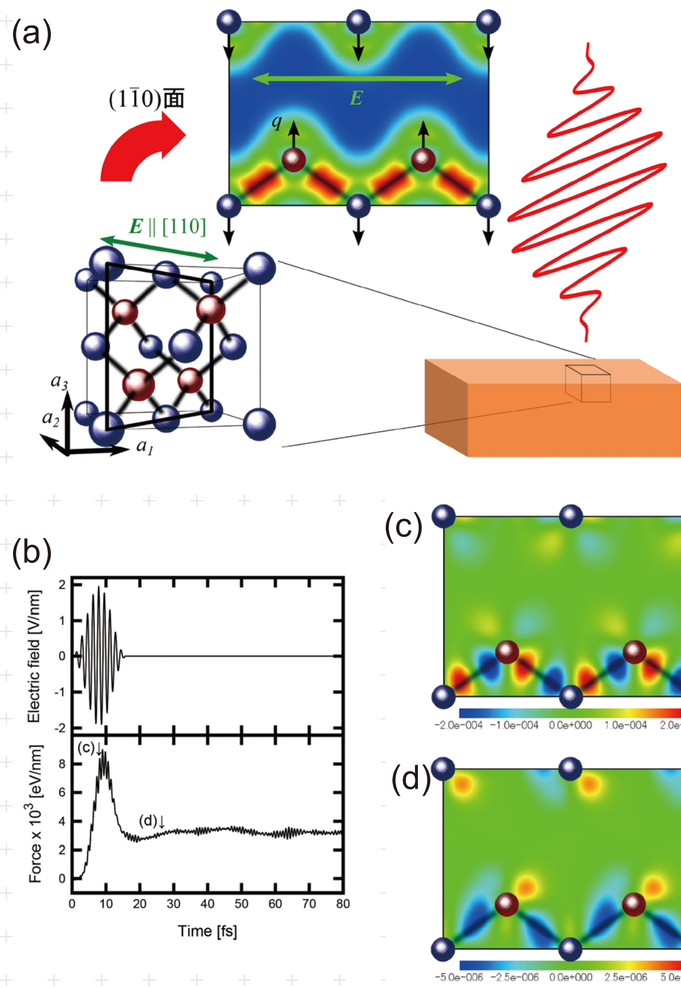
Quantum Condensed Matter Physics

Time-Dependent Density Functional Theory for Ultrafast Dynamics Induced by Laser Pulses

Time-dependent density functional theory (TDDFT) is widely used to describe electron dynamics in atoms, molecules, and solids induced by light fields. We have been developing a real-time and real-space computational code to solve the time-dependent Kohn-Sham equation in crystalline solid, the basic equation in TDDFT. The code is efficiently parallelized in k-points and orbitals in the time evolution calculations. The code may be used to calculate dielectric function, ultrafast electron-phonon dynamics of femto- and attosecond time scale induced by intense and ultrashort laser pulses, and coupled dynamics of electrons and electromagnetic fields in the multiscale implementation of the code.

Recently we have applied the real-time TDDFT method to the coherent phonon generation in GaAs, a well-known compound semiconductor [see Fig. (a)], induced by ultrashort laser pulses. Figure (b) shows laser electric field (top) and the force (bottom) acting on phonon mode induced by the laser pulse. The electron density change from the ground state is shown when the laser field is maximum, and when the laser field ended, as shown in Figs. (c) and (d) respectively.



Numerical Simulation on the Multiple Exciton Generation in Single-Walled Carbon Nanotubes

The multiple exciton generation (MEG) in single-walled carbon nanotubes (SWNTs) is theoretically investigated. We perform the calculation of the MEG rate by considering the direct photogeneration of multiple excitons. In particular, we only consider the generation of two-exciton states. A single photon does not excite two excitons since it is forbidden by the selection rule. However, it is possible to consider the situation where superposed states of multiexciton states are realized because the Coulomb interaction can resonantly couple among multiexciton states. This process was originally proposed for the MEG in nanocrystal. The direct photogeneration produces many excitons by a single photon absorption and thus it is the simultaneous process [see Fig. (e)].

By solving the Bethe-Salpeter equation within the single orbital tight-binding approximation, our calculation shows that it is possible to generate multiexcitons by a single photon as a consequence of the resonant coupling between optically active single-exciton state and the multiexciton states. Generation rates for one exciton shows the usual structure of the linear absorption spectrum with the lowest exciton state, E_{11}, and higher states including the continuum of excitons above the band gap [Fig. (f)]. On the other hand, the generation rate of the two-exciton abruptly begins at the threshold excitation energy, which is two times larger than the lowest exciton energy, i.e. $2E_{11}$. This fact corroborates the fact that the single photon can generate multi excitons. This is a consequence of the strong Coulomb interaction between excitons and of the van Hove singularity of the density of states for the multiexcitons. This is very unique feature of SWNTs that have quasi-one-dimensional structures. Furthermore, our calculation predicts the threshold energy for MEG as $2E_{11}$, which is consistent with the recent experimental results. The present work clarifies the microscopic process of MEG process in SWNTs and sheds light on the underlying physics of the MEG.