

Charge-Density Wave Driven Giant Thermionic-Current Switching in 1T-TaS₂/2H-TaSe₂/2H-MoS₂ Heterostructure

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1T-TaS₂ exhibits several resistivity phases due to the modulation of charge density wave (CDW). The fact that such phase transition can be driven electrically has attracted a lot of attention in the recent past toward *active-metal* based electronics. However, the bias-driven resistivity switching is not very large (less than five-fold), and an enhancement in the same will highly impact such phase transition devices. One aspect that is often overlooked is that such phase transition is also accompanied by a significant change in the local temperature due to the low thermal conductivity of 1T-TaS₂. In this work, such electrically driven phase transition induced temperature change is exploited to promote carriers over a thermionic barrier in a 1T-TaS₂/2H-TaSe₂/2H-MoS₂ T-junction, achieving a 964-fold abrupt switching in the current through the MoS₂ channel. The device is highly reconfigurable and exhibits an abrupt reduction in current as well when the biasing configuration changes. The results are promising for several electronic applications, including neuromorphic chips, switching, nonlinear devices, and industrial electronics such as current and temperature sensing.

by electrical driving,^[12–14] has led to several electronic and optoelectronic device applications.^[10,15–17] The resistance switching ratio plays an important role in determining the performance of these devices. However, the degree of the resistance switching is often weak, less than five,^[12,13,16] depending on the flake thickness, measurement temperature, and crystal quality. Any technique that can enhance the ratio of the resistance switching will be of great importance in such phase transition-based device applications. In this work, we propose a method using a 1T-TaS₂/2H-TaSe₂/2H-MoS₂ T-junction, where we achieve a gate-controllable current switching ratio up to 964 driven by CDW phase change in 1T-TaS₂ – which is more than 190-fold enhancement in the switching ratio compared to existing reports. The principle of operation of the device exploits the abrupt

1. Introduction

The charge density wave (CDW) driven phase transition in layered materials like TaS₂, TaSe₂, NbSe₂, and TiSe₂ has attracted a lot of attention in the recent past due to their structural, thermal, magnetic, electrical, and optical properties.^[1–5] Out of all these, 1T-TaS₂ exhibits polymorphic states with distinctive resistive phases, that can be achieved by thermal,^[6,7] mechanical,^[8,9] optical,^[10,11] and electrical^[12,13] excitation. On heating, 1T-TaS₂ transits from commensurate (C) to triclinic (T) phase at 223 K, T to non-commensurate (NC) phase at 283 K, NC to incommensurate (IC) phase at 353 K and IC to metallic phase at 550 K. On the other hand, 2H-TaSe₂ that undergoes C to NC phase transition at 90 K and NC to metallic phase at 120 K shows a slight change in slope of resistance versus temperature curve. Unlike 1T-TaS₂, 2H-TaSe₂ does not exhibit a sharp change in the resistance during the phase transitions.

The fact that in 1T-TaS₂, the resistance switching through such phase transitions can be obtained through Joule heating

change in the junction temperature due to an abrupt change in the current resulting from Joule heating induced CDW phase transition in TaS₂. This, in turn, helps to promote carriers over a thermionic barrier, modulating the net probe current. Interestingly, the proposed device is electrically reconfigurable between a positive jump (enhancement in current) and a negative jump (reduction in current). Such reduction in current can be modeled as an effective negative differential resistance (NDR), as shown earlier.^[17]

2. Results and Discussion

The 3D schematic diagram of the proposed triple layered T-junction is shown in **Figure 1a**. The inset of **Figure 1b** shows the optical image of the device D_a fabricated by exfoliating a few layer MoS₂ flake on Si/SiO₂ substrate followed by layer-by-layer dry transfer of TaSe₂ and TaS₂ flakes, respectively (see Experimental Section for complete fabrication steps). The current (*I*)–voltage (*V*) characteristics of the TaS₂/TaSe₂ junction of the device D_a probed between terminals T and T_S (with terminal M kept open) at 296 K are shown in **Figure 1b**. The abrupt hysteretic jump in the current (≈ 1.24) results from the reduction in the TaS₂ resistance owing to electrically driven NC–IC CDW phase transition. Note that TaSe₂ being highly conductive, carries ample current to drive NC–IC phase transition of TaS₂ in TaS₂/TaSe₂ junction.^[3] Also, the low thermal conductivity of TaSe₂ compared to Au contact further aids in raising the local

