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Electronic dynamics in helium nanodroplets studied via femtosecond XUV pump / UV probe photoelectron imaging MICHAEL ZIEMKIEWICZ, CAMILA BACELLAR, STEPHEN LEONE, DANIEL NEUMARK, OLIVER GESSNER, Ultrafast X-ray Science Laboratory, Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States — Superfluid helium nanodroplets consisting of $\sim$ 2 x 10^6 atoms are examined using femtosecond time-resolved photoelectron imaging. The droplets are excited by a 23.6(2) eV extreme ultraviolet (XUV) pulse in resonance with an electronically excited band associated largely with the 1s3p Rydberg level of free He atoms. Relaxation dynamics are monitored by ionizing transient states with a 3.2 eV probe pulse and measuring the time-dependent photoelectron kinetic energy distributions using velocity map imaging (VMI). A broad, intense signal associated with the initially excited 1s3p band ($E_{kin} \approx 2.5$ eV) appears within the experimental time resolution and decays within 190(70) fs. Concomitantly, a second photoelectron feature with kinetic energies ranging from 0 to 0.5 eV appears on a time scale of $\sim$ 200 fs. The new feature is identified as originating from the 1s2p droplet Rydberg band, indicating the direct observation of a previously suggested interband relaxation within the droplet. This feature also decays within $\sim$ 200 fs, likely due to intraband relaxation within the 1s2p/1s2s manifold to states which are too deeply bound to be ionized by the 3.2 eV probe pulse.

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