ABSTRACT

X-ray Probe Experiments of Atoms and Molecules in Strong Laser Fields

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The Linac Coherent Light Source (LCLS) is an x-ray free-electron laser that is expected to produce a peak intensity ~1034 photons/(sec cm2) at 870 eV when focused to 1 μm. At that intensity, the K-shell photoionization rate of neon (4 × 1015 sec-1) exceeds the K-LL Auger decay rate (4 × 1014 sec-1), so the probability of producing two K holes in Ne with a single pulse approaches 1. Multiple core holes will be produced in the constituent atomic sites of molecules, leading to complex fragmentation patterns. In weak radiation fields using 3rd-generationsynchrotron x rays, multiple hole states appear as minor components due to electron correlation. For example, the KK/K production ratio ≈3 × 10-3 in Ne due to electron correlation. Understanding the production and decay of hole states in atoms and small molecules by LCLS pulses is fundamental to planning experiments on large molecules and materials. Will intense LCLS pulses modify inner-shell processes such as resonant Raman scattering and photoelectron recapture? The physics of photon-matter interactions using LCLS pulses combines aspects of x-ray processes with strong-field phenomena that are presently studied using ultrafast optical lasers. Intense optical radiation fields can ionize atoms, produce high-harmonic radiation, and spatially align molecules. Laser-pump/x-ray-probe experiments will be a major activity at the LCLS. We are using an x-ray beam line at the Advanced Photon Source in combination with a Ti:sapphire ultrafast laser system to develop a micro focused, tunable, polarized, time-resolved x-ray probe of atoms and molecules in the presence of an intense optical radiation field. The first experiments involve single- and double-ionization of one or two 4p electrons of krypton by the strong field of the laser. 1s → 4p resonances are observed in x-ray absorption of Kr+ and Kr2+ near 14.3 keV. The4p resonance is used to monitor the ion density as a function of time delay between laser and x-ray pulses and of spatial separation between the two beams. We also observe the alignment and dealignment of the Kr+ 4p3/2 hole state and have developed a theoretical model that agrees well with experiments. The model includes treatment of spin-orbit interaction in the hole states and quadrupole interactions between ions and photoelectrons. We are currently developing a source of laser-aligned molecules that will be probed by resonant x-ray absorption and an ion-imaging spectrometer.

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