Determination of Schottky barriers in 2D heterophase devices.

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The Schottky barrier of metal-semiconductor junctions can be extracted from an analysis of the density of states, the transmission spectrum, or the temperature dependence of the current. Electronic structure calculations commonly apply one of the first two methods whereas the latter is often used experimentally. We investigate the charge transport in monolayer MoTe$_2$ 1T'-1H junctions using ab-initio quantum transport calculations and find that the calculated barriers deviate significantly when comparing the three methods. Two main mechanisms cause the deviations; non-negligible tunneling current at room temperature and the restriction of momentum conservation perpendicular to the transport direction.

The contact-channel interface is a crucial performance bottleneck in the development of new transistor technologies. The energy barrier which charge carriers must overcome to move from the metal contact to the semiconductor channel, the Schottky barrier, is a key quantity for describing the charge transport. The atomically-thin transition metal dichalcogenides (TMDs) are emerging as a possible alternative to silicon for transistor channels in the next generations of technology nodes.[1, 2] However, the technology suffers from large contact resistance between the TMD and the metallic electrode. The resistance can be reduced by locally inducing the metallic 1T[3, 4] or the semi-metallic 1T' phase[5, 7] of the TMD and pattern the 3D electrodes directly over the resulting 1T/1T' regions. Understanding and quantifying the energy barrier of TMD 1T'-1H interfaces is therefore of great importance for the development of this technology.

Several techniques exist for extracting the Schottky barrier of 2D metal-semiconductor junctions both theoretically and experimentally. Electronic structure calculations most often extract the barrier height from the projected density of states (DOS) along the transport direction[8, 11] but the barrier can also be extracted from the transmission spectrum (TS).[10, 12] Experimental methods include Kelvin probe force microscopy[7], scanning photocurrent microscopy correlated with photoluminescence imaging[13] and application of the thermionic emission (TE) model.[3, 6, 11] The TE model has been utilized to extract barriers of fabricated TMD heterophase devices in the order of a few meV whereas DFT calculations estimate orders-of-magnitude larger barriers.[8, 12]

In this letter, we analyze the Schottky barrier height of pristine monolayer 1T'-1H MoTe$_2$ heterophase devices using density functional theory (DFT) and non-equilibrium’s Green’s function (NEGF) transport calculations. We extract the barriers from the projected DOS, the TS and using the TE model and find that the barrier heights differ significantly between the three methods. This highlights the challenges of comparing a barrier from an equilibrium electronic structure calculation with a measured barrier since barrier extraction using the TE model requires a calculation of the current. We study both a n- and p-type device. For the n-type device, tunneling through the barrier causes a significant deviation between the barriers extracted from the projected DOS or the TS and the barrier extracted using the TE model. For the p-type device, symmetry-forbidden transport energies cause a significant deviation between the barrier extracted from the projected DOS and the two other barriers. The extracted TE barriers remain an order-of-magnitude larger than the corresponding experimentally measured barriers, which suggests that the improved charge transport measured in the heterophase devices is not related to the intrinsic properties of the 1T'-1H interface.

We choose a free-standing monolayer interface between the MoTe$_2$ 1T' and 1H phase as our model system. Even though a transistor will have two Schottky barriers, one at the source and one at the drain, a forward bias will effectively create a single barrier at the source which will dominate the device behavior.[10] We do not include any substrate or gate but investigate the behavior of the isolated heterophase interface. A substrate below the 2D TMDs may have several effects: a small change of the band gap[17], longer depletion widths[18], and a modulation of the work function or doping level[19]. A longer depletion width would result in a lower tunneling current but wouldn’t change our conclusions. An estimate of this effect can be found in the Supplementary Material.[20]

We use a doping level of $N_{D/A} = 4.9 \times 10^{11}$ cm$^{-2}$ consistent with the estimated p-doping reported by Sung et al.[6].

We investigate three methods for the Schottky barrier extraction. The DOS barrier, $\Phi^{\text{DOS}}$, is extracted from the projected DOS as the distance between the Fermi
level and the maximum (minimum) of the conduction (valence) band for the n-type (p-type) device. The TS barriers, $\Phi_{\text{TS}}$, are defined as the distance between the Fermi level and the energy at which the device experience full transmission.