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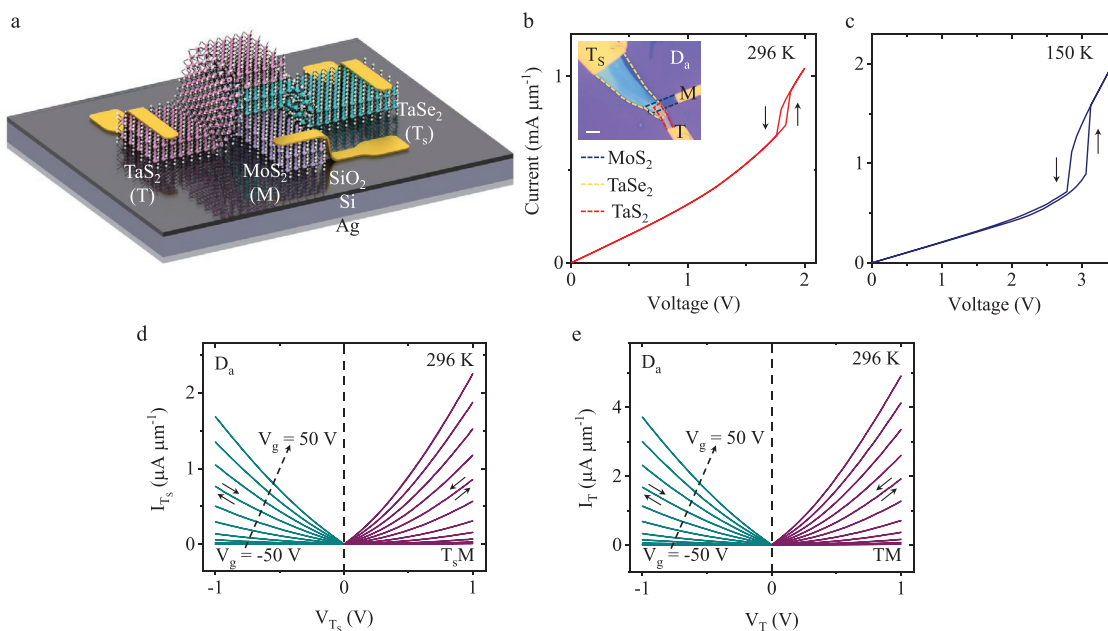


Figure 1. Electrical characterization of individual materials and heterojunctions of the T-junction. a) Schematic representation of triple layered T-junction. The Ag contact is used as a global back gate. b) Current–voltage characteristics of two-probe TaS₂/TaSe₂ junction (probed between terminals T_S and T of D_a) indicating the NC-IC phase transition of TaS₂ at 296 K. Inset: the optical image delineating each layer of the fabricated triple layered T-junction (D_a) (scale bar: 5 μm). c) Current–voltage characteristics of TaS₂/TaSe₂ junction at 150 K. Forward and reverse sweeps are indicated by black arrows. d) *I*–*V* characteristics of TaSe₂/MoS₂ junction, probed between terminals T_S and M keeping terminal T open, for V_g varying from –50 to 50 V in steps of 10 V at 296 K. e) *I*–*V* characteristics of TaS₂/MoS₂ junction, probed between terminals T and M keeping terminal T_S open, as a function of V_g varying from –50 to 50 V in steps of 10 V at 296 K.

temperature of the channel.^[18,19] The current jump can be further enhanced at low temperatures by invoking phase transition through multiple metastable states of TaS₂ using the external electric field.^[10,16] Figure 1c depicts the *I*–*V* characteristics of the TaS₂/TaSe₂ junction of the device D_a at 150 K showing a current jump of ≈1.77. Note that when a material with lower conductivity (such as SnSe₂) replaces the series resistance material (i.e., TaSe₂), the overall current is suppressed, which reduces the Joule heating. Hence, the phase transition in TaS₂ does not occur anymore (Figure S1, Supporting Information).

The current–voltage characteristics of TaSe₂/MoS₂ junction (probed between terminals T_S and M keeping terminal T open) and TaS₂/MoS₂ junction (probed between terminals T and M keeping terminal T_S open) as a function of back-gate voltage (V_g) varying from –50 to 50 V in steps of 10 V at 296 K are outlined in Figure 1d,e, respectively. The forward and reverse sweep directions of the voltage during measurement (indicated by the black arrows in Figure 1d,e) show negligible hysteresis, suggesting excellent interface quality in the device. The corresponding gate-dependent current–voltage characteristics of both the junctions at 150 K are depicted in Figure S2a,b, Supporting Information, respectively.

