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Unidirectional Assembly on Distorted Two-Dimensional Crystal Substrates

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2D crystals possess surface without dangling bonds and the assembly processes on 2D crystals are mainly driven by relatively weak van der Waals interactions between deposited component and 2D substrate. The self-assembly on the surface of two-dimensional (2D) layered crystals have gained significant research interest for fabrication of well-ordered nanostructures and vertical heterostructures based on 2D crystals. The assembled structure obtained by van der Waals interaction is generally difficult to control and often display heterogeneous configurations. On the other hand, the structure and quality at the 2D heterointerface have great influence on the properties and performance of heterostructures. The direct observation of the interfacial structure of 2D heterostructure can provide insight for understanding the electrical and chemical properties of the system, which can lead to a better design of a new 2D heterostructures [1].

In this contribution, we present various self-assembly phenomena on 2D crystals observed by various transmission electron microscopy (TEM) imaging and electron diffraction. In particular, the unidirectional assembly phenomena on various distorted 2D crystals, namely black phosphorus (BP) and distorted transition metal dichalcogenides (TMDCs) are discussed. These atomically puckered 2D crystals can induce the unidirectional assembly along the directions of the crystal distortion.

As a first example, we examine the interfacial structures and properties of physically-deposited metals of various kinds on BP [2]. We find that Au, Ag, and Bi form single-crystalline films with (110) orientation through guided van der Waals epitaxy. TEM imaging and X-ray photoelectron spectroscopy confirm that atomically sharp van der Waals metal-BP interfaces forms with exceptional rotational alignment. Under a weak metal-BP interaction regime, the BP's puckered structure play an essential role in the adatom assembly process and can lead to the formation of a single crystal, which is supported by our theoretical analysis and calculations. The experimental survey also demonstrate that the BP-metal junctions can exhibit various types of interfacial structures depending on metals, such as the formation of polycrystalline microstructure or metal phosphides.

We also investigate the assembly behavior of one-dimensional (1D) AgCN microwires on various distorted TMDC crystals, namely 1T'MoTe2, T_d-WTe2, and 1T'-ReS_2 [3]. The unidirectional alignment of AgCN chains is observed on these crystals, reflecting the symmetry of underlying distorted TMDCs. Polarized Raman spectroscopy and transmission electron microscopy directly confirm that AgCN chains display the remarkable alignment behavior along the distorted chain directions of underlying TMDCs. The observed unidirectional assembly behavior can be attributed to the favorable adsorption configurations of 1D chains along the substrate distortion, which is supported by our theoretical calculations and observation of similar assembly behavior from different cyanide chains. The aligned AgCN microwires can be harnessed as facile markers to identify polymorphs and crystal orientations of TMDCs [4].

Atomic scale structural transformation guided by the lattice distortions of 2D substrates can be visualized using in situ heating experiments. We present in situ TEM results on nanoparticle transformation and substrate etching at elevated temperatures.

References
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