Ferroelectric Polars, Belgian Waffles, and Principles for “Perfect” Semiconductors

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Lead halide perovskites have been demonstrated as high performance materials in solar cells and light-emitting devices. These materials are characterized by coherent band transport expected from crystalline semiconductors, but dielectric responses and phonon dynamics typical of liquids. Here we explain the essential physics in this class of materials based on their dielectric functions and dynamic symmetry breaking on nano scales. We show that the dielectric function in the THz region may lead to dynamic and local ordering of polar nano domains by an extra electron or hole, resulting a quasiparticle which we call a ferroelectric large polaron, a concept similar to solvation in chemistry. Compared to a conventional large polaron, the collective nature of polarization in a ferroelectric large polaron may give rise to order(s)-of-magnitude larger reduction in the Coulomb potential. We show that the shape of a ferroelectric polaron resemble that of a Belgian waffle. Using two-dimensional coherent phonon spectroscopy, we directly probe the energetics and local phonon responses of the ferroelectric large polarons. We find that that electric field from a nascent e-h pair drives the local transition to a hidden ferroelectric order on picosecond time scale. The ferroelectric or Belgian waffle polarons may explain the defect tolerance and low recombination rates of charge carriers in lead halide perovskites, as well as providing a design principle of the “perfect” semiconductor for optoelectronics.