

A first-principles simulation method for ultrafast nano-optics

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Abstract. We develop a computational approach for ultrafast nano-optics based on first-principles time-dependent density functional theory. Solving Maxwell equations for light propagation and time-dependent Kohn-Sham equation for electron dynamics simultaneously, intense and ultrashort laser pulse interaction with a dielectric nano-structure is described taking full account of nonlinear effects. As an illustrative example, irradiation of a pulsed light on silicon nano-sphere system is presented.

1 Introduction

Computational approaches solving Maxwell equations have been recognized as basic and indispensable tools in wide areas of optical science. In current frontiers of ultrafast optical science, however, we often encounter phenomena that require descriptions going back to quantum electron dynamics that takes place in microscopic spatial scale.

We develop a theoretical and computational method of electron dynamics based on time-dependent density functional theory (TDDFT). We invented a computational method to solve the time-dependent Kohn-Sham (TDKS) equation, a basic equation of the TDDFT, in real time and real space [1]. We further combine microscopic electron dynamics calculations with macroscopic Maxwell equations that describe light-wave propagation using a multiscale strategy [2]. We applied the method to ultrafast electron dynamics in dielectrics induced by few-cycle femtosecond laser pulses [3,4]. Our computer code is distributed a free, open source software SALMON (Scalable Ab-initio Light-Matter simulator for Optics and Nanoscience) [5,6].

In this paper, we describe our recent attempt to extend our approach to ultrafast three-dimensional (3D) nano-optics, solving 3D-Maxwell and 3D-TDKS equations simultaneously. We first describe the formalism and then present, as an illustrative example, a calculation of an intense and ultrashort laser pulse interaction with a layered material composed of silicon nano-spheres.

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2 Formalism

To make use of different spatial scales between the laser pulse and the electron dynamics, we develop a multiscale formalism [2]. We utilize two coordinate systems, \mathbf{R} and \mathbf{r} , as shown in Fig. 1. In the macroscopic scale, the light-wave is described using a vector potential $\mathbf{A}_R(t)$ that satisfies the Maxwell equation:

$$-\nabla_{\mathbf{R}} \times \nabla_{\mathbf{R}} \times \mathbf{A}_R(t) + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{A}_R(t) = \frac{1}{4\pi} \mathbf{J}_R(t), \quad (1)$$

where $\mathbf{J}_R(t)$ is the electric current density. At each grid point \mathbf{R} , we describe electron dynamics in dipole approximation using Bloch orbitals $u_{Rkb}(\mathbf{r}, t)$ orbitals that satisfy the following TDKS equation:

$$i\hbar \frac{\partial}{\partial t} u_{Rkb}(\mathbf{r}, t) = \left\{ \frac{1}{2m} \left(\mathbf{p} + \hbar \mathbf{k} + \frac{e}{c} \mathbf{A}_R(t) \right)^2 + V_{R,\text{ion}}(\mathbf{r}) + V_{R,\text{Hxc}}(\mathbf{r}, t) \right\} u_{Rkb}(\mathbf{r}, t), \quad (2)$$

where \mathbf{k} is the crystalline wave number, b is the band index, and $V_{R,\text{Hxc}}(\mathbf{r}, t)$ is a sum of Hartree and exchange-correlation potentials. The electric current density $\mathbf{J}_R(t)$ that appears in Eq. (1) is obtained from the Bloch orbitals $u_{Rkb}(\mathbf{r}, t)$. Eqs. (1) and (2) are solved simultaneously to evolve the coupled dynamics of electrons and electromagnetic fields. We typically use 10 nm for macroscopic, 0.4 Å for microscopic grid spacing, and 1 as for time step. We note that nonlinear interactions are fully incorporated in the calculation since we solve the optically-induced electron dynamics without any perturbative approximations.

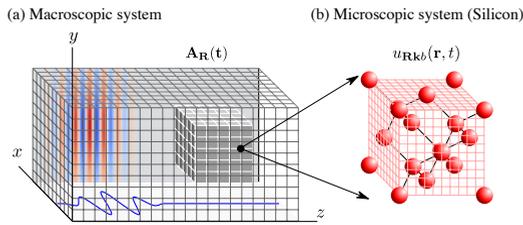


Fig. 1. Schematic illustration of 3D Maxwell + 3D TDDFT multiscale simulation. (a) Macroscopic grid system to solve Maxwell equation. (b) Microscopic grid system to calculate electron dynamics.

3 Application to Silicon Nano-sphere

As an illustrative application, we show an irradiation of an intense and ultrashort laser pulse on silicon nano-spheres. As illustrated in Fig. 2(a), silicon nano-spheres of about $r=250$ nm radius are periodically placed in yz -plane. A linearly polarized laser pulse irradiates normally on them. The average frequency and the pulse duration are set as $\omega=1.55\text{eV}/\hbar$ and 7 fs, respectively. Two cases of different maximum intensities, strong ($I = 10^{12}\text{W}/\text{cm}^2$) and weak ($I = 10^{10}\text{W}/\text{cm}^2$) cases, are considered.

In the calculation, the silicon nano-sphere is expressed by 32,752 macroscopic grid points. We use more than 8,000 nodes to carry out a large number of electron dynamics calculations simultaneously, utilizing a massively parallel supercomputer, Oakforest-PACS at JCAHPC. Computational aspects of our calculations are described in [7,8].

We show snapshots of the calculated electric field around the silicon nano-sphere in Fig. 2 (b), and of the electronic excitation energy inside the nano-sphere in Fig. 2(c). In Fig. 2(b), a generation of whispering-gallery wave as well as a focus is observed inside the sphere.

The electromagnetic fields look weakened for stronger pulsed light. This is because the lifetime of the confined light modes becomes short by the nonlinear absorption of the strong light. In Fig. 2(c), intense electronic excitations are seen inside the sphere. The intensity of the excitation scales approximately linear to the field intensity, indicating that this is not due to the two-photon absorption across the silicon bandgap. Since the figure is drawn at a time when the whispering-gallery wave exists inside the nano-sphere, the electronic excitation energy is mainly associated with the polarization motion of electrons. In the case of stronger pulsed light, the electronic excitations are mixture of two-photon absorption and polarization motion. A long-time simulation is required to separate two excitation mechanisms.

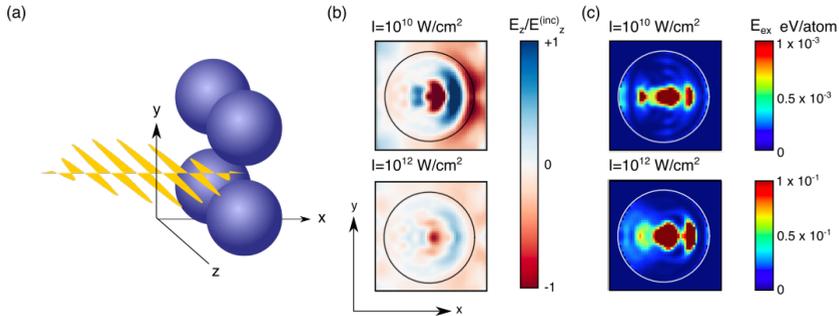


Fig. 2. (a) Schematic illustration of the considered system; a linearly polarized pulsed light irradiate normally on silicon nanospheres that are periodically placed in yz -plane. (b) Electric field E_z and (c) electron excitation energy E_{ex} , at $t=18.4$ fs for two incident intensities ($I = 10^{10} \text{ W/cm}^2$ and $I = 10^{12} \text{ W/cm}^2$).

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