Extending the tool set for non-adiabatic molecular dynamics to describe excited states in solids

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Describing excited states in extended systems, i.e. solids, surfaces, or condensed-phase systems, poses a challenge to state-of-the-art quantum chemistry: Photo-chemical processes in crystals or networks often depend crucially on the system's topology [e.g. 1], highlighting the importance of implying periodic boundary conditions for theoretical simulations. However, periodicity in combination with often extended unit cell sizes standardly restricts investigations to time-dependent density functional theory (TDDFT) ansätze, entailing inherent short-comings of the method. Motivated by the purpose to extend the state-of-the-art applicability of TDDFT [2], with the aim to create a sophisticated tool set for the static and dynamic description of excited states, recent TDDFT method developments will be presented including a short summary on perturbative spin-orbit coupling corrections [3] and extensions to describe semi-local core states in transition metals within the Gaussian and augmented plane wave method [4], as well as a detailed overview on the interface of the program packages CP2K and NEWTONX, extending the electronic-structure code towards efficient non-adiabatic excited-state dynamics [5,6], featuring a smeared occupation ansatz to capture static correlation.

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