Interaction of molecules with ultrashort intense laserpulses
A nonadiabatic quantum molecular dynamics approach

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We present a nonadiabatic quantum molecular dynamics approach, which is suitable to study the response of large polyatomic systems to intense laser pulses. Starting from a quantum-classical Lagrangian, coupled equations of motion for electrons and ions are derived. The electronic wavefunction is propagated according to the Kohn-Sham equation of time dependent density functional theory. We implemented this scheme using a density functional based tight-binding method [1] where the necessary matrix elements are determined within a confined LCAO basis in a two-center representation. Forces on the nuclei are obtained from a generalized Ehrenfest theorem and the external radiation field is incorporated via minimal coupling, where following Graf and Vogel the field dependent matrix elements may be rewritten in a numerical feasible way [2]. Results show the dominate photoexcitation of the breathing mode in C_{60} and the red-shift of this mode due to ultra-intense laser pulses with fluences on the order of 4.0 KJ/m^2. These findings agree well with pump-probe experiments on thin films recently performed by Dexheimer *et al.* [3].

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