Femtosecond non-equilibrium dynamics of clusters irradiated with short intense VUV pulses

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Abstract. The kinetic Boltzmann equation is used to model the non-equilibrium ionization phase that initiates the evolution of atomic clusters irradiated with single pulses of intense vacuum ultraviolet (VUV) radiation. The duration of the pulses is $\leq 50$ fs and their intensity in the focus is $\leq 10^{14}$ W cm$^{-2}$. This statistical model includes various processes contributing to the sample dynamics at this particular radiation wavelength, and is computationally efficient also for large samples. Two effects are investigated in detail: the impact of the electron heating rate and the effect of the plasma environment on the overall ionization dynamics. The results for the maximal ion charge, the average ion charge and the average kinetic energy per ion are compared to the experimental data obtained at the free-electron-laser facility FLASH at DESY. Our analysis confirms that the dynamics within the irradiated samples is complex, and the total ionization rate is the resultant of various processes. In particular, within the theoretical framework defined in this model, the high-charge states as observed in the experiment cannot be obtained with the standard heating rates derived with Coulomb atomic potentials. Such high-charge states can be created with the enhanced heating rates derived with the effective atomic potentials. The modification of ionization potentials by plasma environment is found to have less effect on the ionization dynamics than the electron heating rate. We believe

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that our results are a step towards better understanding the dynamics within the samples irradiated with intense VUV radiation.

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1. Introduction

The unique properties of the short-wavelength free-electron lasers (FELs) [1]–[4] emitting coherent radiation in ultra-intense femtosecond pulses enable probing dynamic states of matter, transitions and reactions happening within tens of femtoseconds, with wide-ranging implications for solid-state physics, material science and femtochemistry. The focused FEL beam is an excellent tool to generate and probe extreme states of matter [5, 6]. X-ray FELs (XFEL) will initiate novel structural studies of biological systems with single particle diffraction imaging. It is expected that single particle imaging will be applicable in the studies of non-repetitive biological samples that cannot be performed with standard crystallographic methods [7]–[12].

The rapid development of the research on FELs and the emerging experimental results has given strong motivation for theoretical studies on the ionization dynamics within the irradiated samples. Various processes are involved in this dynamics, and their contribution strongly depends on the radiation wavelength. Whereas the mechanisms of energy absorption and ionization within irradiated samples have been identified in the infrared (IR) regime [13]–[17], this is not the case in the vacuum ultraviolet (VUV) regime. The electrons resulting from photoionization of atoms with intense VUV radiation form cold, strongly coupled electron plasma. The dynamics of these electrons is strongly influenced by their dense interacting...
surroundings. This effect is known as plasma screening, and its contribution depends on the charge densities and their temperatures. It leads to a lowering of the ionization potentials of ions and atoms, and also influences the cross-sections for interactions of charges within the plasma.

Full \textit{ab initio} calculations of the charge dynamics within strongly coupled plasmas are not available [18]. Therefore various approximate theoretical approaches are applied [19, 20]. Estimates of the plasma effects derived with these approximate models may differ significantly (e.g. screening models discussed in [20]). Dedicated experiments could be helpful to sort out the relevant mechanisms. Among others, the data from the cluster experiments performed at the FLASH facility at DESY are available for theoretical analysis [21]–[27]. They cover the wavelength range from 100 nm ($E_\gamma = 12.7$ eV) down to 13 nm ($E_\gamma = 95.4$ eV). In this paper, we will refer only to the first experiment, where xenon clusters were irradiated with photons of energy $E_\gamma = 12.7$ eV. Pulse duration did not exceed 50 fs, and the maximal pulse intensity was $I \leq 10^{14}$ W cm$^{-2}$. Highly charged Xe ions (up to +8) of high kinetic energies were detected, indicating a strong energy absorption that could not be explained using the standard approaches [22, 26, 28]. More specifically, the energy absorbed was almost an order of magnitude larger than that predicted with classical absorption models, and the ion charge states were much higher than those observed during the irradiation of isolated atoms under similar conditions. This indicates that at these radiation wavelengths some processes specific to many-body systems are responsible for the enhanced energy absorption.

Several interesting theoretical models have been proposed for describing the evolution of clusters exposed to intense VUV pulses [29]–[35]. Below we give brief characteristics of some of them. A comprehensive review of the work performed up to 2006 is given in [36]. The physics underlying the dynamics within the irradiated clusters is complex. The first theoretical studies started with new ideas but introduced also some simplifications. In [29, 30], the strong energy absorption within an irradiated atomic cluster resulted from the enhanced inverse bremsstrahlung (IB) heating of quasi-free electrons. This rate was estimated with an effective atomic potential [37], which represents the attraction of the nucleus and the average screening effect of bound electrons surrounding the nucleus. Therefore the distribution of bound electronic charge around the nucleus is smooth. An energetic electron that passes through the inner part of an atom/ion is then scattered by an effective positive charge, $Z_{\text{eff}}$, larger than the net charge of the ion. This effect leads to enhancement of the total IB rate that is proportional to the squared charge of the scatterer. This mechanism was first explored in [29]. It leads to the production of high charges within the irradiated clusters. These high charges were created in a sequence of electron impact ionizations. Relative distributions of ion charges were similar to those observed in the experiment [21]. However, this first study considered the ionization within an infinitely extended homogeneous cluster, and did not take into account the dynamics of charges. The IB rate was calculated perturbatively. Also, impact ionization was treated approximately with a simplified rethermalization scheme.

This model was improved by the same group in [30]. A model of cluster expansion was added. The IB rate was recalculated with the Debye-screened Herman–Skillman potential, using a non-perturbative approach. Recombination and impact ionization processes were treated explicitly. Simulations performed with this improved model again showed the formation of highly charged ions within the irradiated clusters.

