Supporting Information

Strain and Charge Doping Fingerprints of the Strong Interaction between Monolayer MoS_2 and Gold

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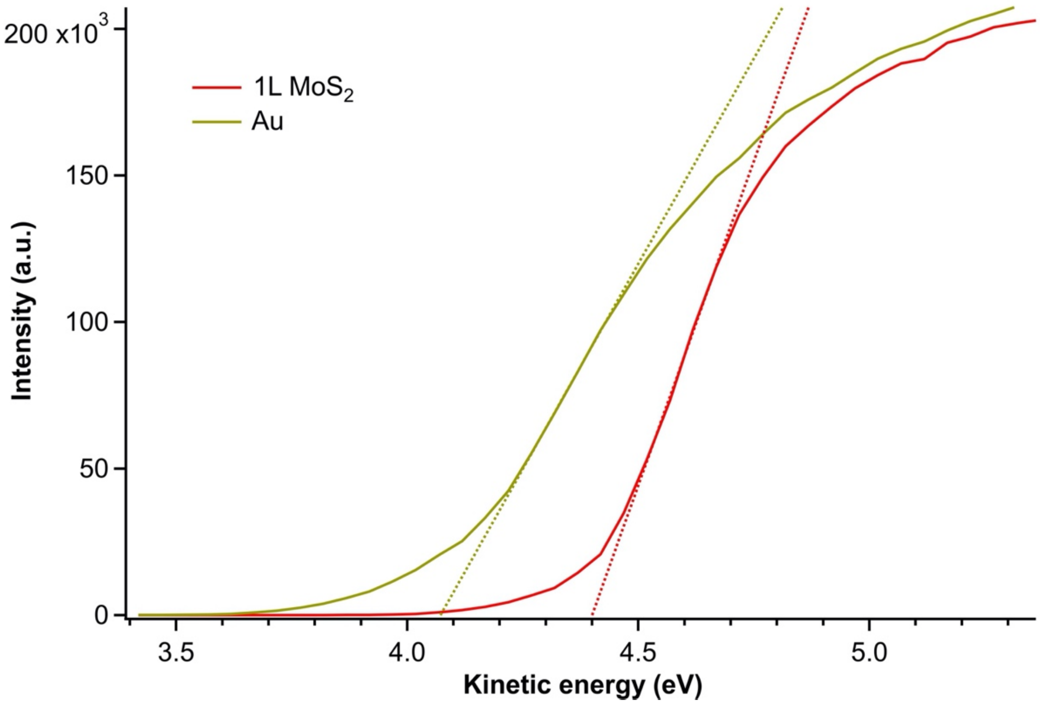
METHODS

