Electron-nuclear dynamics in irradiated systems: Ehrenfest and beyond

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The process of irradiation of matter by high-energy charged particles or EM radiation starts with the ionization of the material and the consequent generation of secondary electrons, holes and radicals. These species then diffuse through the sample experiencing inelastic collisions and losing energy to the medium until they find an opportunity to react, and produce chemical modifications that lead to various types of damage and, eventually, trigger a macroscopic response in the form of failure (swelling, fracture, explosion), or death (in the case of biological matter). The theoretical description of this process requires a coupled treatment of electronic and nuclear dynamics, at a quantum-mechanical level. Needless to say that, even resorting to a minimal mathematical description of the electronic and nuclear components, this is a monumental task. Fortunately, in many cases of interest the various stages of this process happen in different, non-overlapping time scales. Therefore, the problem can be attacked using a multiscale (or many-scale) strategy.

The initial stage involves electronic excitation and ionization. This occurs un the atto- to femtosecond time scale, so that nuclei do not react and can be taken as fixed, classical particles. This regime is quite well described by time-dependent density functional theory (TDDFT) with fixed nuclei, except for the projectile which moves with practically constant velocity while depositing a small fraction of its energy in electronic excitations. I will discuss the extensive work that has been carried out during the past decade in the computation of electronic stopping power via TDDFT [1-10]. In the following stage, electrons (and holes) equilibrate between themselves via e-e collisions, which are absent TDDFT. To go beyond, one can resort to non-equilibrium Green's functions techniques [11] or, alternatively, one can supplement TDDFT with (incoherent) e-e collisions in a stochastic manner [12]. Assuming that, during electronic equilibration, the nuclear degrees of freedom remain unresponsive, this stage does not need to be dealt with explicitly, and can be replaced by an initial Fermi-Dirac electronic distribution corresponding to an electronic temperature that depends on the energy absorbed in the initial stage. If, however, nuclei do respond in this time scale, then e-e collisions must be treated together with electron-nuclear interactions. If we ignore e-e collisions we can implement this coupled dynamics by combining mean-field TDDFT for the electrons with a classical description of the nuclei where the forces are calculated for the time-evolving electronic density. This is the *Ehrenfest* approximation, which works generally well, except for the fact that it does not include spontaneous phonon emission, and hence cannot lead to equilibration between electrons and nuclei [14]. Several methods have been proposed to go beyond Ehrenfest. Here I will focus on the *Correlated Electron-Ion Dynamics* approach (CEID) [13] and more specifically on a recently proposed cost-effective alternative that limits the nuclear motion to harmonic vibrations, named *Effective CEID* (ECEID) and applied to thermalization [15] and to the inelastic electron transport in water chains [16].

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