We stress here that the derivation of the IB rate with the effective atomic potential as performed in [29, 30] is in contrast to the standard approaches that assume Coulomb potentials of point-like ions [38]–[40]. A heating mechanism similar to that in [29] was successfully tested
in the IR regime recently [41]. It was applied to model the heating of quasi-free electrons in large rare-gas clusters irradiated with IR laser pulses. These electrons were heated during elastic large-angle backscattering on ion cores. The potentials of ions were modelled with a parametrized atomic potential similar to that in [29]. The absolute x-ray yield obtained with this effective atomic potential was in better agreement with the experimental data than that obtained with the bare Coulomb atomic potential. Results obtained in [41] are an interesting contribution to the ongoing discussion on the heating mechanisms within clusters irradiated with IR laser pulses [42].

A different mechanism of strong energy absorption was proposed in [31, 32]. According to this model, high charges within small clusters can be created in a sequence of single photoionization processes. Collisional ionization via electron impact and recombination is neglected. For isolated Xe atoms and ions, only single photoionization: \( \text{Xe} + \gamma \rightarrow \text{Xe}^+ \) occurs. This is due to the low energies of the incoming photons, \( E_\gamma = 12.7 \text{ eV} \), that slightly exceed the ionization potential of a neutral Xe atom, \( E_{+1} = 12.1 \text{ eV} \). However, within a cluster the situation changes drastically. Due to the ion environment, interatomic potential barriers are suppressed [16, 31], facilitating the inner ionization of bound electrons into the cluster and the creation of higher charge states due to single photon absorptions.

The electrons released during the photoionization processes are confined within the cluster (inner ionization). They are heated with the classical IB process enhanced by the presence of highly charged ions. The effective heating rate obtained with point-like ions is similar to that of [38, 39]. When the electrons are hot enough, they start to escape from the sample. This initiates its expansion.

Molecular dynamics simulations were performed in order to test this model. Distribution of ion charges obtained for a xenon cluster consisting of 80 atoms was in good agreement with experimental data. The model has not been tested for larger clusters (\( N_{\text{at}} \geq 200 \)) yet. The non-homogeneous distribution of charges within the cluster (consisting of a positively charged outer shell and a neutral core) predicted with these simulations is confirmed by the recent experimental findings for the mixed cluster systems [27].

Another heating process, alternative to IB, was proposed in [34]. This many-body process, called many-body recombination, may occur within dense, strongly coupled electron–ion systems. Electrons are heated in a sequence of recombination and photoionization events. They collide with atoms and ions, creating higher charges via impact ionization. Ions of charge up to +7 were predicted with this model for the Xe_{1000} cluster.

Among other models of laser–cluster interaction, we mention a quasi-classical model of Bauer [33] and the Thomas–Fermi calculations [35]. Results obtained with these models followed qualitatively the experimental findings.

So far the models describing the interaction of the rare-gas clusters with intense VUV radiation were characterized. A model for the absorption of VUV photons in metals and warm-dense-matter was proposed in [43]–[46]. The basis for this model was the microscopic theory of IB that used the IB rates calculated by Krainov [38, 39] for slow and fast electrons. The predictions obtained with this model were in good agreement with the data from the transmission experiments.

As we have shown above, various theoretical models have been developed in order to explore the strong absorption and the presence of high-charge states observed in the first VUV experiment. However, we can expect that if all enhancement factors proposed with these models were included within one model, it would probably lead to the absorption rates being much higher than those observed experimentally.
With this theoretical study, we aim to test the influence of two effects: (i) the impact of the IB heating rate, (ii) the impact of the modification of the ionization potentials (due to the plasma environment) on the non-equilibrium ionization dynamics within the large Xe clusters (\(N = 2500\) atoms) irradiated with a flat pulse of intense VUV radiation. The parameters of the pulse are: photon energy \(E_\gamma = 12.7\) eV, intensity \(10^{12} - 10^{14}\) W cm\(^{-2}\) and duration < 50 fs. We will consider two different IB rates: (i) the IB rate calculated by Krainov for point-like ions in \([38, 39]\) and (ii) the enhanced IB rate proposed by Santra and Greene \([29]\). In order to estimate the effect of the plasma screening and the charged ion environment, we will treat two limiting cases: (i) the case when atomic energy levels are shifted due to the plasma effects and (ii) the case when no energy level shifts are assumed. Atomic potentials then correspond to the potentials of isolated atoms/ions.

2. Simulation method

2.1. Ionization dynamics modelled with a Boltzmann solver

In order to follow the cluster evolution, we will use the statistical Boltzmann approach proposed in \([47, 48]\). Our Boltzmann code solves the full kinetic equations for electron and ion densities within the irradiated sample. Particles (represented as particle densities) interact with the mean electromagnetic field created by all charges and also with the laser field. The microscopic interactions: photoabsorptions and collisional processes (also IB) enter these equations as rates. These rates are included into the two-body collision terms, and are estimated either from experimental data or with theoretical models.

Below we write the general form of kinetic equations within an irradiated sample. The coupled semi-classical Boltzmann equations for a single electron, \(\rho^{(e)}(r, v, t)\), and ion densities, \(\rho^{(i)}(r, v, t)\), where \(i = 0, 1, \ldots, i_{\text{max}}\) denotes the ion charge and \(i_{\text{max}}\) is the maximal ion charge, are

\[
\begin{align*}
\partial_t \rho^{(e)}(r, v, t) + v \cdot \partial_r \rho^{(e)}(r, v, t) + \frac{e}{m} \left( E(r, t) + v \times B(r, t) \right) \cdot \partial_v \rho^{(e)}(r, v, t) & = \Omega^{(e)}(\rho^{(e)}, \rho^{(i)}, r, v, t), \\
\partial_t \rho^{(i)}(r, v, t) + v \cdot \partial_r \rho^{(i)}(r, v, t) - \frac{ie}{M} \left( E(r, t) + v \times B(r, t) \right) \cdot \partial_v \rho^{(i)}(r, v, t) & = \Omega^{(i)}(\rho^{(e)}, \rho^{(i)}, r, v, t).
\end{align*}
\]

These equations include the total electromagnetic force acting on ions and electrons. Collision terms, \(\Omega^{(e,i)}\), describe the changes of the electron/ion densities with time. These changes are due to short-range microscopic processes. The type of processes involved in the sample dynamics depends on the radiation wavelength.

