

Optical spectrum of semiconductors by time-dependent density-functional theory with the exact-exchange method

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July 22, 2002

Abstract

Unlike electronic ground-states for which standard density-functional theory (DFT) approximations work well for most purposes, study of electronic optical excitations remains as a challenging area for first-principles calculations. Recently, realization of Green's function approaches solving the Bethe-Salpeter equation starting from GW quasiparticles appeared as a successful scheme for such purpose, but due to its high computational cost looking for an efficient alternative is still desirable. DFT approach in principle represents a rigorous alternative of optical spectrum calculation with the time-dependent (TD) extension. However, standard approximations to the exchange-correlation potential and kernel have so far failed to describe electronic excitations of solids. In this workshop, I will report a successful TDDFT calculation of optical spectrum of bulk silicon including excitonic features based on the novel exact-exchange (EXX) method: (1) The EXX at the one-particle level without any post-DFT modification such as the quasiparticle shift already gives a much improved spectrum compared with previous local and semilocal approximations. This indicates the importance of employing a qualitatively correct (self-interaction free) Kohn-Sham (KS) potential and corresponding KS eigenvalues. (2) Remaining excitonic effects are obtained at the TD level with the incorporation of the EXX kernel. Analysis of the EXX kernel shows that inclusion of the coupling of electron-hole pairs at different \vec{k} -points and the correct long-wavelength behavior in the kernel is crucial for the proper description of excitonic effects in semiconductors, while the adiabatic approximation can be justified.