Electronic Supplementary Material (ESI) for Nanoscale.
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Electronic supplementary information for: Control of the Metal/TMDC Contact Properties Using 2-Dimensional Buffer Layers†

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1 Bi- and multi-layer graphene on WSe_2

For the graphene on WSe_2 system, we were able to resolve the difference between BL graphene and FL graphene buffer layer using a boron-doped diamond tip. The $I(V)$ curves obtained for the tip/BL-graphene/WSe_2 and tip/8L-graphene/WSe_2 are compared in Figure S1. Similar to Figure 3 in the main text, for the BL graphene the curve is clearly non-linear, while for ML graphene the characteristics are ohmic (possible deviations from ohmic behaviour could not be resolved). This shows that the effect described in the main text is general for the discussed band alignment scheme (n-type Schottky contact, p-doped graphene).

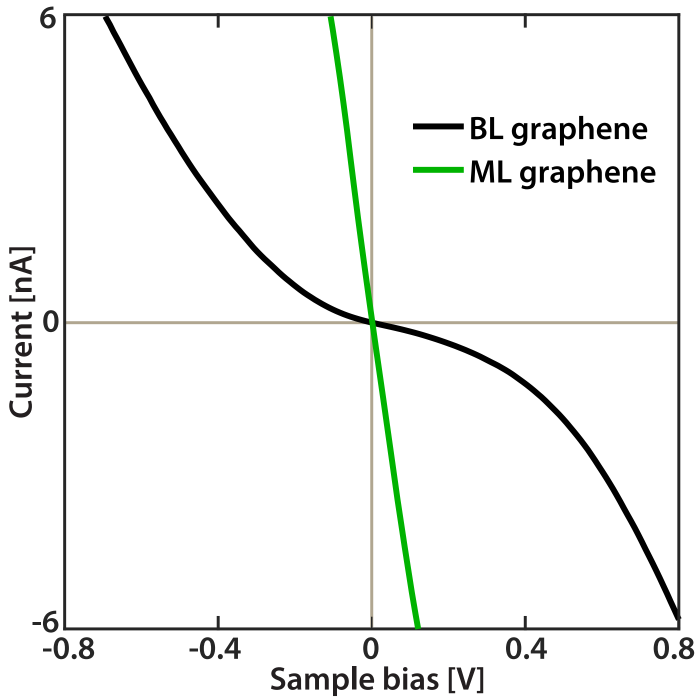

Fig. S1 $I(V)$ curves for tip/BL-graphene/WSe_2 and tip/8-layers-graphite/WSe_2 obtained using the boron-doped diamond tip.

2 Force-Distance measurements

Figure S2 presents an exemplary Force-Distance (FD) curve obtained for a Pt tip on the WS_2 substrate, taken while performing the experiments shown in Figure 2 of the main text. The measurement is taken while approaching the tip to the substrate, *i.e.*, from right to left. After snapping into contact (around 130 nm) the tip is further pressed into the substrate. The force is calculated as $F = k \cdot \Delta V \cdot a^{-1}$, where k is the spring constant of the cantilever, ΔV is the deflection of the cantilever measured by the photodiode and a is the slope of the force-distance curve in the left part of the figure Figure S2. Pressure P at the contact is calculated as $P = F/A$, where A is the area of the contact obtained from the Hertzian model[1]. The electrical measurements with the Pt tip in Figure 2 of the main text were performed at the pressure of around

