Junction-Structure-Dependent Schottky Barrier Inhomogeneity and Device Ideality of Monolayer MoS₂ Field-Effect Transistors

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Supporting Information

ABSTRACT: Although monolayer transition metal dichalcogenides (TMDs) exhibit superior optical and electrical characteristics, their use in digital switching devices is limited by incomplete understanding of the metal contact. Comparative studies of Au top and edge contacts with monolayer MoS₂ reveal a temperature-dependent ideality factor and Schottky barrier height (SBH). The latter originates from inhomogeneities in MoS₂ caused by defects, charge puddles, and grain boundaries, which cause local variation in the work function at Au–MoS₂ junctions and thus different activation temperatures for thermionic emission. However, the effect of inhomogeneities due to impurities on the SBH varies with the junction structure. The weak Au–MoS₂ interaction in the top contact, which yields a higher SBH and ideality factor, is more affected by inhomogeneities than the strong interaction in the edge contact. Observed differences in the SBH and ideality factor in different junction structures clarify how the SBH and inhomogeneities can be controlled in devices containing TMD materials.

KEYWORDS: MoS₂–metal contacts, edge contact, contact resistance, Schottky barrier inhomogeneity, ideality factor, Fermi level pinning

1. INTRODUCTION

Transition metal dichalcogenides (TMDs) have been studied for decades because of their wide ranges of chemical, mechanical, and electrical properties.† However, among them, semiconducting TMDs (s-TMDs) are being revisited as a potential alternative to gapless graphene for two-dimensional (2D) nanoscale electronics. The 1–2 eV direct band gap of monolayer s-TMDs, the absence of dangling bonds on the surface, and the diversity of atomic composition and their various hybrid structures provide advantages for nanoscale devices that has not yet been preceded by graphene, such as visible-range optical sensors,† light-emitting diodes,‡ n-type and p-type field-effect transistors (FETs),§,∥ and logic circuits.¶ Despite their promising features for application to digital electronics, poor understanding of the contact nature between metals and s-TMDs is the main factor limiting device performance.

Most metal electrodes used in s-TMD devices are in physical contact with the channel from the top side. The surface of TMDs, which has no dangling bonds, is chemically inert and produces weak atomic bonding to the electrode metals at the junction. Although the determination of the Schottky barrier height (SBH) is complicated by several factors, such as the defect density,§ Fermi-level pinning,¶ dipole field,∥∥ and interfacial structure,‡ it is generally accepted that the physical top contact between the metal and monolayer MoS₂ (1L-MoS₂) forms an SB with a height ranging from 0.1 to 0.4 eV.¶ This, previous studies have shown that the SBH of MoS₂ did not follow the Schottky–Mott relation. The Fermi level is often pinned near the bottom of the conduction band regardless of the metal used for the electrodes.

In addition to these complexities in the SBH of TMDs, most TMDs contain or are exposed to disorders that can influence the metal–semiconductor junction.§ This may explain the broad range of SBH values reported in the literature. If the metal–TMD contact is made through physical contact, it is quite subject to changes due to the surface potentials exerted by impurities. Although many theoretical predictions have been made on the nature of the junction and the resulting SBH at the metal–s-TMD junction,∥∥∥ the effect of inhomogeneities in TMDs on the SBH and ideality factor and their correlations to the junction geometry of MoS₂ FETs have not been explored yet.

In this study, two types of Au contacts with 1L-MoS₂ are compared: (i) an edge contact, with strong covalent bonding and (ii) a top contact, with weak physical bonding. In both
contacts, the SBH changes as a function of temperature. The result indicates the existence of numerous SBs with different heights in both junctions. Therefore, at a given temperature, a certain patch with a low work function dominates the thermionic or field emission at the junction, giving rise to the change in the SBH with temperature. However, the degree of inhomogeneity in an SB depends on the nature of the atomic bonding at the junction. The SBH and its inhomogeneity at metal–MoS 2 junctions are observed to be lower in edge-contact 1L-MoS 2 devices (ECMs) than in top-contact 1L-MoS 2 devices (TCMs), and the ideality factor is closer to unity in ECMS than in TCMs. The stronger atomic binding in ECMS than in TCMs causes the metal–MoS 2 junction to be less affected by the surface potential from disorders in our device.

