although the intensity of 2H E', 2H B', and 2H A'(L) peaks decrease, the 2H A'(R) peak becomes stronger (Supporting Information S4). Due to the anharmonic effect, the Stokes Raman peak intensity usually degrades with temperature elevation.<sup>[25]</sup> Therefore, the unusual intensity enhancement of 2H A'(R) suggests the appearance of new phase (1T'). Based on previously reported reversible MoTe<sub>2</sub> phase transition, the peak positions of 2H A' mode (170–171 cm<sup>-1</sup>) and 1T' A<sub>g</sub> (167–168 cm<sup>-1</sup>) are very close,<sup>[20,26]</sup> and the redshift of 2H A' peak results in its overlap with 1T' A<sub>g</sub> peak. When the voltage approaches 6.9 V, the 2H A'(L) disappears, and thus starting at this point, the peak around 165 cm<sup>-1</sup> could be regarded as a pure 1T' A<sub>g</sub> peak. With voltage further increasing, the intensity of 1T' A<sub>g</sub> mode continuously grows while other 2H peaks degrade (2H E' mode)

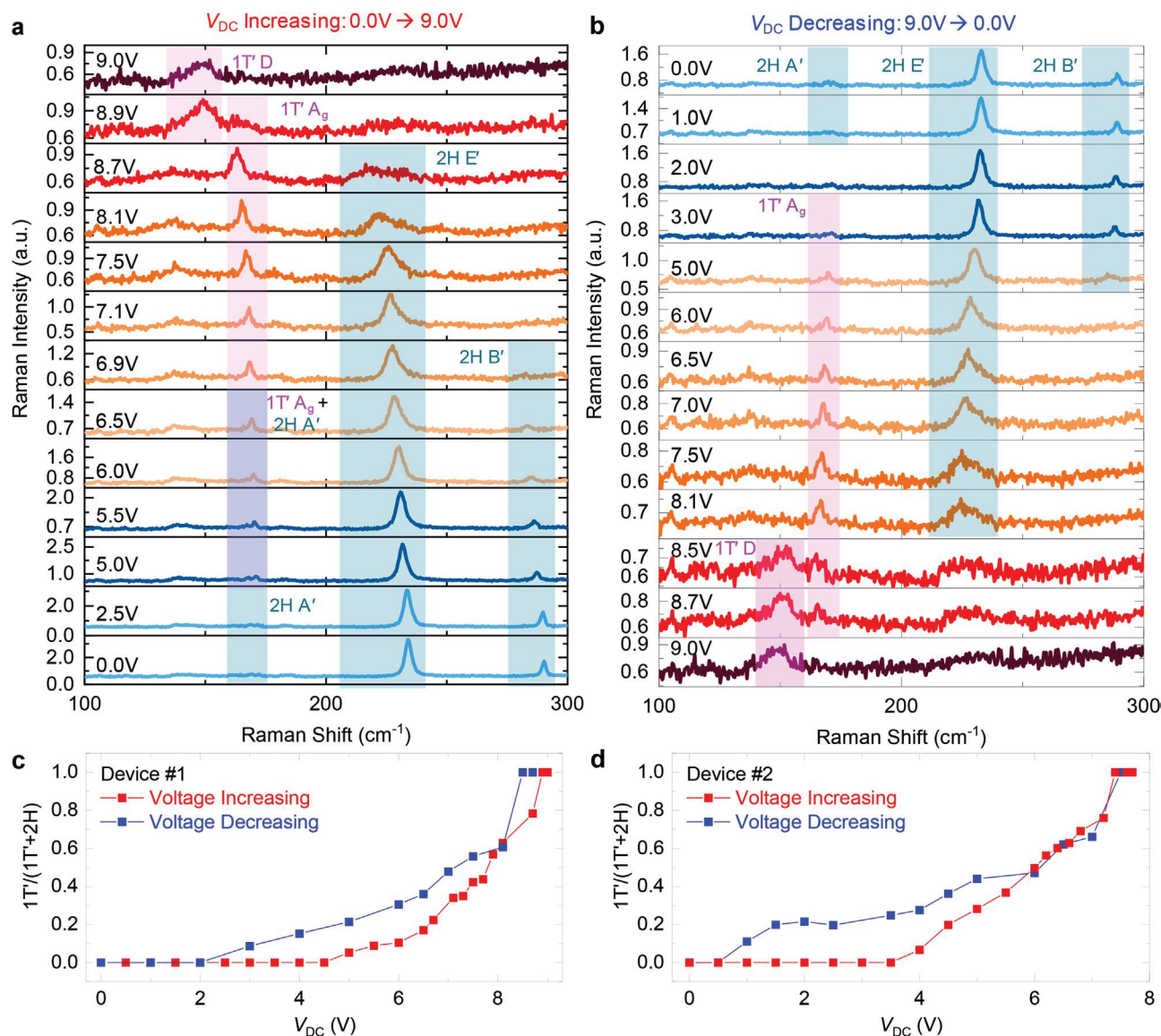
even vanish (2H B' and 2H A' modes). These observations clearly demonstrate the 2H→1T' phase transition. At 8.9 V, all 2H peaks disappear, indicating that the sample has completely transformed to 1T' phase. It should be noted that another new peak, 1T' D peak, appears at this point ( $V_{DC} = 8.9$  V), with even higher intensity than the 1T' A<sub>g</sub> peak. The appearance of 1T' D peak is related to the defect generation at high temperature.<sup>[27]</sup> With bias voltage increased to  $V_{DC} = 9.0$  V, only 1T' D peak remains, and A<sub>g</sub> peak is invisible because of the strong anharmonic effect.

