Analysis and Control of Electronic Motion in the Time Domain

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This lecture is about how electronic motion can be monitored, analyzed and, ultimately, controlled, in real time. In particular:

(i) A novel approach to describe **electronic transport** through single molecules or atomic wires, sandwiched between semi-infinite leads, will be presented. The basic idea is to propagate the time-dependent Kohn Sham equations in time upon ramping up a bias between the metallic leads. In this way, genuinely time-dependent phenomena, not accessible in the standard Landauer approach, can be addressed. For example, employing an Anderson model, we demonstrate that Coulomb blockade corresponds, in the time-domain, to a periodic charging and discharging of the quantum dot [1].

(ii) With modern pulse-shaping facilities, the **control of electronic motion** is becoming more and more realistic. By combining quantum optimal control theory with TDDFT, we calculate shaped laser pulses suitable to control, e.g., the chirality of currents in quantum rings [2], the location of electrons in double quantum dots, as well as the enhancement of a single peak in the harmonic spectrum of atoms and molecules.

(iii) In all practical TDDFT calculations, approximate forms of the exchange-correlation potential need to be employed. One of the most popular approximations, the adiabatic local-density approximation (ALDA) will be analyzed as to whether the main error comes from the adiabaticity assumption, i.e. locality in time, or from the LDA, i.e. locality in space. For an exactly solvable model where the **exact adiabatic approximation** can be extracted, we find the surprising fact, that the adiabaticity assumption can be an excellent approximation even in highly intense laser fields [3].

(iv) Finally, the coupling between electronic and nuclear motion will be addressed. As a first step towards a full ab-initio treatment of the coupled electron-nuclear motion in time-dependent external fields, we deduce an exact factorization of the complete wavefunction into a purely nuclear part and a many-electron wavefunction which parametrically depends on the nuclear configuration. We derive formally exact equations of motion for the nuclear and electronic wavefunctions [4]. These exact equations lead to a rigorous definition of **time-dependent potential energy surfaces** as well as **time-dependent geometric phases**. With the simple example of the hydrogen molecular ion in a laser field we demonstrate the significance of these concepts in understanding the full electron-ion dynamics. In particular, the time-dependent potential energy surfaces are shown to represent a powerful tool to analyse and interpret different (direct vs. tunneling) types of dissociation processes.

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