The last method applies the TE model, which assumes that the current is dominated by coherent transport of thermally excited electrons above the Schottky barrier. The most commonly used expression for the current is,\[^1\] \[^6\] \[^14\] \[^21\] \[^22\]

$$I_{\text{TE}} \approx I_{2D} e^{-\frac{E_{\text{sd}}}{k_B T}}. \quad (1)$$

$V_{\text{sd}}$ is the voltage drop between the source and drain, $e$ is the electron charge, $k_B$ is the Boltzmann constant and $T$ is the temperature. $I_{2D}$ is the reversed saturation current density,

$$I_{2D} = A^{*}_{2D} T^{3/2} e^{-\frac{\Phi_{\text{TE}}}{k_B T}}. \quad (2)$$

$A^{*}_{2D}$ is the Richardson constant and $\Phi_{\text{TE}}$ is the barrier height. This relation can be utilized to make an Arrhenius plot of $\ln(I/T^{2/3})$ vs. $1/T$ and extract the barrier from the slope. Note, that these expressions assumes the limit of where $eV_{\text{sd}} \gg k_B T$ whereas the opposite limit would result in a $T^{1/2}$ dependence in the current. We have investigated the effect of varying the temperature exponent and found that the results depends only weakly on this parameter. Following the available experimental results, we choose the dependence from equation (1) and (2).

If the electrons are transmitted across a tunneling barrier, the total current will have contributions from both the thermal excitation of the electrons and the tunneling. However, if the tunneling contribution can be ignored, the barrier extracted from the Arrhenius plot will equal the Schottky barrier. This regime is attempted to be reached experimentally either by fitting the current response at high temperatures or by applying a gate voltage to reach the flat band condition. We extract TE barriers in a temperature range between 300 and 450 K and using $V_{\text{sd}} = \pm 0.01$ V for the n- and p-type device respectively. Our setup is representative of a FET setup at zero gate voltage, in order to compare as closely as possible with the measurements.

The calculations are carried out using DFT\[^23\] \[^24\] and the non-equilibrium Green’s Function method as implemented in QuantumATK.\[^25\] We apply the Perdew-Burke-Ernzerhof (PBE)\[^26\] exchange-correlation functional and a linear combination of atomic orbitals using PseudoDojo pseudopotentials\[^27\] to expand the wave functions. The occupations are described by using a Fermi-Dirac occupation function with an electronic temperature of 300 K.

The generalized gradient approximation (GGA) functionals are known to produce bandgaps and work functions which are too small for the free standing TMD monolayers\[^28\]–\[^30\]. Our PBE calculations show a 1H phase bandgap of 1.03 eV which should be compared to the value using the GW formalism of 1.56 eV.\[^31\] However, since we are interested in qualitative trends and orders-of-magnitude differences, we expect the PBE functional to perform reasonably well. We do not include the spin-orbit coupling which would open a small gap in the 1T’ phase. This is justified by previous calculations\[^32\] showing that the barrier in TMD monolayer heterojunctions changes very little when including this effect.

The band diagrams of both phases can be seen in Figure 1 including the relative placement of the Fermi level for the n- or p-doped case. The doping is created by modifying the atomic charge density which corresponds to adding or removing electrons to the system.\[^33\] The doping is added to those atoms which belong to the 1H phase before relaxation. These are colored cyan and orange in Figure 2.

**FIG. 1.** Bandstructure of the monolayer a 1T’ phase and b 1H phase of MoTe$_2$. $\varepsilon_{F,n}$ and $\varepsilon_{F,p}$ show the Fermi level of the 1H phase with n- and p-doping concentrations of $N_{D/A} = 4.9 \times 10^{11}$ cm$^{-2}$.