2. RESULTS AND DISCUSSION

2.1. Basic Electrical Properties. The 1L-MoS 2 samples used in this study were grown by chemical vapor deposition (CVD) 20 and transferred onto a SiO 2 (300 nm)/Si (500 µm) substrate with exfoliated hexagonal boron nitride (h-BN, 16 nm in thickness) for device fabrication. Metal/1L-MoS 2 edge contacts were constructed by etching the poly(methyl methacrylate) (PMMA)-coated h-BN/1L-MoS 2/h-BN stack with SF 6 plasma, followed by Cr/Au (2/60 nm) deposition, in which a thin Cr layer was deposited only as an adhesion layer. Figure 1a schematically illustrates a four-terminal ECM; the inset shows an optical image of the device. Figure 1b shows the drain−source current as a function of the backgate voltage, I d−V BG , for selected temperatures on logarithmic (left axis) and linear scales (right axis) for the ECM. The maximum two-terminal field-effect mobility μ ff is estimated to be ∼5 cm 2 V −1 s −1 below ∼20 V BG at 280 K. The current tends to increase with a smaller slope above 20 V BG (right axis). This early indication of current saturation originates from the charge transport being limited by the dominant contact resistance, which will be discussed later. At 280 K, the basic I d−V BG curves (see Figure S1b) measured at various V BG values show linear and symmetric behavior, implying that both contacts of the device are ohmic at this temperature.

For comparison, a conventional TCM on a SiO 2 substrate and a TCM on a h-BN/SiO 2 substrate (TCMBN) were also fabricated (see Figure S1c for the TCM and Figure S1e for the TCMBN). The TCM shows contact qualities similar to those of ECM (see Figure S1d) with linear behavior at 280 K; however, it exhibits mobility higher than that of ECM (unbounded mobility above 40 cm 2 V −1 s −1 at room temperature) without any indication of current saturation within the experimental backgate voltage range, as shown in Figure 1c. Similar linear behavior in I d−V BG is also observed in the TCMBN (see Figure S1f), but μ ff is further enhanced to above 60 cm 2 V −1 s −1 because of reduced Coulomb scattering by the underlying h-BN substrate. 21, 22 It is also important to note that the I d −V BG crossover is observed only in the TCMBN at 40 V BG during the temperature sweep, as indicated by an arrow in Figure 1d. This metal−insulator transition observed in the TCMBN is not found in the ECM and TCM for a large contact resistance and the absence of hBN, i.e., hBN provides more flat surface and cleaner interface by suppressing the interface trap charge density, 23 respectively, which will be discussed in the next section in more detail. The small hysteresis in I d−V BG in our devices (S2 in the Supporting Information) indicates that the effects of polar adsorbates such as water molecules 24 on the carrier transport are negligible.

Figure 2 shows the dependence of the total (R T ), contact (R c ), and channel (R ch ) resistances on the backgate voltage (V BG ) and temperature (T ) for the ECM (a, b), TCM (c, d), and TCMBN (e, f), where R T = V d/BG R c = R T − R ch , and R ch = (V d/ID) (W/L) μ, L is the total length of the channel, W is the width, V BG is the voltage drop between the inner two probes, and L in is the length between the voltage probes. The contribution of R c or R ch to R T differs noticeably depending on the metal–MoS 2 junction geometry. For the ECM, R ch constitutes the major portion of R T for the entire range of V BG T, and R ch makes a negligible contribution, as shown in Figure 2a. Figure 2b shows that R c dominates R T regardless of temperature and V BG T. At −10 V BG T, the channel exhibits recognizable semiconductor behavior, whereas at 70 V BG T, the channel behaves as a metal, but the metallic behavior is obscured in R T by the high R ch.