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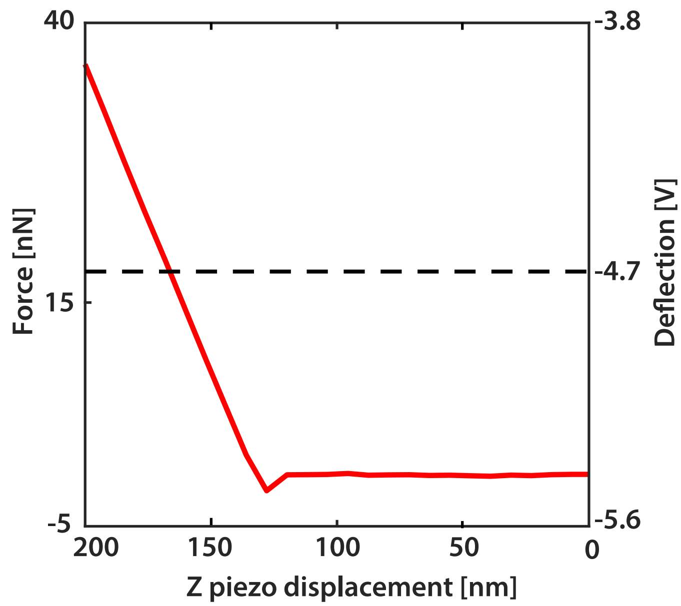

Fig. S2 A Force-Distance curve of Pt tip on WS_2 substrate. The operating set point is marked by the dashed line. Deflection of the cantilever measured by the photodiode (right side of the plot) is translated to the force between the tip and the substrate (left side of the plot). The X axis shows the extension of piezoelectric element controlling the cantilever position. (Negative force means that the tip is 'glued' to the substrate by adhesion forces.)

4 GPa between the metal and the semiconductor.

3 Height profiles

The topography pictures and height profiles, corresponding to Figures 2, 3 and 4 of the main text are shown in Figures S3, S4 and S5, respectively. The number of layers in the main text was obtained by examining the extracted height profiles. Height values for SL graphene Figure S4a and Figure S5a are close to the value expected for inter-planar distance between atomic sheets in graphite ($h = 0.335\,\mathrm{nm}$)[2]. The cantilever of the Pt tip was not designed to measure in such an extreme height resolution regime. Therefore, although we are able to identify the SL and BL graphene in Figure S4, the number of layers and the exact height of the thicker part of the flake (left of the image in Figure S4a), cannot be determined with certainty. Figure S4b shows the lateral force microscopy (friction) image, obtained simultaneously with the topography picture of Figure S4a. The lateral force microscopy highlights the sharp edges of the flake that might appear blurry in the topography image (especially the edge from SL to BL graphene in Figure S4a).

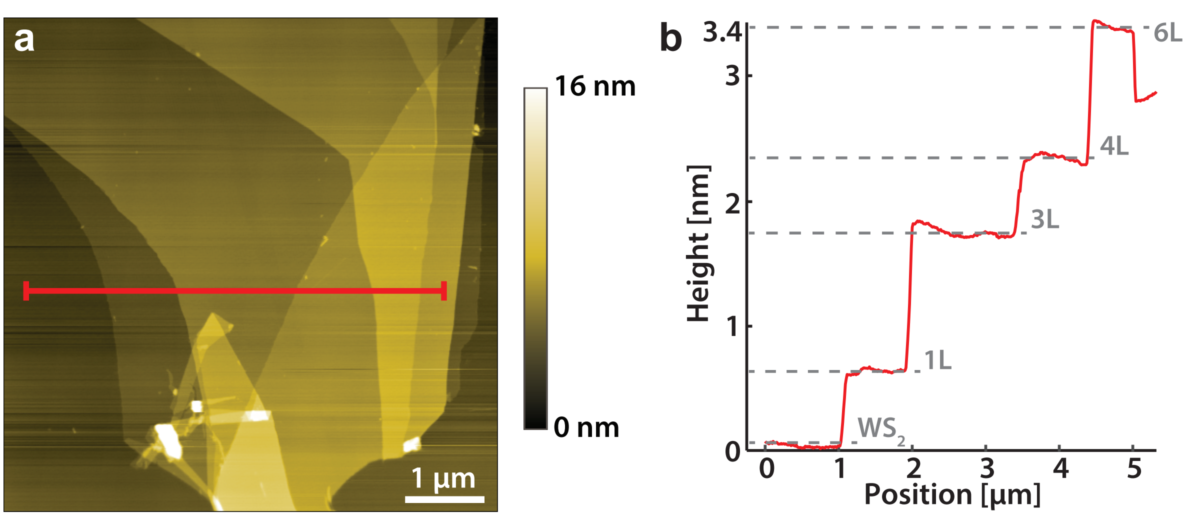

Fig. S3 Topography (a) and the extracted height profile (b), taken along the line shown in (a), for the $MoSe_2$ flake corresponding to Figure 2 of the main text. The measurement was taken with a highly n- doped silicon tip (see methods section of the main text for the tip details). The measured heights are (0.61 ± 0.04) nm for SL, (1.72 ± 0.05) nm for 3L, (2.32 ± 0.05), nm for 4L and (3.37 ± 0.5), nm for 6L.

