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Electron Kinetics in Femtosecond X-Ray Irradiated SiO₂

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A theoretical study of the ultrafast electron kinetics in solid SiO₂, irradiated with a femtosecond X-ray laser pulse, is presented. A Monte Carlo code for event-by-event simulations of individual particles is applied to model the electron kinetics within the irradiated SiO₂ bulk. The simulation includes photoionization, elastic and inelastic scatterings of electrons, Auger decays of core holes, and electron-hole recombination via exciton self-trapping mechanism. Transient electron density is followed, at different photon energies and pulse durations. The change of the optical properties (reflectance, transmittance) of the material is estimated with the help of the Drude model. The analysis of the results allows us to conclude that within the X-ray excited dielectric, the holes in the valence band give the predominant contribution to the transient changes of optical properties within the material. The increase rate of the free electron density is limited by the duration of secondary electron cascading.

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1 Introduction

When a dielectric is irradiated with an ultrashort X-ray laser pulse, various physical processes are initiated. First, photoabsorption occurs during the laser pulse, which, with modern free electron lasers, can be as short as a few femtoseconds [1–4]. The photoabsorption is dominantly exciting deep shell electrons, since the cross section for X-ray photon absorption by a deep shell is much larger than the one for the valence band electrons [5]. The electrons, excited into a high energy state, start to scatter on the target atoms: (i) elastically, by exchanging only the kinetic energy with atoms, and (ii) inelastically, by impact ionizations, exciting atomic electrons from the valence band or deep shells into the conduction band [6]. The following ‘cascading’ production of secondary electrons starts within a characteristic time defined by the electron inelastic scattering cross-section in a solid, typically, on a femtosecond timescale [6, 7]. Electronic subsystem might be still non-thermalized at this stage [7–9]. The cascading process stops when the electron energy decreases below the impact ionization threshold, E_{min} [10–12]. Depending on the initial electron energy, provided by the absorbed photon, this can take up to tens of femtoseconds.

The inverse process to the impact ionization, the three body recombination, strongly depends on the free electron density. Its rate is comparable to the impact ionization rate at high electron densities, and negligible at low electron densities [13]. Radiative recombination usually takes much longer times ($\sim$ ps) [14] and therefore can be neglected. In wide-band gap insulators, another recombination process is possible. This is the recombination via self-trapped excitons [15, 16], which assumes that an electron and a hole get trapped together and create a complex object, exciton, of the total energy less than the energy of the unbound electron and hole. This object then decays (recombines), releasing the excess energy via emission of a photon or phonons. Such a recombination typically takes place on sub-picosecond scales [16–18]. Consequently, at such short times the transient material properties are then defined by the transient state of the electron distribution within the solid.

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On longer time scales (pico- to nano-seconds) a significant amount of energy is transferred from electrons into the lattice via electron-phonon energy exchange [19–21]. This lattice heating can even damage the material (phase transitions, melting, ablation), if the laser fluence was high enough [21].

Recently developed X-ray free electron lasers (such as FLASH [1], LCLS [2], SACLA [3], European XFEL [4] etc.), have a pulse duration shorter or comparable with the characteristic times of the electron kinetics within the material. This allows to probe directly transient states of the material excitation on femtosecond scales, giving an access to investigation of fundamental properties of irradiated dielectrics up to the regime of the non-equilibrium electron-hole plasma [22–27]. This has stimulated fundamental: theoretical [6, 7, 9, 12, 26] and experimental [2, 25, 27–30] investigation of non-equilibrium states of matter. However, many theoretical models, proven useful and widely applied for picosecond and longer laser pulses (e.g. the Two Temperature Model [19–21]), are not applicable on femtosecond timescales [31–33]. Fully nonequilibrium methods are then required to describe the ultrafast electron-hole plasma excitation and kinetics [7, 8, 31, 34].

With the 'X-ray pump - visible light probe' experimental technique [22, 23], a target is excited with a femtosecond X-ray pulse and then probed with visible light. The reflection/transmission of the visible light gives an information on the transient state of the electronic subsystem of the material [35]. Therefore, this technique is ideal for studying the ultrafast processes occurring in excited dielectrics.

In the present contribution a Monte Carlo method of individual-particle event-by-event simulations [6, 7, 11, 12, 36] is applied to describe the femtosecond X-ray irradiation of solid SiO₂. The electron photoexcitation and the subsequent secondary electron cascades are modeled, following the excitation of individual particles to high energy states. That allows us to calculate the transient distribution function of excited electrons and holes, their densities and energy, and the corresponding optical properties. The optical properties can be followed by dedicated pump-probe experiments [22, 23].

