7th Asia-Pacific Conference on Plasma Physics, 12-17 Nov, 2023 at Port Messe Nagoya

First-principles calculations for ultrafast and nonlinear dynamics of light pulses and electrons

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Ultrashort and high-intensity laser pulses yield novel ultrafast and nonlinear phenomena of electrons in condensed matter. These phenomena are crucial for understanding the generation of warm dense matter (WDM). To describe such phenomena, we are developing combined computational methods of the Maxwell equations for light propagation and the time-dependent density functional theory (TDDFT) for electron dynamics [1,2]. In this talk, we introduce application examples of the combined methods and recent studies such as high-order harmonic generation in semiconductors [3,4]. Also, we introduce a calculation example of the laser intensity dependence of the reflection and transmission coefficients in laser irradiated Si films. This calculation considers the transition from the multi-photon absorption regime to the classical plasma regime. These computational methods are implemented in our free-to-use open-source software SALMON [5].

Figure 1(a) shows schematic illustration of our method. We consider an irradiation of a free-standing Si thin film (gray area) in a vacuum by an ultrashort light pulse of a linearly polarized plane wave at the normal incidence. The light propagation is described by the one-dimensional wave equation on a real-space grid. For each grid point in the material, a TDDFT system for electrons in a periodic unit cell of bulk Si is attached. In Fig. 1(a), the electron density changes driven by the light pulse are illustrated for the first three grid points. The time evolution of the electronic wavefunctions and light field is solved simultaneously by exchanging the electron current and vector potential in each time step. We call this method "multiscale Maxwell-TDDFT."

Figure 1(b) and (c) show snapshots of the electric field for the incident and scattered light pulses, respectively, as a typical calculation example. The Si film has the thickness of 3 μm. In Fig. 1(b), the initial pulse (t = 0 fs) that locates in front of the film is shown where the film is exhibited as a thin gray area. In Fig. 1(c), we display results corresponding to initial pulses of two different maximum intensities: a strong pulse ($I = 5 \times 10^{12}$ W/cm^2, red solid line) and a weak pulse ($I = 10^9$ W/cm^2, blue dotted line). For the weak pulse, linear propagation is expected. In the figure, the weak pulse is multiplied by a factor of $\sqrt{5000}$ so that the differences of two lines manifest nonlinear effects in the stronger pulse. From these calculations, we can elucidate the mechanisms of nonlinear phenomena in laser-irradiated materials.

As shown by the above example, our computational method is a powerful tool for elucidating extreme phenomena in the latest optical science.

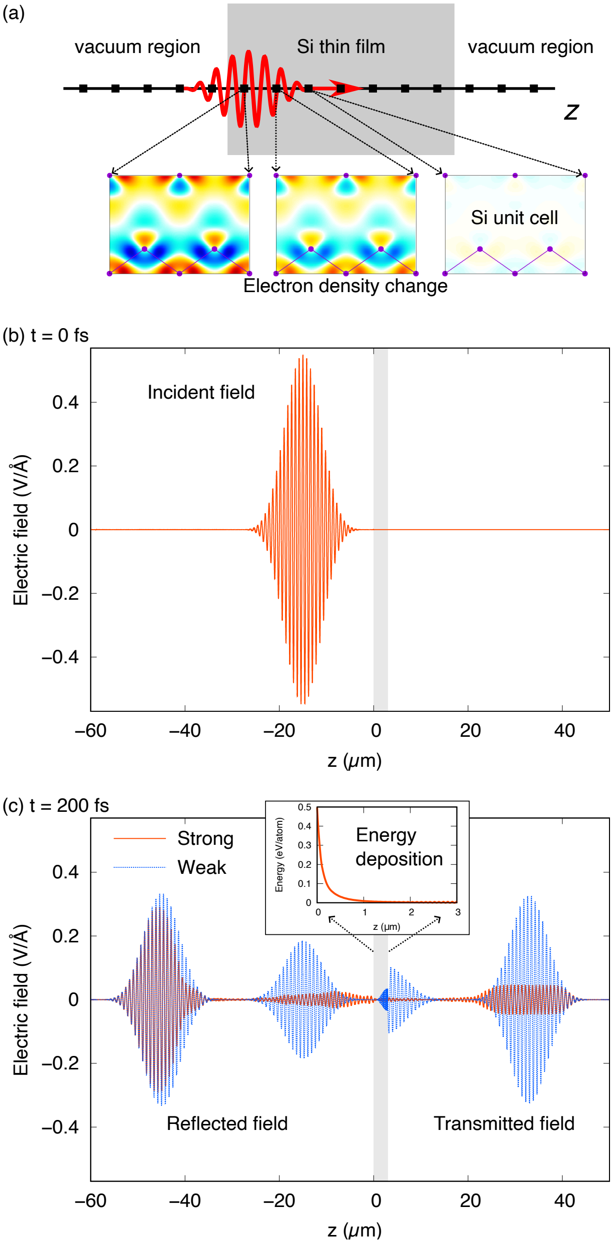

Fig. 1: (a) Overview of the multiscale Maxwell-TDDFT method for a light propagation through a Si thin film. (b) Electric field at t = 0. The incident pulse is prepared in front of the Si thin film, which is exhibited as a gray area. (c) Electric field at t = 150 fs. In the inset, the energy deposition is shown.

References
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