Materials Science & Engineering Doctoral Defense

Synthesis of Monolayer Janus Transition Metal Dichalcogenides

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Abstract

2D materials with reduced symmetry have gained great interest in the past decade due to the arising quantum properties introduced by the structural asymmetry. A particular example is called 2D Janus materials. Named after Roman god Janus with two faces, Janus materials have different chemical compositions on the two sides of materials, leading to a structure with broken mirror symmetry. Electronegativity difference of the facial elements induces a built-in polarization field pointing out of plane, which has driven a lot of theory predictions on Rashba splitting, high-temperature ferromagnetism, Skyrmion formation, and so on. Previously reported experimental synthesis of Janus 2D materials relies on high temperature processing, which limits the crystallinity of as produced 2D layers.

In this dissertation, I present a room temperature selective epitaxial atomic replacement (SEAR) method to convert CVD grown transition metal dichalcogenides (TMDs) into Janus structure. Chemically reactive hydrogen plasma is used to selectively etch off the top layer of chalcogen atoms and the introduction of replacement chalcogen source in-situ allows for achievement of Janus structures in one step at room temperature. It is confirmed that the produced Janus monolayers possess high crystallinity and good excitonic properties. Moving forward, fabrication of lateral and vertical heterostructures of Janus materials is shown, which are predicted to possess exotic properties because of the intrinsic polarization field.

To efficiently screen other kinds of interesting Janus structures, a new plasma chamber is designed to allow in-situ optical measurement during the SEAR process. Successful conversion is seen on mechanically exfoliated $MoSe_2$ and WSe_2, and insights into reaction kinetics are gain from Raman spectra evolution. Using the monitoring ability, Janus SNbSe is synthesized for the first time. It's also demonstrated that the overall crystallinity of as produced Janus monolayer SWSe and SMoSe are correlated with the source of monolayer TMDs.

Overall, the synthesis of the Janus monolayers using the described method paves ways to production of highly crystalline Janus materials, and with the in-situ monitoring ability, a deeper understanding of the mechanism is reached. This will accelerate future exploration of other Janus materials synthesis, and confirmation and discovery of their exciting quantum properties.

April 15, 2021; 1 PM;

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