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Ultrafast Dynamics of Polaron Formation[1]

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The formation of localized electronic states reflects the fundamental physics of coupling between electronic and lattice dynamics, as first noted by Landau who in 1933 described the process of polaron formation as "the electron 'digs its own hole' and is trapped there." Localization of electronic states plays a critical role in determining the properties of a wide range of materials: polaron formation has a profound impact on charge transport properties of electronic materials, and formation of self-trapped excitons, or exciton-polarons, dramatically changes optical properties and energy transport mechanisms. I will present femtosecond time-resolved studies of the dynamics of the localization process, focusing on the formation and evolution of self-trapped excitons and polarons. The experiments are carried out in quasi-one-dimensional materials in which the strength of the electron-phonon coupling that drives the dynamics can be systematically tuned by varying the material composition. Experiments using femtosecond vibrationally impulsive excitation, in which the system is excited with an optical pulse short compared to the periods of the relevant vibrational modes, allow us to time-resolve the coupled electronic and lattice dynamics as the system evolves from the initially photoexcited delocalized electronic state to form a self-trapped exciton, revealing rapid dynamics involving both optical and acoustic phonon modes. Polaron dynamics are probed using time-resolved terahertz spectroscopy, in which short pulses of far-infrared light are used to monitor the fast photoinduced carrier response, and show localization on the time scale of a single vibrational period of the lattice.

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