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Spin-Polarized Nonadiabatic Dynamics with Local Basis Sets

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— Accurate simulations of electron transfer at the solid-electrolyte interphase (SEI) are critical for understanding and predicting electrochemical reactions. Density-Functional Theory (DFT) has been widely applied to study the ground-state structure of novel materials for electrochemical energy storage and for adiabatic molecular dynamics. Unfortunately, many chemical reactions take place on femtosecond time scales where assuming adiabatic electron density propagation is invalid. To resolve this, we have developed the ability to perform time-dependent DFT calculations using nonadiabatic propagation of the electron density along with Ehrenfest dynamics for the ions to better capture the complex interactions that occur during surface-electrolyte electron transfer and chemical reactions. Spin polarization is also implemented to improve the accuracy of the simulations and their suitability for studying a wider range of chemical reactions. Local basis sets are implemented via linear combinations of atomic orbitals to reduce the size of the DFT basis for computational efficiency.

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