

Abstract Submitted
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Direct Observation of Electron–Phonon Coupling and Slow Vibrational Relaxation in Organic–Inorganic Hybrid Perovskites¹ SEBASTIAN HURTADO PARRA, DANIEL STRAUS, NATASHA IOTOV, BRYAN FICHERA, JULIAN GEBHARDT, ANDREW RAPPE, JOSEPH SUBOTNIK, JAMES KIKKAWA, CHERIE KAGAN, University of Pennsylvania — Quantum and dielectric confinement effects in Ruddlesden-Popper 2D hybrid perovskites create excitons with a binding energy exceeding 150 meV. We exploit the large exciton binding energy to study exciton and carrier dynamics as well as electron–phonon coupling (EPC) in hybrid perovskites using absorption and photoluminescence (PL) spectroscopies. At temperatures <75 K, we resolve splitting of the excitonic absorption and PL into multiple regularly spaced resonances every 40–46 meV, consistent with EPC to phonons located on the organic cation. We also resolve resonances with a 14 meV spacing, in accord with coupling to phonons with mixed organic and inorganic character. These assignments are supported by density-functional theory calculations. Hot exciton PL and time-resolved PL measurements show that vibrational relaxation occurs on a picosecond time scale competitive with that for PL. At temperatures >75 K, excitonic absorption and PL exhibit homogeneous broadening. While absorption remains homogeneous, PL becomes inhomogeneous at temperatures <75K, which we speculate is caused by the formation and subsequent dynamics of a polaronic exciton. (JACS 138, 13798 (2016))

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