Interatomic Coulombic decay in helium nanodroplets

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Helium (He) nanodroplets are generally considered as an ultracold, inert spectroscopic matrix for embedded, isolated molecules and clusters. Upon ionization using intense or energetic radiation, however, He droplets turn into a highly reactive medium inducing reactions and secondary ionization processes of the embedded species [1]. The dynamics initiated by photoionization of pure and doped He nanodroplets is currently being explored in view of using He nanodroplets to prepare molecular complexes for photoelectron spectroscopy and for x-ray diffraction imaging. Recent examples include the collective autoionization of multiply excited pure He droplets [2] and the creation of doubly charged species by one-photon ionization of doped He droplets [3].

An important correlated ionization process occurring in many weakly-bound molecules and condensed phase systems exposed to XUV and X-ray radiation is interatomic Coulombic decay (ICD) [4]. Surprisingly, even in the extremely diffuse He dimer He_2 (bond length 52 Å), following the simultaneous ionization and excitation of one of the constituent He atoms, ICD is prominent [5]. Here we present a study of ICD in He nanodroplets using synchrotron radiation. Using coincidence imaging, we fully characterize the product ion mass spectra as well as electron and ion kinetic energies. Photoelectron spectra measured in coincidence with He_n^+, n=1,2,3 ions closely match those observed for He_2 implying that ICD proceeds in a binary process and interaction of the emitted electrons with the He droplet is weak. In contrast, He_n^+ ion kinetic energy distributions are substantially perturbed by the presence of the He droplet. Fig. 1a) shows two features in the kinetic energy spectra of He^+ measured for various droplet sizes. The peak around 4.5 eV reflects free Coulomb explosion of the pair of He^+ ions as observed for ICD occurring in He_2 (pink line, [5]). The prominent feature around 1 eV we attribute to billard-like collisions between the exploding He^+ ions and neighboring He atoms in the droplets, which effectively lead to delayed Coulomb explosion starting from a larger distance. Various scenarios of ICD taking place inside or on the surface of droplets are identified [Fig. 1b)].

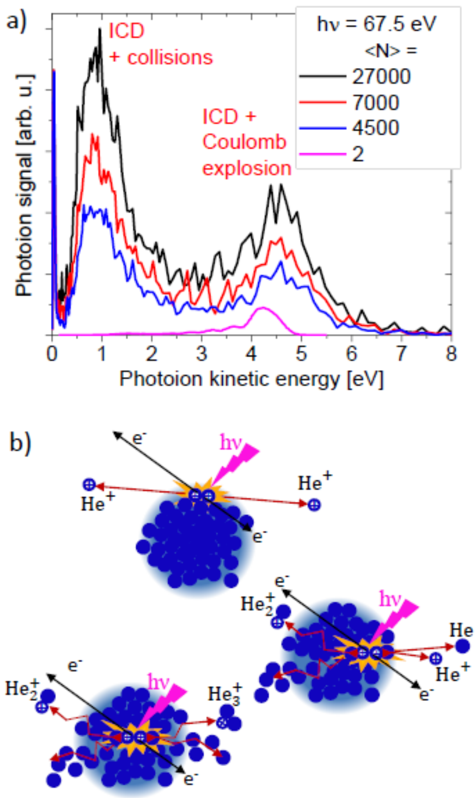

Figure 1. a) He^+ ion kinetic energy distributions measured upon irradiation of He droplets of given sizes by XUV radiation at hν=67.5 eV. **b)** Illustration of possible scenarios of ICD occurring at the surface or inside He nanodroplets.

References

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