Light-induced ultrafast dynamics of molecules in helium nanodroplets

Jian Wu[1]

1) State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai 200241, China
Email: jwu@phy.ecnu.edu.cn

The newly developed reaction microscopy of helium-nanodroplet (He_N) target recoil ion momentum spectroscopy (He_NTRIMS) allows us to explore the ultrafast dynamics of in-droplet molecules driven by femtosecond laser pulses [1,2], where the electrons and ions ejected from an in-droplet molecule are measured in coincidence.

By performing pump-probe experiments in the He_NTRIMS, we capture in real time the collision-induced ultrafast dissipation of vibrational nuclear wave packet dynamics of D_2^+ ion embedded in a He_N [3]. Our results show that, differing from the behaviors of the in-droplet neutral molecules, the charged ions in the He_N interior strongly couple to the He solvent via ion-He collisional interactions, leading to the extremely fast collisional dissipation dynamics within ~140fs. Our findings underscore the crucial role of ultrafast collisional dissipation in shaping the molecular decoherence dynamics during solution chemical reactions, particularly when the solute molecules are predominantly in ionic states.

Furthermore, we explore the spatial scale of the matter wave of a cold molecule with respect to the He_N by diagnosing the angular nodal structures in the photoelectron momentum distributions (PMDs). The preserved and nodal structures in the PMDs allows us to identify and characterize the delocalization of the lightest H_2 and localization of heavier D_2 and O_2 molecules, whose de Broglie wavelength is comparable to or smaller than droplet size.

[1] J. Qiang et al, "Femtosecond Rotational Dynamics of D_2 Molecules in Superfluid Helium Nanodroplets", Phys. Rev. Lett. **128**, 243201 (2022).

[2] L. Zhou et al, "Enhancing Strong-Field Dissociation of H_2^+ in Helium Nanodroplets", Phys. Rev. Lett. **130**, 033201 (2023).

[3] J. Qiang et al, "Femtosecond Collisional Dissipation of Vibrating D_2^+ in Helium nanodroplets", Phys. Rev. Lett. **132**, 103201 (2024).