Photodissociation and Photoassociation inside He Nanodroplets

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Helium nanodroplets (He_N) provide promising opportunities to explore the dynamics of photochemical reactions in the time domain, in particular for aggregates that are otherwise difficult to form. Time-resolved photoelectron spectroscopy has proven to be a valuable method for following the electronic population dynamics of dopants inside He_N. In contrast, insight into nuclear structure dynamics of dopants is usually veiled in He_N due to the large solvation energy of ions which prevents their detection. Ion solvation also impedes electron-ion coincidence techniques, which would provide unique insight into fragmentation processes and the underlying conversion of electronic into nuclear energy.

Ion ejection from droplets can, however, be facilitated by supplying additional kinetic energy, for example, through resonant excitation of nonbonding ionic states, as recently demonstrated for In_2^+ [1]. Ionization to dicationic states with strong Coulomb repulsion presents another way to supply the ions with sufficient kinetic energy to overcome the droplet solvation potential. This approach naturally leads to Coulomb explosion imaging (CEI), a technique that directly measures the interatomic distance of two cations. We apply CEI to study the photodissociation of iodine (I_2) excited above the A-state dissociation limit inside helium droplets. The time-dependent ion velocity distributions inform about the kinetics of the dissociating I atoms within the droplet, including deceleration in the neutral and cationic state and respective solvation energies.

In a third example, the photoinduced formation of van der Waals clusters from a metastable, foam-like aggregate of Mg atoms is presented, with focus on underlying energy transfer processes. Energy pooling of several photoexcited Mg* atoms populates highly-excited Mg states and yields energy transfer to kinetic degrees of freedom. The additional kinetic energy released during the cluster formation process in combination with probe photon energy facilitates cluster fragmentation and ion fragment ejection from the droplet. The covariance detection of electrons with ejected ions allows us to separate the photoinduced cluster formation process from the concurrent photoexcitation dynamics of initially dense clusters, which are present in some of the droplets. Only due to this separation we gain insight into parameters like cluster formation time, energy flow to highly excited states and electronic-to-nuclear energy conversion.

References:
[1] PCCP 24, 24727 (2022)

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