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Markus Koch

Institute of Experimental Physics, Graz University of Technology, Graz, Austria

über

Ultrafast photoexcitation dynamics of atoms and molecules inside helium nanodroplets

Superfluid Helium nanodroplets (He_N) offer fascinating opportunities for spectroscopic studies of atoms, molecules and aggregates because of their low temperature of 0.4 K and the high degree of control in loading the droplets. Systems investigated with frequency-domain spectroscopy include charge and energy transfer in tailor-made or weakly bound aggregates, and the investigation of solvent influence in a growing microsolvation environment. Here, we present the development of He_N for time-domain studies, aiming at a broad application of these nano-cryo-reactors to investigate the dynamics of photophysical and photochemical processes in real time.

In a first step, we were able to observe the photoexcitation dynamics of a single atom (In) solvated inside a He_N with femtosecond time-resolved photoelectron spectroscopy [1,2]. This experiment yields three important findings: (i) it demonstrates that photoelectrons are a good observable for ultrafast processes inside He_N , (ii) it enabled a characterization of the response of superfluid He to photoexcitation of solvated atoms/molecules and, (iii) it showed that these processes can be modeled with time-dependent helium density functional theory simulations.

In a second step, we investigated the He_N influence on coherent nuclear dynamics by exciting a vibrational wave packet (WP) in dimer molecules (In_2) [3]. Very surprisingly, we find that the coherent WP signal can be observed for tens of picoseconds, demonstrating that the perturbation imposed by this quantum liquid can be lower by a factor of 10-100 compared to any other solvent.

Finally, in a recent effort to characterize the propagation of free electrons within the helium nanodroplets, we were able to demonstrate laser-assisted electron scattering, an energy transfer process between strong laser fields and free electrons, for the first time in the condensed phase [4].

References

- [1] Nat. Commun. **9**, 4006 (2018)
- [2] J. Chem. Phys. **152**, 014307 (2020)
- [3] Phys. Rev. Lett. **124**, 115301 (2020)
- [4] Nat. Commun. **12**, 4204 (2021)

ZOOM Link

<https://jku.zoom.us/j/92739438925?pwd=YU1hSSt0RFY3Ny9mVUs4eUdKcVZLUT09>

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