After the MoTe<sub>2</sub> has completely transformed to 1T' phase, we gradually reduce the DC voltage (Figure 2b). Because device cooling is much slower than heating in vacuum, we always wait for 5 to 10 mins till the Raman signal is stabilized before further reducing the voltage. With the voltage decreased to 8.1 V, 1T' A<sub>g</sub> peak and 2H E' peak appear, indicating the initialization of the reverse 1T'→2H phase transition. When the voltage is reduced to 7.0 V, both 1T' A<sub>g</sub> peak and 2H E' peak become stronger compared with those at  $V_{DC} = 8.1$  V, with, however, different reasons. The intensity increment of 1T' A<sub>g</sub> peak only results from decreasing temperature and thus reducing anharmonic effect. In contrast, the enhancement of 2H peak intensity is attributed to both reverse 1T'→2H phase transition and reducing anharmonic effect. This conclusion could be further supported by the trend when voltage drops from  $V_{DC} = 7.0$  to 2.0 V. Due to the reversible phase transition, 1T' A<sub>g</sub> peak disappears completely while the 2H E' peak becomes dominant. The vanishing of 1T' peak shows the completion of reversible phase transition. When  $V_{DC}$  reduces to 0.0 V, as all the 2H peaks reappear and no 1T' peak exists, MoTe<sub>2</sub> converts back to the original 2H pristine state completely, clearly demonstrating the MoTe<sub>2</sub> phase transition is a controllable and reversible process. To see the phase transition clearly, we extract and plot Raman peak intensity with voltage variation. The peak intensity ratio of 1T' A<sub>g</sub> peak over the sum of 1T' A<sub>g</sub> peak and 2H E' peak is used to characterize the 1T' ratio of the MoTe<sub>2</sub> sample during phase transition (Figure 2c,d). The approach to determining 1T' peak intensity is described in Supporting Information S5.<sup>[20]</sup> It is noticed that there is a small phase transition hysteresis between the voltage increasing and decreasing branches, which might be attributed to the temperature variation hysteresis, that is, under the same voltage, there are still temperature differences between the voltage increasing and decreasing processes. Figure 2d shows the 1T' ratio with voltage sweeping in a 14-layer MoTe<sub>2</sub>-based vdW device (Device #2). The two devices exhibit a similar phase transition trend (the 1T' ratios in these devices are determined from data in Supporting Information S3–S5).

Besides Raman peak intensity variation, another phenomenon during voltage sweeping is the Raman peak shift, that is, all Raman peaks redshift to the lower frequency regime and blueshift to higher frequency with temperature increases and decreases, respectively. The Raman peak position variation allows us to estimate the device temperature. The MoTe<sub>2</sub> and graphene temperature are calculated using the equation

$$\omega_T = \omega_{300K} + \chi(T - 300K) \quad (1)$$

where  $T$  is temperature,  $\omega_T$  is Raman peak position at temperature  $T$ ,  $\omega_{300K}$  is Raman peak position at room temperature (300K), and  $\chi$  is first-order temperature coefficient



**Figure 2.** Evolution of Raman spectra measured during reversible phase transition in MoTe<sub>2</sub> vdW heterostructure devices. Evolution of MoTe<sub>2</sub> Raman spectra with voltage  $V_{DC}$  a) increasing and b) decreasing in an 8-layer MoTe<sub>2</sub>-based vdW device. 1T' phase ratio of MoTe<sub>2</sub> with the voltage sweeping in c) Device #1: an 8-layer MoTe<sub>2</sub>-based vdW device and d) Device #2: a 14-layer MoTe<sub>2</sub>-based vdW device.

