Unraveling Interlayer Interactions in Multilayered Ferecrystalline Thin Films via Photoemission Spectroscopy

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The stacking of two-dimensional sheets provides a pathway towards the study of interlayer interactions, emergent properties, and subsequently novel applications for 2D materials. Common approaches to create these heterostructures are the manual stacking of exfoliated sheets, or sequential layer-by-layer growth via chemical or physical vapor deposition. However, the achievable complexity of the layering sequence is often limited by available stable constituent layers or compatible growth conditions.

In contrast, the self-assembly of artificial layered precursors, which is known as the Modulated Elemental Reactants (MER) technique, offers unparalleled flexibility in the composition and layering sequence of thin films. In this two-step synthesis, amorphous elemental precursors are first deposited via sequential physical vapor deposition. By calibrating the thicknesses and layering sequence of the constituents to mimic the targeted structure, it is possible to crystallize the precursors into a layered thin film via annealing in inert atmosphere. Due to the structuring of the precursors on an atomic level, the necessary energy input as well as layer intermixing during crystallization is reduced, enabling the synthesis of metastable structures with almost arbitrary complexity. The increased structural freedom of MER thin films comes at the expense of epitaxial alignment, as layers are generally showing turbostratic rotational disorder along the crystals *c*-axis. [1]

We will discuss how by controlling the nanoarchitecture of the precursor, one can create systematic series of heterostructures to facilitate the growth of certain metastable constituent layers such as rock-salt structured BiSe or metallic 1T-$MoSe_2$ [2], and investigate the effects of modulation doping to tune charge transport [3], superconductivity, or charge density waves. The contribution will focus on recent results on two classes of heterostructures built up from BiSe, Bi_2Se_3, and $MoSe_2$ [2], as well as PbSe and VSe_2 [3], respectively.

While the thickness of MER thin films is usually on the order of 50 nm spanning multiple unit cells, we were able to push the MER synthesis towards the two-dimensional limit by preparing a one monolayer thick, nanocrystalline layer of $MoSe_2$ on a substrate of epitaxial graphene on silicon carbide [4]. Building on these results, we are now exploring the growth of more sophisticated, one unit cell thick heterostructures by also incorporating bismuth selenide. Preliminary results suggest that these films are at least partially aligned with respect to the graphene, indicating that the main drawback of the MER synthesis could be eliminated by the choice of an appropriate growth substrate.

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