Absence of a Band Gap at the Interface of a Metal and Highly Doped Monolayer MoS$_2$

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Supporting Information

ABSTRACT: High quality electrical contact to semiconducting transition metal dichalcogenides (TMDCs) such as MoS$_2$ is key to unlocking their unique electronic and optoelectronic properties for fundamental research and device applications. Despite extensive experimental and theoretical efforts reliable ohmic contact to doped TMDCs remains elusive and would benefit from a better understanding of the underlying physics of the metal−TMDC interface. Here we present measurements of the atomic-scale energy band diagram of junctions between various metals and heavily doped monolayer MoS$_2$ using ultrahigh vacuum scanning tunneling microscopy (UHV-STM). Our measurements reveal that the electronic properties of these junctions are dominated by two-dimensional metal-induced gap states (MIGS). These MIGS are characterized by a spatially growing measured gap in the local density of states (L-DOS) of the MoS$_2$ within 2 nm of the metal−semiconductor interface. Their decay lengths extend from a minimum of ~0.55 nm near midgap to as long as 2 nm near the band edges and are nearly identical for Au, Pd, and graphite contacts, indicating that it is a universal property of the monolayer semiconductor. Our findings indicate that even in heavily doped semiconductors, the presence of MIGS sets the ultimate limit for electrical contact.

KEYWORDS: Transition metal dichalcogenide, scanning tunneling microscopy/spectroscopy, 2D semiconductor, band mapping, ohmic contact, molybdenum disulfide

Since the onset of mechanical exfoliation as a means to isolate thin layers of van der Waals materials, a wide array of research has been conducted on characterization, synthesis and device applications. In particular, extensive efforts have been directed toward transition metal dichalcogenides (TMDCs) due to their electronic$^{1,2}$ and optoelectronic$^{3-6}$ properties. Low-resistance ohmic contacts are critical for investigating and utilizing these material properties. Ohmic contacts enable ambipolar conduction, enable high “on” current,$^{7,8}$ and allow efficient extraction of photoresponse in optoelectronic devices.$^{3,9}$ A number of methods for achieving low-resistance contacts have been employed in the past:$^{10}$ optimizing contact geometry (top/edge contacts),$^{11}$ optimizing contact material,$^{12-14}$ doping underneath contacts,$^{15,16}$ gating contacts,$^{17}$ phase engineering,$^{18}$ insertion of tunnel barriers between the metal and semiconductor,$^{18}$ and so forth. However, despite extensive experimental and theoretical$^{19,20}$ efforts, reliable high-quality contact to these materials still remains elusive and efforts toward

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The large n-type carrier concentration is highlighted by the yellow dotted box in the zoom-in inset. The large n-type carrier concentration is highlighted by the yellow dotted box in the zoom-in inset. (b) Schematic of the device and experimental setup for Au−MoS2 or Pd−MoS2 junctions made by evaporating thin sharp Au or Pd contacts through a shadow mask 5 μm from the monolayer MoS2 surface. Topographies and line spectroscopies are taken across the edge of the contact as highlighted by the yellow dotted box in the zoom-in inset. (c) A representative STM topographic image of the edge of a graphite electrode atop the monolayer MoS2 film. The image shows the sharpness of the junction as well as the uniformity of both the MoS2 and graphite adhering to the SiO2 substrate. The insets show atomic resolution topographies for each material, which have been used to confirm the lattice constants of each respective material. The white scale bar in the insets is 1 nm and the intensity bars represent 0 to 150 pm for graphite and 0 to 200 pm for MoS2. STM topography set points are 1 V, 300 pA for the large area junction, 3 V, 100 pA for the graphite inset, and 4 V, 500 pA for the MoS2 inset.