( $-0.015 \text{ cm}^{-1} \text{ K}^{-1}$  for graphene G peak<sup>[28]</sup> and  $-0.0164 \text{ cm}^{-1} \text{ K}^{-1}$  for MoTe<sub>2</sub> 2H E' peak<sup>[29]</sup>). Because of the disappearance of the 2H peak E' after MoTe<sub>2</sub> transitioning to 1T', the 1T' A<sub>g</sub> peak position shift is used to calibrate temperature of the phase transition completion point (Supporting Information S6). **Figure 3** shows the measured lattice temperature of graphene and MoTe<sub>2</sub> at the device center region (Raman measurement spot) for two devices during reversible phase transition, tuned by applied bias voltage  $V_{DC}$ . For the 8-layer MoTe<sub>2</sub> device (Device #1, Figure 3a,b), the 2H→1T' phase transition starts at 440 K ( $V_{DC} = 5.0 \text{ V}$ ). Above this point, two phases coexist together and 1T' phase ratio keeps increasing. At around 1180 K ( $V_{DC} = 8.9 \text{ V}$ ), the MoTe<sub>2</sub> sample is fully converted to 1T' phase. Because the phase transition completion temperature is very close to previously reported values (1200 K, purple dashed line in Figure 3<sup>[16,30]</sup>), tension does not play a crucial role in phase

transition and phase transition is mainly driven by temperature change in Device #1. The temperature of graphene is 2000 K when  $V_{DC} = 8.9 \text{ V}$ . This large temperature difference between graphene and MoTe<sub>2</sub> at this point could be explained by the significantly different thermal conductivity between graphene (5000 W/(m·K)<sup>[31]</sup>) and MoTe<sub>2</sub> (42 W/(m·K)<sup>[32]</sup>), and different interfacial thermal conductance between graphene/h-BN (52.2 MW/(m<sup>2</sup>·K)<sup>[33]</sup>) and MoTe<sub>2</sub>/h-BN (170 MW/(m<sup>2</sup>·K)<sup>[33]</sup>). Considering that MoTe<sub>2</sub> and MoS<sub>2</sub> have very similar crystal structure, it is reasonable to speculate that interfacial thermal conductances at MoS<sub>2</sub>/h-BN and MoTe<sub>2</sub>/h-BN are similar. After the device has fully transformed into 1T' phase, we reduce the applied voltage (Figure 3b). When the temperature is reduced to 925 K ( $V_{DC} = 8.1 \text{ V}$ ), the MoTe<sub>2</sub> flake starts to transition to 2H phase and this reversible phase transition process completes at 386 K ( $V_{DC} = 2.0 \text{ V}$ ). The phase transition of a 14-layer

MoTe<sub>2</sub> device (Device #2) is described in Figure 3c,d. The 2H to 1T' phase transition process starts at 455 K and completes at 1050 K. Although two devices exhibit similar phase transition initialization temperature, the completion temperature in Device #2 (1050 K) is noticeably lower than the one in Device #1 (1180 K), suggesting that the other energy input (should be tension in this scenario) plays an important role in the phase transition in Device #2. The reversible phase transition (1T' to 2H) starts at 810 K and completes at around 460 K.

In order to estimate the possible impact from the strain effect induced by Joule heating, we measure the resonance frequency under different V<sub>DC</sub>, which allows us to extract the average thermal strain experienced by the heterostructure. The resonance frequency of the heterostructure can be written as follows

$$f = \frac{(k_2^{mn} na)}{2\pi} \sqrt{\frac{D}{\rho_{\text{Hetero}} \zeta a^4} \left[ \frac{(\gamma_0 + \gamma_{\text{th}}) a^2}{D} + (k_2^{mn} a)^2 \right]} \quad (2)$$

where  $D = \frac{E_{Y,\text{Hetero}} t^3}{12(1-\nu^2)}$  is the flexural rigidity,  $\zeta$  is coefficient of adsorbed mass,  $t$  is the total thickness of the heterostructure,  $\gamma_0$  is the initial tension, and  $a$  is the radius of the microtrench.<sup>[34]</sup>  $E_{Y,\text{Hetero}}$  and  $\rho_{\text{Hetero}}$  are effective Young's modulus and density of the heterostructure, which can be calculated as below

