

FUNDAMENTAL UNDERSTANDING OF TWO-DIMENSIONAL ORGANIC SEMICONDUCTOR-INCORPORATED PEROVSKITES AND HETEROSTRUCTURES

Two-dimensional (2D) perovskite semiconductors are an emerging family of hybrid materials featuring a built-in quantum well architecture which has gained much interest due to its potential as a promising candidate for next-generation photovoltaic and optoelectronic applications. To successfully integrate 2D perovskites as efficient devices, it is imperative that a thorough understanding of the fundamental properties these materials possess and how their complex heterostructures behave is established. However, to date, the synthetic challenges regarding high-quality crystals of these materials due to the structural complexity and the hybrid nature have impeded further progress in this area. Thus, we demonstrate a general method to construct tunable 2D organic semiconductor-incorporated perovskites (OSiP) by simultaneously manipulating slab thickness of the inorganic layers and conjugation length of the organic substituents. The energy band offsets and exciton dynamics at the organic-inorganic interfaces were elucidated using computational means and ultrafast spectroscopy, while lattice dynamics were quantified via temperature-dependent spectroscopy and X-ray diffraction studies. Results show that longer and more planar π -conjugated organic ligands induce a more rigid inorganic crystal lattice, which leads to suppressed exciton-phonon interactions and superior optoelectronic properties such as efficient lasing.

Furthermore, understanding ion migration in two-dimensional (2D) perovskite materials is key to enhancing device performance and stability as well. However, prior studies have been primarily limited to heat and light-induced ion migration. To investigate electrically induced ion migration in 2D perovskites, we construct a high-quality single crystal 2D perovskite heterostructure device platform with near defect-free van der Waals contact. While achieving real-time visualization of directional ion migration, we also uncover the unique behavior of halide anions inter-diffusing towards the opposite direction under prolonged bias. Confocal microscopy imaging reveals a halide migration channel that aligns with the crystal and heterojunction edges. After sustained ion migration, stable junction diodes exhibiting up to ~ 1000 -fold forward to reverse current ratio are realized. Unraveling the fundamental properties of 2D OSiPs as well as ion migration in 2D perovskite heterostructures paves the way towards stable and efficient devices.