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Coherent motion of an electron hole near a conical intersection: Using time-resolved XUV spectroscopy to study charge dynamics in polyatomics[1]

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The quantum mechanical motion of charge across a molecule is at the heart of complex biological and chemical processes occurring in nature. Ultrafast pump-probe studies of valence electron phenomena in neutral molecules suggest that such charge dynamics are often mediated through coupled electronic and nuclear motion or through electronic correlations. However, the real-time evolution of an electron hole in a photoionized molecule remains a relatively unexplored facet of the charge transfer phenomena. Investigation of such processes in time-domain requires preparation and monitoring of a superposition of quantum states in the ionized molecule. We report our recent investigations of the coherent motion of an electron hole wavepacket created near a conical intersection in CO2 molecule. Using extreme ultraviolet (XUV) attosecond pulse train as a pump and femtosecond near-infrared (NIR) pulse as a probe, we resolved the oscillation of the electron hole density between sigma and pi orbitals. We found that these charge dynamics are driven by the coupled bending and asymmetric stretch vibrations of the molecule. We also quantified the mixing between electron hole states and found that the degree of electronic coherence decreases with time due to thermal dephasing. The experimental and theoretical results we obtained for the linear triatomic molecule represent the first steps in elucidating the inner workings of coherent charge migration processes and pave the way for the application of attosecond and femtosecond XUV spectroscopies in the measurement and control of charge dynamics in complex biochemical systems.

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