Ultrafast electronic dynamics in Helium nanodroplets studied by femtosecond time-resolved EUV photoelectron imaging

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Synopsis: We have performed the first femtosecond EUV-pump, IR-probe experiment to study the photoionization dynamics of pure Helium nanodroplets below the atomic Helium IP in real-time. Using Velocity-Map Imaging (VMI) photoelectron spectroscopy we were able to discern processes with associated timescales ranging from tens of femtoseconds to tens of picoseconds. The results will be discussed in the light of complementary energy-domain studies and theoretical models of the droplet's electronic and nuclear dynamics.

The emerging field of ultrafast x-ray science is at a point at which its true potential for revealing electronic and structural dynamics in the chemical and material sciences has to be demonstrated. In particular the study of complex systems will provide critical benchmarks for the transition from a specialists' discipline to a widely-used tool.

Helium nanodroplets constitute a unique cryogenic matrix for the creation, isolation and spectroscopy of regular and exotic species, such as free radicals and molecules in high-spin states. Comprised of thousands to millions of atoms at cryogenic temperatures of ~0.4 K the droplets readily pick up atoms and molecules but interact only very weakly with the respective dopants due to their superfluid nature. The electronic structure and relaxation dynamics of Helium droplets have been subject of a number of synchrotron-based studies which have revealed rich but so far largely unexplained structures in EUV absorption and photoemission spectra [1].

We have set up a high harmonic generation based femtosecond EUV-pump, IR-probe photoelectron imaging experiment, Fig. 1, to extend EUV photoemission studies into the time-domain. In the first series of experiments we have monitored the electronic relaxation dynamics of large ($N\sim10^6$) pure Helium droplets from the initial EUV excitation at 23.7 eV to the emergence of both neutral and charged products in real-time. Transient photoelectron energy- and angular-distributions recorded by means of VMI photoelectron imaging reveal rich dynamics with associated timescales ranging from the femtosecond to the picosecond regime. Clear correlations between spectral features, angular distributions and dynamics timescales have been established.

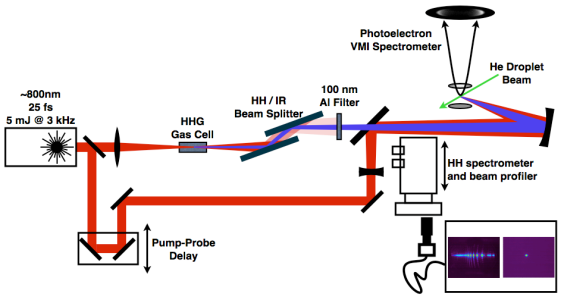

Fig. 1. Schematic of the experimental setup.

We observe transient spectral features similar to atomic Rydberg state photoionization spectra but which display dramatically different dynamics compared to free Rydberg atoms. Despite the complexity of the system, these and other findings can be explained by a relatively simple model that involves contributions from atoms that reside either on the droplet surface or in the bulk. The model will be discussed along with possible alternatives and the results of *ab-initio* calculations on the EUV excited states of Helium clusters.

References

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