Figure 1. (a) Schematic of the device and experimental setup for graphite−MoS2 junction. Vg is the sample-probe bias voltage and Vc is the gate voltage. Topographies and line spectroscopies are taken across the edge of the few-layer graphite contact, as highlighted by the yellow dotted box in the zoom-in inset. (b) Schematic of the device and experimental setup for Au−MoS2 or Pd−MoS2 junctions made by evaporating thin sharp Au or Pd contacts through a shadow mask 5 μm from the monolayer MoS2 surface. Topographies and line spectroscopies are taken across the edge of the contact as highlighted by the yellow dotted box in the zoom-in inset. (c) A representative STM topographic image of the edge of a graphite electrode atop the monolayer MoS2 film. The image shows the sharpness of the junction as well as the uniformity of both the MoS2 and graphite adhering to the SiO2 substrate. The insets show atomic resolution topographies for each material, which have been used to confirm the lattice constants of each respective material. The white scale bar in the insets is 1 nm and the intensity bars represent 0 to 150 pm for graphite and 0 to 200 pm for MoS2. STM topography set points are 1 V, 300 pA for the large area junction, 3 V, 100 pA for the graphite inset, and 4 V, 500 pA for the MoS2 inset.

Ultrapure vacuum scanning tunneling microscopy (UHV-STM), the probe used in this study, provides the atomic-scale resolution necessary to investigate the lateral properties and precise band alignment but is experimentally more challenging for a number of reasons. Ultraclean, conducting samples are necessary, which has been hindered by residue from photoresist and standard polymer transfer techniques, as well as the difficulty of performing STM on an insulating substrate. A sharp contact edge is also imperative for an abrupt metal−semiconductor junction. Furthermore, optical resolution limitations resulting from the large optical working distance from outside a UHV chamber to samples within the UHV chamber also make it difficult to approach STM probes to small-area TMDC samples.

In this work, we fabricate <10 nm thick top contacts with nanometer-scale edge sharpness atop high-quality, large-area, heavily n-type monolayer MoS2 films allowing UHV-STM and scanning tunneling spectroscopy (STS) atomic-scale characterization of the interface. The large n-type carrier concentration is used in order to minimize Schottky barrier effects and examine contacts near the ohmic regime. We also investigate the impact of different metal properties by studying three different types of metal−MoS2 top contact junctions. Graphite and gold (Au) were chosen as metals of interest due to their use as reasonably low-resistance top contacts to MoS2.26 Palladium (Pd) was also chosen due to its high work function and previous works achieving p-type contact to MoS2 using Pd.27 Monolayer MoS2 films were grown directly on Si/SiO2 substrates with nearly uniform growth across 4-in. wafers.28 More than 95% of the film area consisted of monolayer MoS2 with an occasional patch of bilayer or trilayer MoS2 (such patches were avoided in this study). Optical absorption, photoluminescence (PL), and Raman spectroscopy were used for preliminary sample quality characterization. Monolayer films showed a PL peak at 1.87 eV, in confirmation with previous PL on high quality exfoliated and CVD samples in other works. After preliminary film quality characterization, wafers were cleaved into 3 mm × 10 mm pieces and split into three batches for fabrication with different contact metals.

In Batch 1, a graphite−MoS2 top contact junction was created by depositing a thin exfoliated flake of graphite (about 2 nm-thick), providing a naturally sharp contact edge, onto monolayer MoS2 using a polymer dry transfer technique (Supporting Information). To enable collection of the STM tunneling current, contact to the graphite and MoS2 was formed by Au evaporation through a shadow mask to preserve sample cleanliness (see Figure 1a).

Note that for Batch 1, in contrast to Batches 2 and 3, the evaporated metal is merely for collecting tunneling current because the graphite−MoS2 junction of interest is atop an insulating substrate of SiO2. Figure 1a shows...
a schematic of the fabricated structure as well as the electrical connections for the STM experiment. For Batches 2 and 3, 8 nm of Au or Pd, respectively, was directly deposited atop the MoS₂ film (without exfoliated graphite), as contacts of interest for metal−MoS₂ junction characterization. The metals were evaporated through a shadow mask that was designed and positioned to lie only ~5 µm above the MoS₂ film (Supporting Information), producing sharp Au−MoS₂ and Pd−MoS₂ junctions, while preserving the cleanliness of the samples. Figure 1b is a schematic of the Au−MoS₂ and Pd−MoS₂ sample batches.

