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HYBRID 2D AND 3D NANOSTRUCTURED PEROVSKITES: FROM UNDERSTANDING FUNDAMENTAL PHYSICS TO OPTOELECTRONIC APPLICATIONS

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Hybrid organic–inorganic perovskites (HOPs) have demonstrated an extraordinary potential for clean sustainable energy technologies and low-cost optoelectronic devices. This talk overviews the main features of three dimensional (3D) and layered two-dimensional (2D) HOPs by combining solid-state physics concepts with simulation tools based on density functional theory. A comparison between layered and 3D HOPs highlights differences and similarities such as spin-orbit effects, quantum and dielectric confinements and excitonic properties. In 3D HOPs we study in depth the effects of electron-phonon coupling leading to polaron formation across the broad range of materials. Calculated electronic structure, charge density, changes the geometry, and reorganization energies are further related to experimentally measured specific vibrational modes, Huang-Rhys parameters and Jahn-Teller like distortions. These effects lead to formation of meta-stable deep-level charge states, which potentially responsible for photocurrent degradation in thin-film perovskite devices. The photophysics of 2D materials is defined by an interplay of strongly bound excitons and lower-energy states associated with the edges of the perovskite layers. The latter provide a direct pathway for dissociating excitons into longer-lived free carriers that substantially improve the performance of optoelectronic devices. Our theoretical simulations rationalize specifics of electronic structure of these materials, dynamics and a role of interfacial states. We also outline specific ways to rationally control geometry of edges in 2D HOP materials via external fields, contact interfaces and composition of organic compound. Overall, our results provide insights towards the material design for various applications.

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