Our model predicts the ultrafast change of the electron density that induces the change of optical properties of the material. The scattering of the probe pulse occurs mostly on the valence band holes, because the holes are much slower than the highly excited electrons. We also demonstrate that a decrease of the laser pulse duration below the characteristic duration time of the secondary electron cascading does not further influence the transient optical properties of the irradiated material. The material transmission is following the increase of the electron/hole density, which is dominantly stimulated by secondary ionizations, and not by direct photoabsorption. The drawn conclusions on the parameters optimization of a X-ray pump - visible light probe scheme can be useful for future experiments [22, 23].

2 Model

We apply a Monte Carlo method to trace individual particles with an event-by-event simulation scheme [6, 7, 11, 12, 36]. Accounting for the fact that the typical laser spot ($\sim 10\mu\text{m}$) is much larger than the simulation box which can be used in our model ($\sim 10\text{ nm}$), we apply periodic boundary conditions to our simulation box. In this case, all produced electrons are confined within the box, and the spatial propagation can be neglected. This is a fairly good assumption on a sub-picosecond timescale, before a significant electron diffusion takes place on micrometer scales [37]. The electron distribution function can then be reduced to a function of two variables only: energy and time.

The MC algorithm details can be found in Refs. [12, 36]. The simulation scheme is following. First, we estimate photoabsorption rate within the simulation box. The number of sampled photons is proportional to the temporal profile of the Gaussian pulse envelope. Considering the specific attenuation length of the X-ray photon [5], we sample the realized free path with the Poissonian distribution [38]:

$$l = -\langle \lambda_{\hbar\omega}(\hbar\omega) \rangle \ln(\gamma), \quad (1)$$

where l is the realized free path, $\langle \lambda_{\hbar\omega} \rangle$ is the mean free path of a photon with the energy $\hbar\omega$ [5], and γ is a random number, uniformly distributed in the interval $[0,1)$. If the realized path lays within the simulation box, we consider the photon to be absorbed. An atomic shell from which an electron is excited is then chosen, accounting for the photoabsorption probabilities by all shells for which the ionization potential is smaller than the photon energy. The selected electron then gets ionized with the initial energy $E_e = \hbar\omega - I_p$, where I_p is the corresponding ionization potential.

Created free electrons then scatter elastically or inelastically. In the first case, they only lose a minor part of their kinetic energy, transferring it into a kinetic energy of atoms/phonons [31]. We use the Mott cross-section [36, 39] for the description of the elastic scattering on atoms, while for the description of the phonon scattering we apply the scattering rate extracted from Ref. [40]. In case of inelastic scattering, the electron excites another electron by an impact ionization. To describe this process, we use the complex dielectric function formalism [41]. In this method, the total impact ionization cross-section, $\sigma_i(E_e) = \int_{E_{min}}^{E_e/2} d(\hbar\omega) (d\sigma_i(E_e, \hbar\omega)/d(\hbar\omega))$ is obtained from the differential cross section which is modeled in the following form [38, 41, 42]:

$$\frac{d\sigma_i(E_e, \hbar\omega_e)}{d\hbar\omega_e} = \frac{2e^2}{\pi n_e \hbar^2 v_e^2} \int_{q_-}^{q_+} \frac{dq}{q} \text{Im} \left(\frac{-1}{\epsilon(\omega_e, q)} \right), \quad (2)$$

where the integration limits are: $q_{\pm} = \sqrt{2m_e/\hbar^2} (\sqrt{E_e} \pm \sqrt{E_e - \hbar\omega_e})$ [38]. The cross-section depends on the transferred energy $\hbar\omega_e$, and is integrated over the transferred momentum, q . The charge, e denotes the electron charge; n_e is the density of valence band electrons; the constant, $\hbar$, is the Planck constant; v_e is the incident electron velocity corresponding to the energy E_e ; and m_e is the electron mass. The loss function is parameterized as in [38, 41, 43]:

$$\text{Im} \left(\frac{-1}{\epsilon(\omega_e, q)} \right) = \sum_j \frac{A_j \Gamma_j \hbar\omega_e}{\left(\hbar^2 \omega_e^2 - (E_{0j} + \hbar^2 q^2 / (2m_e))^2 \right)^2 + (\Gamma_j \hbar\omega_e)^2}. \quad (3)$$

The coefficients A_j , Γ_j and E_{0j} for SiO₂, presented in Table 1, are taken from Ref. [43], where they were adjusted to reproduce the experimentally known optical parameters.

