Describing excited states in solids: from static excited-state properties to trajectory surface hopping

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The plethora of photochemical processes that need to be unraveled when aiming for a comprehensive description of excited states in solids becomes apparent when considering the process of upconversion: Converting two low energy photons to a high energy one is of vital importance for both catalytical as well as bio-medical applications, holding the promise to enhance both sensing, phototherapy, and theranostics as well as photocatalysis under low solar irradiance. The challenge of overcoming the systematically too low quantum yields demands an atomistic understanding of the full range of relevant photochemical mechanisms, emphasizing the need for an advanced quantum-mechanical tool set for the description of excited states within time-dependent density functional theory implying periodic boundary conditions. This presentation summarizes first steps towards establishing such a tool set within the electronic structure code CP2K and the mixed quantum classical dynamics code Newton-X, with recent developments ranging from semi-empirical and approximate hybrid density functional kernels for static absorption and emission spectroscopy [1], the extension of the Gaussian and plane wave ansatz to the Gaussian and augmented plane wave ansatz [2], the inclusion of perturbative spin-orbit coupling [3], establishing an interface between CP2K and Newton-X, enabling surface hopping dynamics for extended systems implying periodic boundary conditions [5, 6] to capturing static and dynamic correlation [7].

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