Spectral Propagation Schemes for TDDFT Calculations with Applications to Molecules and CNTs

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In nanoelectronic applications, efficient time-dependent simulations have become increasingly important for characterizing the electron dynamics under time dependent external perturbations such as electromagnetic fields, pulsed lasers, and particle scattering. Reliable modeling approaches in time domain, however, are often limited in term of trade-off between robustness and performances, and a direct numerical treatment is difficult.

In this work, we aim to go beyond these limitations by introducing high-performance numerical propagation schemes to compute the solution of the time-ordered evolution operator. The numerical treatment of these evolution operators often gives rise to the matrix exponential, commonly treated using approximations such as split-operator techniques. In contrast, the efficiency of the time-domain propagation techniques described here, is enhanced by reliance on the capabilities of the FEAST algorithm for solving the eigenvalue problem [1]. Using FEAST the eigenvalue problem is reformulated into solving a set of well-defined independent linear systems along a complex energy contour. Obtaining the spectral decomposition of the matrix exponential (i.e. direct diagonalization of the Hamiltonian) becomes then a suitable alternative to PDE based techniques such as Crank-Nicolson schemes, since larger time-intervals can now be considered and all the resulting linear systems can be solved in parallel. Additionally, FEAST has the ability to re-use the basis of a precomputed subspace as suitable initial guess for solving the series of eigenvalue problems that are close one another all along the time-domain propagation. In order to reduce even further the number of large-scale eigenvalue problems to be computed, two highly optimized propagation schemes (Gauss and BTPS) [2] have also been proposed and implemented.

Within RT-TDDFT (ALDA) framework, our model consider all-electron calculations, real-mesh techniques (finite-element method), and real time propagation of the Kohn-Sham orbitals. In order to obtain the optical responses, we apply a weak delta kick to the system, let it evolve over time, then Fourier transform the dipole moment to get the optical oscillator strength. The spectroscopy results shows very good agreement with the experiments for various molecules ranging from H_2 to C_{60}. The time-dependent framework has also been used to obtain the THz response of metallic (5,5) CNT (isolated) devices and reproduce the experimental results of the Fermi-velocity and kinetic inductance [3].

[1] E. Polizzi, Phys. Rev. B **79**, 115112 (2009).

[2] Z. Chen and E. Polizzi, Phys. Rev. B **82**, 205410 (2010).

[3] Z. Chen, S. Yngvesson, and E. Polizzi, 11th IEEE-NANO 2011, Conf. Proceed. 1339 (2011).

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