

The substrate effects on the properties of single-layer MoS₂: enhanced Valley Helicity and enhanced HER catalytic activity

Lun Dai

Yi Wan, Jun Xiao, Jingzhen Li, Xin Fang, Kun Zhang, Lei Fu, Pan Li, Zhigang Song, Hui Zhang, Yilun Wang, Mervin Zhao, Jing Lu, Ning Tang, Guangzhao Ran, Xiang Zhang, Zeyao Zhang, Xiaolong Xu, Zhihong Zhang, Kai Yuan, Kaihui Liu, Yan Li, Yu Ye

State Key Lab for Mesoscopic Physics, School of Physics, State Key Laboratory of Rare Earth Materials Chemistry and Applications, College of Chemistry and Molecular Engineering, Peking University, Peking University, Beijing 100871, China; NSF Nanoscale Science and Engineering Center, University of California, Berkeley, CA 94720, USA

lundai@pku.edu.cn

We demonstrated that epitaxially grown single-layer MoS₂ on a lattice-matched GaN substrate, exhibits strong substrate-induced interactions. The phonons in GaN quickly dissipate the energy of photogenerated carriers in the MoS₂ through electron-phonon interaction, resulting in a short exciton lifetime and an enhanced valley helicity at room temperature (0.33 ± 0.05). The findings highlight the importance of substrate engineering for modulating the intrinsic valley carriers in ultrathin 2D materials. We also demonstrated that fractal-shaped single-layer MoS₂ with large tensile strain synthesized on fused silica is superior to the triangle-shaped MoS₂ grown on SiO₂ for catalyzing the hydrogen evolution reaction (HER). The optimal HER electrocatalyst of the fractal-shaped single-layer MoS₂, which has an edge-to-substrate ratio of about $0.33 \mu\text{m}^{-1}$, exhibits superior HER catalytic activities, such as a low overpotential, a low Tafel slope of 45 mV/dec, a large exchange current density of $50.9 \mu\text{Acm}^{-2}$ etc. The study provides new ways to design 2D HER electrocatalysts, including controlling the

geometry, strain, and modulating the electrical conductivity.

References

- [1] Yi Wan, Xiang Zhang, Yu Ye*, and Lun Dai* etc., *Advanced Materials*, DOI: 10.1002/adma.201703888, 2017.
- [2] Yi Wan, and Lun Dai* etc., *ACS Appl. Mater. Interfaces* 8, 18570 (2016).
- [3] Yi Wan, Yan Li*, Yu Ye*, and Lun Dai* etc., *Nano Energy*, accepted.

Figures

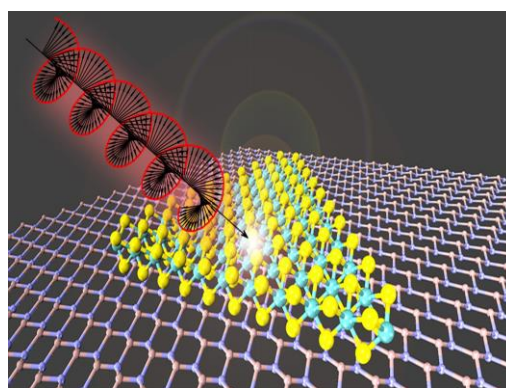


Figure 1: Epitaxial single-layer MoS₂ on GaN with enhanced valley helicity

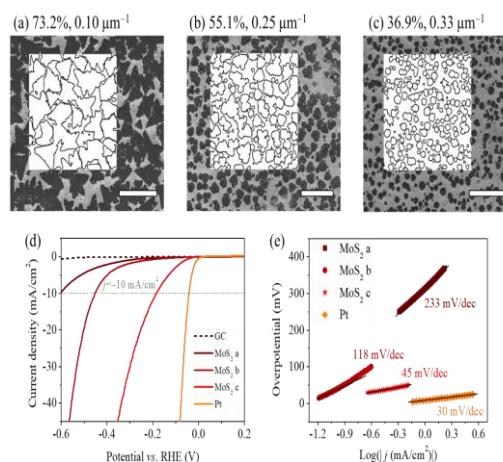


Figure 2: The HER catalyzing performance of the fractal-shaped single-layer MoS₂.