The samples were then loaded into a UHV scanning tunneling microscope and annealed at 100 C for a minimum of 2 h, to minimize surface contaminants. To ensure sharp, high quality tips by preparing and calibrating tips on single crystalline Au(111). During UHV-STM measurements, the samples (metal contacts and MoS₂ films) were grounded and the probe was biased to establish tunneling current. All measurements were taken at room temperature since contact resistance becomes very large at cryogenic temperatures preventing accurate measurement of STM tunneling current. To map the spatial local density of states (L-DOS) of the junctions, STS dI/dV₅s were taken at 204 equally spaced points across each metal−MoS₂ junction (341 for 2.5 µm line profile). For each dI/dV, a sample-probe voltage difference of 2 V and initial set point of 500 pA for graphite (100 pA for Au and Pd) was used to adjust the sample-probe distance after which the feedback was frozen and sample probe voltage was ramped from 2 to −2 V while measuring tunneling current. The dI/dV₅s, proportional to the L-DOS, were calculated by taking numerical derivatives of the STS tunneling current profiles. Note that dI/dV only provides a value proportional to the L-DOS rather than the actual magnitude. Thus, the method can be used to determine band gaps and the L-DOS functions up to a multiplicative constant. This constant can be treated as nearly uniform within the individual materials due to the uniformity of the L-DOS within each material at the voltage set-point of 2 V (which determines the constant), thus the spatially varying L-DOS within each individual material can be compared; however, because of the difference in L-DOS at 2 V between different materials only the L-DOS shapes can be compared between different materials. By allowing the system to stabilize over several days and minimizing thermal sources, lateral drift was minimized to less than a nanometer per dI/dV line profile, after which it was retroactively eliminated using a reference topography profile taken immediately before the line profile.

Figure 1c shows an STM topographic image at the edge of the graphite contact atop the MoS₂ film. The image reveals the sharpness of the graphite−MoS₂ junction, as well as the uniformity of the monolayer MoS₂ film and the graphite top contact. The two materials adhere to the natural roughness of the SiO₂ substrate. The MoS₂ roughness is comparable to the roughness of single-layer graphene on SiO₂, while the graphite roughness is significantly less due to the stiffness of a larger number of stacked layers. The sharp, abrupt boundary at the graphite−MoS₂ interface, as well as the cleanliness of the surfaces of both materials, are ideal for spatial L-DOS characterization. Topography of the Au−MoS₂ and Pd−MoS₂ junctions are provided in the Supporting Information. The evaporated Au and Pd top contacts are inherently a coalescence of grains from the evaporation deposition. Thus, they do not have straight edges like exfoliated graphite. Given this limitation, the closely placed shadow masks through which the Au and Pd were evaporated ensured abruptly ending edges defined by the local grains at the edges, allowing L-DOS characterization across the Au−MoS₂ and Pd−MoS₂ junctions as well.

We first investigate the material properties of monolayer MoS₂ and graphite at large length scales by analyzing a 2.5 µm dI/dV line profile across the graphite−MoS₂ junction (Figure 2a). The figure shows a color map of the dI/dV₅s as a function of position and sample-probe voltage difference (all normalized to the same value at 2 V in order to put all spectra on the same colorscale), spatially aligned with a concurrently measured sample-probe displacement below the color map. This long dI/dV spatial profile provides the L-DOS of graphite and MoS₂ far away from the junction, which allows prediction of the graphite−MoS₂ band alignment and verifies the spatial uniformity of the L-DOS at the junction, which allows prediction of the graphite−MoS₂ band alignment and verifies the spatial uniformity of the L-DOS at

![Figure 2](image-url)
Figure 3. (a–c) Color maps of short (~9 nm) dI/dV line profiles taken across the (a) graphite–MoS$_2$, (b) Au–MoS$_2$, and (c) Pd–MoS$_2$ junctions consisting of 204 equally spaced points. Below the color map is a simultaneously measured STM topography profile revealing the approximate location of the contact edge. The color map reveals a metallic L-DOS profile up until the edge of the electrode. Immediately to the right of the interface into the MoS$_2$ side, there is still no observed L-DOS gap. A L-DOS gap emerges at a finite distance into the MoS$_2$ side and grows for about 2 nm before reaching the far from junction magnitude. (d) Individual dI/dV line spectra taken at 0, 0.5, 1, 1.5, and 2 nm from the junction, as shown by the dashed lines in panel a. The individual line spectra confirm the apparent evolution seen in the color map, as the conduction and valence band edges are gradually defined with distance from the junction, reaching the far from junction, full MoS$_2$ band gap several nanometers from the junction.

large length scales. It is clear from the dI/dV line profile that there is an abrupt change in the dI/dV at the graphite–MoS$_2$ junction, signifying a change in the L-DOS, after which there is long-range uniformity of L-DOS of both graphite and MoS$_2$ individually. Small fluctuations in the L-DOS can be attributed to defects in the materials.

