Computational first-principle real-time spectroscopy with applications to nanoplasmonic in 1D Carbon-based materials

Eric Polizzi

Department of Electrical and Computer Engineering,
Department of Mathematics and Statistics,
University of Massachusetts, Amherst, USA

e-mail: polizzi@ecs.umass.edu

First-principle calculations are becoming critical to provide evidences of collective plasmon excitations in 1D Carbon-based materials along with quantitative results for the relevant physical properties (e.g. plasmon velocity). The chief signature of 1-D plasmons is a high-frequency excitation (i.e. energy resonance) that depends inversely on the length of the conductor. These simulations are known to be challenging, since they should be capable to provide reliable information on the many-body excited-states (beyond ground state theory), and also to address large-scale computational needs of finite dimensional systems (beyond the solid-state unit-cell). The time dependent density functional theory (TDDFT) [1] provides a practical (i.e. numerically tractable) path for investigating the dynamics of quantum systems and performing excited states calculations. Our numerical methodology is making use of a real-time TDDFT framework [2] which is well-suited to accurately capture various nanoscopic effects including band-to-band transitions, single particle excitations and the various collective plasmon excitations. In essence, once all the occupied ground-state wave functions solution of the DFT Kohn-Sham equations are obtained, they are propagated (non-linearly) in time using a time-dependent Schrödinger-type equation and after a short-polarized impulse is applied in any given direction of the molecular system. Using the solution of the electron density at each time step, the induced dipole can then be computed in time-domain and the imaginary part of its Fourier transform provides the dipole strength function and the absorption spectra. While the design of advanced time-dependent exchange-correlation functionals is still a challenging task, ALDA (adiabatic local density approximation) for TDDFT has been very successful for providing accurate absorption spectra of a large number of complex molecular systems which can be quantitatively compared with the experimental data. In addition, the capabilities of the new FEAST-based framework [3, 4, 5] have considerably broadened the perspectives for ever higher level of accuracy and high-performance in these large-scale calculations. From atom and benzene-like chain structures to finite-length carbon nanotubes, our numerical results will be thoroughly discussed [6].

References

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