Table 1 Coefficients of the effective complex dielectric function entering Eq.(3), as presented in Ref. [43].

Shell	j	E_{0j}	A_j	Γ_j
Valence band	1	10.6	0.6	0.45
	2	12.5	2	1.2
	3	15	3.5	1.8
	4	19	5	4
	5	22	100	9
	6	26	200	7
	7	26.1	5.5	1
	8	40	180	40
L-shell of Si	1	103	4.5	5
	2	115	6	15
	3	120	7	20
	4	125	15	30
	5	145	100	120
	6	165	18	50
	7	175	150	175
K-shell of O	1	500	150	350
K-shell of Si	1	1700	65	1050

The cross-sections for the valence band and the deep shell ionizations of SiO₂, with the coefficients obtained in [43], are presented in Fig. 1. They raise from zero at the energies corresponding to the ionization potentials of different shells: for the valence band the band gap of SiO₂ is $E_{gap} = 9$ eV, for the L-shell of Si the ionization potential is $I_p(L, Si) = 103.7$ eV, for the K-shell of O it is $I_p(K, O) = 538.25$ eV, and for the K-shell of Si it is $I_p(K, Si) = 1844.1$ eV [44]. The energy transfer in a collision is calculated with the differential cross-section (2).

The MC method in its standard classical version assumes that the individual scattering events of any particle are uncorrelated with scattering events of other particles within the system. However, following the recently introduced combined Monte Carlo - kinetic equation approach [11, 12], we can include some correlation effects through Pauli exclusion principle. This affects the probability of electron scattering which now depends on the

occupation of electronic states within the system. The corresponding mean free path, calculated from the total inelastic scattering cross-section, accounts for the Pauli exclusion principle in the following way [36]:

$$\langle \lambda_e \rangle = 1 / (F \sigma_i(E_e) n_e), \quad (4)$$

where the factor $F = (1 - f_e(E_i))(1 - f_e(E_j))$ calculates the number of vacancies within the energy interval E_j , where the primary electron is incoming to, and multiplies it by the number of vacancies within the energy interval, E_i , to which the secondary electron is excited; $f_e(E)$ is the electron distribution function, e.g. Fermi function in case of thermal equilibrium. At each time-step of the simulation we trace the distribution function of the electrons in the conduction band and of the holes in the valence band, directly checking the occupation of the energy intervals with respect to the density of states (DOS, $g(E)$) of the material [45].

Fig. 1 Impact ionization cross-section of different atomic shells within solid SiO₂, obtained with Eqs. (2) and (3).

The energy level from which an electron is being excited, is chosen according to the probabilities of ionization from different shells, including all shells with ionization energy smaller than the energy transferred in this collision, $\hbar\omega_e$. During the impact ionization of the valence band, the energy state, from which an electron is being excited, is chosen according to a probability proportional to the current occupation of the energy states within the valence band [45].

If an electron is excited from the level, E_i , its energy after photoexcitation is then: $E_e = \hbar\omega_e - E_i$. The following propagation of this electron and its elastic and inelastic scattering are traced, as well as the propagation of all secondary electrons produced by impact ionization, succeeding the impact of the primary electron. The secondary electron cascading stops when the energy of all electrons decreases below the limiting energy, E_{min} [10] which is the impact ionization threshold. The specific value of this energy depends on the band structure of the material. For SiO₂, $E_{min} = 13.5$ eV [12].

Deep shell holes produced by the photoionization or by secondary electron impact ionization relax via Auger decays. We describe such a relaxation with Poissonian law, using the characteristic Auger-decay times taken from Ref. [14]. This way, all deep holes pop up into the valence band within ~ 10 fs.

The most efficient mechanism of conduction electron - valence hole recombination in wide band gap dielectrics is the exciton self-trapping [17, 18]. The characteristic time of this recombination is ~ 150 fs in SiO₂ [17]. We model this process, using the Poissonian law for the time of the self-trapping. This leads to a fast reduction of the free electron density (see the Results section).

The distribution functions of free electrons, $f_e(E_e)$, and of valence holes, $f_h(E_h)$, are updated at each time step and can be used to obtain the averaged quantities:

$$n_e = \int_0^\infty f_e(E_e)g(E_e)dE_e, \quad n_h = \int_{E_{VB}}^{E_{gap}} f_h(E_h)g(E_h)dE_h, \\ \langle E_e \rangle = \int_0^\infty E_e f_e(E_e)g(E_e)dE_e, \quad \langle E_h \rangle = \int_{E_{VB}}^{E_{gap}} E_h f_h(E_h)g(E_h)dE_h. \quad (5)$$

Here, $n_{e,h}$ are the densities of the conduction band electrons and of the valence band holes; $\langle E_{e,h} \rangle$ are the mean free electron and the mean valence hole energies, respectively. The integration over the energy, $E_{e,h}$, includes

either the whole conduction band $(0; \infty)$, or the valence band from its bottom, E_{VB} , defined by the density of states of SiO_2 [45], up to the uppermost valence state which is separated by the bandgap, E_{gap} , from the conduction band.

