Ab-initio description for the interaction of intense laser pulses with solids

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We have been developing a first-principles description for electron dynamics in crystalline solids based on the time-dependent density functional theory (TDDFT). In my presentation, I first explain theoretical and computational aspects of our method, and then show applications to electron dynamics in crystalline solids induced by intense and ultrashort laser pulses.

We consider electron dynamics in infinitely periodic system under a spatially-uniform, time-dependent electric field. To achieve it in practice, there are two issues, the choice of gauge and the treatment of polarization. As for the gauge, the velocity gauge expressing the spatially-uniform electric field by the vector potential as a function of time is convenient [1]. It allows us to use periodic Bloch wave function at any time. As for the polarization, we need to specify macroscopic shape of the solid to define the electric field inside materials. We also consider recently a multiscale implementation for the coupled dynamics of macroscopic electromagnetic fields and microscopic Kohn-Sham orbitals [2].

We have applied the formalism to dielectric function [1], coherent phonon generation [3], dense electron-hole pair generation and optical breakdown [4], and propagations, reflections, and transmissions of intense laser pulses at solid surfaces [2].

In the figures below, we show calculated results of a propagation of linearly polarized intense laser pulse irradiating normally on bulk Si. The mean frequency of the pulse is 1.55 eV, and is smaller than the direct bandgap of Si (2.4 eV in the calculation). The maximum intensity of the pulse is set to 10^{11}W/cm^2. In the left three panels of Fig 1, we show a propagation of the macroscopic vector potential. The incident pulse (top) reaches the surface of bulk Si (middle) and splits into reflected and transmitted pulses (bottom). In the right three panels of Fig. 1, we show electronic excitation energy per atom at each time. The bottom panel shows that electrons at the surface are strongly excited by the two-photon absorption process.

We also achieve a pump-probe calculation. In Fig. 2, we show reflectivity of the probe pulse by filled circles as a function of the intensity of the pump pulse. As seen in the figure, the reflectivity once decreases and then increases almost up to unity. This is caused by a formation of the dense electron-hole pairs at the Si surface.

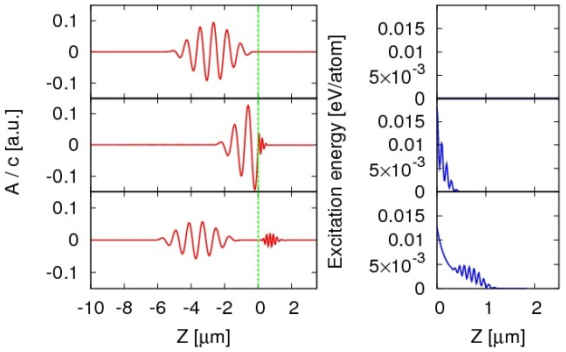

Fig. 1: (Left) Laser pulse propagation calculated by Maxwell + TDDFT multiscale calcualtion. (Right) Electronic excitation energy per atom. Bulk Si is placed at Z>0 and a vacuum at Z<0.

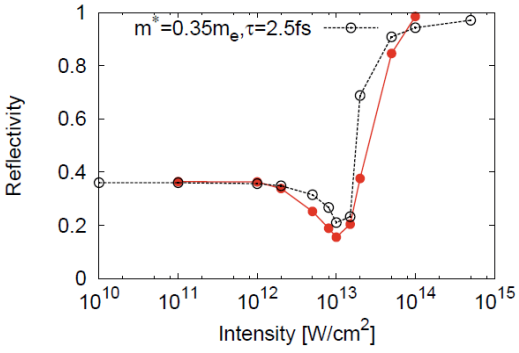

Fig. 2: Reflectivity of probe pulse in the pump-probe calculation, as a function of pump pulse intensity is shown by filled circles. Fit by Drude model is shown by open circles..

References

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