2.1. Reconfigurable Abrupt Current Switching in T-junction

We now show the characteristics of 1T-TaS₂/2H-TaSe₂/2H-MoS₂ T-junction that is reconfigurable between abrupt current increment and decrement by utilizing the modulation of the MoS₂

resistance through an external gate voltage. In this device, we pass a high current through a voltage bias through the TaS₂/TaSe₂ path and use the branched out MoS₂ channel current as a probe. The equivalent circuit diagram of the four terminal device D_a is shown in Figure 2a wherein R₁, R₂, and R₃ correspond to the effective resistance of TaSe₂, TaS₂, and MoS₂ flakes, respectively. Here, R₃ includes both the TaSe₂/MoS₂ Schottky junction resistance and the MoS₂ channel resistance, and both the components are tunable by the gate voltage. A global back gate is connected to the fourth terminal of the device. Figure 2b shows the variation of MoS₂ channel current (I_M) with V_{T_s} of the device D_a when we apply the bias at T_S terminal while keeping T and M terminals grounded. The characteristics show current decrement behavior for both V_{T_s} < 0 V and V_{T_s} > 0 V for V_g varying from –50 to 50 V in steps of 10 V at 296 K. When the external bias exceeds the threshold voltage for the NC-IC phase transition of TaS₂, the TaS₂ resistance abruptly reduces resulting in the abrupt increment in the TaS₂ current, and hence a simultaneous reduction in the MoS₂ current. The peak-to-valley current ratio (PVCR) obtained for V_{T_s} < 0 V and V_{T_s} > 0 V at 296 K are shown in bottom panel of Figure 2g. A simplistic way to further improve the PVCR values is by increasing the switching ratio of the TaS₂ resistance, which directly regulates the abrupt change in the MoS₂ current. A base temperature lower than the C-T phase transition temperature can be utilized to achieve such enhancement. We operate the device (D_a) at a temperature below the C-T transition temperature to invoke field-driven metastable states of TaS₂ that increase the abrupt current jump by 1.42 × in comparison to room temperature. Figure 2c depicts I_M versus V_{T_s} characteristics delineating current decrement at