To model the change of optical properties probed by the pulse of visible light, we calculate the transient dielectric function with help of the Drude model [35, 46]:

$$\epsilon = \epsilon_0 - \frac{\omega_{p,e}}{\omega_L} \frac{1}{1 + i/(\tau_e \omega_L)} - \frac{\omega_{p,h}}{\omega_L} \frac{1}{1 + i/(\tau_h \omega_L)}, \quad (6)$$

where ω_L is the probe pulse frequency; $\omega_{p,e,h} = \sqrt{n_{e,h} e^2 / m_{e,h}}$ are the conduction electron or valence hole plasma frequencies, respectively, with the effective electron mass, $m_e = 0.26 m_0$ and hole mass, $m_h = 0.33 m_0$ [47, 48] in the free electron approximation in SiO_2 ; m_0 is the electron rest mass; times $\tau_{e,h}$ are the mean scattering times of free electrons or valence holes.

The plasma frequencies are obtained at each timestep with the electron and hole densities calculated with Eqs. (5). The mean scattering times are calculated with the averaged scattering times, according to the Matthiessen's rule [49]:

$$\begin{aligned} 1/\tau_e &= 1/\tau_{ph} + 1/\tau_{el} + 1/\tau_{imp}, \\ \tau_\alpha &= \int_0^\infty f_e(E_e) dE_e / (n_e \sigma_\alpha(E_e) v_e), \end{aligned} \quad (7)$$

where $\tau_\alpha = \tau_{ph,el,imp}$ is either the phonon scattering time, τ_{ph} , or elastic scattering time, τ_{el} , or the impact ionization scattering time, τ_{imp} , which are averaged over the electron energy, E_e , with the corresponding distribution function $f_e(E_e)$; $\sigma_\alpha(E_e)$ is the cross-section of the corresponding scattering process; v_e is the electron velocity, corresponding to the energy E_e . For valence holes, we assume the same scattering cross-sections as for the free electrons. The difference in the dynamics is introduced by the different effective masses of electrons and holes. Thus, the knowledge of the electron and hole transient distribution functions allows us to obtain all necessary parameters for the dielectric function, and, in turn, the transmission coefficients.

3 Results and discussions

We considered the irradiation of SiO_2 with X-ray photons of energy of 7.1 keV, as in the recently performed experiments at LCLS [22]. A Gaussian X-ray pulse of the 30 fs duration at full width of half maximum (FWHM) and of the fluence of 0.1 mJ was shining on a spot of 500 μm . For the probing, an optical laser pulse of 800 nm wavelength (visible light) was used.

Fig. 2 Calculated transmission of the optical laser pulse of 800 nm wavelength within SiO_2 bulk, irradiated with the 7.1 keV X-ray pulse of energy, 0.1 mJ, focussed to 500 μm laser spot.

The calculated transmission coefficient is presented in Fig. 2. The total transmission is decomposed into the contribution of the conduction band electrons and of the valence band holes (two terms in Eq. (6)). The initial decrease of the transmission, accompanied by the increase of the absorption coefficient, corresponds to the increase

of the free electron density, as it can be seen in Fig. 3. It occurs during the pump pulse and slightly after, following the impact ionization cascades. Later, the transmission tends to restore its original value exponentially. This is due to the decrease of the free electron density and the density of the valence holes via the exciton recombination with a characteristic time of 150 fs.

The contribution of valence holes dominates over the free electron contribution. That is due to the fact that the electrons created by the X-rays are excited to high energies of ~ 7 keV. Such fast electrons cannot couple efficiently to 800 nm photons. The valence holes, on the contrary, are slow, since their kinetic energy lays always within the valence band width, which is ~ 20 eV. They then 'couple' to visible light much more efficiently. This indicates that in the models attempting to describe the scattering of visible light on excited insulators, the contribution of the valence holes must be taken into account.

Fig. 3 Calculated conduction electron density in SiO₂ bulk, irradiated with 7.1 keV X-ray pulse of total energy, 0.1 mJ, focussed to 500 μ m laser spot at different pulse durations: 1 fs, 30 fs, and 80 fs at FWHM.

