Density parameterizations for quantum fluids: From unimolecular reactions to multi-adsorbate dynamics

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Tailored representations of the density distribution for the electronic and the nuclear degrees of freedom are benchmarked on model systems (e.g., atomic photoelectron spectra, vibrational predissociation of van der Waals triatomic complexes [1-3]), and subsequently applied to more challenging phenomena (e.g., adsorbate dynamics, elementary excitations in doped helium droplets, interatomic coulombic decay (ICD) in paired quantum dots, non-adiabatic photoisomerization of molecular switches [4-5]). The parameterized density-based simulations target nanoscale systems which constitute promising candidates for technological applications in hydrogen production and storage in nanomaterials, high-resolution spectroscopy, and optoelectronics. We assess the numerical convergence, accuracy and stability of the proposed algorithms for quantum molecular dynamics simulations. On the one hand, the computed observables (such as carrier-envelope phase effects onto the photoelectron spectra, autoionization rates, branching ratio between bound and dissociative reaction channels, unimolecular reaction rates, volumetric and gravimetric hydrogen storage capacities of nanoporous materials, and of the probability density distribution of the molecular wave packet) exhibit a close correspondence with complementary experimental and theoretical results for the target systems. On the other hand, the parameterized densities provide further insight into the mechanisms underlying the target phenomena.

Index Terms: quantum molecular dynamics, wave packet propagation, photoelectron spectra, photoisomerization, adsorbate dynamics, interatomic coulombic decay, liquid density-functional theory.

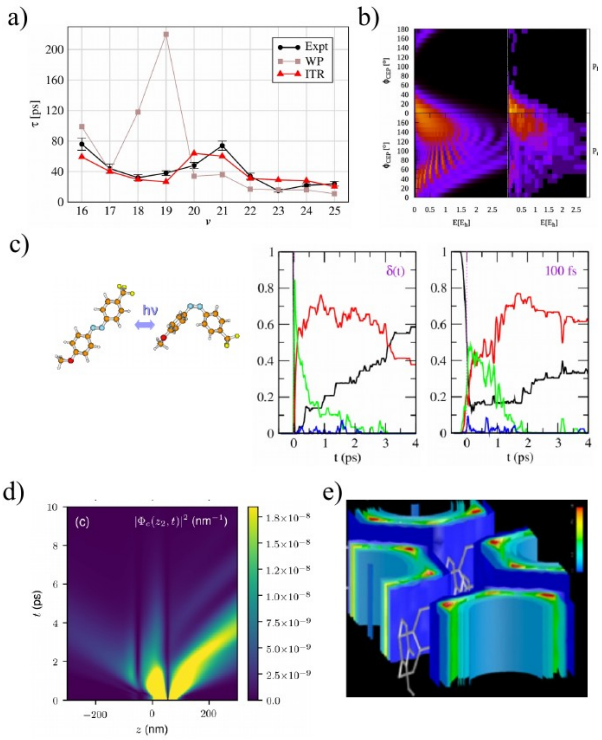

Figure 1: *a) Lifetimes of predissociating $ArBr_2(B,v)$ van der Waals complexes. b) Density map of photoelectron spectra. c) Isomerization dynamics of an azobenzene derivative. d) ICD dynamics in paired gallium arsenide quantum dots. e) Contour plots of the molecular hydrogen density in nanoporous carbon foams.*

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