On the MoS$_2$ side of the dI/dV color map in Figure 2a, the region of nearly zero L-DOS (red color) indicates the band gap of the material, clearly not present on the graphite side. To analyze the details of the dI/dV profile of each material far from the junction, Figure 2b displays a spatially averaged dI/dV curve of the monolayer MoS$_2$ and in inset displays the graphite. These averaged dI/dVs can be interpreted as the DOS of the graphite and MoS$_2$ when not modified by junction physics. The graphite DOS exhibits metallic properties, as there exists no region with zero DOS as a function of sample-probe voltage, hence there is no band gap. All three contact metals (Pd, Au, graphite) studied show a similar metallic DOS, consistent over many different samples. The MoS$_2$ DOS shows a considerable band gap, as the DOS plunges to nearly zero from about −1.75 to about +0.35 V alluding to a quasiparticle gap size of about 2.10 eV, similar to that of previous reports on MoS$_2$. In this conversion of quasiparticle band gap energy size in eV from dI/dV sample-probe voltage thresholds, it has been calculated that tip-induced band bending and image charge potential are nearly equal and opposite and thus allow direct conversion of the observed gap in the dI/dV spectrum to quasiparticle band gap. It is clear from the dI/dV that the conduction band is closer to the Fermi level (represented by zero sample-probe voltage) than the valence band implying that the MoS$_2$ film is n-type. The n-type behavior is in agreement with typical CVD MoS$_2$ samples that have been studied both by transport and STM measurements. Further, we observe band-tails (characterized by ill-defined band edges beyond what is expected by the Fermi–Dirac broadening) signifying large effective sheet charge concentrations, which we use to study contacts near the ohmic regime. It is tempting to determine the precise carrier concentration based on the apparent conduction band edge location with respect to the Fermi level, however, the presence of an unknown amount of contact doping caused by the probe makes this inaccurate.

Next, we investigate the local properties of graphite–MoS$_2$ junction by analyzing a short spatial dI/dV profile across the junction. Figure 3a shows a color map of the dI/dV calculated from a ~9 nm STS line scan of across the junction with the concurrently measured sample-probe displacement. The sample-probe displacement shows that the contact edge is rounded for several nanometers which we primarily attribute to real physical rounding of the imperfect step edges. We have verified this by...
obtaining consistent profiles with a variety of different STM tips carefully prepared on Au(111). A comparison of line cuts taken with different tips is also shown in the Supporting Information. We define the approximate junction position, the zero on our plots, as where the sample-probe displacement flattens out to that of the MoS₂ layer, signifying that the STM probe is definitively tunneling only into MoS₂. For analysis of effects in the MoS₂, this is a useful definition of the junction location because it sets a clear boundary beyond which there is no possibility that finite tip radius causes partial tunneling into the metal. Any uncertainty in location due to tip convolution would place the real junction closer to the metal, not further, thus data analyzed beyond this position is not affected by finite tip radius interaction with the metal. Continuing to analyze the dI/dV color map in Figure 3a, while the probe is above the graphite the color map shows that the electronic spectra remain metallic, similar to the graphite region observed in the long dI/dV line profile in Figure 2a. Once the probe crosses onto purely MoS₂, a finite gap begins to grow over the next 2 nm. This can be seen as both the valence and conduction band edges begin near 0 V and gradually shift toward their long-range MoS₂ film values, observed earlier in Figure 2a. No clear band-bending and depletion region is observed in our measurements. Color maps of Au–MoS₂ and Pd–MoS₂ junctions in Figure 3b,c reveal the same behavior. To ensure that this effect is not a color map/plotting artifact, Figure 3d shows several individual dI/dV profiles at different spatial positions in the region of the evolution from the graphite–MoS₂ interface. Comparing the dI/dV profiles, we see a gradual formation of the valence band edge based on the negative voltage tails, which begin metallic and gradually become more and more flat, approaching the profile seen earlier in Figure 2b. We can also see the conduction band edge forming, although masked by its proximity to the Fermi level. Thus, the L-DOS profiles show that the dI/dV gap grows with increasing distance from the edge of the graphite–MoS₂ junction. The same effect is observed in Au–MoS₂ and Pd–MoS₂ on similar length scales, as can be verified by Figure 3b,c.