Our simulation tool follows the non-equilibrium femtosecond dynamics within spherically symmetric samples of large or moderate size. As it evolves the particle densities, the computational costs do not scale directly with the number of atoms within the sample. During the sample evolution, no assumption of local thermodynamic equilibrium (LTE) is made \([49]\). Therefore this code can be applied to describe the dynamics of samples irradiated with ultra-short pulses of duration less than a few femtoseconds, i.e. less than or comparable with the thermalization timescale. Techniques for generating such ultra-short pulses have already been
discussed in [50]–[53]. The non-equilibrium treatment of sample evolution is an advantage when comparing our program to the hydrodynamic codes. These codes are efficient for large samples but they include simplifying assumptions on the dynamics of charges such as LTE conditions or the collective movement of charges. If the thermalization timescales are short compared to the pulse length, hydrodynamic models are reliable tools to follow the evolution of irradiated samples. However, at shorter pulses, sample evolution should be treated with non-equilibrium models.

Compared with the state from [47, 48], our model has been significantly extended and improved. More interactions are now treated and included in the program. We will discuss them in the next section.

2.2. Interactions

We will now specify the physical processes that have been included in our model of the charge dynamics within the irradiated cluster:

1. **Photoionizations, collisional ionizations and elastic scatterings of electrons on atoms/ions.** As in [47], the cross-sections for these interactions were estimated with the experimental data on the atomic cross-sections.

2. **Long-range Coulomb interactions of charges.** Interactions with external laser field are treated within the dipole approximation. This approach is justified by the small spatial size of the irradiated cluster of radius \( \sim 36 \, \text{Å} \) when compared to the wavelength of laser radiation \( \sim 100 \, \text{nm} \). Following our estimates from [47], we expect that the attenuation of the laser beam is small, and we neglect it. Interactions of a charge with the internal field are modelled as the electrostatic interaction of this charge with the mean field created by all charges. This mean field is estimated with the densities of positive and negative charges.

3. **Heating of electrons due to the IB process.** The heating rate is estimated either: (i) with the Krainov heating rates calculated for slow and fast electrons [38, 39] or (ii) with the quantum mechanical cross-section obtained with the Born approximation [54], using the effective atomic potential proposed in [29].

4. **Modification of atomic potentials by electron screening and ion environment.** In order to calculate the energy level shifts due to the electron screening, we use the hybrid potential proposed in [20]. This potential was constructed to match the ion-sphere picture (limit of strongly coupled plasma) at small distances and the Debye–Hückel picture (limit of weakly charged plasma) at large distances. Therefore it can adapt to the changing conditions during the evolution of an irradiated cluster. This hybrid potential extends the standard treatment proposed by Stewart and Pyatt [55], as it may additionally account for degeneracy effects. Modification of the ionization cross-sections due to the plasma effects was estimated as in [19] by including the shifted ionization potentials into these cross-sections. This is the first-order approximation that may underestimate the magnitude of the cross-sections, if the energy level shifts are large [56].

Following the ideas proposed in [16, 31, 32], we considered also the influence of the charged environment of an ion within the plasma on the ionization potential of this ion. We included an estimate of the ionization potential shift due to the lowering of the interatomic potential barriers. As the quasi-free electrons within the cluster screen the ion charge, this shift was calculated from the overlap of the screened (Debye–Hückel) potentials of
the neighbouring ions. If the screening by electrons is efficient, as e.g. in the interior of the cluster, the reduction of the ionization potentials due to the barrier suppression will be smaller than that obtained with the bare Coulomb potentials [16, 31]. In the case of low electron screening, e.g. at the surface layer of the cluster, the estimated shifts should approach those obtained with the bare Coulomb potentials.

5. Shielded electron–electron interactions. They induce fast thermalization of the sample. The respective Fokker–Planck term [57] representing this interaction was added to the right-hand side of the Boltzmann equation for the electron density.

The following processes were neglected within our model: excitation and de-excitation of bound electrons, multi-ionization processes, \( \text{Xe}^{+q} + e/γ \rightarrow \text{Xe}^{+(q+n)} \) at \( n > 1 \), as subleading to the single electron impact ionization. Ionization by the internal electric field at the cluster edge could also be neglected. In particular, we have estimated the field at the cluster edge from figure 8(b) to be \( \sim 3.5 \times 10^{10} \) V m\(^{-1}\). According to the estimates of the critical field by [58] that also take into account the Stark shift of the energy levels, this field is not even sufficient for the over-barrier ionization of the \( \text{Xe} \) atom (critical field \( \sim 6.1 \times 10^{10} \) V m\(^{-1}\)). The respective critical field for the \( \text{Xe}^{+1} \) ion is \( 1.4 \times 10^{11} \) V m\(^{-1}\). Further results of our work show that the energy level shifts due to the plasma environment are small. Therefore we can neglect field ionization within our model.