In the TCM (Figure 2c), R c and R ch are comparable during the V BG sweep. This tendency is reproducible at each temperature, and the channel exhibits no metallic phase (Figure 2d). The situation is quite different for the TCMBN: R c is reduced substantially in the on state (in the high-I d region), and R T is determined primarily by R ch (Figure 2e). The absolute value of the channel resistance is, however, still smaller than the R ch value of the TCM (see Figure S3). At 0 V BG T, R ch is dominant, with negligible R c, and decreases in proportion to the temperature, revealing semiconducting behavior. At 50 V BG T, R ch is still dominant but increases with increasing temperature, revealing metallic behavior (Figure 2f). The observed resistance changes show good agreement with the I d−V BG characteristics shown in Figure 1.

2.2. Origin of Large Contact Resistance in ECM. Now we discuss the origin of the large R ch in the ECM in Figure 2a and 2b. The large contact resistance of the ECM is entirely different from that in the case of graphene; the remarkably low contact resistance observed in edge contacted graphene devices is ascribed to the shorter bonding length or larger atomic orbital overlap than those of top contacted devices. 25 However,
the bond between the metal electrode and the channel does not seem to be the only reason for the low contact resistance in graphene because such strong atomic bonding presumably also occurs in edge-contacted MoS2. The main cause for this difference in R contacted graphene and MoS2 devices is the large transfer length L_T for MoS2. The transfer length is defined as the distance from the contact edge, x = 0 in Figure 3, to the point toward L where the voltage or current level becomes 1/e of the value at x = 0. It is determined by the relative magnitude of the top contact resistivity (ρct) at the metal–MoS2 interface and the sheet resistivity of MoS2 (ρsh) below the metal. Thus, it can be expressed as L_T = (ρct/ρsh)^1/2.

In graphene, L_T is very short, i.e., ρct >> ρsh; therefore, current flows mainly through the metal–graphene contact edge, near x = 0. In contrast, L_T is reportedly rather large for a metal–1L-MoS2 contact, ranging from 0.6 to 1.2 μm, depending on VBG. Thus, for the ECM, a longer contact length than L_T is required for efficient charge injection, which is not feasible for an ECM consisting of a one-dimensional (1D) atomic contact. However, considering the current density, the ECM has a tremendous advantage over the TCM. It is noteworthy that the TCM has a contact area at least 3 orders of magnitude larger than that of the ECM but has a current level only 1 order of magnitude larger (see S4 in the Supporting Information for a comparison of the contact resistivity of the three devices as characterized by the contact area). Even if current flows through the edge in the TCM, it would be minor compared to the current flow through the top because ρct/1/W >> ρct/LTW, as can be inferred from Figure 2, where t is the MoS2 monolayer thickness, and ρct is the contact resistivity between the metal and the edge.

2.3. Ideality Factor and Schottky Barrier Height. To analyze the effect of the junction structure on the device parameters, we further investigate the temperature-dependent ideality factor (n) and the effective SBH (Φb) at a given VBG. In general, a MoS2 FET with two identical source and drain contacts can be viewed as back-to-back Schottky diodes (Figure S5a). In this device geometry, most of the voltage drop occurs in the reverse-biased side. Therefore, the following single-diode equation for a 2D system can be used to extract the ideality factor n and SBH Φb for the negative bias side as shown in Figure S5b.

\[
I = I_o \exp \left( \frac{qV}{n k T} \right) \left( 1 - \exp \left( -\frac{qV}{k T} \right) \right)
\]

(1)

where \(I_o = \frac{A^* e^{\frac{qV}{k T}}}{n k T} \), A is the junction area, \(A^*\) is the Richardson constant, \(k\) is the Boltzmann constant, and \(q\) is an electrical charge. Rearranging eq 1 gives

\[
\ln \left( \frac{I \exp(qV/k T)}{\exp(qV/k T) - 1} \right) = \ln(I_o) + \frac{qV}{n k T}
\]

(2)