We find that the observations are best explained by the continuum of metal-induced gap states (MIGS). Heine showed that the presence of metal at the interface creates...
evanescent states inside the semiconductor band gap which decay exponentially from the junction. These tailing states could be visualized as virtual gap states of the complex band structure of the semiconductor as shown in Figure 4a. For states that lie within the band gap, only the imaginary part of wavevector $k$ exists, resulting in exponential decay characterized by the decay length, $\delta = \frac{1}{q}$, where $q = i k$ and $k$ is the standard plane wave vector. Mönch developed a one-dimensional virtual gap state model to quantify this virtual gap state decay length (Supporting Information). The model reveals that as a function of energy the decay lengths diverge at the band edges, whereas the decay lengths are minimum at the charge neutrality level (CNL), as plotted in Figure 4b. The CNL is at the center of the band gap in this model but shifts when effective electron and hole masses have any mismatch, which is the case of MoS$_2$. The minimum wave function decay length in this model is $\delta_{\text{min}} = \frac{2 \pi E_F}{m_e a E_F}$, where $h$ is the Planck constant, $m_e$ is the free electron mass, $a$ is the lattice constant, and $E_F$ is the semiconductor band gap. The decay length of the L-DOS is half the value of the wave function decay length, as the charge density is proportional to the wave function squared. Using the experimental values of in-plane lattice constant $a = 0.32$ nm and single particle band gap $E_F = 2.1$ eV for monolayer MoS$_2$, the theoretical minimum decay length for metal–MoS$_2$ interfaces is 0.36 nm.

To confirm the presence of MIGS, we investigate the spatial evolution of the MoS$_2$ $dI/dV$s within the gap. The inset of Figure 4b shows the logarithm of $dI/dV$ intensity at a series of fixed energies within the band gap, as a function of distance from the graphite–MoS$_2$ interface. The linear decrease in $\log(dI/dV)$ with distance from the junction signifies the exponential decay of the MIGS in confirmation with theory. Linear regression fits on position versus $\log(dI/dV)$ at each voltage within the band gap allow determination of the experimentally observed decay length of the MIGS. It is important to keep in mind that determination of these exponential decay lengths does not require the ability to probe the precise junction edge thus is not inhibited by tip convolution. Figure 4b shows the experimental decay lengths as a function of energy for the three different metal–MoS$_2$ junctions, Au–MoS$_2$, Pd–MoS$_2$, and graphite–MoS$_2$, as well as the decay lengths predicted by the previously mentioned theoretical model. The MIGS decay lengths for the three contact metals are almost identical, as the decay length is determined solely by semiconductor parameters, as in Mönch’s model (Supporting Information). The minimum decay length of 0.55 ± 0.10 nm occurs around $-0.6$ V, slightly shifted toward the conduction band edge from midgap. This experimental decay length is quite short compared to the 0.55 nm minimum decay length of the MIGS, we observe the decay length can vary from a minimum of 0.55 ± 0.10 nm to greater than 1 nm near the band edges, in good agreement with theory. The MIGS decay length is shown to be independent of the contact metal and solely due to the mismatch in the periodic potential of the lattice. The shift of the minimum decay length in energy from midgap is due to the mismatch in the effective electron and hole masses in MoS$_2$, which shifts the CNL from midgap. It has been found that the CNL in monolayer MoS$_2$ is shifted toward the conduction band, the same direction as the shift in minimum decay length in our experimental results. At the conduction band edge, we observe the expected divergence in the MIGS decay lengths, in concurrence with the theoretical curve. Approaching the valence band edge, there is a gradual increase in decay length, although the MIGS persist into the valence band before diverging. This continuation of MIGS into the valence band, in disagreement with models, has been previously observed. The inability of the simple model to capture this effect is likely because the model fails to account for the precise nature of the valence band and the localization of carriers near the valence band edge, which is affected by defects and film quality. MIGS have been experimentally observed in 1D$^{15}$ and 3D$^{36}$ systems but to our knowledge this is the first experimental confirmation of MIGS in 2D materials.