We estimated the contribution of multiphoton processes in detail. Using the cross-sections for multiphoton ionization of \( \text{Xe} \) ions, calculated in [59], we estimated the two-photon and three-photon ionization rates at \( I = 10^{14} \) W cm\(^{-2}\) and \( E_γ = 12.7 \) eV. They were: \( R_{2γ} = σ_2(I/E_γ)^2 = 1.1 \) fs\(^{-1}\) for the \( \text{Xe}^+ \rightarrow \text{Xe}^{+2} \) process, and \( R_{3γ} = σ_3(I/E_γ)^3 = 0.02 \) fs\(^{-1}\) for the \( \text{Xe}^{+2} \rightarrow \text{Xe}^{+3} \) process accordingly. For comparison, the average collisional ionization rate for the process \( \text{Xe}^+ \rightarrow \text{Xe}^{+2} \), estimated after \( \sim 2 \) fs of exposure, when all atoms are singly ionized, was \( R_c \sim 0.5 \) fs\(^{-1}\), and for the process \( \text{Xe}^{+2} \rightarrow \text{Xe}^{+3} \) it was \( \sim 0.2 \) fs\(^{-1}\). This implies that the multiphoton process can contribute only early in the exposure, and at later times the collisional ionization dominates. Therefore the contribution of the multiphoton processes to the total ionization rate is of minor importance. The other multiphoton processes, \( \text{Xe}^{+q} \rightarrow \text{Xe}^{+(q+1)} \), where \( q > 1 \), have even lower rates, and are therefore negligible within the cluster environment.

We also estimated the three-body recombination rate, using the Zeldovich–Raizer formula for singly charged plasma in LTE [60]. This formula was derived assuming the detailed balance principle. It was estimated as \( \sim 1 \) fs\(^{-1}\) early in the exposure (low electron temperatures) and \( \leq 0.04 \) fs\(^{-1}\) later in the exposure (high electron temperatures). Higher ion charges may lead to the enhancement of these recombination rates [61]. As the simulation times are less than 100 fs, the recombination processes are omitted within the present model.

Additional pressure on ions due to the recoil effects during electron–ion collisions was neglected due to a large mass difference between electrons and ions, \( m_e/M_{\text{Xe}} \sim 10^{-5} \), and the short simulation timescales.

At present, our simulation follows the ionization phase of the cluster evolution. The expansion phase will be treated in forthcoming papers (see section 5).

3. Evolution of samples exposed to intense VUV radiation

We will now demonstrate the ionization dynamics within an irradiated cluster using the following example. We will study the evolution of the \( \text{Xe}_{2500} \) cluster exposed to a flat FEL pulse

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of intensity $I = 6 \times 10^{13} \text{ W cm}^{-2}$ and duration $\Delta t = 10 \text{ fs}$. The IB process is modelled with the enhanced IB rate from [29]. Atomic potentials correspond to those of the isolated atoms.

We define the integrated charge densities, $n_j(v, t)$, $n_j(r, t)$, which will further be used to analyse the simulation predictions:

$$n_j(v, t) \equiv \int \rho^{(j)}(r, v, t) \, d^3r,$$

$$n_j(r, t) \equiv \int \rho^{(j)}(r, v, t) \, d^3v.$$

The densities, $\rho^{(j)}(r, v, t)$, are charge densities in phase space. Indices are: $j = e$ for the electron density and $j = 0, \ldots, i_{\text{max}}$ for the ion densities. These densities are evolved with equations (1) and (2). Integrated densities are then obtained with equations (3).

Our Boltzmann solver solves equations (1) and (2) in phase space within the simulation box of a finite size. The limits are: $0 < r < 120 \text{ Å}$ and $0 < v < 140 \text{ Å fs}^{-1}$. The number of grid points is correspondingly 60 in real space and 140 in velocity space. Time steps are of the order of a few attoseconds at most.

The initial configuration is given by a smooth uniform density function representing a spherically symmetric cluster consisting of 2500 neutral xenon atoms. The edges of the sample are smoothed in order to facilitate computation. The density in the centre corresponds to that of the xenon cluster. The radius of this cluster is $\sim 36 \text{ Å}$. The initial velocity distribution of atoms is given by a delta function, $\delta(v)$, approximated by the narrow Gaussian distribution function.

In order to check how our results are influenced by the choice of the width of this Gaussian distribution, we performed a test simulation at the ten-times smaller width. The results obtained agreed with the previous ones. This confirms that our results are not biased by this specific parametrization of the initial ion velocity distribution at the considered simulation timescales.

We can distinguish two main phases of the sample evolution: ionization phase and expansion phase (not discussed here in detail). The non-equilibrium ionization phase starts when the sample is exposed to the laser radiation and lasts until the saturation of ionizations is reached. Its duration depends on the pulse length and the pulse intensity. We have checked that this phase may last up to several tens of femtoseconds at the maximal pulse length considered, $\Delta t = 50 \text{ fs}$, and the pulse intensities in the range, $I \sim 10^{12} - 10^{14} \text{ W cm}^{-2}$.

### 3.1. Photoionization

The ionization phase starts with single photoionizations that release single electrons from the outer shell of Xe atoms: $\gamma(E_\gamma) + \text{Xe} \rightarrow e(E_e) + \text{Xe}^{+1}$, where $E_\gamma$ denotes the energy of the incoming photon, and $E_e$ is the energy of the released photoelectron. In this case, the photoelectron energy will be $\sim 0.6 \text{ eV}$, as the ionization threshold for Xe is $E_{+1} = 12.1 \text{ eV}$. Early in the exposure, only single photoionizations are possible, due to the low energies of the incoming photons. When the density of emitted electrons grows, plasma effects lead to a lowering of the ionization potentials within the sample. If these energy shifts are sufficiently large, further ionizations of Xe ions via single photon absorption can occur. The photoabsorption process: $\gamma + \text{Xe}^{+1} \rightarrow \text{Xe}^{+2}$ is treated within our model.

Figure 1(a) shows the photoionization peak in the electron velocity distribution (on the left) after $\sim 0.5 \text{ fs}$. The second peak (on the right) corresponds to the single photoabsorption during the IB process. It is clearly visible that photoionization remains the dominating process.
Figure 1. Photoionization phase: (a) strong photoionization peak at the electron energy distribution at \( t = 0.5 \) fs, (b) number of electrons released and gross number of ions, \( N_{\text{ion}} = \sum_{i=1}^{i_{\text{max}}} i \cdot N_i \), created as a function of time. Up to \( \sim 2 \) fs of the exposure, the electron population is dominated by photoelectrons. The density \((2m_eE)^{1/2} n_e(E,t)\) corresponds to the density \(v^2 n_e(v,t)\) obtained from the Boltzmann equations that was rewritten in terms of the kinetic energy of electrons.