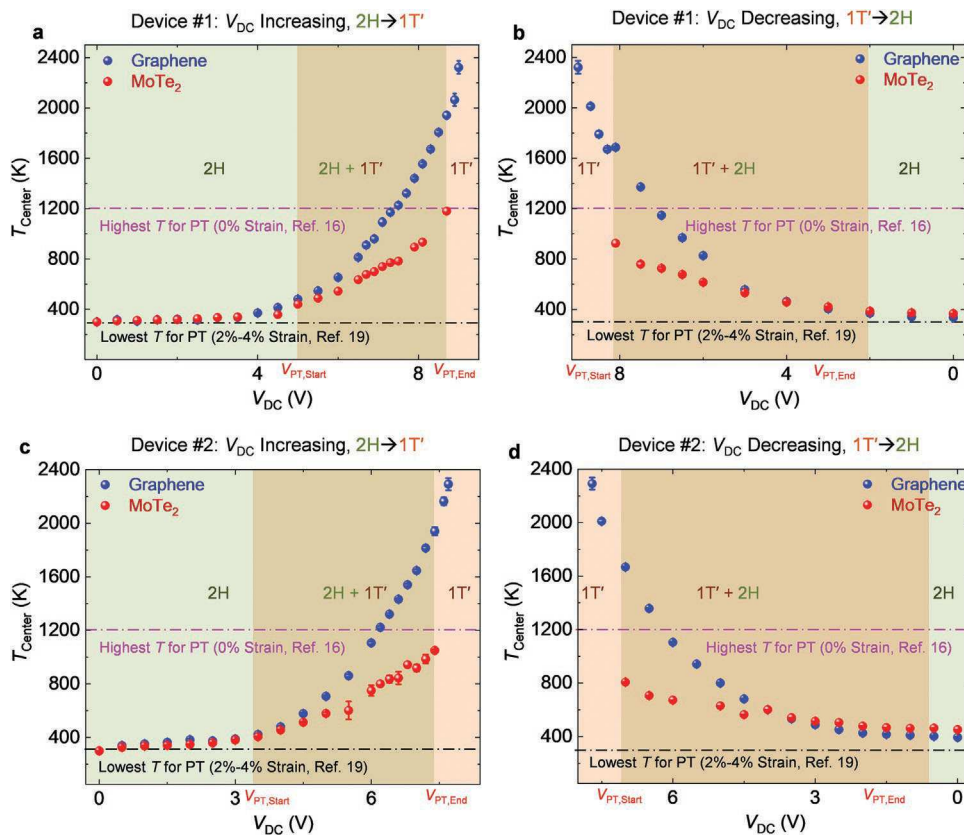
$$E_{Y,\text{Hetero}} = \frac{E_{Y,\text{Gr}} t_{\text{Gr}} + E_{Y,\text{BN}} (t_{\text{BN,bottom}} + t_{\text{BN,top}}) + E_{Y,\text{MoTe}_2} t_{\text{MoTe}_2}}{t_{\text{Gr}} + t_{\text{BN,bottom}} + t_{\text{BN,top}} + t_{\text{MoTe}_2}} \quad (3)$$

$$\rho_{\text{Hetero}} t_{\text{Hetero}} = \rho_{\text{Gr}} t_{\text{Gr}} + \rho_{\text{BN}} (t_{\text{BN,bottom}} + t_{\text{BN,top}}) + \rho_{\text{MoTe}_2} t_{\text{MoTe}_2} \quad (4)$$

Electrothermally induced tension ( $\gamma_{\text{th}}$ ) can be obtained from measured frequency by using Equation 2. After obtaining electrothermally induced tension, we calculate the average thermal strain ( $\epsilon_{\text{th,avg}}$ ) experienced by the suspended heterostructure using the following equation

$$\epsilon_{\text{th,avg}} = \frac{\gamma_{\text{th}}}{E_{Y,\text{Hetero}} t} \quad (5)$$

Figure 4a shows the measured resonance frequency  $f_{\text{res}}$  under increasing voltage for the 8-layer MoTe<sub>2</sub>-based vdW device. With voltage V<sub>DC</sub> increasing, due to the tension increase upon heating from the negative TECs of graphene and h-BN, the frequency of the device increases. It can be seen that in the low voltage regime (V<sub>DC</sub> = 0 to 4 V), because the temperature does not increase significantly in this regime, the frequency is almost invariant with the applied voltage. From V<sub>DC</sub> = 3.5 to 8.0 V, the frequency rises with applied voltage, which could be attributed to the significant temperature elevation (corresponding to MoTe<sub>2</sub> temperature increasing from 400 to 900 K).

