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Dynamics of Exciton and Polaron Formation in Molecular Electronic Materials[1] JASON LEICHT, JASON MANCE, SUSAN DEXHEIMER, Washington State Univ — We present measurements of the coupled electronic and vibrational dynamics of exciton and polaron formation using femtosecond wavepacket techniques. The experiments are carried out on the halide-bridged mixed-valence linear chain complex PtCl(en), a Peierls insulator with strong electron-phonon coupling. Earlier studies on longer time scales showed that excitation well above the optical gap energy can result in the formation of charged polarons in addition to the self-trapped excitons that form following excitation near the band edge, though the formation mechanism for polarons had not previously been established. Our measurements reveal formation of both types of excitations in $\sim$ 200 fs. Two distinct vibrational frequencies associated with the self-trapping process are observed: 176 cm^{-1}, associated with formation of self-trapped excitons, and upon excitation at higher energy, an additional component appears at 240 cm^{-1}, associated with the formation of polarons. The rapid formation of polarons, on the time scale of a single vibrational period following photoexcitation, together with the observation of accompanying vibrational coherence strongly suggests that the polarons form directly from the initial photoexcitation, rather than by dissociation of primary excitons.

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