First-Principles Investigation of Electronic Excitation Dynamics in Water under Proton Irradiation

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A predictive and quantitative understanding of electronic excitation dynamics in water under proton irradiation is of great importance in many technological areas ranging from proton beam therapy to preventing nuclear reactor damages. Despite its importance, an atomistic description of the excitation mechanism has yet to be fully understood. Identifying how a high-energy ion dissipates its kinetic energy into the electronic subsystem is crucial for predicting atomistic damages, which result in the formation of various radiolytic products.

In this work, we use our new large-scale first-principles Ehrenfest dynamics method [1,2] based on real-time time-dependent density functional theory to simulate the non-adiabatic electronic response of bulk water to a fast proton. In particular, we will discuss the topological nature of the electronic excitation. We will employ maximally-localized Wannier functions to bridge our quantitative findings from our first-principles simulations to the conceptual understanding held in the field of water radiolysis.

[1] “Plane-wave Pseudopotential Implementation of Explicit Integrators for Time-Dependent Kohn-Sham Equations in Large Scale Simulations” A. Schleife, E. W. Draeger, Y. Kanai, A. A. Correa, J. Chem. Phys., 137, 22A546 (2012)

[2] “Quantum Dynamics Simulation of Electrons in Materials on High-Performance Computers” A. Schleife, E. W. Draeger, V. Anisimov, A. A. Correa, Y. Kanai, Computing in Science and Engineering, 16 (5), 54 (2014).