Nonlinear resonant core-excitations in molecules.

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The resonant interaction of x-rays with matter is central for exploiting site-selectivity in molecular systems. However, this interaction remains largely unexplored when subjected to intense x-ray pulses, which can trigger subsequent processes like stimulated emission and stimulated x-ray Raman scattering [41], as well as create induced resonances and multiple resonant excitations. We demonstrated experimentally that fully resonant sequential multi-core excitations to exotic neutral multi-core excited states ($K^{-1}K^{-1}V^{2}$) are feasible[1]. Notably, the resonant transition energies are only slightly affected by a preceding resonant excitation at a different atomic site in the molecule (∼ 1-2 eV), in contrast to sequential ionization energies (≥10 eV).Thus, multiple resonances at different atomic sites of the same element can in fact be covered within the spectral content of a single XFEL pulse. We have experimentally confirmed the formation of double-core-excited molecule in the Nitrogen molecule at the SQS endstation of the EuXFEL, by comparing electronic decay spectra obtained at high and low x-ray intensity. The high intensity signal included distinct peaks corresponding to the unique decay channels where one or both of the core-excited electrons take an active role (so-called participator-spectator and participator-participator decays). The observed features are in very good agreement with calculated decay spectra obtained using a more realistic approach for continuum states involved in the processes.

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