Real-time TDDFT calculation in molecules and solids

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We have been developing a computational method to solve the time-dependent Kohn-Sham equation in real time and real space to describe electron dynamics induced by optical field. The method may be applied to both isolated molecules and bulk periodic solids. In the presentation, I would like to present our recent progresses with the method taking examples from both molecules and solids.

In molecules, we have recently developed a real-time computational method for magnetic circular dichroism (MCD) [1]. In our framework, we solve the time-dependent Kohn-Sham equation under a finite magnetic field. We may thus obtain the MCD spectra of both discrete and continuum region without any perturbative expansion.

In periodic solids, we may describe nonlinear electron dynamics in dielectrics induced by an ultrashort laser pulse. We show our recent application to explore a generation mechanism of coherent phonon in typical dielectrics, Silicon [2]. It has been invoked that there are two mechanisms in coherent phonon generation, the impulsively stimulated Raman scattering induced by a virtual electronic excitation during the irradiation of the laser pulse, and the displacive excitation through a real excitation of electrons. We show that the time-dependent density functional theory describes both mechanisms in a unified way.

[1] K.M. Lee, K. Yabana, G.F. Bertsch, J. Chem. Phys. 134, 144106 (2011).
[2] Y. Shinohara, K. Yabana, Y. Kawashita, J.-I. Iwata, T. Otobe, G.F. Bertsch, Phys. Rev. B82, 155110 (2010).