Here V is the voltage drop across the junction; hence, V = Vsh - IR where R is the series resistance, the largest contribution to which comes from the channel resistance. By using I-V measurements in the plot of ln(I) vs V for the reverse bias at each temperature, n and Φb can be determined from the slope and y intercept, respectively (Figure S5b). To avoid the confusion, we note again that fittings are performed for the negative bias region to account for the reverse biased side. Fairly good linear fits were obtained for the ECM for a given gate bias (−10 VBG) at several temperatures. To calculate Φb, from the y intercept, \(ln(I_o)\), we used 54 A K^-2 cm^-2 for \(A^*\). For the junction area (A), the MoS2 monolayer thickness of 0.72 nm for the ECM and Lx ≈ 1 μm for the TCM and TCMBN were multiplied by the width of each device. Because \(Φ_b\) is not very sensitive to \(A^*\) and A (e.g., doubling or halving these values changes \(Φ_b\) by approximately 10%; see Figure S6 in the Supporting Information), interpretation of the transport mechanism using \(Φ_b\) is qualitatively valid.

The resulting values of n for all three devices in the selected VBG region are presented in Figure 4a. In general, the current...
through the SB is determined by two competing processes: tunnelling and thermionic emission. In pure thermionic emission at a given $\Phi_b$, the ideality factor $n$ is unity. As the temperature decreases, the thermionic emission current is suppressed. At the same time, the field emission current becomes dominant. Thus, $n$ is expected to increase from unity.\(^{34}\) The dashed line in Figure 4a represents this trend. Here the $n$ values of the TCM and TCMBN are larger than unity near room temperature and decrease as the temperature goes down. The $n$ value for the ECM shows a similar trend to those of the TCM and TCMBN, but it is much smaller than the latter. Decreasing $n$ with descending temperature means that the field emission does not contribute at the low temperature in Figure 4a. In contrast, the convergence of $n$ to unity in all three devices strongly suggests that thermionic emission plays a dominant role even at low temperature. We attribute $n > 1$ at that temperature to the distribution of SBs with various $\Phi_b$ values, that is, SBH inhomogeneity. The MoS$_2$ in our device probably consisted of numerous patches with different surface potentials and spatial sizes that may be caused by impurities, grain boundaries, charge puddles, and adsorbates. Therefore, when MoS$_2$ with such disorders comes into contact with the metal electrode, the corresponding SBH between MoS$_2$ and Au is expected to vary locally because of the inhomogeneities, as shown in Figure 4b. The jagged sharp edge at the interface of Au$-$MoS$_2$ in Figure 4b indicates SBs with different heights and widths resulting from the defects mentioned above. Therefore, SB dips and peaks congregate together. Depending on the distribution of the inhomogeneities, the SB shapes may vary dramatically.

2.4. Schottky Barrier Height Inhomogeneity. Inhomogeneous SBs in the theoretical models indicate various situations.\(^{35}\) In general, the relative work function difference, spatial separations between the patches, temperature, and applied bias all contribute to the complexity of SBH inhomogeneity. Figure 4b shows a model that fits our case; it consists of an ensemble of a few wide patches of low SBH (blue arrow) and numerous narrow patches of high SBH (red arrow). Because of the large patch area and narrow energy window for thermionic emission at low temperature, the homogeneity increases, giving rise to an ideality factor close to unity, as shown in Figure 4a. However, as the temperature rises, even small and high-work-function patches participate in thermionic emission. This results in the variation of work function and area among the patches. Therefore, the inhomogeneity of the SBH increases, resulting in an ideality factor much greater than unity.\(^{35}\) In Figure 4a, all our devices show an ideality factor affected by SBH inhomogeneity, although the degree of unevenness of the SBH varies depending on the structural configuration of the metal$-$MoS$_2$ junction. The ECM seems to show the least effect from disorders at the metal$-$MoS$_2$ interface. The mean and standard deviation of $\Phi_b$ will be further discussed later.

