Femtosecond coherent spectroscopy of doped helium droplets

Frank Stienkemeier[1]

Institute of Physics, University of Freiburg, Hermann-Herder-Str. 3, 79104 Freiburg, Germany

Femtosecond time-resolved spectroscopy has revealed insight into the dynamics of processes induced by laser excitation of doped helium nanodroplets. On the one hand, pump-probe action spectroscopy allows a direct observation, e.g. of the desorption of dopants from the droplets' surface or exciplex formation of alkali-doped droplets. Even XUV femtosecond pulses can be used to study the dynamics upon electronic helium excitations. On the other hand, coherent femtosecond methods as e.g. wave packet interferometry or 2-dimensional spectroscopy have opened new directions to study ultrafast dynamics in complex quantum systems. While most experiments so far have addressed condensed phase samples, the application of such methods to dilute gas phase samples or doped helium droplet beams is challenging because of the low-density targets. However, isolation in helium droplets at millikelvin temperatures provides unique opportunities to synthesize well-defined complexes, to prepare specific rovibronic states, and to study their dynamics. To account for the small densities in our samples, we implemented a phase-modulation technique in order to reach enough sensitivity and a high spectral resolution in electronic wave packet interferometry experiments. The combination with mass-resolved ion detection enabled us e.g. to characterize vibrational structures of excimer molecules. By extending this technique we have observed collective resonances in samples of very low density. Results on multiple quantum coherences in atomic vapors and supersonic doped helium droplet beams are discussed in the context of weakly interacting many-body quantum systems. With a variant of this method, we are currently elaborating the implementation of nonlinear all-XUV spectroscopy.

[1]E-mail: stienkemeier@uni-freiburg.de