Physics Letters A 376 (2012) 3327–3331

Contents lists available at SciVerse ScienceDirect

Physics Letters A

www.elsevier.com/locate/pla

Transient localized electron dynamics simulation during femtosecond laser tunnel ionization of diamond

C. Wang [a], L. Jiang [a,*], F. Wang [a], X. Li [a], Y.P. Yuan [a], L.T. Qu [b], Y.F. Lu [c]

[a] Laser Micro/Nano Fabrication Laboratory, School of Mechanical Engineering, Beijing Institute of Technology, Beijing 100081, China
[b] Key Laboratory of Cluster Science, Ministry of Education, School of Chemistry, Beijing Institute of Technology, Beijing 100081, China
[c] Department of Electrical Engineering, University of Nebraska-Lincoln, Lincoln, NE 68588-0511, USA

ARTICLE INFO

Article history:
Received 8 May 2012
Received in revised form 2 July 2012
Accepted 6 July 2012
Available online 27 August 2012
Communicated by R. Wu

Keywords:
Time-dependent density-functional theory
Tunnel ionization
Electron dynamics

ABSTRACT

A real-time and real-space time-dependent density-functional theory (TDDFT) is applied to simulate the nonlinear electron–photon interactions during femtosecond laser processing of diamond when tunnel ionization dominates. The transient localized electron dynamics including the electron excitation, energy absorption and electron density evolution are described in this Letter. In addition, the relationships among average absorbed energy, Keldysh parameter and laser intensity are revealed when the laser frequency is fixed.

1. Introduction

Compared with a long pulse, an ultrashort pulse laser in some aspects fundamentally changes the laser–material interaction mechanisms, which introduces new phenomena, requiring new theoretical and experimental understanding. Among the various photon absorption mechanisms induced by ultrashort laser field, it is critical to understand ionization mechanisms. Ionization is an interesting physical process showing a crossover of the mechanisms including electron impact ionization, avalanche ionization [1], multiphoton ionization [2] and tunnel ionization [3]. Avalanche ionization strongly depends on free electron density and is sometimes assumed as linearly proportional to laser intensity [4]. Its efficiency is determined by competitions between energy gain through inverse Bremsstrahlung and energy loss through phonon emission [5]. Avalanche ionization is responsible for the ablation of wide bandgap materials at the laser intensities below 10^{12} W/cm^2 [4,5]. At high intensities (typically $> 10^{13}$ W/cm^2), multiphoton ionization becomes significantly strong [6]. However, when intensities are higher than 10^{15} W/cm^2, tunnel ionization becomes significant [7]. The key characteristic parameter of ionization mechanisms is the Keldysh parameter $\gamma = \omega\sqrt{2I_P}/F$, where ω is the laser frequency, I_P is the ionization potential and F is the field strength [8]. The multiphoton ionization is expected to dominate when $\gamma \gg 1$, while tunnel ionization dominates when $\gamma \ll 1$.

As a fundamental quantum phenomenon that challenges classical intuition, tunneling has been a testing ground for the understanding of quantum mechanics [9]. Since it influences the electronic structure of atoms, molecules and solids and is responsible for radioactive decay, intensive research interests have been attracted to investigate the tunneling experimentally and theoretically [10–19]. For example, based on the quasistatic approximation, the Ammosov–Delone–Krainov (ADK) model [14] has been applied for tunnel ionization of atoms with success. The Keldysh–Faisal–Reiss model [8,15,16] is an alternative approach which takes into account the time dependence of the external field. The PPT theory (Perelomov, Popov and Terent'ev) [17] and the S-matrix theory developed by Faisal and Becker [18,19] excellently fit experimental results. Besides the aforementioned analytical approaches, various numerical approaches have been rapidly developed to describe the electron dynamics under the intense ultrashort laser irradiation [20–29], among which the time-dependent density-functional theory (TDDFT) [30] is the most feasible theoretical framework to treat the electrons quantum mechanically. TDDFT combined with the linear response theory has been successfully applied to the optical responses of atoms, molecules, and solids [31–34]. It has also been used to describe the nonlinear and nonperturbative electron dynamics induced by intense ultrashort laser pulses [35–39].

