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Atomic and molecular ionization by ultraintense hard x-rays

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The development of x-ray free-electron lasers (XFELs) opened up a new extreme regime of light-matter interactions and provide unique novel capabilities for studying the structure and dynamics of biological systems, complex materials, and matter under extreme conditions [1,2]. Particularly intriguing is 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 during the x-ray pulse.

In this talk I will discuss how rare gas atoms and small polyatomic molecules respond to femtosecond hard x-ray pulses at the intensities exceeding 10^{19} W/cm² [3,4]. 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 [5]. Combined experimental and theoretical analysis demonstrates the importance of resonant and relativistic effects in multiphoton hard x-ray ionization [3]. For molecules containing a single high-Z element, like iodomethane and iodobenzene, it is found that under ultraintense, hard x-rays, the ionization of a molecule is considerably enhanced compared to an individual heavy atom with the same absorption cross section [4], which is qualitatively different from earlier observations in the soft X-ray domain [6,7] or with weaker hard X-rays [8,9]. 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 [10], which provides a detailed time-dependent description of molecular dynamics under ultraintense x-ray pulses [4]. Pulse duration and pulse energy dependence of the data reveal further subtleties of the interplay between x-ray absorption, electronic relaxation and nuclear motion, yielding a comprehensive picture of XFEL interactions with small molecules.

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Primary author: RUDENKO, Artem (Kansas State University)

Presenter: RUDENKO, Artem (Kansas State University)

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