Ab-initio theory for propagation of strong optical pulse in solids

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When we theoretically investigate interaction of an intense and ultrashort laser pulse with solids, there are two aspects that should be considered: the strong electric field of the light pulse induces extremely nonlinear electron dynamics in solids that cannot be described by perturbation theory. The nonlinear electron dynamics manifests as a nonlinear polarization in macroscopic scale that determines the propagation of the light pulse. Therefore, to describe the propagation of the intense light pulse, it is necessary to treat the coupled dynamics of the light electromagnetic fields and the electron dynamics. The scheme should also take into account the separation of the spatial scale of two dynamics. We have developed a multiscale framework based on ab-initio time-dependent density functional theory [1], and have applied the method to various phenomena related to strong optical pulses in solids.

When an intense light pulse irradiates on transparent materials, nonlinear electronic excitations such as multiphoton absorption and tunneling ionization take place. Above a certain critical intensity, the material is damaged permanently. We have applied our multiscale description for the electronic excitations in alpha-quartz. Comparison with measurements using attosecond streaking method have revealed that the onset of the energy transfer can be described reasonably [2]. The result indicates that our method can provide a spatial distribution of energy deposition after the intense and ultrashort light pulse passes through the transparent materials.

We further consider a coupling with the lattice dynamics using Ehrenfest molecular dynamics [3]. Solving the Maxwell, the time-dependent Kohn-Sham, and the Newton equations simultaneously, light propagation together with microscopic dynamics of electrons and ions are described. As a demonstration, we show a simulation of a pump-probe measurement of coherent phonon in diamond. After the pump pulse generates the coherent optical phonon, a generation and amplification of the impulsively stimulated Raman wave is described in the probe stage.

When we consider interaction of a pulsed light with an extremely thin films such as mono-atomic layer materials, the multiscale description is no more useful. For such situations, we have invented a description solving the Maxwell and the time-dependent Kohn-Sham equations in a common spatial grid. This scheme provides a comprehensive description, applicable to any thickness of films from atomic monolayer to bulk surface and to field weak and strong intensities, although it comes computationally expensive as the thickness increases.

We are further considering an extension of the method for interactions of an intense light pulse with meta-surfaces, 2D array of nano-particles. When the radius of nano-particles is comparable to light wavelength, there occurs complex interplay between the light propagation and the electron dynamics. Furthermore, when the distance between nano-particles becomes less an a few tens of nm, quantum tunneling affects the optical response. I would like to mention the progress for such directions.

[1] K. Yabana et.al, Phys. Rev. B85, 045134 (2012).
[2] A. Sommer et.al, Nature 534, 86 (2016).
[3] A. Yamada et.al, arXiv:1810.06168.
[4] S. Yamada et.al, Phys. Rev. B98, 245147 (2018).