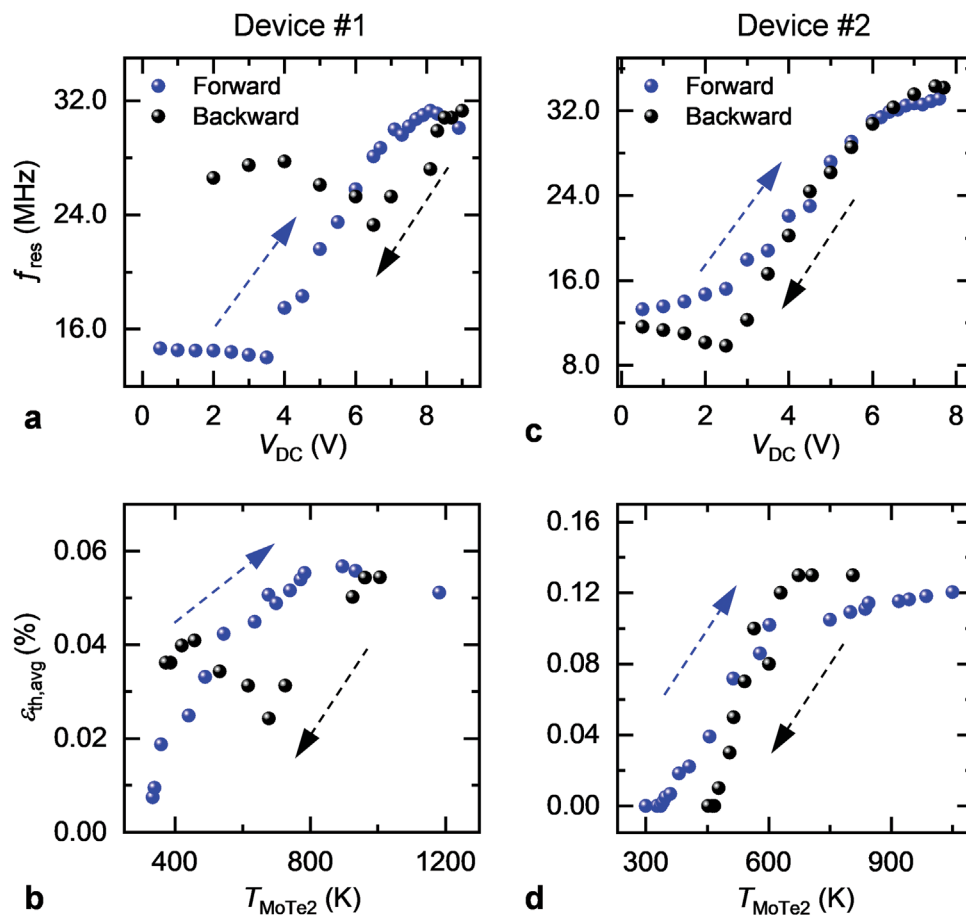


**Figure 3.** Temperature variation of MoTe<sub>2</sub> and graphene measured during reversible phase transition. Temperature a) increasing and b) decreasing of Device #1 (8-layer MoTe<sub>2</sub> device) with the DC voltage increasing and decreasing, respectively. Temperature c) increasing and d) decreasing of Device #2 (14-layer MoTe<sub>2</sub> device) with the DC voltage increasing and decreasing, respectively. The green, orange, and overlapped green/orange regions represent 2H phase, 1T' phase, and 2H+1T' mixed phase, respectively.

