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Understanding the synthesis, structure, and physics of hybrid 2D perovskites for efficient optoelectronics

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Presentation language is English. Guests are welcome

Abstract
Organic-inorganic two-dimensional halide perovskites (2DPKs) are soft semiconductors formed by combining organic and inorganic moieties, which self-assemble in solution to form highly ordered periodic stacks. They exhibit a large compositional and structural phase space, which has led to novel and emergent physical properties offering potential for applications in optoelectronics, energy conversion, photonics, spintronics, and quantum devices. To achieve this paradigm, there is a need to understand both the fundamental physics and the growth of both 2DPK crystals and thin films used for integration in devices. In this seminar, I will describe three exciting results.
First we overcome a long-standing challenge in 2D perovskites of realizing phase pure thin-films. Briefly, we demonstrate a scalable approach involving dissolution of single-phase crystalline powders with homogeneous perovskite layer thickness in desired solvents, to fabricate 2D perovskite thin-films with high phase purity. In-situ characterizations reveal the presence of sub-micron-sized seeds in solution that preserve the memory of the dissolved single-crystals, and dictates nucleation and growth of grains with identical layer thickness of the perovskite layers in thin-films. Photovoltaic devices fabricated with such films, yields an efficiency of 17.1% and 1.20V open-circuit voltage, while preserving 97.5% of their peak-performance after 800 hours under illumination without any external thermal management.
Second, we report sunlight-activated anisotropic lattice contraction in 2DPKs, which is reversible and strongly dependent of both the nature of the interlayer organics and the details of the structure. Our theory suggests that light-generated charge accumulation results in the build-up of a bulk compressive strain, which induces a continuous lattice contraction over minutes. In-situ structural measurements on photovoltaic devices directly correlate the light-induced lattice contraction to an increase in the photovoltaic efficiency of Dion-Jacobson 2DPK solar cells from 13.8% to 16.4%.
Third, we employed ultrafast electron diffraction (UED) to capture at the picosecond time scale how light generated hot carriers dissipate their energy to the lattice and affect the 2DPK structure. The carriers cooling dynamic in 2DPKs yields a fast lattice re-ordering that is distinct from 3DPKs, followed by classic slow thermal dissipation of carriers excess energy to the lattice. The former process suggest the light-induced activation of an incipient phase transition, toward a hypothetical undistorted phase due charge-lattice interactions. The effect of anisotropy and the nature of the bulky organic cations were also confirmed to play a role in tailoring carrier-phonon interactions.

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