

# The Exact Mechanisms of Ultra-Large Area Metal Assisted Exfoliation

Jakob Ziewer

Fumin Huang

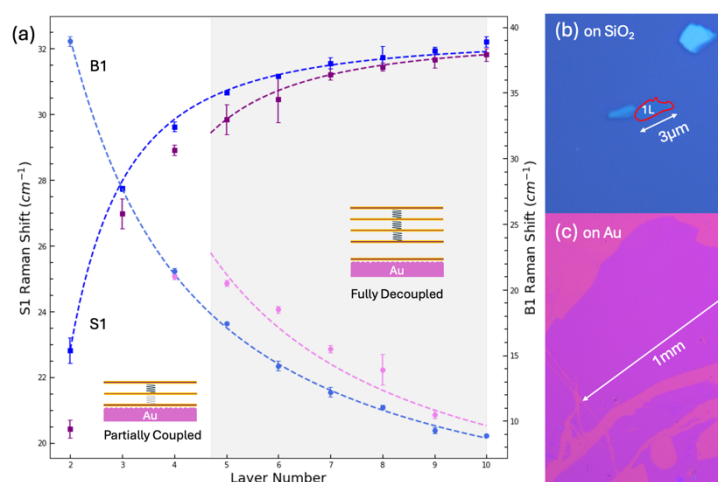
Centre for Quantum Materials and Technologies, Queens University Belfast, Belfast, United Kingdom  
jziewer01@qub.ac.uk

Obtaining large area high quality monolayers of 2D materials is of key importance to both academic and industrial applications. Metal assisted mechanical exfoliation can produce ultra large, high-quality monolayers with sizes up to centimetre scale (only limited by the parent crystal size) thousands of times larger than flakes produced from traditional exfoliation technique using SiO<sub>2</sub> substrates (Fig.1) [1]. The advent of such a ground-breaking technique has significantly advanced the research in 2D materials [2-3]. One intriguing characteristic of metal-assisted exfoliation is that it always preferentially produces large-size monolayer films, whereas multilayers are scarce and small in sizes. The mechanisms for this peculiar phenomenon are unclear and largely unexplored. Here we report experimental investigation of the interlayer forces of exfoliated 2D films through ultra-low frequency Raman spectroscopy [4]. The results provide clear evidence showing that metal substrates significantly weaken the binding force between the bottommost contact layer and the remaining body of 2D materials, leading to the predominant exfoliation of monolayer films. The decoupling effect is dependent on film thickness. For films above 5 layers, the bottom layer is completely decoupled from the top block (Fig.1a). Understanding the key principles of metal-assisted exfoliation is of paramount importance to devise novel strategies of exfoliating 2D films on different substrates and generating films of controllable thickness.

References

- [1] Velický, M.; *et. al.* Mechanism of Gold-Assisted Exfoliation of Centimeter-Sized Transition-Metal Dichalcogenide Monolayers. *ACS Nano* 2018, 12 (10), 10463–10472.
- [2] Hus, S. M.; *et. al.* Observation of single-defect memristor in an MoS<sub>2</sub> atomic sheet. *Nature Nanotechnology* 2021, 16 (1), 58-62.
- [3] Huang, Y.; *et. al.* Universal Mechanical Exfoliation of Large-Area 2D Crystals. *Nat Commun* 2020, 11 (1).
- [4] Huang, S.; *et al.* Low-Frequency Interlayer Raman Modes to Probe Interface of Twisted Bilayer MoS<sub>2</sub>. *Nano Letters* 2016, 16, 1435–1444.

Figures



**Figure 1:** (a) Measured first order breathing modes (B1, circles) and shear modes (S1, squares) of few-layer MoS<sub>2</sub> flakes exfoliated on SiO<sub>2</sub> substrates (blue) and Au substrates (pink). The results on SiO<sub>2</sub> substrates are well fitted with a standard linear chain model (dashed line), but those on Au substrate can only be modelled with decoupled force constants. (b) and (c) Optical images of exfoliated MoS<sub>2</sub> monolayer films.