Hypotaxy of Wafer-scale Single Crystal Transition Metal Dichalcogenides

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Abstract (Century Gothic 11)

Wafer-scale single crystal transition metal dichalcogenides (TMDs) have been synthesized on a crystalline substrate, such as c-plane sapphire, with epitaxial relation by chemical vapor deposition (CVD). The limited substrates for epitaxial growth of TMDs necessitate the transfer process onto desired substrates for device fabrication, leading to unavoidable damage and wrinkles. Here, we report hypotaxial (downward arrangement) growth of TMDs (MoS₂, MoSe₂, WS₂, and WSe₂) below an ultrathin 2D template (graphene and hBN) by chalcogenization of transition metal thin films. The chalcogen atoms diffuse through nanopores of graphene, which are generated during the chalcogenization process, leading to highly crystalline and layered TMDs below graphene with crystalline orientation alignment with high controllability of thickness. The grown single crystal TMDs show remarkably high thermal conductivity and carrier mobility comparable to those of exfoliated ones. Our hypotaxial growth method enables to overcome the substrate constraints of conventional epitaxial growth and to fabricate 4-inch single crystal TMDs suitable for monolithic 3D integration.

References

- [1] Kang, K. et al. High-mobility three-atom-thick semiconducting films with wafer-scale homogeneity. *Nature* **520**, 656-660 (2015).
- [2] Liu, L. et al. Uniform nucleation and epitaxy of bilayer molybdenum disulfide on sapphire. *Nature* **605**, 69-75 (2022)
- [3] Kim, K. S. et al. Non-epitaxial single-crystal 2D material growth by geometric confinement. *Nature* **614**, 88-94 (2023).

[4] Jayachandran, D. et al. Three-dimensional integration of two-dimensional field-effect transistors. *Nature* **625**, 276-281 (2024).

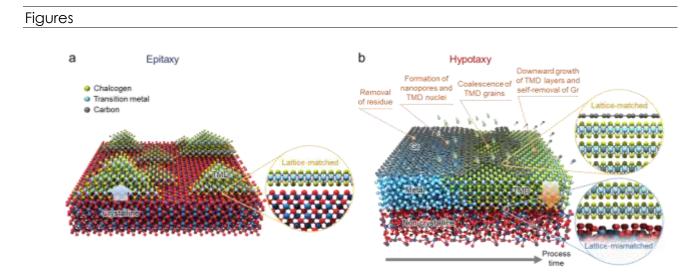


Figure 1: Growth mechanisms of epitaxy and hypotaxy. Schematics of growth mechanisms for a, conventional epitaxy and b, proposed hypotaxy.