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Femtosecond time-resolved imaging of Rydberg atom emission from electronically excited helium nanodroplets OLIVER GESSNER, OLIVER BUENERMANN, OLEG KORNILOV, STEPHEN LEONE, DANIEL NEUMARK, Lawrence Berkeley National Laboratory — The relaxation dynamics of electronically excited helium nanodroplets are investigated using femtosecond time resolved extreme ultraviolet photoelectron- and ion-imaging spectroscopy. The novel time-domain measurements provide new insight into one of the most prominent droplet relaxation channels in which the excitation energy is removed from the cluster by emission of Rydberg atoms. Droplets consisting of $\sim 2 \times 10^6$ helium atoms are excited with 23.6 eV pump-pulses from a high-order harmonic generation (HHG) light source. Photoelectrons and ions are produced by subsequent ionization using 1.6 eV probe-pulses. Strong indications for a very fast (<120 fs) emission of 1s4p Rydberg atoms and a delayed (~ 200 fs) emission of 1s3d Rydberg atoms with significantly different kinetic energies are revealed. The findings are interpreted within a description of excited droplet states by perturbed atomic Rydberg states, leading to a close correlation between the energy and the location of the excitation. The relatively simple model yields surprisingly accurate predictions of the droplet absorption band structure and the emission dynamics of Rydberg fragments, providing a detailed physical picture of a previously proposed intraband relaxation mechanism.

Oliver Gessner
Lawrence Berkeley National Laboratory

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