**FIG. 2.** The 1T’-1H interface of ML MoTe$_2$ observed by Sung et al. \[^6\] seen from, a, the side and, b, the top. Note, that only the region around the interface is shown. The total cell size is (25.3, 0.352, 24.0) nm. The black lines show the unit cells of the two phases.
We set up the interface in the geometry found by Sung et al. [6] using tunneling electron microscopy. The interface is between the (100)-plane of 1T' and the (01-10)-plane of 1H and is shown in Figure 2 along with the unit cells of the two phases. The size of our computational cell for the NEGF calculations is (25.3, 0.352, 24.0) nm. The length/height ratio close to one ensures that the out-of-plane fields due to the 2D interface are properly accounted for and that their effect on the size of the barrier is minimized.[34–36] The heterophase device is created using 4 steps. First, the infinite 1T' and 1H monolayers are relaxed, then the interface is created using the interface builder in QuantumATK[37] and by straining the 1T' monolayer by 2.1 % to match the 1H phase. A nanoribbon of this interface is created to relax the atomic positions in the interface region. Finally, a device configuration consisting of two electrodes and a central region is created in order to perform the NEGF calculations. The central region is composed of 1T' and 2H segments with lengths of 6 nm and 19 nm, respectively. All the structure relaxations use a force tolerance of 0.02 eV/Å and the k-point grid for the isolated 1H phase is (7, 7, 1) while it is (k_x, 11, 1) for all other calculations. k_x = 5 for the isolated 1T' phase, k_x = 1 for the nanoribbon calculation, and k_x = 401 for the NEGF calculation.

For each device, we calculate the projected DOS and the transmission spectrum in equilibrium. The projected DOS of the n-doped device can be seen on Figure 3a showing a tunneling barrier due to significant band bending. The barrier height is 0.82 eV and the depletion width, x_D, is found by to be 7.05 nm assuming a band bending following CB(x) \propto e^{-x/x_D}. The corresponding transmission spectrum can be seen on Figure 3b showing significant contributions from tunneling. We consider the device to experience full transmission when the transmission becomes 1-10% of the maximum transmission. This corresponds to a TS barrier between 0.83 and 0.92 eV which agrees reasonably with the DOS barrier. The barrier corresponding to 1% of maximum transmission is illustrated on Figure 3b. Figure 3c and d show the transmission spectrum and the energy barrier (green) corresponding to 1% of maximum transmission. The n-doped device shows a significant tunneling contribution whereas the p-doped device shows no tunneling and a TS barrier significantly below the DOS barrier.

For the TE barrier extraction, we perform self-consistent calculations of the current and use the Landauer-Büttiker expression to calculate the temper-
The temperature dependence,

\[
I = \frac{2e}{\hbar} \int T(E, \mu_L, \mu_R) \times \left[ \frac{f \left( E - \mu_L \right)}{k_BT} - \frac{f \left( E - \mu_R \right)}{k_BT} \right] dE. \tag{3}
\]

\(\hbar\) is Planck’s constant, and \(\mu_L\) and \(\mu_R\) is the chemical potential of the 1T’ and 1H electrode respectively. The current can be separated into a tunneling and thermionic contribution by dividing the energy integral into a tunneling part running from the center of the band gap to the Schottky barrier height and a thermionic part running from the barrier to infinity. The resulting Arrhenius plot is seen on Figure 4a showing the total, tunneling and thermionic current of each device. The n-doped device shows a dominating tunneling behavior below 410 K and thermionic behavior above, which can be identified by the two distinct slopes above and below this temperature. These two regimes are a footprint of a tunnelling barrier and agrees qualitatively with the behavior predicted by the two distinct slopes above and below this temperature. These two regimes are a footprint of a tunnelling barrier and agrees qualitatively with the behavior predicted by the two distinct slopes above and below this temperature.

The p-doped device has negligible contributions from the tunneling current and the thermionic current is seen to be coinciding with the total current on Figure 4a. The TE barrier is found to be 0.33 eV which agrees with the barrier extracted from the TS. The temperature dependence of the barrier is seen on Figure 4b and shows a small variation with temperature around the value of the TS barrier. The small variation might be a result of the approximate nature of equation 1 and 2 or of the non self-consistent treatment of the temperature dependence. To shed light on the deviation between the TS and TE barrier and the barrier extracted from the projected DOS, we investigate the density of states available in each electrode.