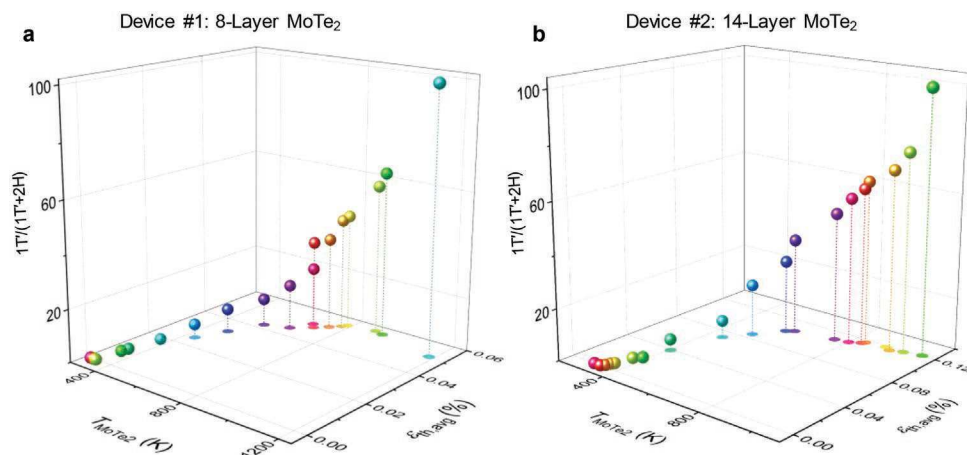
Beyond  $V_{DC} = 8.0$  V, because of the transition from negative TECs to positive TECs of graphene and h-BN at high temperature ( $>1600$  K),<sup>[35]</sup> the frequency decreases slowly with applied voltage. In the reversed (backward) phase transition process of Device #1, with the voltage decreasing from  $V_{DC} = 9.0$  to 2.0 V, the frequency variation shows three regimes. In the first regime (from  $V_{DC} = 9.0$  to 6.5 V), because of significant temperature lowering, the frequency decreases with applied voltage. In the second regime ( $V_{DC} = 6.5$  to 4.0 V), the frequency shows an unusual gentle rise with voltage decreasing. One possible reason for this variation might be the formation and movement of tiny bubbles at the interfaces in the heterostructure (Supporting Information S8). In the third regime ( $V_{DC} = 4.0$  to 2.0 V), the temperature effect becomes dominant again and frequency reduces with voltage decreasing. Utilizing the frequency variation results, we further extract the corresponding  $\epsilon_{th,avg}$ , shown in Figure 4b, and we obtain that at the phase transition (2H to 1T') completion point the thermal strain  $\epsilon_{th,avg} \approx 0.055\%$  for this Device #1. We notice that there is a tension difference (frequency difference) between the final state and initial state for Device #1. This difference could be attributed to the annealing effects or morphology change (wrinkles) at the interfaces in the device. Device #2 (Figure 4c,d) exhibits overall similar frequency variation trends as those in

Device #1, but with two differences. First, the tension level of Device #2 at phase transition (2H to 1T') completion point is  $\epsilon_{th,avg} \approx 0.15\%$ , much higher than that in Device #1, which is consistent with the fact that the phase transition temperature of Device #2 is much lower than that of Device #1. Second, the frequency and tension level of the initial state are close to those of the final state, demonstrating that there is no measurable morphology change (wrinkles) during the heating and cooling process.

With the temperature calibration and strain estimation, we plot the 1T' ratio in 2H to 1T' phase transition process versus coupled effects of MoTe<sub>2</sub> lattice temperature and thermal strain (Figure 5). In Device #1 (Figure 5a), the phase transition begins at  $T_{MoTe_2} = 440$  K with a strain level around  $\epsilon_{th,avg} \approx 0.01\%$ . As the voltage  $V_{DC}$  increases, the temperature and thus thermal strain level elevates; consequently, the 1T' phase ratio becomes increasingly dominant. When the temperature approaches  $T_{MoTe_2} = 1180$  K and a strain level around 0.055%, the MoTe<sub>2</sub> flake has completely transitioned to 1T' phase. In Device #2 (Figure 5b), the 2H to 1T' phase transition starts at  $T_{MoTe_2} = 455$  K, with tension level around  $\epsilon_{th,avg} \approx 0.039\%$ ; and it completes at 1050 K with tension around 0.12%. The lower phase transition completion temperature in Device #2 is attributed to the relatively higher thermal tension induced by Joule heating.



**Figure 4.** Frequency and strain evolutions measured for two MoTe<sub>2</sub>-based vdW devices with varying voltage and temperature during the reversible phase transition. Frequency evolution of a) Device #1: an 8-layer MoTe<sub>2</sub> device and b) Device #2: a 14-layer MoTe<sub>2</sub> device with varying applied voltage. Thermal strain of c) Device #1 and d) Device #2 with varying temperature.



