FEMTOSECOND PHOTOEXCITATION DYNAMICS OF DIMERS SOLVATED IN HELIUM NANODROPLETS: VIBRATIONAL WAVEPACKETS AND PREDISSOCIATION

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Ultrafast pump-probe photoionization experiments were conducted on dimers solvated inside liquid helium nanodroplets (He_N) to gain insights into the influence of the quantum solvent on nuclear and electronic dynamics. We first present coherent vibrational wave packet dynamics of Al_2 inside He_N, where coherence is preserved for tens of picoseconds, in analogy to our previous work on In_2 [1] . The short revival time of ~ 30 ps of Al_2 will allow us to investigate the influence of additional solvent molecules on the wavepacket motion.

Time-resolved photoion spectroscopy of In_2 revealed the detachment of $InHe_N^+$ ions from the helium droplet following probe-pulse ionization [2]. Detection of the ion fragments, which are typically trapped inside the droplet, is enabled by electronic excitation of the ion during the ionization process. The excess energy provides the ion fragments with sufficient kinetic energy to overcome the attractive He_N potential. Ion detection enables electron-ion correlation spectroscopy within liquid helium droplets, which provides deeper insight into reaction pathways, particularly in cases where significant kinetic energy is released during fragmentation.

Time-resolved photoion spectroscopy was furthermore employed to investigate iodine molecules inside He_N. Excitation to the bound B-state did not yield wave packet oscillation, as observed in the gas phase, but instead exhibited immediate ground-state bleach. With velocity map imaging, we observe IHe_N^+ fragments appearing after 1 ps and remaining at least up to 150 ps. Comparison with A-state dissociation of gas-phase I_2 suggests rapid and efficient predissociation of the molecule from the B-state via droplet-mediated internal conversion. Large velocities partially exceeding those of Coulomb explosion of gas phase iodine indicate that the fragment iodine atoms are confined within the droplet after dissociation.

[1] doi:10.1103/physrevlett.124.115301, B. Thaler, M. Meyer, P. Heim, and M. Koch, Phys. Rev. Lett. **124**, 115301 (2020)

[2] doi:10.1039/d2cp03571e, M. Stadlhofer, B. Thaler and M. Koch, Phys. Chem. Chem. Phys. **24**, 24727-24733 (2022)

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