Exotic forms of matter in atoms and molecules driven by free-electron lasers

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We discuss novel forms of matter created in atoms and molecules interacting with FELs, compare with experiments and provide theoretical support for experimental results.

Free-Electron Lasers (FEL) are new Light Sources which are promising to bring a revolution in our understanding of electron and nuclear dynamics inside driven complex molecules. FEL-pulses are orders of magnitude more intense than the pulses provided by conventional synchrotron radiation sources. FELs have short duration and short-wavelength, ranging from XUV pulses of a few eV to hard Xrays of a few thousand eV. These properties cause the laser to boil away the electrons from the inside out, a fascinating aspect of the interaction of matter with FEL-radiation. That is, when molecules interact with FELs, inner-shell electrons are ionized by single-photon absorption, creating an inner-shell hole. As a result, an Auger process takes place with an outer-shell electron dropping to fill in this hole and another outer-shell electron ionizing. Multiple sequential single-photon ionization (SPI) processes and cascades of Auger transitions take place. This fast electronic arrangement leads to the formation of exotic forms of matter, that is, novel, far from equilibrium, states with multiple inner-shell holes.

Employing accurate continuum molecular orbitals we identify the FEL parameters for the highest contribution of double core hole states to the formation of atomic ion fragments in N_2 for high photon energies [1]. Moreover, for Ar and small photon energies we identify the main single-photon and direct multi-photon pathways contributing to the formation of Ar ion states and provide theoretical support for experimental results [2]. In addition, unlike to common expectation, we predict that for 93 and 115 eV photon energies and attosecond duration pulses the Xe^{5+} atomic ion is formed by direct two-photon absorption [3]. Finally, we discuss very recent work that supports experimental FEL results.

References

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