For energy transfer in bulk materials, there is a good fit of $\delta E \simeq CI^N$ between average absorbed energy δE and the laser intensity I when multiphoton ionization dominates, where N is the number of photons absorbed to free from the valence band [38]. In

* Corresponding author.
E-mail address: jianglan@bit.edu.cn (L. Jiang).

0375-9601/$ – see front matter
http://dx.doi.org/10.1016/j.physleta.2012.07.039

the intermediate case between multiphoton ionization and tunnel ionization, there is almost a linear relationship between average absorbed energy and laser frequency at the same laser intensity [39]. But for tunnel ionization, less research has been reported to describe the electron dynamics especially the energy transfer in bulk materials irradiated by intense ultrashort pulses, which needs further investigations. In this Letter, a real-time and real-space time-dependent density-functional theory is employed to describe the transient localized electron dynamics including the photon energy absorption, generation of electrons and energy transfer of diamond under femtosecond laser irradiation in the high laser intensity regime when tunnel ionization dominates. Fundamental understanding of ionization process and electron dynamics is critical to control and improve micro/nanoscale processing of materials.

2. Theory

The time-dependent density-functional theory is a reformulation of time-dependent quantum mechanics, where the fundamental variable is no longer the many-body wave function but the density, which gives the advantage in computational speed, allowing study of larger systems compared with traditional wave-function methods. This time-dependent density is determined by solving an auxiliary set of non-interacting Schröinger equations, the Kohn–Sham equations [30]. To describe the motion of electrons, the time-dependent Kohn–Sham (TDKS) equation for single particle orbitals $\psi_i(\vec{r},t)$ is applied as follows,

$$i\hbar\frac{\partial}{\partial t}\psi(\vec{r},t) = H_{KS}(\vec{r},t)\psi(\vec{r},t) \tag{1}$$

$$n(\vec{r},t) = \sum_i \left|\psi_i(\vec{r},t)\right|^2 \tag{2}$$

where $n(\vec{r},t)$ is the electron density, $H_{KS}(\vec{r},t)$ is the Kohn–Sham Hamiltonian which is conventional separated in the following way,

$$H_{KS}(\vec{r},t) = \frac{1}{2m}\left(\vec{p} + \frac{e}{c}\vec{A}_{tot}(t)\right)^2 + V_{ion}(\vec{r},t) + e^2\int dr'\,\frac{n(\vec{r},t)}{|\vec{r}-\vec{r}'|} + V_{xc}(\vec{r},t) \tag{3}$$

where e is an elementary charge, $V_{ion}(\vec{r},t)$ is the electron–ion potential, $V_{xc}(\vec{r},t)$ is the exchange-correlation potential and $\vec{A}_{tot}(t)$ is a time-dependent spatially-uniform vector potential which is composed of the external and induced vector potentials, $\vec{A}_{tot}(t) = \vec{A}_{ext}(t) + \vec{A}_{ind}(t)$. The external vector potential $\vec{A}_{ext}(t)$ is related to the electric field of the applied laser pulse, $\vec{E}_{laser}(t) = -d\vec{A}_{ext}(t)/dt$. The induced vector potential $\vec{A}_{ind}(t)$ expresses the electric field caused by the polarization, which can be described as follows,

$$\frac{d^2\vec{A}_{ind}(t)}{dt^2} = \frac{4\pi}{c}\vec{i}(t) \tag{4}$$

where $\vec{i}(t)$ is the average electric current density [40]. The time profile of the electric field from the laser pulse is parameterized by

$$E_{laser}(t) = E_0\sin^2\left(\frac{\pi t}{T}\right)\sin(\omega t) \quad (0 < t < T) \tag{5}$$

where E_0 is the magnitude of the peak electric field, ω is the laser frequency and T is the pulse duration. In practical calculations, the total electric field is proportional to the applied field, $E_{tot}(t) \approx \varepsilon^{-1}E_{ext}(t)$, where ε is the static dielectric constant of the material [40–42].