To confirm the effects of SBH inhomogeneity on transport, $I_{ds}$ vs $V_{ds}$ measurements of the TCMBN at $0 \ V_{BG}$ and several temperatures are displayed in the top panel of Figure 4c. When each trace is normalized by its current at $1 \ V_{ds}$ ($I_{ds} = I_{ds}(V) / I_{ds}(1)$), all the traces almost collapse onto a single line, as shown in the bottom panel. This behavior is unlikely to be explained by the thermionic emission through a single SBH because, as the temperature decreases, the field emission gradually dominates the thermionic emission, so the current is suppressed appreciably at low bias, and the tunneling current starts dominating at high bias and increasing at a fast rate. In this case, normalization of $I_{ds}$ by $I (V_{ds} = 1 \ V)$ would not superimpose all the $I_{ds}$ vs $V_{ds}$ curves. However, if the thermionic emission is a result of multiple SBs, then during the temperature sweep, the patches with low work function inject many carriers and emerge as the new SB. Therefore, in each $I_{ds}$ vs $V_{ds}$ curve in Figure 4c, the thermionic emission is expected to be strongly sustained. This leads to the same $I_{ds}$ vs $V_{ds}$ behavior, that is, the same slope with normalization. This implies that the effective SBH at a given $V_{BG}$ changes with temperature, which is the conceptual essence of SBH inhomogeneity.

To assess the SBH unevenness quantitatively using the $y$ intercept, $\ln(I_{ds})$, in eq 2 (see also Figure S5b), which is a function of $\Phi_b$, the SBHs of our devices were estimated as a function of temperature for selected $V_{BG}$ values, as shown in Figure 5a. One common feature in the $\Phi_b$ vs $T$ plots for all the devices is that the SBH changes with temperature, supporting the concept of SBH inhomogeneity. Among various patches, those with a higher SBH become inactive for thermionic emission.

Figure 5. (a) Temperature dependence of the SBH ($\Phi_b$) at selected $V_{BG}$ values. The inset displays the examples of linear fits for the high-temperature region in $\Phi_b$ vs $q/k_B T$ for TCM and TCMBN at $50 \ V_{BG}$ and ECM at $70 \ V_{BG}$; (b) $\Phi_b$ vs $V_{BG}$ for TCM, TCMBN, and ECM.
emission as the temperature decreases. Thus, thermionic emission is available only from patches with low SBH. Consequently, the SBH decreases as the temperature decreases. In contrast to the reduction in the SBH at low temperature, the contact resistance increases, as shown in Figure 2b.d,f because the number of thermally excited carriers decreases rapidly when the temperature decreases. A strong similarity is seen between n in Figure 4a and Φ_b in Figure 5a among the devices, the effective SBH Φ_eff and n are smallest for the ECM. The mechanism for this behavior will be discussed later.

2.5. Comparison of Mean Values and Inhomogeneity of the Schottky Barrier Height. Figure 5a supports the existence of SBH inhomogeneity. To assess the mean (Φ_eff) and standard deviation (σ_b) of inhomogeneity, we plot Φ_eff vs 1/T from which the mean (y intercept) and standard deviation (slope) of SBH can be extracted based on eq 3. According to Werner and Güttler, assuming a Gaussian distribution of the SBH, the effective SBH Φ_eff at a certain V_BG is represented as a function of temperature by

\[ \Phi_{\text{eff}}(V_{\text{BG}}) = \Phi_{\text{bo}}(V_{\text{BG}}) - \frac{\sigma_b^2(V_{\text{BG}})}{2k_B T/q} \]  

(3)