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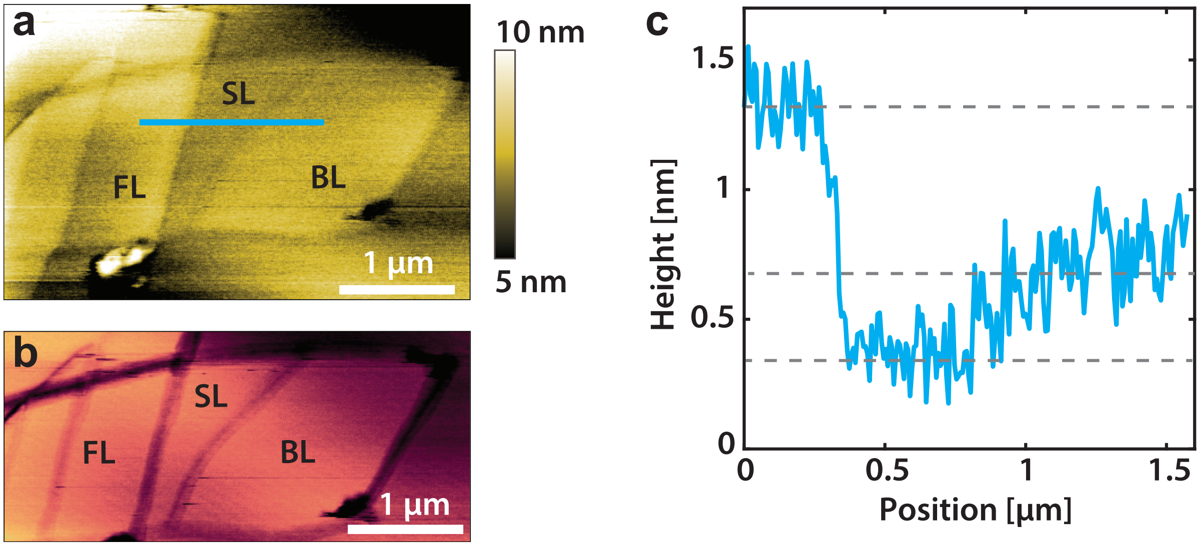

Fig. S4 Topography (a) and the extracted height profile (c), taken along the line shown in (a), for the graphene flake corresponding to Figure 3 of the main text. (b) The simultaneously recorded lateral force microscopy (friction) image, which highlights the edges of graphene layers of various thickness. The measurement was taken with the Pt tip (see methods section of the main text for the tip details). The measured heights are (0.35 ± 0.13) nm for SL, (0.70 ± 0.12) nm for BL and $(1.33 \pm 0.12)\,$,nm for FL (corresponding to 4 layers).

