Atomic-scale materials design: vdW and non-vdW 2D materials

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High-throughput computations based on ab-initio electron theories are likely to become an integral part of future programs for accelerated materials development. Here the strategy is used to identify novel 2D Janus materials[1] and self-intercalated transition metal dichalcogenides (TMDs)[2], respectively. We uncover some of the unique features of the discovered materials and point to potential applications and future studies.

The identified MXY Janus materials, consisting of a middle layer of metal atoms (M) sandwiched between different types of chalcogen, halogen, or pnictogen atoms (X,Y), are found to exhibit diverse and fascinating properties including finite out-of-plane dipole moments, giant Rashba-splittings, direct and indirect band gaps ranging from 0.7 to 3.0 eV, large exciton binding energies, and strong light matter interactions. By embedding the Janus layers in vdW heterostructures, it is possible to engineer the internal electric field and thereby control band alignment and charge transfer processes between the layers[3].

Filling of the octahedral vacancy sites in the vdW gap of TMD bilayers by self-intercalation of metal atoms creates a new class of ultrathin 3D materials whose properties can be tuned by varying the coverage and spatial arrangement of the filled sites. The self-intercalation of TMDs by their native transition metal atoms is found to be highly facile under growth conditions of low chalcogen chemical potential. Using high throughput DFT calculations, we found that 14 self-intercalated TMDs should exhibit ferromagnetism, including Mo_8Se_{12}, Nb_7S_{12}, Ti_8S_{12} and V_7S_{12}, and several of these were experimentally confirmed. Our work establishes self-intercalation as a new approach to grow ultrathin 3D materials from 2D bilayers with stoichiometry or composition-dependent properties.

References

[1] A. Riis-Jensen, T. Deilmann, T Olsen, and K. S. Thygesen, ACS Nano, 13 (2019) 13354.
[2] X. Zhao et al., accepted in Nature
[3] M. Palsgaard, T. Gunst, T. Markussen, K. S. Thygesen, and M. Brandbyge, Nano Letters 18 (2018) 7275

Figures

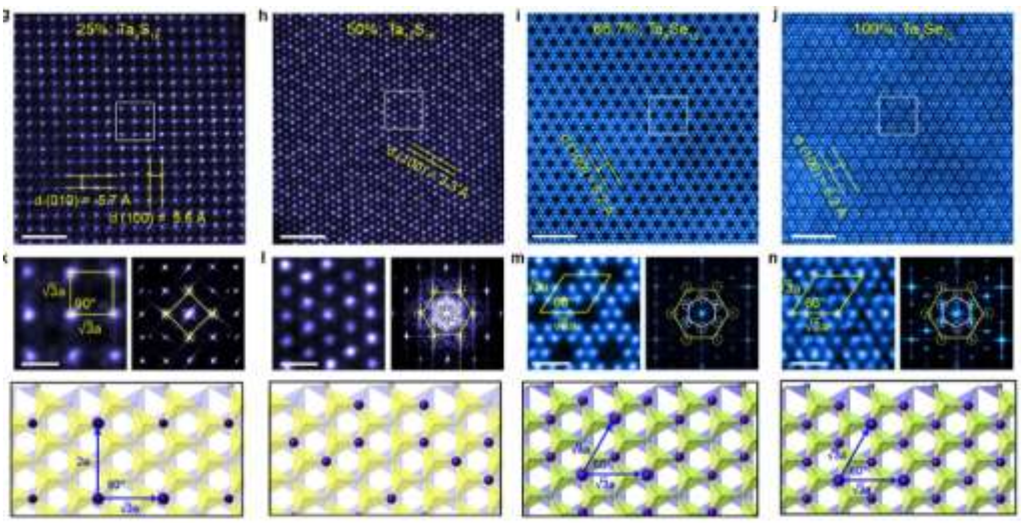

Figure 1: Engineering properties by self-intercalation. From vdW bilayer $TaSe_2$ to covalently bonded Ta_9Se_{12}. STEM-ADF image of the MBE grown self-intercalated compounds.