Dynamics in helium nanodroplets studied by ultrafast XUV and X-ray techniques

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Our work focuses on a deeper understanding of the electronic structure and, in particular, the coupled electronic and nuclear dynamics that occur in excited and/or ionized helium droplets.Two complementary projects are discussed. In a series of femtosecond high-harmonic generation based experiments, an XUV pump pulse excites droplets containing N_{He}~$2x10^6$He atoms up to ~24 eV above the ground state. Excited droplets and relaxation products are ionized by a femtosecond infrared(IR) or UV probe pulse. Transient electronic configurations and the release of energy into atomic motion are probed by recording pump-probe delay dependent photoelectron angular and energy distributions as well as mass-resolved ion momenta. Distinctly different dynamics are observed with IR and UV probe photons. While IR probing provides a detailed picture of the ejection of Rydberg atoms and molecules, UV probing gives direct access to interband relaxation dynamics and trajectories that are associated with the formation of covalently bound molecular complexes inside the droplet. The results are discussed in the context of surface vs. bulk processes and connections to ab-initio molecular dynamics simulations are presented. A second project employs a pair ofintense femtosecond X-ray pulses (~840 eV, ~10^{16} - 10^{17} W/cm^2) from the Linac Coherent Light Source (LCLS) to interrogate the coupled flow of charge and energy in highly ionized pure and doped He droplets.For each pulse pair, an ion mass spectrum and an X-ray diffraction patternassociated with a single He droplet are recorded. The results are analyzed as a function of the delay between the two X-ray pulses.Doping large (N_{He}~10^6-10^{10}) helium droplets with ~1% of Xe atoms leads to dramatically different dynamic trends compared to both pure helium droplets and pure Xe clusters.The observations are discussed in the context of possible charge- and energy-transfer mechanisms between the host matrix andthe embedded clusters.