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First-principles method for propagation of ultrashort pulsed light in thin films

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Abstract. We develop a first-principles method to simulate the propagation of intense and ultrashort pulsed light in crystalline thin films solving the Maxwell equations for light electromagnetic fields and the time-dependent Kohn-Sham equation for electrons simultaneously using common spatial and temporal grids. As a demonstration, we apply the method to silicon thin films.

1 Introduction

In current frontiers of optical science, we often encounter cases in which macroscopic electromagnetism is not sufficient and it is required to go back to quantum descriptions of electron dynamics. We develop a first-principles computational method for the propagation of intense and ultrashort laser pulses through thin films based on the time-dependent density functional theory (TDDFT). In the method, we solve the Maxwell equations for light electromagnetic fields and the time-dependent Kohn-Sham (TD-KS) equation for electron dynamics simultaneously, in real space and real time. We expect the method will be useful to explore interactions of pulsed light with thin films, 2D-materials, and surfaces of metals.

Before this work, we have developed a multiscale theory using two kinds of spatial grids for macroscopic electromagnetic fields and microscopic electron dynamics [1]. The method here is different from the previous one in that the electromagnetic fields and electron dynamics are solved in the same spatial scale, using a common spatial grid. This is important for systems of thickness much less than the light wavelength and/or for systems with strong absorption.

2 Formalism

We consider an irradiation of a pulsed light normally on a crystalline thin film. We utilize electron Bloch orbitals $u_{\boldsymbol{k}n}(\boldsymbol{r},t)$ and scalar and vector potentials for the electromagnetic fields, $\phi(\boldsymbol{r},t)$ and $\boldsymbol{A}(\boldsymbol{r},t)$, respectively, We treat the whole system as a periodic problem in two-dimensions: these orbitals and fields are periodic in two-dimension, and $\boldsymbol{k}$ represents two-dimensional crystalline momentum.

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The Bloch wave function $u_{kn}(\boldsymbol{r},t)$ satisfies the following TD-KS equation:

$$i\frac{\partial u_{kn}(\boldsymbol{r},t)}{\partial t} = \left\{\frac{1}{2}\left[\frac{1}{i}\nabla + \boldsymbol{k} + \boldsymbol{A}(\boldsymbol{r},t)\right]^2 - \phi(\boldsymbol{r},t) + V_{\mathrm{ion}}(\boldsymbol{r}) + V_{\mathrm{xc}}(\boldsymbol{r},t)\right\} u_{kn}(\boldsymbol{r},t). \quad (1)$$

Maxwell equations in the Coulomb gauge are written as

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2\right)\boldsymbol{A}(\boldsymbol{r},t) + \frac{1}{c}\nabla\dot{\phi}(\boldsymbol{r},t) = -\frac{4\pi}{c}\boldsymbol{j}_e(\boldsymbol{r},t), \quad (2)$$

$$\nabla^2\phi(\boldsymbol{r},t) = 4\pi n_e(\boldsymbol{r},t), \quad (3)$$

where $\boldsymbol{j}_e$ and n_e are the electron current and the electron density, respectively. They are calculated from the Bloch orbitals $u_{kn}(\boldsymbol{r},t)$. We solve these equations simultaneously in real time and real space, employing a three-dimensional uniform grid to express physical quantities. In practical calculations, we only treat valence electrons employing norm-conserving pseudopotentials for electron-ion interactions.

The apparent advantage of the method is that it does not assume a coarse graining of the microscopic scale, in other words, the spatial scale of electromagnetic fields is treated on the same footing as the scale of electron dynamics. Therefore, the photo-electron interaction in very thin films or atomic layers can be described by our method. It should also be noted that it is possible to describe extremely nonlinear interaction may without any perturbation expansions, since we solve quantum electron dynamics without any approximation.

3 Results

As an example, we describe an irradiation of a high-intensity ultrashort pulsed light on a 5.43 nm silicon thin film of 20 layers. In the left panel of Fig. 1, the thin film is placed at Z = 0, and a right-going incident wave is prepared on the left side of the film. The photon energy of the incident pulse is set to 3 eV, the pulse width (FWHM) is about 5 fs, and the intensity is 5×10^{12} W/cm^2. This intensity is close to the threshold of silicon permanent damage [2]. The right panel of Fig. 1 shows the electric field at the time after the pulsed light passes through the thin film and is composed of the reflected and transmitted waves. For comparison, the electric field corresponding to a weak incident pulse is also indicated by a blue dashed line. It shows the electric field at the intensity of 1×10^9 W/cm^2, multiplied by a factor 100. As is readily recognized, the reflected wave of high intensity pulse is significantly reduced. Also, the shape of the pulse changes greatly from the case of the weak intensity pulse. Regarding the transmitted wave, there is not so big change, although it is somewhat weakened and delayed.

Fig. 2 shows the electron density distribution. The upper panel shows the density in the ground state and the lower panel show the density change from that in the ground state during the penetration of the pulse. It can be seen that bonds between Si atoms are weakened and electrons spread in interatomic space.

4 Summary

We have developed a new simulation method that describes the interaction between ultrashort pulsed light and thin films based on first-principles time-dependent density functional theory. The method will be useful for various phenomena in which the medium is so thin that macroscopic electromagnetism is no more useful and/or the field is so strong that the interaction is highly nonlinear. We expect to apply our method to ultrafast dynamics in 2D materials, nonthermal laser processing at metal surfaces, chemical reactions on material surfaces, and so on. We also plan to implement the method here to the open-source software SALMON [3,4] that is developed in our group.

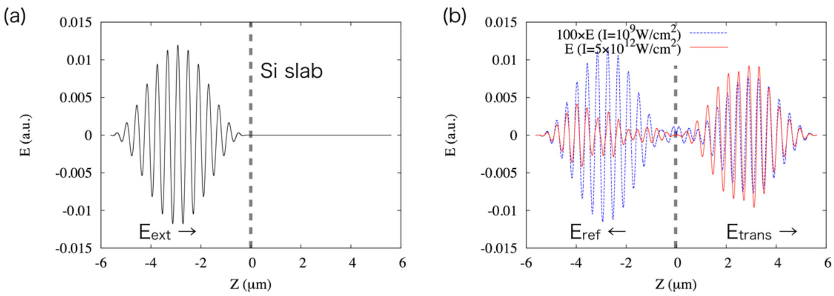

Fig. 1. Snapshots of the electric field averaged over the x-y plane (parallel to the surface) of the typical Si slab calculation. (a) Right-going incident field with the intensity I=5×10^{12} W/cm^2. (b) Reflected and transmitted fields with the intensity I=5×10^{12} W/cm^2 (red solid line). The I=1×10^9 W/cm^2 fields are scaled up by factors of 100 (blue dashed line).

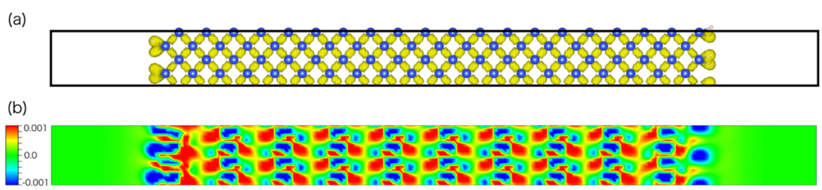

Fig. 2. (a) Structure and static electron density of the Si slab of Fig. 1. (b) Density difference with the static density at T= 8 fs by slicing in the [1,-1,0] plane.

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