where Φ_bo is the mean value, and σ_b is the standard deviation. This equation states that the effective SBH Φ_eff is always smaller than the mean SBH Φ_bo unless σ_b/k_B T ≈ 0. This is because thermionic emission preferentially occurs through lower barriers. Thus, Φ_eff converges to Φ_bo at very high temperature, at which all the SBHs are actively involved in carrier injection. If the SBH inhomogeneity follows a Gaussian distribution closely, we may expect a linear relationship in the Φ_eff vs q/k_B T plot. Such agreement between the model and our experimental data is well supported above ~160 K (inset of Figure 5a), where numerous patches are involved in thermionic emission. For direct comparison, the different V_BG values for each device are adjusted to V_BG values by subtracting 40, 20, and 25 V for the ECM, TCM, and TCMBN, respectively, at which the device turns on in logarithmic plots in Figure 1b–d (V_on) at room temperature (V_BG = V_BG − V_on). The inset shows examples for the TCM, TCMBN at 50 V_BG and ECM at 70 V_BG. The linear fit for ECM gives Φ_bo ≈ 0.124 V and σ_b ≈ 49 mV. The Φ_eff bo values obtained in this way for the other devices are displayed in Figure 5b and Table 1 for several backgate voltages. The table clearly shows that the ECM has a smaller SBH than the TCM and TCMBN. This result indicates that the SBH is not determined simply by the metal work function, i.e., the Schottky–Mott relation, which should yield an SBH of 0.6–0.8 V or a bit lower due to the thin Cr layer, but by more complex processes. Note that application of the Richardson plot for determining the SBH is not appropriate here because the SBH is not unique regardless of the temperature (see S7 in the Supporting Information). Furthermore, σ_b becomes much higher for the TCM and TCMBN than for the ECM, as shown in Table 1. This indicates that depending on the metal–MoS_2 contact, the SBH inhomogeneity can vary. Because of the larger σ_b in the TCM and TCMBN, the ideality factor of these devices should be larger than that of the ECM, as shown in Figure 4a. Using h-BN as a substrate reportedly improves the device performance by reducing carrier scattering from charge puddles. Here this effect is demonstrated via the SBH inhomogeneity. However, we see only a minute effect from the h-BN substrate. Instead, the SBH inhomogeneity is much more suppressed in the ECM.

To appreciate why the SBH and its inhomogeneity are influenced by the junction structure, that is, the ECM or TCM, it is important to recognize the characteristics of binding between Au and MoS_2 in different junction structures. Binding between MoS_2 and Au is expected to differ greatly between the edge and top contact structures. What differentiates the binding characteristics of these two junction structures is the interaction strength between Au and Mo or S (Mo/S) atoms. In the ECM, dangling atoms of Mo and S are expected to form covalent bonds with Au atoms. Therefore, the atomic distance between Au and Mo/S atoms in the ECM (1.7 Å) is much shorter than that of the top contact (2.7 Å). Such strong interaction among these atoms at the edge of the MoS_2 significantly modifies the atomic coordinates, producing a large wave function overlap, which in turn causes redistribution of charges, an interface dipole. The band bending due to the interface dipole field contributes to Fermi-level pinning near the conduction band edge, resulting in a low SBH (see S8 in the Supporting Information for more details).

However, in the TCM, there are no covalent bonds between Au and MoS_2. The interaction energy of Au on MoS_2 is reportedly much weaker than that of other metals such as Ti, Pd, and Mo. Thus, a weak overlap of atomic orbitals between Au and MoS_2 should exist. Owing to the weak interaction between Au and MoS_2 in the TCM structure, weak pinning is expected through an unusual mechanism, contributing to the formation of a higher SBH than that of the ECM. The above theoretical predictions are well reproduced in Figure 5b. The Φ_eff value of the TCM is almost three times higher than that of the ECM. The Φ_eff value for the TCMBN is slightly lower than that of the TCM, which is attributed to the reduction of p-doping and increase of dipole field by h-BN layer which blocks the neutralization of fixed positive charges in SiO_2. Cr, which is used as a buffer layer in our devices, is reported to form a chemical bond with MoS_2. Supposing that Cr (2 nm in thickness) interacts chemically with MoS_2, the junction properties of the ECM and TCM are expected to be similar. However, a large difference in the SBH and inhomogeneity between the two types of contact implies that the Au layer, rather than the Cr layer, plays the dominant role in the ECM and TCM (see S9 and S10 in the Supporting Information for further discussion with SEM images).

