Femtosecond response of atoms and molecules to ultra-intense hard x-rays

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The start-up of the Linac Coherent Light Source (LCLS) a decade ago made a new extreme regime of x-ray interactions with matter experimentally accessible. Since then, numerous novel capabilities for studying the structure and dynamics of biological systems, complex materials, and matter under extreme conditions have been demonstrated at the LCLS and other X-ray Free-Electron Laser (XFEL) facilities. Particular attention has been drawn to the domain of hard x-ray wavelengths and highest available intensities since this combination holds potential for imaging applications to reach atomic spatial resolution. Design and interpretation of such experiments to a large extent relies on our quantitative understanding of how individual atoms within an extended polyatomic system respond to multiphoton x-ray absorption, and by how much the distances between the atoms change within the duration of the x-ray pulse.

For hard x-rays, the interaction of light with a polyatomic system is often dominated by the presence of heavy (high-Z) elements since their absorption cross-section is typically two to three orders of magnitude larger than for light atoms such as carbon, nitrogen, and oxygen. Here we report experimental and theoretical results quantitatively characterizing the response of isolated heavy atoms and small polyatomic molecules containing a single heavy atom to the irradiation by femtosecond hard x-ray pulses at the intensities exceeding 10^{19} W/cm^2. For atoms, experimental data in the 5.5–8.3 keV range manifest surprisingly structured charge state distributions, which can be accurately described by the newly developed theoretical model [1]. This combined experimental and theoretical analysis demonstrates the importance of resonant and relativistic effects in multiphoton hard x-ray ionization at certain wavelengths. For molecules containing a single high Z element, like iodomethane and iodobenzene, it is found that under ultra-intense, hard x-rays, the ionization of a molecule can be considerably enhanced compared to an individual heavy atom with a similar absorption cross section [2], which is qualitatively different from earlier observations in the soft X-ray domain [3] or with weaker hard X-rays [4,5]. This enhancement is driven by ultrafast charge transfer within the molecule, which refills the core holes created in the heavy atom, providing further targets for inner-shell ionization and resulting in the emission of more than 50 electrons during the XFEL pulse. For iodomethane, such extreme ionization and fragmentation process can be simulated by the recently developed XMOLECULE package [6], which provides a detailed time-dependent description of molecular dynamics under ultraintense x-ray pulses [2]. Analysis of the pulse duration and pulse energy dependence reveals further subtleties of the interplay between x-ray absorption, electronic relaxation and nuclear motion, yielding a comprehensive picture of XFEL interactions with small molecules [7].

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