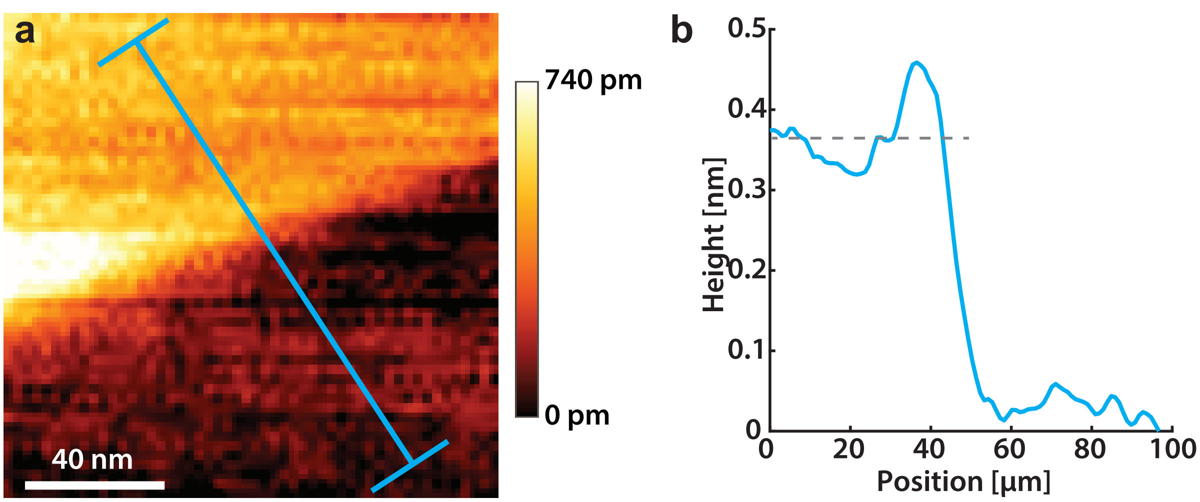

Fig. S5 Topography (a) and the extracted height profile (b), taken along the line shown in (a), for the SL graphene flake corresponding to Figure 4 of the main text. The measurement was taken with the boron doped diamond-tip (see methods section of the main text for the tip details). The height of SL graphene is $h = (0.37 \pm 0.1)$ nm.

4 Fits to $\ln(I)$ vs V curve

Figure S6 shows the thermionic emission model fitting for Figure 2 of the main text. The lines in Figure S6 show the thermionic emission current, given by Equation 1 of the main text, for the obtained Φ_B and n values (Table 1 of the main text).

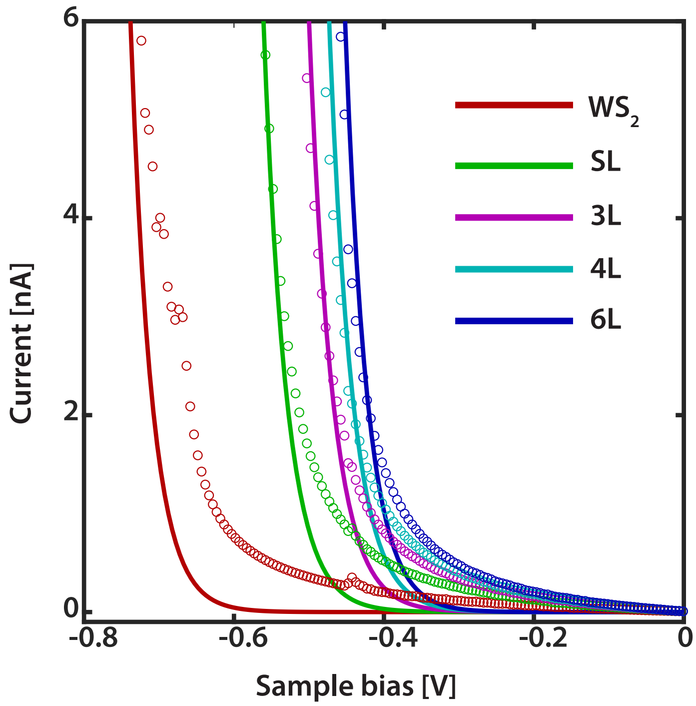

Fig. S6 Average $I(V)$ curves for the WS_2 substrate and the $MoSe_2$ buffer layers of different thickness (open circles). The lines are a thermionic emission model prediction using the extracted Φ_B and n parameters described in the text.

Notes and references

1 B Cappella and G Dietler. Force-Distance Curves by Atomic Force Microscopy. *Surf. Sci. Rep.*, 34(1-3), 1999.

2 Cameron J Shearer, Ashley D Slattery, Andrew J Stapleton, Joseph G Shapter, and Christopher T Gibson. Accurate thickness measurement of graphene. *Nanotechnology*, 27:125704, 2016.

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