DAMOP14-2014-000908

Abstract for an Invited Paper
for the DAMOP14 Meeting of
the American Physical Society

Imaging charge and energy transfer in molecules using free-electron lasers[1]

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Charge and energy transfer reactions drive numerous important processes in physics, chemistry and biology, with applications ranging from X-ray astrophysics to artificial photosynthesis and molecular electronics. Experimentally, the central goal in studies of transfer phenomena is to trace the spatial localization of charge at a given time. Because of their element and site sensitivity, ultrafast X-rays provide a promising tool to address this goal. In this talk I will discuss several experiments where free-electron lasers were employed to study charge and energy transfer dynamics in fragmenting molecules. In a first example, we used intense, 70 femtosecond 1.5 keV pulses from the Linac Coherent Light Source (LCLS) to study distance dependence of electron transfer in laser-dissociated methyl iodide molecules. Inducing well-localized positive charge on the heavy iodine atom, we observe signature of electron transition from the separated methyl group up to the distances of 35 atomic units. In a complementary experiment, we studied charge exchange between two partners in a dissociating molecular iodine employing a pump-probe arrangement with two identical 90 eV pulses from the Free-Electron LASer in Hamburg (FLASH). In both cases, the effective spatial range of the electron transfer can be reasonably described by a classical over-the-barrier model developed for ion-atom collisions. Finally, I will discuss a time-resolved measurement on non-local relaxation mechanism based on a long-range energy transfer, the so-called interatomic Coulombic decay.

[1]This work was supported by Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, US Department of Energy and by the Kansas NSF “First Award” program.