The experimentally extracted TE barriers for the heterophase devices extracted from the projected DOS, the TS and using the TE model. The TS barriers assume full transmission at 1% of the maximum transmission and the TS barriers are extracted in a temperature range of 300-450 K.

| Doping | \(\Phi_{\text{DOS}}\) | \(\Phi_{\text{TS}}\) | \(\Phi_{\text{TE}}\) |
|--------|----------------|----------------|----------------|
| \(N_D = 4.9 \times 10^{11}\) cm\(^{-2}\) | 0.82 eV | 0.83 eV | 0.60 eV |
| \(N_A = 4.9 \times 10^{11}\) cm\(^{-2}\) | 0.18 eV | 0.32 eV | 0.33 eV |

TABLE I. Calculated barriers for 1T’-1H heterophase devices extracted from the projected DOS, the TS and using the TE model. The TE barriers assume full transmission at 1% of the maximum transmission and the TS barriers are extracted in a temperature range of 300-450 K.

Figure 5 shows the JDOS and shows an onset much below the barrier extracted from the DOS.

\[
\rho_{L,R}(\varepsilon) = \int \rho_L(\varepsilon, k_L) \rho_R(\varepsilon, k_R) \delta(k_L^\perp - k_R^\perp) d\varepsilon d\delta \tag{4}
\]

\(\rho_L\) is the density of states of a pure 1T’ device and \(\rho_R\) is the density of states of a 1H device. Figure 5a shows the calculated JDOS of the p-doped device. It is seen that no states are available for transport at the DOS barrier height. Figure 5a and c compare the \(k_y\)-dependence of the joint DOS and transmission. The \(k_y\)-dependence of the transmission clearly follows the joint density of states. This illustrates how the TE of a pristine interface can be limited by quantum mechanical effects resulting in a TE barrier which is larger than the barrier seen in the projected DOS. The joint density of states of the n-doped device can be found in the Supplemental Material.

The experimentally extracted TE barriers for the heterophase devices at zero gate voltage are 10 meV at 300-450 K for a n-type device with no estimation of the doping level and 24 meV at 150-300 K for a p-type device with an estimated doping level corresponding to our calculations. The gap between experiments and calculation can therefore by no means be bridged by these calculations alone. However, the illustrated discrepancy between the three methods for barrier extraction highlights the difficulty in comparing a barrier extracted from an equilibrium calculation and a barrier extracted using the thermionic emission model. The fabricated devices differ from our devices since they are few layer devices.
rather than a monolayer devices, since they are placed on a substrate, and since we must expect that defects will be present. The presence of defects has been shown to lower the Schottky barrier for metal contacted 2H MoS$_2$[39] and to lower the contact resistance in 1T-2H MoS$_2$ heterojunctions.[40] Furthermore, we note that our calculations do not include electron-phonon interactions which could lead to phonon assisted tunneling. Inelastic transport has previously been shown to have a large effect on the transmission, for instance, it strongly dominates the restriction of momentum conservation causes a large deviation of the joint density of states of the barrier from even a metal contacted 2H MoS$_2$[41].

One might speculate that a combination of these effects results in the very low measured barriers.

In conclusion, we have extracted the Schottky barriers of monolayer MoTe$_2$ 1T’-1H heterostructure devices with either n- or p-doping using three different methods. Our results show that the extracted barrier heights are sensitive to the extraction method and varies significantly between methods. This highlights the challenges of comparing Schottky barriers between different extraction methods. For the n-type device, tunneling lowers the barrier extracted by the TE model compared to the barrier extracted from the projected DOS and TS. The barrier lowering persists up to 600 K. For the p-type device, the restriction of momentum conservation causes a large deviation between the barriers extracted from either the TS or the TE model and the barrier seen in the projected DOS. This effect illustrates how new challenges appear when dealing with pristine devices and will become increasingly relevant as the quality of 2D samples improves.

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