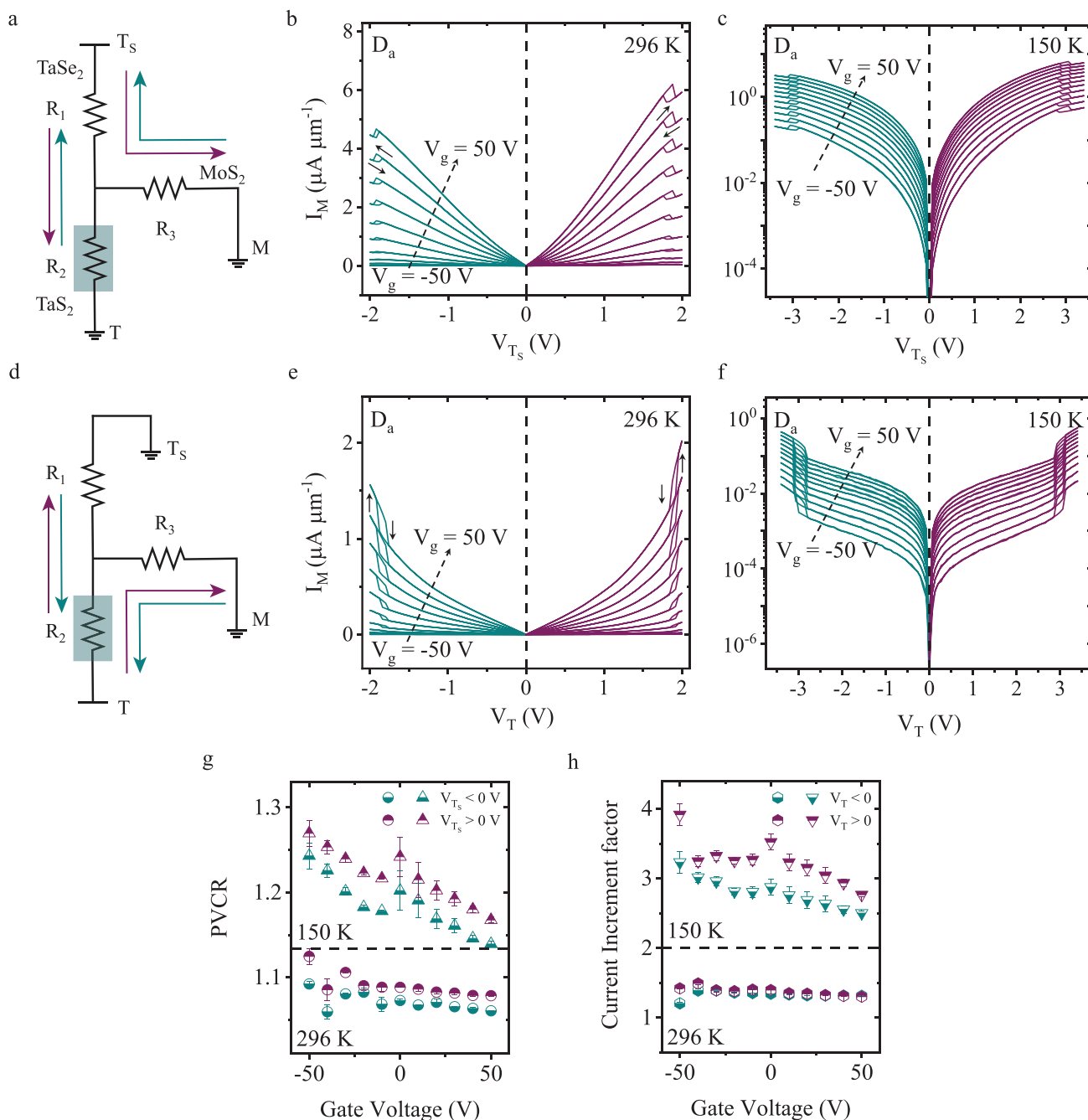


Figure 2. Gate-tunable, reconfigurable abrupt current increment and negative differential resistance. a,d) The three branches of the device D_a depicted by three effective resistances R_1 , R_2 , and R_3 , corresponding to TaSe₂, TaS₂, and MoS₂, respectively, for TaSe₂ (T) biasing (in (a)) and TaS₂ (T_S) biasing (in (d)). R_3 includes the MoS₂ branch resistance and the TaSe₂/MoS₂ Schottky junction resistance. The cyan box (in the TaS₂ branch) indicates the branch where CDW phase transition occurs. The arrows indicate the direction of the current flow in different biasing configurations. b,c) Current through the MoS₂ branch (I_M) versus V_{T_S} as a function of V_g varying from -50 to 50 V in steps of 10 V at 296 and 150 K depicting current decrement for both $V_{T_S} > 0$ and $V_{T_S} < 0$. Forward and reverse sweeps are indicated by black arrows. e,f) I_M versus V_T as a function of V_g varying from -50 to 50 V in steps of 10 V at 296 and 150 K depicting abrupt current increment for both $V_T > 0$ and $V_T < 0$. Forward and reverse sweeps are indicated by black arrows. g) Peak-to-valley current ratio (PVCRR) as a function of V_g at 296 and 150 K in bottom and top panel, respectively. h) Current increment factor as a function of V_g at 296 and 150 K in bottom and top panel, respectively.

150 K. The corresponding PVCRR values at 150 K are shown in the top panel of Figure 2g reaching a maximum value of 1.27 at $V_g = -50$ V for $V_{T_S} > 0$ V which is higher in comparison to the room temperature values.

Suppose we apply the bias at the TaS₂ terminal while keeping terminals T_S and M grounded as depicted in the equivalent circuit in Figure 2d. In that case, the MoS₂ current exhibits an abrupt increment instead of a decrement at the phase

transition. When we apply a high electric field across the TaS₂/TaSe₂ junction, the TaS₂ phase transition occurs, which reduces the resistance of the TaS₂ branch resulting in the abrupt increment in the TaS₂ current as well as the MoS₂ current. Here, the total current flow through R₂ instead of R₁ (in the case of TaSe₂ biasing) as shown by the arrows in Figure 2d. Figure 2e,f shows the gate voltage (V_g) dependent current increment characteristics for both V_T < 0 V and V_T > 0 V at 296 and 150 K, respectively. The corresponding current increment factor as the function of V_g for both 296 and 150 K is shown in Figure 2h. Current increment factor reaches a maximum value of ≈4 at 150 K for V_T > 0 V which is ≈2.75 × higher than the room temperature value.

The low-temperature current increment factor of four cannot solely result from phase transition induced resistance switching of TaS₂ as it is limited to a factor of ≈1.77 as shown in Figure 1c. In addition to 1T-TaS₂, 2H-TaSe₂ also exhibits CDW phase transitions at 90 and 120 K. However, it does not cause any abrupt discontinuity in the TaSe₂ resistance apart from a slight slope change in resistance versus temperature curve.^[3,20,21] The overall change in resistance of the TaSe₂ branch from the estimated temperature values (discussed later) is small and rules out the contribution of TaSe₂ resistance change to the current jump. Also, the heat equation solution of the channel and the in situ Raman measurement exclude the possibility of structural phase transition of MoS₂ from the semiconducting (2H) to metallic (1T) phase. The abrupt current increment due to the TaS₂ phase transition increases the local temperature along the device channel. The 1D heat equation solution (using FEM) for the TaS₂/TaSe₂ channel estimates that the local temperature increment due to Joule heating is sufficient to invoke TaS₂ phase transitions (see Figure S3, Supporting Information), depending upon the base temperature as well as the external bias. However, it is not enough to induce structural 2H to 1T phase transition in MoS₂.^[22] This argument is further supported by bias-dependent Raman measurement (see Figure S4, Supporting Information), wherein j-peaks have not been observed, which could otherwise confirm the presence of the 1T-phase of MoS₂.

