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Mapping Local Structure, Electronic and Excitonic Properties at the 2D/3D Interface

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Two-dimensional (2D) materials provide a promising platform to engineer future opto-electronic and quantum devices [1]. Metal integration with 2D materials (2D/3D integration) is routinely required for 2D device fabrication, for example in electronic gating and control [2], as well as to enable exotic light-matter coupling [3]. However, such metallic contacts locally affect the underlying 2D material via orbital hybridization, the creation of mid-gap states, and Fermi level pinning [4]. This can drastically alter the local opto-electronic properties [5]. Further knowledge of the impact of metal deposition on the structural, electronic, and excitonic properties of 2D materials is required for their continued integration into functional devices. As a variety of 2D devices emerge with nanoscale dimensions, it is moreover necessary to understand the influence of 2D/3D integration at high spatial resolution. This is crucial for composite systems where local environment greatly affects the resulting opto-electronic properties [3, 6].

We have explored the local structural, electronic, and excitonic properties of the 2D/3D interface using a combination of atomic resolution STEM, *in situ* ultra-high vacuum (UHV) TEM, and monochromated high-energy resolution electron energy-loss spectroscopy (EELS). We achieve low defect-density 2D/3D interfaces using slow metal evaporation rates onto ultra-clean suspended 2D materials in UHV (Figure 1a). We demonstrate epitaxial, facetted nanoisland growth for several metals (Au, Ti and Nb) on a variety of 2D materials, controlling the temperature of deposition. Islands self-assemble into their equilibrium Winterbottom shapes [7], as confirmed by heating *in situ* in UHV-TEM. This allows a quantitative estimate of the 2D/3D quasi-vdW interface energy (Figure 1b), which is similar to that of a van der Waals bond. We describe how these suspended interfaces, with low defect density and weak quasi-vdW bonds, open novel opportunities for 2D/3D device fabrication.

Using these well-defined interfaces, we study the effect of 2D/3D integration on the local excitonic spectrum of MoS_2. Here, the epitaxial arrangement at the MoS_2/Au interface can be accurately determined via moiré contrast (Figure 2a), and electronic charge density is also modulated according to the moiré period [8]. Monochromated STEM-EELS allows local changes of excitonic spectra of MoS_2 to be mapped on the nanometer scale. We utilize low dose EELS acquisition to obtain a high signal-to-noise ratio by recording multiple spectral images; the spectral data can then be aligned through non-rigid registration [9]. We observe a variety of exciton states arising from local n-doping of MoS_2 in contact with Au. These vary in spatial location across the sample (Figure 2b) and can be compared with EELS simulations of the MoS_2/Au system. This allows us to map the effect of Au metal contacts on the local opto-electronic response of MoS_2 with both spectral and spatial resolution. We discuss mechanisms by

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which optimizing this 2D/3D interface is crucial for improving existing devices as well as generating novel 'mixed- dimensional' 2D/3D devices [10].

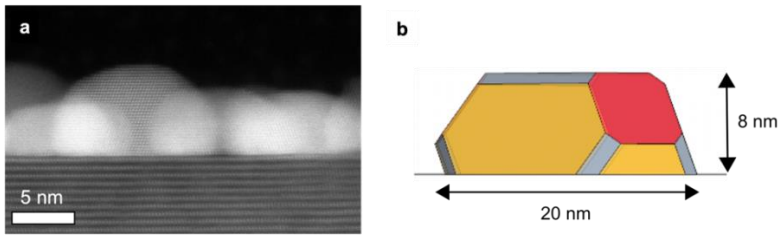

Figure 1: a, Cross-sectional STEM image showing facetted Au/MoS_2 islands as deposited at ultra-low rate in UHV. **b**, Winterbottom construction with estimated interface energy ~0.16 J/m^2, similar to that of the interface between van der Waals layers.

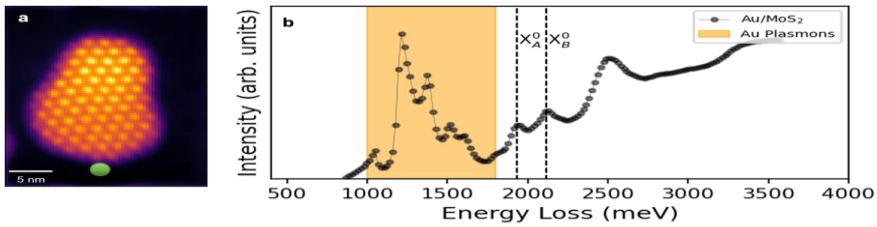

Figure 2: a, HAADF image showing Au island on suspended MoS_2. Epitaxially aligned Au and MoS_2 lattices are noticeable through the well-defined moiré pattern. **b**, Calculated EEL spectrum for the beam position indicated in green.

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