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Monte Carlo simulation of electron dynamics in liquid water

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HIGHLIGHTS

- We use a direct Monte Carlo algorithm and give a detailed description (pseudocode).
- The energy loss of VUV laser-excited electrons is modeled by a pure jump process.
- The majority of the laser pulse energy is stored in holes of water molecules.
- The creation of secondary electrons occurs almost entirely during laser irradiation.
- We show the number and kinetic energy of primary and secondary electrons over time.

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ABSTRACT

We present a stochastic model for the energy loss of low-energy electrons (<100 eV) in water in the liquid phase. More precisely, we treat the electrons as independent particles and are thus able to model the time evolution of the kinetic energy of a single electron as a so-called pure jump process. Free electrons are created due to irradiation of an extreme ultraviolet femtosecond laser pulse. In our model, free electrons may interact with water molecules via elastic scattering and impact ionization. Moreover, we present numerical results for the kinetic energy of electrons during and after laser irradiation. Furthermore, we distinguish between primary and secondary electrons, where the latter are created by impact ionization. The numerical results show that creation of secondary electrons due to impact ionization occurs almost entirely during laser irradiation. After irradiation, only a small amount of the laser pulse energy remains in the electron system, while the majority is stored in holes of water molecules.

1. Introduction

As the major constituent of the biological cell, water plays a crucial role in understanding damage to biological tissue caused by radiation or charged particles [1,2]. More precisely, ionizing radiation leads to a vast amount of secondary electrons responsible for molecular damage due to energy deposition. Especially low-energy electrons play a major role in producing clustered DNA damage. The desire for a better understanding of these effects gives rise to the analysis of track structure caused by charged particles. For this task, a Monte Carlo (MC) algorithm is a suitable choice and various attempts have been made so far, see Refs. [3–5,1] in the case of liquid water or [6] for a general overview.

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Table 1
Binding energies of the water molecule.

Shell	I_i (eV)
$1b_1$	10.9
$3a_1$	13.5
$1b_2$	17.0
$2a_1$	32.2

The new free electron laser in Hamburg (FLASH) provides highly brilliant ultrashort laser pulses in the extreme ultraviolet range [7,8]. At this wavelength experiments are realized to study the photolysis of water-radicals [9] or the intermolecular reactions in and on thin ice layers [10].

We investigate the electron dynamics in liquid water irradiated by an ultrashort laser pulse with parameters possible by FLASH, e.g., a temporal intensity profile with a full width half maximum (FWHM) of 10 fs and a photon energy of $E_{\mathrm{ph}} = 100$ eV.

The electron dynamics is studied on a femtosecond time scale similar to Refs. [11,12]. In this paper electrons are treated as independent particles. Consequently, we model the time evolution of the kinetic energy of a single electron as a so-called pure jump process [13,14]. This means that the kinetic energy of an electron remains constant up to isolated jumps, where a jump represents the energy loss during an interaction with a water molecule. This leads to a direct MC algorithm, where we simulate the time between two successive interactions and the subsequent energy loss step by step. Since we treat electrons as independent particles, the algorithm is particularly suitable for parallelization.

This paper is organized as follows. In Section 2 we describe the creation of free electrons due to photon absorption. In Section 3 we explain the electron dynamics in detail. We discuss the time between two successive interactions for which we take into account elastic scattering and impact ionization. The energy loss essentially depends on the corresponding differential cross section. In Section 4 we describe the underlying mathematical model. Moreover, we present a direct MC algorithm in Section 4.4. Finally, in Section 5 we provide numerical results for the electron energy at distinct times.

2. Photon absorption

As the initial physical process, we consider irradiation of a sample of liquid water by an ultrashort laser pulse produced by FLASH. Based on the possible pulse parameters we assume a temporal intensity profile of Gaussian shape with a FWHM of 10 fs and a photon energy E_{ph} of 100 eV.

During laser irradiation a water molecule absorbs a photon and sets free an electron. The instant of time of this event is distributed according to the temporal intensity profile of the laser pulse.

Furthermore, we take into account four molecular orbitals of the water molecule from which the free electron can be ejected. The corresponding binding energies of the three outermost shells are taken from photoelectron spectroscopy at liquid water surfaces [15]. For the fourth shell we apply the binding energy from the gas phase [16, Table 2]. The binding energies are summarized in Table 1.

A newly created free electron starts with the initial kinetic energy

$$E_i^{\mathrm{init}} = E_{\mathrm{ph}} - I_i,$$

which leads to four distinct initial energies $E_1^{\mathrm{init}} = 89.1$ eV, $E_2^{\mathrm{init}} = 86.5$ eV, $E_3^{\mathrm{init}} = 83$ eV, and $E_4^{\mathrm{init}} = 67.8$ eV. Finally, all initial energies are assumed to be equally likely.

3. Electron dynamics

After the photon absorption has created a free electron we investigate its dynamics through the sample of liquid water. The underlying model of the electron dynamics is based on a classical trajectory approach. Concerning the validity of such an approach we refer to Ref. [17] and the references therein. At first, we examine the time evolution determined by the time between two successive interactions in Section 3.1. Secondly, we study the two possible interactions, namely elastic scattering and impact ionization, in Section 3.2.

3.1. Time evolution

According to classical collision theory, see Ref. [18], the average time $\langle t \rangle$ between two successive interactions is given by

$$\langle t \rangle = \frac{\lambda}{v},$$

with the mean free path λ and the velocity v of the considered electron. The latter can be derived from its kinetic energy E according to the classical formula $E = m_{\mathrm{e}} v^2/2$ with the electron mass m_{e}. The mean free path of a single interaction can be written as $\lambda = 1/(n_{\mathrm{w}}\sigma)$, where σ is the cross section of the respective interaction and n_{w} is the density of water molecules

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