Such a large current increment can be explained by the enhanced thermionic carrier injection across the TaSe₂/MoS₂ barrier with a sudden increase in the junction temperature resulting from the Joule heating induced phase transitions of TaS₂, as detailed next. The mechanism is schematically illustrated in Figure 3a,b. The abrupt jump in temperature due to the CDW phase transition in TaS₂ causes a corresponding increment in temperature at the TaSe₂/MoS₂ junction due to efficient heat conduction through the TaS₂/TaSe₂ interface. That, in turn, enhances the kinetic energy of the carriers, helping them overcome the TaSe₂/MoS₂ Schottky barrier.

2.2. Giant Current Increment Driven by Thermionic Switching

To explore the thermionic effect due to local temperature switching, we fabricate another triple layered T-junction (D_b) comprising a narrow MoS₂ channel with a reduced junction overlap area (see the optical image in the inset of Figure 4a). Here we choose a small junction overlap area between MoS₂ and the TaS₂/TaSe₂ to further increase the temperature by

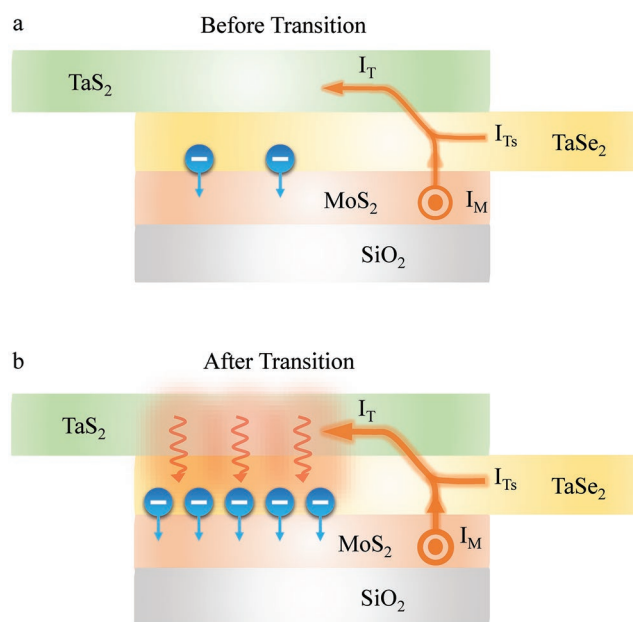


Figure 3. Schematic illustration of the mechanism. Schematic representation of the situation a) before and b) after the CDW phase transition in TaS₂. The orange arrow and the out-of-plane direction symbol indicate the direction of the current flow through the junction and in the MoS₂ channel, respectively. The heat flow from TaS₂ to the TaSe₂/MoS₂ junction in (b) is indicated by the red curved arrows. The abrupt enhancement in temperature promotes a larger number of electrons through the TaSe₂/MoS₂ Schottky barrier, in turn causing an abrupt change in the MoS₂ channel current.

forcing the hot electrons to pass through a smaller region. Apart from weaker heat dissipation efficiency, the smaller junction area helps in two other ways. First, when the junction area is small, the fractional contribution of the junction resistance to the total device resistance increases. That helps to achieve a larger switching ratio since the hot carriers, due to the phase transition-induced temperature change, primarily modulate the junction resistance. Second, a smaller overlap area helps to remove any trapped residue and air from the interface during annealing, which in turn helps to achieve closer proximity between MoS₂ and TaSe₂/TaS₂ heater, helping the MoS₂ layer to achieve a higher temperature effect at the junction during the switching. I_M versus V_T of device D_b for selective V_g values varying from 20 to 50 V (step size: 10 V) at 300 K for V_T < 0 V is depicted in Figure 4a. Similarly, Figure 4b shows the I_M versus V_T for V_T > 0 V at V_g equal to 0, 30, and 50 V. The corresponding current increment factors during the phase transition are plotted in Figure 4c showing a maximum value of ≈3 at V_g = -10 V for V_T > 0 V at 300 K which is 2 × higher in comparison to the current increment factor of device D_a.