Sample Preparation

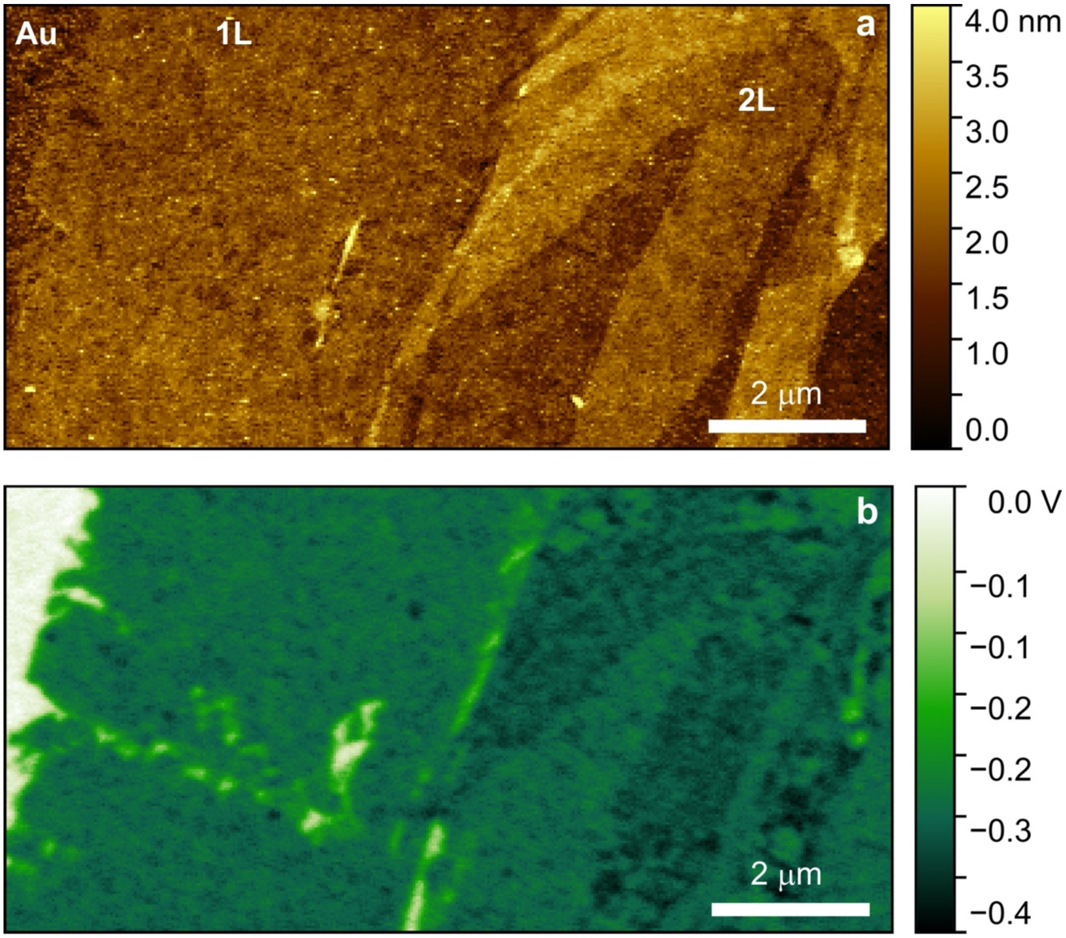
Gold films on 90 nm SiO_2/Si wafers (IDB Technologies Ltd) were prepared by three different methods: magnetron sputtering (CMS-A, Kurt J Lesker Company Ltd), e-beam evaporation (SC4500, CVC Products Inc), and thermal evaporation (DV502-A, Denton Vacuum Inc) followed by peeling from a sacrificial Si substrate.[1] Au thicknesses ranged from 3 to 100 nm and an adhesion layer of 1 nm or 3 nm Ti was employed for the sputtered and e-beam samples, respectively. MoS_2 was exfoliated onto the Au surface from bulk molybdenite crystals (Manchester Nanomaterials Ltd), using a low-stain tape, immediately after the removal of Au from the ultra-high vacuum deposition system. The bulk MoS_2 was peeled from the tape to expose the fresh MoS_2 surface immediately prior to the exfoliation. The samples were not treated in any other way before their characterization and were stored in a vacuum desiccator when not in use.