The choice of the propagator algorithm is crucial for a time-dependent method. This study employs the enforced time-reversal symmetry (ETRS) [43] method to efficiently describe the propagation of electron wave functions in real time. The time evolution of the wave function for a short period Δt is approximately calculated by

$$\psi(\vec{r},t+\Delta t) = e^{-i\hat{H}_{KS}(t+\Delta t)\frac{\Delta t}{2}}e^{-i\hat{H}_{KS}(t)\frac{\Delta t}{2}}\psi(\vec{r},t). \tag{6}$$

The number of excited electrons per unit cell $n_{ex}(t)$ is defined as [40],

$$n_{ex}(t) = \sum_{nn'k}\left(\delta_{nn'} - \left|\langle\phi_{nk}|\psi_{n'k}(t)\rangle\right|^2\right) \tag{7}$$

where k is the Bloch wave number and n is the band index.

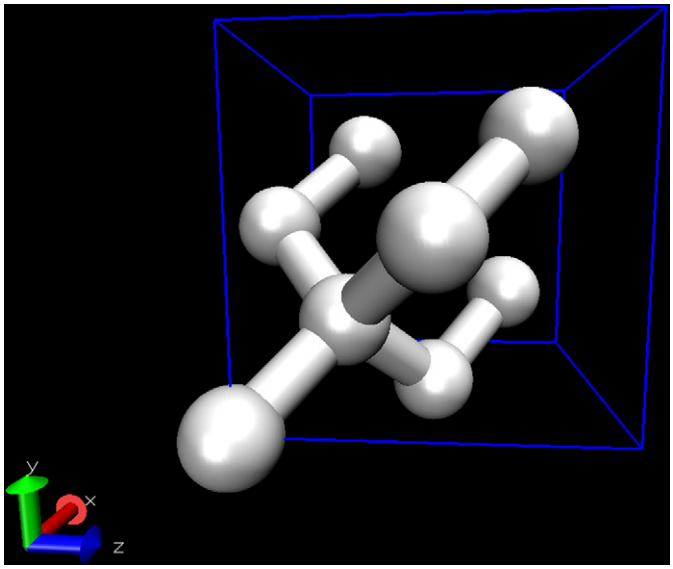

Fig. 1. Geometry of the atoms in the eight-atom unit cell.

3. Results and discussion

The OCTOPUS code [44] is used to carry through our calculations. Here we summarize the essential aspects of our model setting. A face-centered cubic unit cell containing eight carbon atoms is considered to calculate femtosecond laser processing of diamond, as is shown in Fig. 1. The wave functions, densities and potentials are discretized in a real-space grid. Lattice constant is 6.74 a.u., which is discretized into 18 grid points in each direction. The irreducible wedge of the Brillouin zone is represented by 165 k points, which corresponds to sampling the full Brillouin zone by 16^3 k points to represent the Bloch wave functions for the momentum-space integration. Perdew–Zunger's exchange-correlation functional [45] and corresponding Troullier–Martins' pseudopotential [46] are used for electron–nucleus interactions. We represent the laser irradiation by subjecting our system to an external alternating electric field parallel to the z axis. The time step of $\Delta t = 0.02$ a.u. is used. In addition, ion positions are frozen in all calculations.

Fig. 2 shows the electric field of the applied laser pulse, excited electrons and excitation energy as functions of time after the laser termination. The laser pulse is a 400 nm 20 fs Gaussian wave packet. The laser peak intensity of the pulse is 2×10^{15} W/cm^2 and the Keldysh parameter is 0.352, which means tunnel ionization dominates in the case. After the laser termination, the number of excited electrons is 1.274 out of the total electrons in the unit cell; the total absorbed energy is 72.37 eV; and the average absorbed energy per excited electron is 56.81 eV.

Fig. 3 shows the electron density in real space in the $x = 0$ plane. The left panel shows the ground state electron density. The