We also postulate that the SBH inhomogeneity, which appears to different degrees in the ECM and TCM, is associated with the characteristics of binding between Au and MoS_2. In our device, a variety of disorders exists, including charge puddles, randomly distributed trapped charges, S vacancies, and grain boundaries. Imaging of 1L-MoS_2 on a SiO_2 substrate using Kelvin probe force microscopy clarifies the presence of an uneven distribution of the surface potential. The local variation of the surface potential is
strongly driven by the number of MoS₂ layers. Nevertheless, impurities can perturb the local potential of MoS₂ by up to about 100 meV within a single flake. This implies that the local potential contrast can affect the SBH. Owing to the different binding characteristics, the ECM and TCM have different binding energies and thus different SBHs. Thus, the stronger atomic binding in the ECM is thought to be less affected by the inhomogeneous surface potential. In contrast to the ECM, the TCM exhibits binding that is quite vulnerable to local perturbations in the surface potential. This is probably why the SBH inhomogeneity is much stronger in the TCM than in the ECM, as shown in Table 1. Such a large homogeneity in the SBH of the TCM contributes to an ideality factor much greater than unity, as shown in Figure 4a. The effect of the h-BN layer in the TCMBN is noticeable in Table 1; σ at $V_{BG} = 70 \text{ V}$ decreases from 93.8 mV for the TCM to 77.5 mV for the TCMBN. Multilayer h-BN screens the Coulomb potentials from charged impurities on the SiO₂ substrate such that the potential inhomogeneity weakens.

3. SUMMARY AND CONCLUSION

We clarify that the metal—MoS₂ contact consists of patches with different potentials, which contribute to the formation of diverse SBHs. Local potential puddles at the Au—MoS₂ junction originate from disorders in the MoS₂ and SiO₂ substrates. Because the potential due to various impurities is about 100 meV, the TCM, in which Au is in physical contact with MoS₂, is severely modulated by the local potentials, resulting in increased SBH inhomogeneity, whereas the influence of disorders on the SBH is reduced in the ECM, resulting in low SBH inhomogeneity. The local fluctuations of the junction barrier height can be controlled by using edge contacts or choosing an electrode metal that can provide covalent bonds between itself and MoS₂.

4. EXPERIMENTAL SECTION

Device Fabrication. Multilayered h-BN flakes were mechanically exfoliated on a highly doped 300 nm SiO₂/Si substrate. CVD-grown 1L-MoS₂ flakes (see ref 20 for the details of the CVD growth and transfer methods) were then transferred onto it. Optimally stacked MoS₂/h-BN and nearby MoS₂ without h-BN flakes underneath it were obtained for fabrication of the TCMBN and TCM, respectively. For the ECM, additional steps were added to the TCMBN fabrication process. Multilayered h-BN flakes were again exfoliated on another SiO₂/Si substrate, which was spin-coated (5000 rpm, 60 s) with PMMA C4/poly(vinyl alcohol) (0.3 g in 20 mL of deionized water) layers and baked on a hot plate at 70−80 °C. After 10−20 nm-thick h-BN was obtained, the h-BN/PMMA layer was detached from the SiO₂/Si substrate by floating it in water. The h-BN/PMMA was then scooped out and transferred onto the MoS₂/h-BN stack with the PMMA side up. The PMMA was washed out by acetone. Then fresh PMMA was spin-coated (3000 rpm, 50 s) for electron beam lithography (EBL), which was performed at a plasma power of 10 W for 10 s in a 10 sccm flow of SF₆ to define the device area for etching. After etching, the PMMA was washed out by acetone, and new PMMA was spin-coated again, followed by EBL, to define the electrode areas. Cr/Au (2/60 nm) were evaporated for electrodes in high vacuum (∼10⁻⁸ Torr). To avoid deposition of oxidized Cr, several-nanometer-thick Cr was evaporated away and then deposited on the devices. No postannealing was performed for any of the devices, but the ECM was thermal-cycled from room to low temperature.

The channel dimensions for the ECM, TCM, and TCMBN are 3.3 × 3.6, 7.5 × 12, and 7.5 × 12 (width × length) in nm, respectively. The electrical measurements were performed in high vacuum (∼10⁻⁸ Torr) using a commercial semiconductor characterization system (4200-SCS, Keithley).

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