Solving the lead halide perovskite puzzle

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Abstract: Research in my group aims to understand photophysics in nano, molecular, and hybrid semiconductors and interfaces. We focus on fundamental interactions beyond the single particle approximation, and probe the dynamics of quasiparticles (excitons, polarons, polaritons, etc.) that are important to a range of optoelectronic processes, such as solar energy conversion, light emission, and polariton condensation. Following a brief introduction to my research program, I will tell one particular story – the discovery of why lead halide perovskites (LHPs) work so well in solar cells and light emitting materials. A major puzzle from recent studies on LHPs is that optoelectronic performances suggest nearly perfect semiconductors despite the unavoidable presence of defects from room temperature and solution processing. Here we explain the essential physics in this class of materials based on their disordered phonon dynamics and dielectric functions. We show that the dielectric function of a hybrid organic-inorganic lead halide perovskite (LHP) possesses combined characteristics of a polar liquid and a ferroelectric material. The latter response 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*. 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 and introduce potential barriers to charge carrier scattering. The ferroelectric large polaron may explain the defect tolerance, low recombination rates, and slow cooling of charge carriersin lead halide perovskites, as well as providing a design principle for high performance semiconductors from nano, molecular, and hybrid materials.

Bio-sketch: Xiaoyang Zhu is the Howard Family Professor of Nanoscience and a Professor of Chemistry at Columbia University. He received a BS degree from Fudan University in 1984 and a PhD from the University of Texas at Austin in 1989. After postdoctoral research with Gerhard Ertl at the Fritz-Haber-Institute, he joined the faculty at Southern Illinois University as an Assistant Professor in 1993. In 1997, he moved to the University of Minnesota as a tenured Associate Professor, later a Merck endowed professor. In 2009, he returned to the University of Texas at Austin as the Vauquelin Regents Professor and served as directors of the DOE Energy Frontier Research Center (EFRC) and the Center for Materials Chemistry. In 2013, he moved to Columbia University. His honors include a Dreyfus New Faculty Award, a Cottrell Scholar Award, a Friedrich Wilhelm Bessel Award, a Fellow of the American Physical Society, a Vannevar Bush Faculty Fellow Award from DOD, and an Ahmed Zewail Award from A. Among his professional activities, he serves on the editorial/advisory boards of *Accounts of Chemical Research*, *Science Advances*, *Chemical Physics*, and *Progress in Surface Science*, and as a scientific advisor to the Fritz-Haber-Institute of the Max-Planck Society and ShanghaiTech University.