Growth of Single-Crystal Hexagonal Boron Nitride by Chemical Vapor Deposition

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Large-area single-crystal monolayers of two dimensional (2D) materials such as graphene, hexagonal boron nitride (hBN) and transition metal dichalcogenides have been successfully grown. Among them, hBN has been demonstrated to be the "ideal" dielectric substrate for 2D materials-based field effect transistors (FETs) - offering the potential for extending Moore's law. Although hBN thicker than a monolayer is more desirable as substrate for 2D semiconductors, the growth of highly uniform and single-crystal few- or multi-layer hBN has not yet been demonstrated. Here we report the epitaxial growth of wafer-scale single-crystal tri-layer hBN by a chemical vapour deposition method. Uniformly aligned tri-layer hBN islands are found to grow on a 2 cm × 5 cm single-crystal Ni (111) at early stage of growth and finally to coalesce into a single-crystal film. Cross-sectional transmission electron microscopy (TEM) results show that a Ni₂₃B₆ interlayer is formed (during cooling) between the single-crystal trilayer hBN film and Ni (111) substrate by boron dissolved in Ni (111) and that there is epitaxial relationship between tri-layer hBN and Ni₂₃B₆ and between Ni₂₃B₆ and Ni (111). We further find that the tri-layer hBN film acts as a protective layer that remains intact during catalytic evolution of hydrogen – suggesting continuous and uniform single-crystal tri-layer hBN in large area. This tri-layer hBN transferred onto the SiO₂ (300 nm)/Si wafer acts as a dielectric layer to reduce electron doping from the SiO₂ substrate in MoS₂ FETs. Our results demonstrate that it is possible to achieve high quality multi-layered hBN over large areas by CVD – opening up new pathways for making it a ubiquitous substrate for 2D semiconductors and other purposes.