**Figure 5.** Coupled effects of varying temperature and strain in the 2H to 1T' phase transition process in a) Device #1: an 8-layer MoTe<sub>2</sub>-based vdW device and b) Device #2: a 14-layer MoTe<sub>2</sub>-based vdW device.

### 3. Conclusions

In summary, we have demonstrated reversible and controllable MoTe<sub>2</sub> crystalline phase transition via electrothermal-mechanical tuning on a vdW heterostructure resonant nanoelectromechanical systems (NEMS) platform. By electrothermally heating the graphene nano-hotplate, the temperature of each layer in the vdW heterostructure and thus the thermal strain elevates. From measured Raman shift, we observe MoTe<sub>2</sub> transitions from 2H phase to 1T' phase with rising temperature and strain level. In reverse, with voltage decreasing and thus temperature plus tension reducing, the 1T' MoTe<sub>2</sub> converts back to 2H phase. The coexistence of 2H phase and 1T' phase suggests that the MoTe<sub>2</sub> phase transition may be a gradual process. By monitoring the tension level from the resonance frequency, we find that tension could significantly lower the MoTe<sub>2</sub> phase transition completion temperature. This work demonstrates, for the first time, a novel platform for reversible MoTe<sub>2</sub> phase transition, with only two-terminal inputs needed, manufacturable on regular SiO<sub>2</sub>-on-Si substrates. From an engineering perspective, this paves the way toward further integrating reversible MoTe<sub>2</sub> phase transition devices onto CMOS as it requires no other special substrates or additional components. This work opens a new avenue for designing and building phase transition MoTe<sub>2</sub> multiphysical devices that can capitalize on NEMS-enabled tuning and control, for memory and neuromorphic computing applications.

### 4. Experimental Section

**Fabrication of Suspended Graphene/h-BN/MoTe<sub>2</sub>/h-BN van der Waals Heterostructure:** The bottom graphene flakes are first exfoliated from graphite crystals on PDMS and then transferred onto the pre-patterned microcavities on SiO<sub>2</sub>-on-Si substrates using the all-dry transfer method.<sup>[21]</sup> Following the 1<sup>st</sup> transfer, bottom h-BN layer, MoTe<sub>2</sub> layer, and top h-BN layer are transferred on top subsequently, one by one. The bottom h-BN is employed to isolate graphene and MoTe<sub>2</sub>, and the top h-BN is used to encapsulate MoTe<sub>2</sub> and prevent it from degradation. The diameter of the circular microtrench or microcavity is typically around 10 μm with a depth around 2.2 μm.

**Raman Scattering Measurement:** The graphene/h-BN/MoTe<sub>2</sub>/h-BN vdW heterostructure devices are preserved in a vacuum chamber and measured using a custom-built micro-Raman spectrometer. The Raman spectrometer is integrated into an optical interferometric resonance measurement system (Figure S1, Supporting Information). In the Raman measurement, a 532 nm laser is focused upon the center of the vdW heterostructure. The spot size of laser is around ≈1 μm and the power of the 532 nm laser is lower than 200 μW. The electrical power applied to heat up graphene is 6 mW when graphene reaches 1200 K. The laser power is significantly lower than the electrical power applied.<sup>[36]</sup> Raman scattered light from the device is collected in backscattering geometry and then guided to the spectrometer (Horiba iHR550) with a 2400 g mm<sup>-1</sup> grating. Raman signal is recorded using a liquid-nitrogen-cooled CCD.

**Interferometric Resonance Measurement:** The resonance characteristics of the vdW heterostructure devices are measured by an ultrasensitive optical interferometry system that we have built and meticulously engineered for systematically investigating 2D NEMS made of various 2D materials and their vdW heterostructures. The resonance motion is electrothermally excited by applying an RF signal  $\nu_{RF}$  coupled with a strong DC signal to drive the vdW heterostructure membrane. The resonance motion is detected by employing the same 532 nm green laser with power below 300 μW to avoid laser heating,<sup>[36]</sup> and the resonant motion-modulated optical interferometry signal is fed to a photodetector. A network analyzer is used to provide the sweeping RF drive signal, and to measure the resonance signal transduced by the photodetector.

### Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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## Conflict of Interest

The authors declare no conflict of interest.

## Data Availability Statement

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

## Keywords

2D materials, nanoelectromechanical systems, phase transition, van der Waal heterostructures

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