Characterization

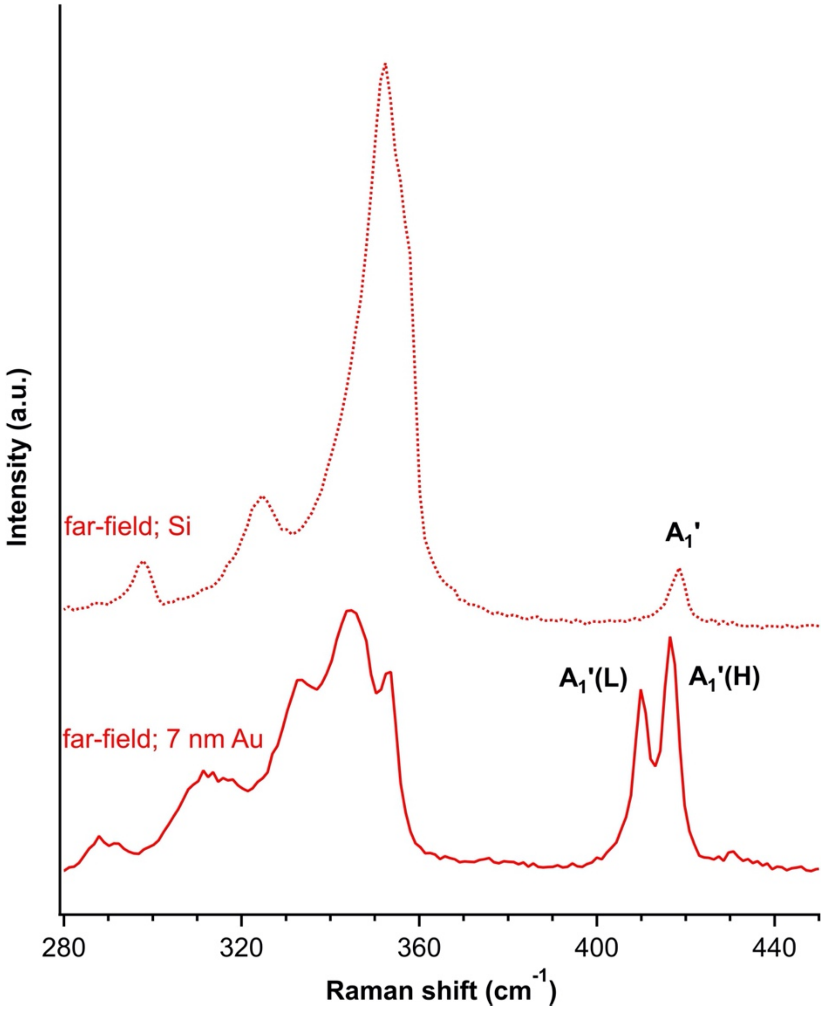
The exfoliated MoS_2 was inspected, and its thickness determined, using a Nikon L200N Eclipse optical microscope. MFP-3D AFM (Asylum Research) in tapping mode was used to determine the surface roughness. An Icon Dimension AFM (Bruker Corp.) in PeakForce tapping mode using Scanasyst-Air probes was employed for the high-resolution characterization of the surface. Far-field micro-Raman spectra were collected using an inVia Reflex confocal spectrometer (Renishaw plc) with a 532 nm laser and 2400 l/mm grating and LabRAM HR (Horiba Ltd) with a 633 nm laser and 1800 l/mm grating, focused to ~1 μm^2 spot size by a 100× objective. Near-field tip-enhanced Raman spectroscopy was measured using a LabRAM Nano system comprised of HR Evolution spectrometer and OmegaScope-R SPM/Optical system (HORIBA Scientific) with a 633 nm laser, 1800 l/mm grating, and Ag-coated Si tips (App Nano), using 1 s (3 s) integration time for mapping (tip force) measurements, respectively, and <300 μW laser power for each pixel. XPS, UPS, and PEEM were measured in NanoESCA microscope (Omicron). The XPS was collected using a monochromated Al Kα source ($h\nu = 1486.7$ eV), and the UPS was carried out using He I discharge lamp ($h\nu = 21.2$ eV). The XPS spectral calibration was done using the Au $4f_{7/2}$ core level at 84 eV along with the Fermi level edge.



Supporting Figure S1. Work function estimation using ultraviolet photoelectron spectroscopy. Low kinetic energy cut-off spectra of 1L MoS_2 on Au (solid red; 15 nm e-beam) and of bare Au substrate (solid green) measured next to the MoS_2, with the linear fits (dashed) through the inflection points of the curves. The work function (Φ) is determined from the value of the fitted line at zero intensity. Φ_{MoS_2} was found to be ~0.3 eV larger in comparison to Φ_{Au}. Although the absolute Φ values are burdened with uncertainty due to the spectrometer response, their difference yields a reliable work function difference estimate.



Supporting Figure S2. Work function estimation using Kelvin probe force microscopy. a, Topography atomic force microscopy image of an area containing 1L and 2L MoS_2 and the bare Au substrate (50 nm e-beam). **b,** Contact potential difference (CPD) map of **a**, obtained by Kelvin probe force microscopy. The CPD of 1L MoS_2 is lower, and therefore its Φ is larger, than that of the Au substrate. The corresponding difference in the CPD of the two materials, averaged over the whole measured area, is 0.22 V. The trend of increasing work function (decreasing CPD) with the number of layers observed here for 1L/2L MoS_2 is consistent with other literature observations.[2,3]



Supporting Figure S3. Raman spectra of 1L WS_2. Micro-Raman spectra of 1L WS_2 on 7 nm sputtered Au (bottom curve) and on Si/SiO_2 (top curve), collected using 532 nm excitation.

Supporting References

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3. Choi, S.; Shaolin, Z.; Yang, W. Layer-number-dependent work function of MoS_2 nanoflakes. *J. Korean Phys. Soc.* **2014,** *64*, 1550-1555.