courtesy Tony Mastres

When do Ultrafast Processes Matter? From Batteries to Strong Localization in Solar Materials

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The Cushing lab focuses on ultrafast instrumentation science ranging from tabletop X-rays, to entangled photons, to new forms of battery spectroscopy. In this talk, I will briefly introduce our research areas, mentioning the increasingly “null” space explored with entangled photons, and then focus on two of the techniques – tabletop X-ray spectroscopy and ultrafast battery dynamics. For the latter, we use our newly developed, laser-driven ultrafast impedance method to investigate many-body ion-hopping mechanism in superionic conductors. Picosecond temporal and spectral correlations differentiate electron-ion, phonon-ion, and potentially ion-ion interactions.

Our first results on LLTO show that superionic conductivity does not occur by random thermal motion but rather by highly correlated ion-phonon modes in the THz, contrary to current ionic conductor design principles. Reducing charge density on the apical O anion using a transient charge-transfer transition also improves ionic conductivity on the picosecond timescale of optical phonons.

Next, we use transient X-ray techniques to explore the complex photodynamics of the Hubbard-Holstein Hamiltonian that describes systems ranging from solar fuel materials to O-LEDs. The ultrafast X-ray pulses measure a mix of electronic and structural dynamics and, using our excited state Bethe-Salpeter equation approach, we can extract time-resolved electron and hole energies, phonon and polaron modes, and transport phenomena. We measure materials with a range of electron-phonon coupling strength versus electronic and spin correlations to map the Hubbard-Holstein Hamiltonian phase space and evaluate its predictive accuracy for new excited state materials design.

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