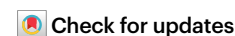


Ultraclean monolayers from a van der Waals confinement

Chang-Hsun Huang, Jui-Han Fu & Vincent Tung



Growing transition metal dichalcogenide monolayers at a buried van der Waals interface simultaneously improves thickness control, preserves ultraclean interfaces, and enables intrinsic patterning and atomically selective Janus structure formation.

For more than a decade, researchers have pursued a deceptively simple goal for two-dimensional (2D) transition metal dichalcogenides (TMDs): make the monolayer limit routine^{1–3}. Yet translating such ‘one layer makes the difference’ physics into reproducible devices is often derailed by many bottlenecks. For example, open-surface growth frequently produces unintended adlayers⁴. Atomically precise Janus conversion, which replaces only one chalcogen plane, is hard to realize without alloy-like disorder⁵. Moreover, transfer-based assembly of monolayers into van der Waals (vdW) heterostructures can trap contaminants at buried interfaces, blurring interlayer coupling and degrading performance⁶. Writing in *Nature Materials*, Ce Bian and

colleagues address these issues by changing where growth happens⁷. Instead of performing chemical vapour deposition on an exposed substrate, they relocate the reaction into a nanometre-confined vdW space – the buried gap between a capping layer (graphene or hexagonal boron nitride, hBN) and the underlying substrate. Precursors diffuse into this vdW gap, nucleate under confinement and crystallize into a monolayer, which is incorporated into an ultraclean heterostructure and sealed in situ⁷.

At a practical level, the method begins by laminating graphene or hBN onto SiO₂/Si to create a shallow interfacial gap. Vapour-phase metal and chalcogen precursors then diffuse into this gap through access points provided by edges, wrinkles or sparse defects. Once the local precursor concentration exceeds the nucleation threshold, a TMD domain forms inside the gap rather than on the exposed surface, as shown in cross-sectional scanning transmission electron microscopy (STEM) images in Fig. 1.

Why does simply ‘closing the lid’ suppress adlayers so effectively? The work of Bian and colleagues sketches a mechanistic picture that combines energetics and kinetics. On the energetic side, density functional theory calculations suggest that a graphene–TMD–substrate stack is more stabilized than open configurations, consistent with a