The slope of the transmission curve, directly related to the increase rate of the free electron density, defines the temporal resolution which can be achieved in pump-probe experiments [22, 23]. Therefore, we have additionally performed studies for short and long pump pulses: 1 fs and 80 fs long ones with the same pulse fluence and photon energy. As it can be seen in Fig. 3, the electron density increases initially, and the transmission decreases accordingly (Fig. 2). For the longer pulse of 80 fs, the increase of conduction electron density follows the X-ray laser pulse profile (time-integrated). Applying short laser pulse of 1 fs duration, however, does not change significantly the slope of the free electron density curve. This can be expected, as the secondary ionization cascades play the dominant role in electron production (as also shown earlier in [11]). The present calculations show that after irradiation of SiO₂ with X-rays of 7.1 keV energy, approximately 440 impact ionized electrons are produced per 1 photoionized electron. Thus, making pump pulse shorter only speeds up the production of the primary photoelectrons, but does not change the time scales for the electron density increase which are determined by the specific cascading time. Specifically, an electron of 7 keV needs a few tens of femtoseconds in order to reduce its energy down to 13.5 eV. Making laser pulse shorter than this characteristic time does not further change the slope of the transmission curve. Therefore, this time defines the limiting resolution that one can achieve at this particular X-ray photon energy.

Further increase of the photon energy would produce primary electrons of even higher energies, which would then need even longer times for cascading. Thus, to increase the slope of the electron density, and correspondingly, to improve the time resolution, one should decrease the photon energy to an energy $\sim 2E_{gap}$ of the material. In this case, each photon would produce only one electron directly, and thus the influx of electrons would follow directly the laser pulse shape, without being affected by the secondary cascades.

In order to confirm this idea, we have analyzed electron density produced at different photon energies. Fig. 4 shows the electron density increase for the 1 fs laser pulse of the same absorbed fluence, but for different photon energies. The same absorbed fluence means that the incoming pulse fluence was adjusted to compensate the differences in the photoabsorption cross-section for different photon energies [5]. The reference value of the photoabsorbed fluence was the one obtained for the irradiation of SiO₂ with photons of energy, 7.1 keV (see above). The calculated total pulse fluences are then within the range from 0.02 μ J (for 20 eV photons) up to 0.6 mJ (for 10 keV photons) per 500 μ m spot. In this intensity regime the multiphoton absorption is negligible, and only

single photon absorption contributes to the electron production. Inverse bremsstrahlung can still contribute at this photon energy, heating up the emitted photoelectrons, however, we can neglect this subdominant process in our low intensity case. With the decreasing photon energy, the electron density increase occurs faster, as the ratio between the secondary impact-ionized electrons and the primary photoionized electrons decreases. For the photon energy of 20 eV (which is only a little above the value of $2E_{gap} = 18$ eV), the conduction band electrons produced by photoionizations have energy below the energy of E_{min} , thus, they do not have a sufficient energy to perform secondary ionizations. Therefore, the increase of the electron density takes place only within the laser pulse itself.

Fig. 4 Calculated conduction electron density in SiO₂ bulk, irradiated with 1 fs (at FWHM) pulses of different photon energies: 20 eV, 100 eV, 1 keV, 4 keV, 7.1 keV, and 10 keV but of the same value of the total photoabsorbed fluence.

4 Conclusions

In this paper we presented a theoretical study on modeling of the X-ray pump-visible light probe experiments with solid SiO₂. The model is based on the Monte Carlo method tracing individual particles event-by-event. It treats the following processes: (i) photoabsorption, resulting in the production of the primary energetic free electrons and the deep shell holes, (ii) inelastic electron scattering on deep shells and on the valence band of the material, (iii) elastic scattering on target atoms and bands, (iv) Auger decays of the deep holes, producing secondary electrons, and (v) electron-holes recombination via self-trapping of excitons. Calculated electron and hole distribution functions are then used to predict the transient optical properties of the material.

The transmission of the 800 nm visible light by the material irradiated with the 7.1 keV X-ray femtosecond pulse is calculated. It is shown that the valence band holes give the dominant contribution to the visible light scattering, since the released electrons have too high energies to efficiently interact with visible light photons. It is also found that the timescales of the transient changes of the optical properties are limited by the secondary cascading duration for high photon energies. This implies, the higher the photon energy is, the longer it takes for the increase of the electron density (and the corresponding decrease of the transmission). Reducing the photon energy can help to achieve a better time-resolution.

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