The number of neutral atoms decreases exponentially, \( N_{\text{at}}(t) = N_0 \cdot e^{-I \sigma_{\gamma} t / E_{\gamma}}\), as can be seen in figure 3(a). This formula implies that the energy absorbed during photoionization, \( E_{\text{abs}}\), will change linearly with \( t \) at small \( t \) (figure 3(b)).

3.2. Heating through IB

When ion and electron densities are large enough, heating or cooling of electrons with the IB process starts: \( e(E_e) \pm n\gamma \rightarrow e(E_e \pm n\hbar\omega)\). IB is defined as a stimulated emission or absorption of radiation quanta by a free electron in the field of an ion. Within the approximation of [54], if the field strength parameter, \( s = eE_0 \frac{1}{m_0 \hbar \omega} \) is low, and the free electrons are slow, single-photon exchanges dominate. In this regime, IB heating rate is fast, i.e. the relevant timescale of IB dynamics is given by tens of attoseconds. As \( s \) is large, or as the free electrons are fast and may undergo collisions with ions with large momentum transfers, multi-photon exchanges can occur. This latter (limiting) case can be identified with the classical impact picture [54]. The IB rate decreases correspondingly. This is reflected by the temperature curve in figure 2(b).
Figure 2. Global parameters within the irradiated cluster as a function of time: (a) total number of electrons, $N_\text{el}$, and gross number of ions, $N_\text{ion} = \sum_{i=1}^{i_{\text{max}}} i \cdot N_i$, where $N_i$ is the number of ions of charge $i$, (b) temperature of electrons, $T_\text{el}$, and (c) energies within the sample per atom, $E/N_\text{atoms}$. Total energy within the sample per atom, $E_\text{total} = E_\text{kinet} + E_\text{pot} + E_\text{bind}$, is the sum of the kinetic energies of electrons and ions, $E_\text{kinet}$, the potential energy within the electron–ion system, $E_\text{pot}$, and the total energy that was needed to release electrons from atoms and ions during the ionization processes, $E_\text{bind}$.

Figure 3. Energy absorption during photoionization: (a) production of single Xe ions from Xe atoms at $t \lesssim 2$ fs and (b) total energy absorbed by the sample as a function of time. Up to $\sim 1.5$ fs, the total energy absorption is a linear function of time. At later times, it becomes nonlinear due to the IB process.
During the heating, the total energy absorption within the sample can be nonlinear with respect to the exposure time and the pulse intensity: \( \frac{dE_{\text{abs}}}{dt} \propto N_{\text{ion}}(I, t) \sigma_{\text{IB}}(I) N_{\text{el}}(I, t) \), as the total numbers of ions and electrons, \( N_{\text{ion}}(I, t) \) and \( N_{\text{el}}(I, t) \), change with the pulse intensity and the exposure time. Due to these nonlinearities, we may expect a different amount of radiation energy absorbed within the sample at the same radiation energy flux, \( F = \int dt \ I(t) = \text{const} \), but at various pulse intensity shapes, \( I(t) \).

Here, we comment on the possible double counting of the classical and quantum IB effect during our simulations. In fact, during the simulations we also take into account the contribution of the classical IB effect as we include elastic scatterings of electrons on ions. This is important for maintaining isotropic distribution of fast electrons within the sample, which is a necessary condition for using the angular momenta expansion of Boltzmann equations [47]. However, heating by the classical effect is small compared to the enhanced heating described by the quantum rate. We have checked that in order to increase the temperature of electrons by 1 eV during IB heating within the Xe\(_{2500}\) cluster, we need an integrated radiation flux of: (i) 0.57 J cm\(^{-2}\) classically and (ii) 0.02 J cm\(^{-2}\) using the quantum cross-section; that is, the flux needed to increase the temperature by 1 eV classically is 30 times larger than in the quantum case. Therefore we claim that we model quantum IB heating with a good accuracy.

During the simulations with the Krainov IB rate that is much smaller than the IB rate calculated with effective atomic potentials, we did not include the elastic scattering of electrons on ions; that is, we did not have double counting of classical IB rates.

### 3.3. Collisional ionization

Heated photoelectrons can collide with ions, releasing secondary electrons: \( e(E_e) + \text{Xe}^{+q} \rightarrow e(E'_e) + e(E_{\text{sec}}) + \text{Xe}^{+(q+1)} \). These secondary electrons, \( e(E_{\text{sec}}) \), will also be heated, and they can collide with other ions, releasing more electrons. This initiates cascading processes [62]. Due to the hierarchy of ionization cross-sections, ions of higher charges are created consecutively, and the highest charges are created later during the exposure (figure 4). Three important factors influence the collisional ionization rate:

1. **Screening and ion environment within the plasma.** Early in the ionization phase the plasma is formed. Figure 5 shows the Debye length calculated with electrons at time \( t = 0.02 \) fs
of the exposure. At this time the cluster interior is a plasma with the Debye length, \( l_D \sim 3 \text{ Å} \), much less than the cluster size, \( R \sim 36 \text{ Å} \). With increasing number of electrons, the Debye length decreases down to \( l_D \sim 1 \text{ Å} \). Including the effects of electron screening and cluster environment on the ionization potentials leads to increased production of higher ion charges as compared to the case where collisional cross-sections are calculated with the potentials of isolated atoms. As the shifts of the ionization potentials depend on the electron density and electron temperature, and these parameters change with time, this will affect the energy absorption during collisional ionization, leading to nonlinear effects.

2. **Heating rate.** Results of this and previous simulations \[48\] show that the maximal ion charge created within the sample strongly depends on the heating rate applied.