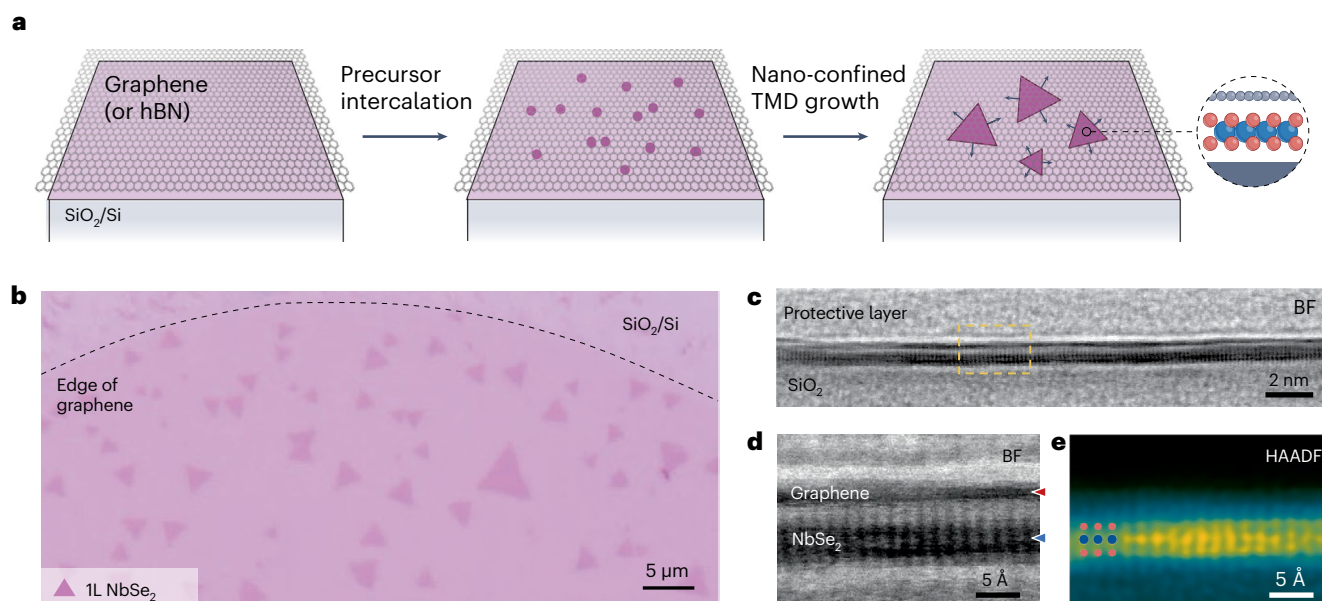


Fig. 1 | Nano-confined growth of NbSe₂ monolayers beneath a graphene cap. **a**, Schematic of intercalation-enabled growth in a buried vdW nanogap with in situ encapsulation. **b**, Optical image of triangular monolayer (1L) domains.

c–e, Bright-field (BF; **c,d**) and high-angle annular dark-field (HAADF; **e**) STEM confirm stacked graphene (upper stripe) and NbSe₂ (lower stripe) in the confined interface. Figure adapted from ref. 7, Springer Nature Limited.

monolayer benefiting from interactions on both its top and bottom interfaces. But thickness selection is ultimately kinetic. With the cap intact, intercalation occurs mainly from the edges, creating a directional diffusion front that supplies precursors towards the interior. Crucially, confinement makes in-plane edge attachment the dominant incorporation pathway, suppressing second-layer nucleation that would otherwise produce adlayers on an exposed surface.

Confinement can do more tricks than picking the layer count; it also defines the device footprint. Under conditions of high deposition flux and intact caps, nucleation preferentially occurs near the cap perimeter, and domains can merge into a continuous monolayer that traces the boundary of the capping flake. Demonstrations show NbSe₂ rings following triangular, square and circular hBN geometries, with Raman mapping supporting continuity along the perimeter. For device fabrication, the appeal is clear: conducting channels can be drawn by cap geometry, potentially avoiding lithography steps that can damage or contaminate ultrathin crystals.

The same one-sided access can also be repurposed for atomically selective chemistry. Janus TMD monolayers break out-of-plane symmetry and can introduce polarity-driven effects; however, atomically precise synthesis is challenging because reactions on the two chalcogen planes tend to compete with the desired one-sided substitution. In the nano-confined scheme, Bian and colleagues first grow MoS₂ under a cap and then use NbSe₂-derived precursors as a selenium source. Lattice mismatch blocks edge incorporation, so NbSe₂-derived species intercalate at the MoS₂/substrate interface and drive selective bottom-plane substitution under cap protection, forming the MoSSe structure.

Growing within a vdW interface ensures sufficient crystal quality with ultraclean interfaces, preserving the intrinsic properties of the heterostructures from the moment they form. In graphene/Janus-MoSSe heterostructures, the vdW spacing is reported to be 3.7 Å, and the optical response depends strongly on the capping material: graphene-capped regions show photoluminescence quenching consistent with interfacial charge transfer, whereas hBN-capped regions exhibit enhanced emission, as expected for an insulating encapsulant. In patterned NbSe₂ devices, the authors observe a sharp superconducting transition with an onset transition temperature around 2.8 K. The transition is quickly suppressed by perpendicular magnetic

fields but remains more resilient under in-plane fields, consistent with spin-orbit-protected pairing in ultrathin NbSe₂ (ref. 8).

The nano-confinement strategy connects synthesis, integration and environmental protection into a single step. The atomically defined confinement volume naturally limits growth to the monolayer regime, preserves clean buried interfaces by eliminating transfer, and can even enable pattern definition through the geometry and continuity of the capping layer. Translating this concept into a general, scalable manufacturing approach, however, will require wafer-scale and reproducible control over precursor intercalation and transport – capabilities that, in practice, hinge on managing the cap's defect density, adhesion and long-range continuity. It will also demand predictive design rules that connect cap geometry, precursor flux and diffusion pathways to device-ready layouts, alongside demonstrations on more industry-relevant substrates and heterogeneous stacks without sacrificing throughput. More broadly, this work reframes chemical vapour deposition as an interface-first process: the material is synthesized directly at its target interface, and that interface becomes the primary lever for controlling nucleation, growth kinetics and encapsulation.

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Published online: 09 February 2026

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Competing interests

The authors declare no competing interests.