Two-dimensional Heterolayer Superlattices

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Novel functional materials prepared by stitching two different components into a heterostructure could possibly lead to symbiosis and optimization of multiple properties. By appropriate selection of 2D layered materials with the inherent properties of interest, and alternatively stacking them via ex-situ or in-situ self-assembly could be a facile synthetic route to fabricating these materials. Transition metal chalcogenides and dichalcogenides (TMC/TMD) are widely studied 2D layered materials with arrays of transition metals sandwiched between two layers of chalcogenide anions. This family of compounds have garnered attention with unique electronic structure, tunable bandgaps, and their applications. Another category of extensively studied inorganic 2D materials are the layered double hydroxides (LDHs). These are inorganic compounds made up of positively charged host layers with anionic guest molecules occupying the interlayer space. Owing to the lamellar structure of both these materials, it is possible to cleave and restack the layers, making them promising candidates for preparation of hybrid multifunctional materials.

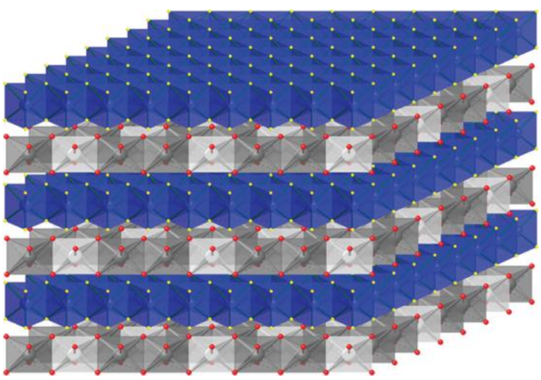

Figure 1: *Representative image of alternating layers of* $[TaS_2]^{0.33-}$ *superconducting layer and* $[Ni_{0.66}Al_{0.33}(OH)_2]^{0.33+}$ *magnetic layer.*

In this work, we address synthesis of these ordered heterolayer materials with two different approaches. First is an ex-situ method, where the components (LDH, TMD) were prepared individually, exfoliated into cationic and anionic layers. Both the solutions containing exfoliated 2D sheets were mixed to prepare the hybrid superlattice via electrostatic self-assembly. Second is an in-situ formation of LDH intercalated tetrahedral transition metal chalcogenides by hydrothermal synthesis.

Crystal chemistry can provide interesting insights into understanding the structure-property relations of these materials. We investigate the structural aspects using X-ray, neutron and electron diffraction techniques. With the most intense peaks in powder XRD being the 00*l* reflections, tells us that the periodicity of heterolayer stacking occurs along the c-axis with an estimate of the basal spacing. Neutron diffraction is the technique with complementary scattering efficiency compared to X-rays, can help locate lighter elements such as hydrogen atoms in the structure. Electron diffraction along the [001] zone axis helps us visualize the symmetry of the heterostructure, more so evidently when the individual layers have contrastingly different symmetries. These results would help devise strategies to synthesize and understand structures of hybrid materials using LDH and TMC/TMD, opening up access to a class of functional materials with interesting properties.

Keywords: Transition metal dichalcogenide, layered double hydroxide, superlattice