3. **Shielded electron–electron interactions.** They strongly influence the distribution of energy among plasma electrons. Fast thermalization induced by this interaction (local thermalization timescale \( \leq 3 \text{ fs} \) in the simulated case) cuts the tail of high electron energies (figure 6). We checked that this effect delays the appearance of higher ion charges within the sample, compared to the case where shielded electron–electron interactions are not treated (not shown).

### 3.4. Charge distribution

At the end of the pulse the charge distribution within the sample evolves into a characteristic layer structure consisting of a neutral core and a positively charged outer shell (figures 7 and 8(a)). The interior of the cluster (core) is dominated by ions of the highest charges (figure 9); however, the net charge of the core remains equal to 0. This is due to the quasi-free electrons bound within the core. The positively charged surface layer consists of ions of various charges.

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**Figure 5.** Formation of plasma within the irradiated cluster. The Debye length, \( l_D \), calculated with electrons at \( t = 0.02 \text{ fs} \) within the whole simulation box. The Debye length is small compared to the cluster size of radius \( R = 36 \text{ Å} \). This is one of the plasma signatures.
Figure 6. Effects of the IB heating rate and the shielded electron–electron interactions on the electron velocity distribution. The solid curve shows the electron distribution and the dashed curve shows the Maxwell–Boltzmann distribution obtained with the instantaneous temperature and the electron density estimated at a given time within the cluster. Evolution of the electron density is shown at times $t = 0.5$, 1.0, 1.5, 2.0, 2.5 and 3.0 fs of the exposure. After 3 fs, the shape of the electron distribution approaches that of the Maxwell–Boltzmann distribution. At later times (g), after the pulse has finished and electrons are no longer heated, these two distributions overlap. The density $(2m_e E)^{1/2} n_e(E, t)$ corresponds to the density $v^2 n_e(v, t)$ obtained from the Boltzmann equations that was rewritten in terms of the kinetic energy of electrons.
Figure 7. Schematic plot of charge distribution within an irradiated large cluster at the end of the ionization phase. A positively charged outer shell coats the neutral cluster core of a net charge equal to zero. Thermalized electrons slowly escape from the cluster.

This inhomogeneous spatial distribution of charges is created in the following way. During the irradiation the most energetic electrons escape from the sample. The remaining ions create a Coulomb potential that keeps the slower electrons within the sample (figure 8(b)). These electrons move freely within the cluster, and have the largest velocities when they are far from the cluster edge. At the edges electrons are stopped by the ion potential. As a result, they do not ionize efficiently at the cluster edge but they do ionize the interior of the cluster. Therefore the highest ion charge is created within the core.

However, let us stress the point that the ion distributions observed within the cluster at the end of the ionization phase will not correspond to those recorded by the detector during experiments. Ions from the outer shell will be the first to escape from the sample, and they will reach the time-of-flight (TOF) detector with unchanged charge distribution. In contrast, the cluster core at the end of the ionization phase is a dense system of quasi-free electrons and ions. Recombinations and ionizations (to and from excited states) still occur within the sample. During the long picosecond expansion phase the charges within the core will have enough time for efficient recombination. As a result, the remnants of the core will be weakly charged or neutral. They will reach the detector late, during the hydrodynamic expansion of the core. The charge distribution recorded in the TOF spectrum will then be modified by increasing the participation of lower charges. The mechanism proposed here should be quantitatively verified with an expansion code, e.g. a hydrodynamic code. This is, however, beyond the scope of the present study.
Figure 8. Electrons and ions during the ionization phase: (a) formation of the outer shell of ions, the charge density is defined as $q_{\text{tot}}(r, t) = \sum_{i=1}^{i_{\text{max}}} i \cdot n_i(r, t) - n_e(r, t)$, and (b) electrostatic attractive potential felt by electrons.

Figure 9. Inhomogeneous spatial distribution of ion charge: atom and ion densities, $n_i(r, t)$ ($i = 0–8$), are plotted as a function of the distance from the centre of the cluster. These densities were recorded at the end of the ionization phase. The interior of the cluster is dominated by the highest ion charges. Low charges can be found only at the edge of the cluster.

3.5. Global parameters

Finally, we discuss global parameters obtained with our model (figures 2(a)–(c)). Ionization (from ground states) saturates within $\sim10$ fs after the pulse is switched off. The electron temperature increases during the pulse. This is due to the heating of electrons within the cluster. The temperature decreases rapidly after the pulse is switched off, as the system cools fast during the collisional ionization. After the saturation of ionization the electron temperature decreases with time much more slowly. This effect is due to the slow escape of the thermalized electrons from the cluster.
The total energy of the system increases nonlinearly with time during the pulse. Photoionization and IB are two mechanisms of energy absorption. After the pulse, the total energy is conserved at the considered short simulation timescales.

Our simulation has been stopped at the end of the ionization phase, i.e. after the saturation of ionization was observed. Although the system has not undergone full evolution yet, we can derive some physical predictions from the simulation results. They are: (i) maximal and average ion charges observed, (ii) distribution of ions within the outer shell, (iii) limits for the average kinetic energy per ion and (iv) thermalization timescales. Predictions (i)–(iii) can be compared to the experimental data.

4. Comparison to experimental data

For further analysis we estimate the total amount of pulse energy transferred through a unit surface during the pulse. This is the time-integrated energy flux, \( F \). For a flat pulse of intensity \( I \) and duration \( \Delta t \), it takes a simple form: \( F = I \cdot \Delta t \).