Additionally, it is interesting that no Schottky barrier behavior is observed. Schottky barriers are normally characterized by spatial band-bending inside the semiconductor which indicates the depletion region inherent due to Fermi level misalignment between the metal and semiconductor, as shown in Figure 4c. This is clearly not observed in our experimental $dI/dV$ spatial line profiles implying that if there is any Schottky barrier, it is within several angstroms of the real junction where we cannot resolve it or is concealed in the MIGS spatial extent, as depicted in Figure 4c. Thus, in these highly doped monolayer semiconductors, MIGS set the ultimate limit for electrical contact. This short upper limit on the depletion width is due to the large effective sheet concentration of our MoS$_2$ film, which we can estimate by employing an analytic model for 2D depletion width ($\delta_{\text{eff}}$) given by Gurugubelli et al., where $\delta_{\text{eff}} = \frac{4 e c_0 (\phi_{bi} - V)}{\pi a N_{\text{sp}}}$.

Here $c_0$ is effective dielectric constant, $\phi_{bi}$ is the built-in potential, $V$ is the applied bias, $q$ is the elementary charge, and $N_{\text{sp}}$ is the effective sheet carrier concentration. As opposed to the standard 3D model, the 2D model is suited for systems with 2D materials such as our metal–MoS$_2$ junctions as it considers the role of the significant out-of-plane electric field which is absent in 3D model. On the basis of the MIGS spatial extent and the tip convolution uncertainty, we can assume a depletion width shorter than 1.5 nm. This implies a lower limit on the effective sheet carrier concentration using the $dI/dV$ width model of at least $\sim 10^{13}$ Carriers/cm$^2$, a reasonable carrier concentration for degenerate n-type MoS$_2$ films. We also find that the application of a back gate voltage from $-20$ to $80$ V (corresponding to a sheet carrier density of $5 \times 10^{12}$ Carriers/cm$^2$) does not significantly shift the $dI/dV$ spectrum, further verifying this degenerate carrier concentration. This agrees with our previously mentioned findings that the MoS$_2$ film has a high carrier concentration due to the presence of band tails in the DOS. Future investigations of MoS$_2$ and other TMDC samples that are more intrinsic will further the understanding of contacts by allowing direct observations of the Schottky barrier, depletion width, and MIGS, which can be used to compare metals, study gate dependence, and investigate novel methods to avoid Fermi level pinning (which has in the past been attributed to MIGS).

In conclusion, we investigated the metal–MoS$_2$ junction of three different top contacts, graphite, Au, and Pd, on heavily n-type monolayer MOCVD grown MoS$_2$ using UHV-STM and STS. By fabricating clean nanoscale sharp contact edges on large area MoS$_2$ films atop SiO$_2$, we have provided subnanometer-scale spatial spectroscopic characterization of the evolution of the $dI/dV$ proportional to the L-DOS, of the MoS$_2$ in the nanoscale vicinity of all three metal–MoS$_2$ junctions. $dI/dV$ line profiles across the junctions reveal a gradually growing L-DOS gap in the MoS$_2$, originating at the metal–MoS$_2$ junction. The effect is attributed to MIGS originating from the contact metals, decaying into the MoS$_2$. By analyzing the energy dependence of the decay length of the MIGS, we observe the decay length can vary from a minimum of 0.55 ± 0.10 nm to greater than 1 nm near the band edges, in good agreement with theory. The MIGS decay length is shown to be independent of the contact metal and solely dependent on the intrinsic mismatch in the periodic potential of the lattice.
determined by the parameters of the semiconductor, as predicted in theory. We also show that in these contacts, no Schottky barrier is observed for as long as the spatial extent of the MIGS indicating that MIGS set the ultimate limit for highly doped monolayer semiconductor electrical contact.

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**ASSOCIATED CONTENT**


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