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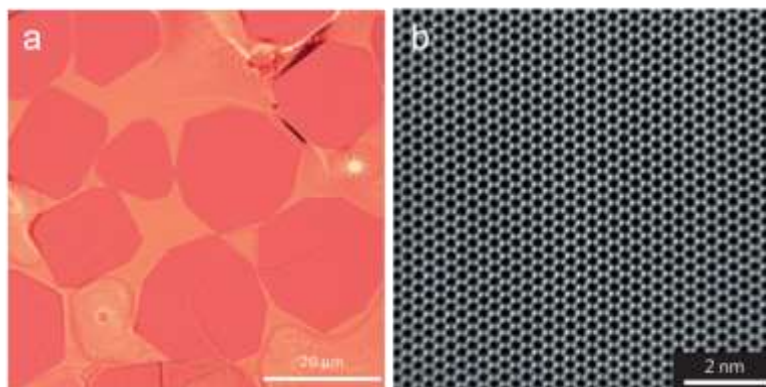
# CVD-Grown High-Quality Ultrathin Mo<sub>2</sub>C Crystals and Their Vertical Heterostructures with Graphene

Transition metal carbides (TMCs) are a large family of materials that combine the properties of ceramics and metals. High-quality 2D TMCs are essential for investigating new physics and properties in 2D limit. However, the commonly used chemical etching method can only produce functionalized 2D TMC nanosheets with abundant defects and functional groups, known as MXenes. Recently, we developed a universal CVD method [1] to fabricate large-area high-quality ultrathin 2D TMC crystals. The obtained  $\alpha$ -Mo<sub>2</sub>C crystals are a few nanometres thick and over 100  $\mu\text{m}$  in size. They show 2D characteristics of superconducting transitions; moreover, the superconductivity is also strongly dependent on the crystal thickness. Further studies show that 2D  $\alpha$ -Mo<sub>2</sub>C crystals have unique domain structure with rotational-symmetry and well-defined line-shaped domain boundaries, and the domain boundaries have a significant influence on 2D superconductivity [2]. We also realized the direct growth of high-quality graphene/2D superconducting  $\alpha$ -Mo<sub>2</sub>C vertical heterostructures with uniformly well-aligned lattice orientation and strong interface coupling by a two-step CVD process [3]. The strong interface coupling leads to a phase diagram of superconducting transition with multiple voltage steps being observed in the transition regime. Furthermore, we demonstrated the realization of highly transparent Josephson junction devices based on these heterostructures.

## References

- [1] C. Xu, et al., *Nature Materials*, 11 (2015) 1135-1141
- [2] Z. Liu, et al., *Nano Letters*, 7 (2016) 4243-4250
- [3] C. Xu, et al., *ACS Nano*, 6 (2017) 5906-5914

## Figures



**Figure 1:** (a) Typical optical image of ultrathin  $\alpha$ -Mo<sub>2</sub>C crystals on a Cu/Mo substrate, showing different regular shapes. (b) Atomic-level HAADF-STEM image of an  $\alpha$ -Mo<sub>2</sub>C sheet, showing highly crystalline quality.