We compare the results of our simulations to the experimental data from the first experiment performed with FLASH at DESY in 2001. Ion fractions and average kinetic energy per ion estimated with averaged TOF spectra were recorded in this experiment at five different pulse energy fluxes: \( F = 0.05, 0.3, 0.84, 1.25 \) and \( 1.5 \) J cm\(^{-2}\) [22]. The error in estimation of the value of \( F \) could be up to a factor of 5. We recall here that the TOF detector could record charged particles (ions) only. There are no data on the neutrals available from these measurements. Experimental predictions that we use here were obtained with the averaged integrated intensities recorded at TOF detectors. For the fluxes \( F = 0.84 \) and \( 1.25 \) J cm\(^{-2}\), those intensities were weighted with relative geometric acceptances and the MCP detector efficiencies. For the fluxes \( F = 0.05, 0.3 \) and \( 1.5 \) J cm\(^{-2}\), only unweighted data are available.

We simulated the non-equilibrium phase of the evolution of Xe\(_{2500}\) clusters exposed to single flat VUV pulses of a fixed flux, \( F \), but of various intensities and pulse durations. Intensities and pulse durations were chosen in order to match the condition: \( I \cdot \Delta t = F \). Pulse intensity was \( \leq 10^{14} \) W cm\(^{-2}\) and pulse length, \( \Delta t \leq 50 \) fs. The predictions obtained from different events were then averaged over the number of events. This procedure enabled us to account for the nonlinear response of the system to the various pulse lengths and pulse intensities at higher radiation fluxes. This scheme followed the experimental data analysis: experimental data were obtained after averaging the single shot data obtained with various FEL pulses of a fixed radiation flux.

Firstly, we show the simulation results obtained with standard IB rates [38, 39]. These rates estimated the heating of the quasi-free electrons during their scattering on the Coulomb potentials of point-like ions. They were calculated separately for slow and fast electrons. We also included the modification of the ionization potentials by the plasma environment. The hybrid model [20] and an estimate of the effect of the surrounding ions (discussed in detail in the preceding section) were used for calculating the energy level shifts within the plasma.

Below we show: (i) ion fractions obtained with the experimental data, (ii) ion fractions obtained within the whole cluster with our model (figure 10). At the fluxes \( F = 0.05 \) and \( 0.3 \) J cm\(^{-2}\), only single charged ions were observed. At higher fluxes, \( F = 0.84 \) and \( 1.5 \) J cm\(^{-2}\), Xe ions up to +2 could be detected. These predictions are in disagreement with the experimental findings that predict much higher charge states at higher radiation fluxes. Obviously, these heating rates were too low to lead to the creation of higher charges within the sample, at least.
with the modification of ionization potentials obtained with the hybrid screening model and
the barrier lowering modelled as described in the preceding section. We explain here that as the
quasi-free electrons within the cluster screen the ion charge, the energy level shift due to the
barrier lowering was calculated from the overlap of the screened (Debye–Hückel) potentials of
the neighbouring ions. If the screening by electrons is efficient, the interatomic potential barriers
obtained with the screened potentials will be higher than those estimated in [16, 31] with bare
Coulomb potentials. As a result, the reduction of the ionization potentials due to the barrier
suppression will be smaller than that obtained with the bare Coulomb potentials. This explains
the discrepancy between the results of [31] and our results.

Secondly, we show the results of the simulations performed with the enhanced IB rate as
proposed in [29] and with the plasma-shifted atomic energy levels. These rates were estimated
with the effective atomic potential. Below we show: (i) the plots of the ion fractions obtained
with the experimental data, (ii) ion fractions obtained within the whole cluster with our
model and (iii) ion fractions obtained within the surface layer (outer shell) with our model
(figure 11).
Figure 11. Results for the enhanced IB rate and the plasma modified atomic potentials: ion fractions within the irradiated xenon cluster at the end of the ionization phase calculated within the whole cluster and within the outer shell. They are compared to the experimental data. In each case (a)–(d), these clusters were irradiated with pulses of different intensities ($\leq 10^{14}$ W cm$^{-2}$) and lengths ($\leq 50$ fs) but of fixed flux. The results obtained were then averaged over the number of pulses. Irradiation was considered at four different radiation fluxes: (a) $F = 0.05$ J cm$^{-2}$, (b) $F = 0.3$ J cm$^{-2}$, (c) $F = 0.84$ J cm$^{-2}$ and (d) $F = 1.5$ J cm$^{-2}$.

At the lowest flux, $F = 0.05$ J cm$^{-2}$, we obtain a large discrepancy with the data. In the experiment, ions of charge up to +3 were found. In the simulation, we find ions up to +2 at most. Also, ion fractions are very different, e.g. the high participation of neutrals predicted within our model cannot be verified with experimental data. Experimental data on the charge distribution at $F = 0.05$ J cm$^{-2}$ can be fitted well with our model at $F = 0.11–0.13$ J cm$^{-2}$ (not shown). This is still within the experimental error of the estimation of the radiation flux.

At the flux $F = 0.3$ J cm$^{-2}$, experimental ion fractions lay between the theoretical fraction histogram obtained within the whole cluster and that obtained within the outer cluster shell. Maximal ion charge is found to be +5 with both experimental data and simulation results.

At higher fluxes, $F = 0.84$ and 1.5 J cm$^{-2}$, the ion fractions predicted within the whole cluster overestimate the experimental data. However, the distribution of ions within the surface layer follows the tendency of data, with the maximum at charge +3. If recombination within the cluster core were efficient during the expansion phase, ion charge within the core should be significantly reduced. The recorded ion spectra from the outer shell would then be corrected by the contributions of the weakly charged remnants of the expanded core. The total charge distributions obtained should then be in agreement with the experimental ones.
First we list our detailed predictions on ion charges. The model predictions on maximal ion charges, $Z_{\text{max}}$, follow the experimental data for higher fluxes, $F = 0.3, 0.84$ and $1.5 \text{ J cm}^{-2}$: $Z_{\text{max}} = +5$ for $F = 0.3 \text{ J cm}^{-2}$, $Z_{\text{max}} \geq +7$ for $F = 0.84 \text{ J cm}^{-2}$, and $Z_{\text{max}} = +8$ for $F = 1.25$ and $1.5 \text{ J cm}^{-2}$. For comparison, if the pulse length is fixed at $\Delta t = 50 \text{ fs}$, the radiation fluxes of $F = 0.3, 0.84, 1.25$ and $1.5 \text{ J cm}^{-2}$ could be achieved with the following intensities, $I \sim 0.6, 1.7, 2.5$ and $3 \times 10^{13} \text{ W cm}^{-2}$.