At low temperature, we could further enhance the current jump as depicted in I_M versus V_T characteristics of device D_b at 77 K (see Figure 4d,e for V_T < 0 V and V_T > 0 V, respectively) for different V_g values. The corresponding current increment factors are plotted in Figure 4f for both V_T < 0 V and V_T > 0 V. We could enhance the MoS₂ current by a factor of ≈964 at V_g = 10 V for V_T > 0 V as represented in Figure 4f. As discussed earlier, such a huge jump can not solely arise from electrically

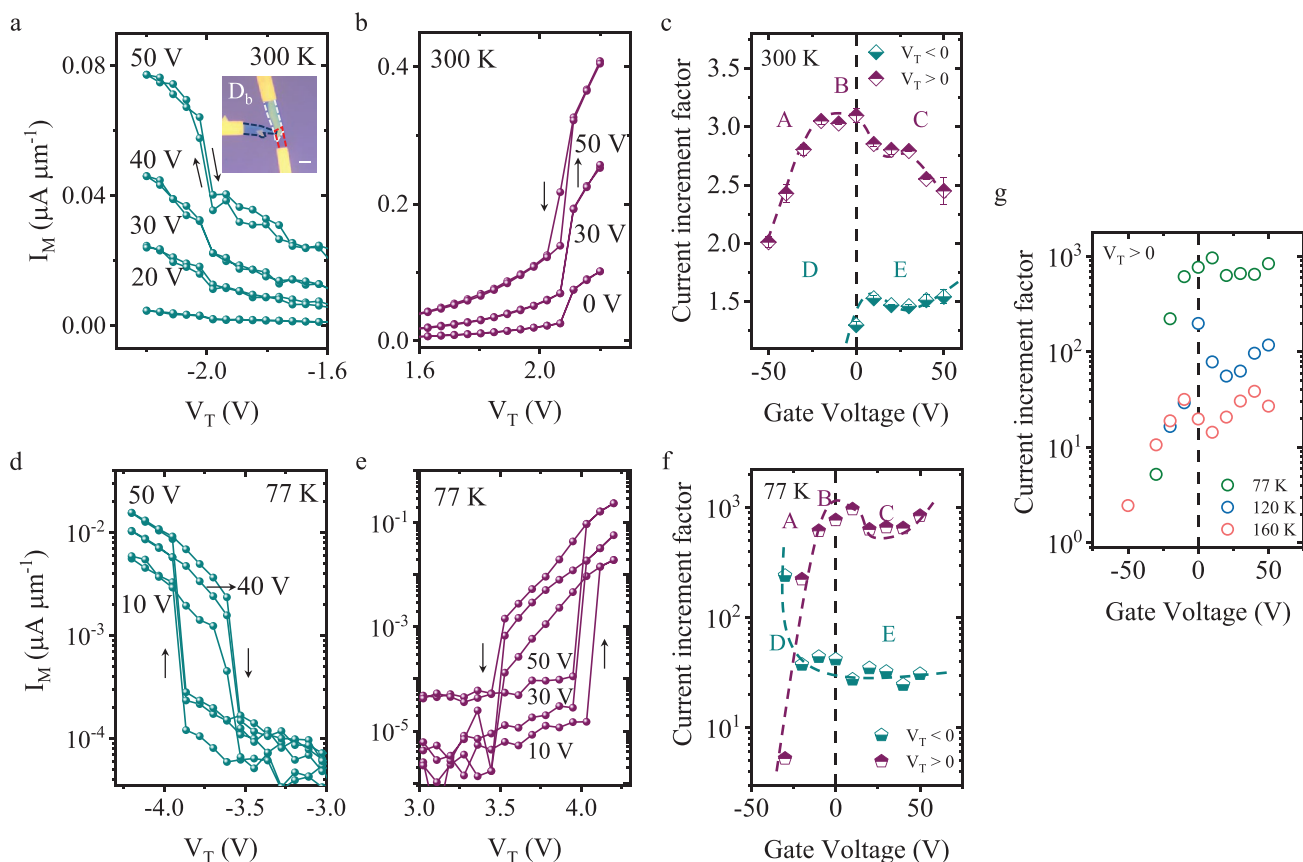


Figure 4. Giant current increment in triple layered junction. a,b) I_M versus V_T for selected V_g at 300 K for $V_T < 0$ (in (a)) and $V_T > 0$ (in (b)) depicting abrupt current increment. Forward and reverse sweeps are indicated by black arrows. The inset of (a) shows the optical image of the fabricated triple layered junction (D_b) (scale bar: 5 μm). d,e) I_M versus V_T for selected V_g at 77 K for $V_T < 0$ (in (d)) and $V_T > 0$ (in (e)) depicting abrupt current increment. c,f) Current increment factor as the function of V_g at 300 K (in (c)) and 77 K (in (f)) extracted from (a,b) and (d,e), respectively. g) Current increment factor as a function of V_g at 77, 120, and 160 K.

