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Understanding x-ray driven impulsive electronic state redistribution using a three-state model¹ MATTHEW R. WARE, PULSE Institute and Stanford University, JAMES CRYAN, PULSE Institute and SLAC, PHILIP H. BUCKSBAUM, PULSE Institute and Stanford University — The natural timescale for electron motion is extremely fast; electrons can move across molecular bonds in less than a femtosecond. To understand this fast motion and the role of electronic coherence, we are interested in creating a superposition of valence excited states through excitation with a broad bandwidth (>5eV) laser pulse. In the x-ray regime, the molecular ground state can couple to valence-excited states through an intermediate autoionizing resonance in a process known as stimulated x-ray Raman scattering (SXRS). X-rays excite electrons from the highly localized K-shells in a molecule, creating a superposition of valence-excited states initially localized around a target atom in the molecule. Coherences between states in the superposition will subsequently drive charge transfer as the wavepacket spreads out across the molecule. We use an effective 3-state model coupling the ground, auto-ionizing, and valence-excited states in diatomic systems to study the cross-section of SXRS as function of x-ray intensity, central frequency, bandwidth, and chirp. We also make observations on how the x-ray parameters affect the degree of initial localization to an atom of the wavepacket created in SXRS.

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