driven TaS₂ phase transition. And, we must invoke the corresponding sudden rise in the junction temperature due to the abrupt enhancement in current through TaS₂/TaSe₂ during the CDW phase transition of TaS₂. Figure 4g depicts the variation of the current increment factor as a function of V_g at a base temperature of 77, 120, and 160 K for $V_T > 0$ V. Such a strong temperature dependence in the current increment factor suggests possible usage of the technique in sensing temperature.

In Figure 4f for $V_T > 0$ V, the current jump ratio at the phase transition exhibits a strong non-monotonic behavior with V_g . Also, the ratio shows an opposite trend for $V_T < 0$ V, particularly when $V_g < 0$ V. The origin of such behavior is explained in Figure 5. In the case of $V_T > 0$ V, that is, when MoS₂ injects electrons into TaSe₂, there are three different regions of device operation (namely A, B, and C as shown in Figure 4c,f) as schematically explained in Figure 5a. Note that, due to the high electrical conductivity of TaSe₂ in comparison to TaS₂ (in particular, at a low temperature), the floating voltage at the triple junction is small; hence the drop across the MoS₂ channel is also small (estimated to be <26 mV). Due to such a small effective drain voltage, the current through the MoS₂ channel strongly depends on the drain barrier as well in the different regimes of operation. In the current situation, where TaSe₂ acts as the

drain contact for $V_T > 0$ V configuration, the drain barrier plays an even more important role. We recently found that due to van der Waals nature of the contact interface, TaSe₂ exhibits strong Fermi level depinning^[23] with layered semiconductors. Accordingly, due to the relatively large work function of TaSe₂, the conduction band offset between MoS₂ and TaSe₂ is large. On the other hand, due to Fermi level pinning, Ni/MoS₂ junction has a relatively small Schottky barrier height.^[24] That leads to a larger drain barrier compared to the source barrier at the small effective drain bias as schematically shown in Figure 5a, and hence the TaSe₂/MoS₂ interface controls the drain current.

In region A, that is, for $V_g \ll 0$ V, electrons do not see any barrier at the drain side and can quickly transfer into TaSe₂ (see top panel of Figure 5a). Thus, the current does not change much with the abrupt increase in temperature during the CDW phase transition. In region B, the electrons from the MoS₂ region require to overcome the drain barrier to be transferred to TaSe₂ (see middle panel of Figure 5a), and thus the current is a strong function of the local temperature at the drain barrier. The current induced Joule heating can increase the local temperature to ≈ 230 K for a base temperature of 77 K (see Figure S3, Supporting Information, for simulated results). The current jump ratio simulated for various barrier heights (ϕ_b varying

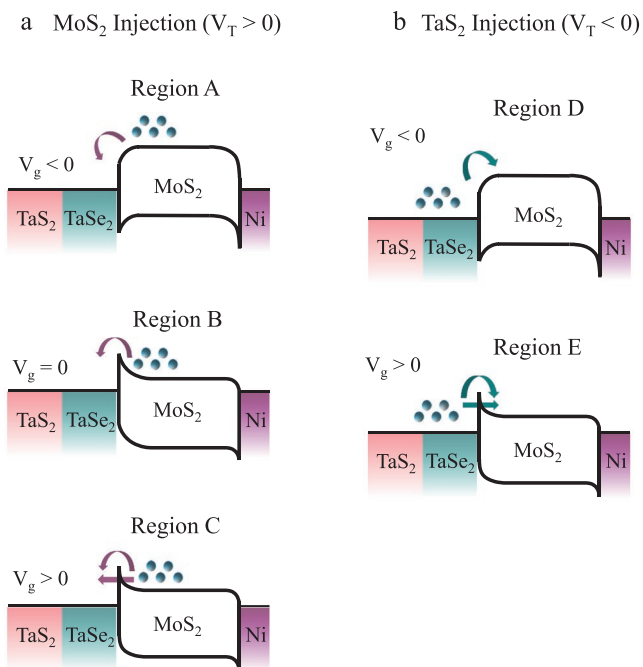


Figure 5. Thermionic switching behavior. a) Band alignment schematics for $V_T > 0$, that is, when MoS₂ injects electron into TaSe₂ through the drain barrier at $V_g < 0$ (in top panel), $V_g = 0$ (in middle panel), and $V_g > 0$ (in bottom panel) corresponding to region A, B, and C in Figure 4. b) Band diagram for TaSe₂ injection into MoS₂ through the source barrier ($V_T < 0$) for $V_g < 0$ (in top panel), and $V_g > 0$ (in bottom panel) illustrating region D and E from Figure 4c,f.

from 30 to 80 meV) by solving modified Richardson's equation^[24] at temperatures of 120, 150, 180, 210, and 240 K with respect to 77 K is shown in Figure S5, Supporting Information. The simulation results clearly show that the current ratio before and after the phase transition can go as high as 10^4 depending upon ϕ_b . That justifies the high current increment factor at $V_g = 10$ V. Finally, in region C for $V_g \gg 10$ V, the band bending increases, and the electrons can tunnel through the Schottky drain barrier, as shown in the bottom panel of Figure 5a. Thus, the local temperature does not affect the current, reducing the current ratio before and after the phase transition. Beyond region C, at an even higher positive V_g , we observe an increment in the current increment factor, the origin of which is yet not very clear and could result from a V_g dependent change in the relative resistance between the source and the drain barriers.

On the other hand, for $V_T < 0$ V, TaSe₂ acts as a source contact, and the electrons are injected from TaSe₂ to the MoS₂ channel. At high negative V_g , that is, region D, the carrier injection is determined by thermionic transport over the Schottky barrier height at TaSe₂/MoS₂ interface, as shown in the top panel of Figure 5b, and hence contributes to higher current jump. However, for $V_g > 0$ V (region E), the current increment ratio decreases monotonically as the carrier injection is dominated by the tunneling phenomenon schematically shown in the bottom panel of Figure 5b.

In similarity to device D_a, the I_M versus V_{T_s} characteristics of device D_b exhibits current decrement for V_{T_s} biasing at both

300 and 77 K (see Figure S6, Supporting Information). The corresponding I_M versus V_T (depicting current increment) and I_M versus V_{T_s} (depicting current decrement) characteristics for device D_b at 120 and 160 K are outlined in Figures S7 and S8, Supporting Information, respectively. The current increment characteristics have been repeatedly observed in several devices, some of which are outlined in Figures S9 and S10, Supporting Information.

3. Conclusion

The technique proposed in this work demonstrates a unique way of significantly amplifying the resistance switching ratio typically obtained from a TaS₂ CDW phase transition. That is achieved by exploiting enhanced carrier injection through a Schottky barrier height by the abrupt increment in the local temperature during the phase transition. Accordingly, the technique can be applied to enhance the device performance in several applications where TaS₂ phase transition is used, for example, in detecting infrared photons and neuromorphic applications. In addition, the gate tunable sharp jump in current can be useful for sensing applications, such as temperature and current. On the other hand, the enhancement in the MoS₂ channel current during the CDW phase transition of TaS₂ provides an excellent probe to monitor the local temperature of TaS₂.

4. Experimental Section

Triple Layered T-junction Device Fabrication and Characterization: 1T-TaS₂/2H-TaSe₂/2H-MoS₂ T-junction was fabricated in the following manner. First, the thin flakes of MoS₂ were mechanically exfoliated on a heavily doped Si substrate coated with 285 nm thick SiO₂ using polydimethylsiloxane (PDMS), followed by dry transfer of TaSe₂ and TaS₂ flakes, respectively. A rotational stage controlled the alignment during each layer transfer to form the T-junction. The complete exfoliation and dry transfer processes were done at room temperature. The substrate was then spin-coated with a high contrast positive resist—polymethyl methacrylate (PMMA) 950 C3 and softly baked for 2 min at 180 °C. Patterns were formed through electron beam lithography with an electron beam dose of 200 $\mu\text{C cm}^{-2}$, an electron beam current of 300 pA, and an acceleration voltage of 20 KV. The pattern development was carried out in 1: 3 MIBK/IPA developer solution followed by IPA wash and blow drying in N₂. Metal contacts were formed by blanket deposition of 10 nm Ni / 50 nm Au using a DC magnetron sputter coating system in the presence of Ar plasma at 6.5×10^{-3} Torr. Excess metal lift-off was carried out by immersing the substrate in acetone for 15 – 30 min, followed by IPA wash for 30 s and blow drying in N₂. Buffered HF solution was used to etch the back oxide from the substrate, and highly conducting silver paste was used for the back gate contact.

The electrical measurements were carried out in a probe station with a base vacuum level of about 1.6×10^{-3} Torr at room temperature and 6.45×10^{-6} Torr at the low temperature with the supply of liquid N₂.

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

1T-TaS₂, 2H-MoS₂, 2H-TaSe₂, charge density waves, current switching, negative differential resistance, phase transitions

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