The average charge is plotted as a function of radiation flux in figure 12. The charges calculated within the outer shell are close to the corresponding experimental values. This indicates that recombination should be efficient during the expansion phase.

Below we also show the average kinetic energy per ion (estimated with our model) as a function of the radiation flux (figure 13). With our model we can only obtain the upper and lower limits for this energy. The lower limit assumes that no recombinations occur during the further expansion of the sample and that the total energy absorbed within this sample minus the sum of the electron binding energies is transferred into the kinetic energy of ions. The upper limit results if we assume the full recombination of the sample during the expansion, that is, the recombination of all quasi-free electrons. In this case, the total absorbed energy can be transferred into the kinetic energy of ions.

We stress here that during the expansion of the sample, electron escapes may also occur. Deriving these limits, we have fully neglected electron escape from the sample. Therefore these limits are rough estimates only and cannot be identified with a realistic physical situation. These limits are compared to the available experimental data on the average ejection energy per atom. The experimental data lay within the model estimates. As expected, the energy absorption shows a nonlinear increase with increasing radiation flux.

Finally, we discuss the results obtained with the enhanced IB rate and the atomic potentials of isolated atoms/ions (plots not shown). As expected, the estimates obtained are lower than
in the previous case in which ionization was facilitated by lowering the ionization potentials. However, the differences are not large, e.g. at the highest flux, $F = 1.5 \text{ J cm}^{-2}$, the Xe$^{+8}$ ion fraction obtained within the whole cluster is: (i) 0.85, when shifts of atomic energy levels are included, and (ii) 0.80 in the case of isolated atomic potentials. The total energy absorbed within the whole cluster differs by $\sim 20\%$ at the highest flux. The average charges differ by $\leq 10\%$ at most.

5. Limitations of the present model

5.1. Single particle densities evolved

The applicability of Boltzmann equations is limited to the classical systems which fulfil the assumptions of molecular chaos and two-body collisions. These assumptions are usually justified by the presence of short-range forces [57, 63]. The single particle density function obtained with Boltzmann equations does not contain any information on the three-body and higher correlations. If the higher order correlations are important, a more fundamental Liouville equation for the $N$-particle density function should be applied [57]. The Liouville equation reduces to the collisionless Vlasov equation [57] in the case of an uncorrelated system. The Fokker–Planck equation [57] can be derived as a limiting form of the Liouville equation for long-range forces (e.g. Coulomb forces). It was shown in [57] that a correct description of many-body Coulomb interactions of plasma electrons and ions as that obtained with the dedicated Fokker–Planck equations can also be obtained with the two-body Boltzmann collision term, assuming the Debye cut-off in the Rutherford scattering cross-section. This simplification does not apply to the electron–electron interactions, where the interacting charged particles...
have identical masses, and the momentum transfer during their collisions cannot be neglected. Therefore we included the respective Fokker–Planck term describing the shielded electron–electron interactions in our equations.

5.2. Classical evolution

We describe the evolution of the irradiated samples, using the classical particle densities. We have checked that the degeneracy parameter $\Upsilon < 0.5$ during the whole evolution of the Xe$_{2500}$ cluster. This implies that the system remains within the classical regime and our classical description is therefore justified.

5.3. Expansion phase

Our Boltzmann solver can also follow the expansion phase: both the electron and ions can move during the sample evolution. However, it becomes computationally inefficient at entering this long semi-equilibrium evolution phase, as it has still to maintain full stability conditions in both velocity and real space that restrict time steps. On the other hand, there is no need to use the full kinetic equation to follow the semi-equilibrium evolution. At this stage, the Boltzmann equation can be conveniently replaced by its hydrodynamic limit. Therefore we use the Boltzmann solver only to follow the non-equilibrium phase and we stop the evolution of the sample when it enters the expansion phase. Simulation of the expansion phase is planned for a forthcoming model. The three-body recombination rate will also be included in this simulation.

5.4. Strong field limit

The Keldysh parameter, $\gamma$, calculated for the maximal field intensity considered in the present work, $I = 10^{14}$ W cm$^{-2}$, and the photon energy, $E = 12.7$ eV, is $\gamma \sim 8$. This indicates that multiphoton processes should dominate at the intensities considered over the tunnelling or over-barrier-ionization processes. According to the estimates of multiphoton rates performed in section 3.1, the multiphoton processes contribute only early in the exposure and therefore can be neglected.

If we increase the intensity to $10^{16}$ W cm$^{-2}$, the corresponding value of the Keldysh parameter is $\gamma = 0.8$. This implies that at these high intensities we enter the tunnelling regime. Correct treatment of the high field limit would then require including the field ionization rates in the model. At present neither multiphoton ionization nor tunnelling (nor over-barrier ionization) are included in our model. This can be done in future when such high intensities have to be considered.

5.5. Many-body effects

As heating is efficient in our model, the irradiated cluster usually enters the weak coupling regime early in the exposure. For instance, in the case of the Xe$_{2500}$ cluster irradiated with a pulse of intensity $I = 6 \times 10^{13}$ W cm$^{-2}$, we used the values of electron temperature and electron density obtained with our model to calculate the Coulomb coupling parameter and degeneracy parameter for electrons. Our estimates show that the sample remains within the classical regime throughout the evolution ($\Upsilon < 0.5$ ), and it leaves the strong coupling regime after $\sim 1.4$ fs of
exposure. The many-body effects such as many-body recombination could then manifest only within this short time interval.

During the irradiation of the Xe\textsubscript{2500} cluster with the intensity of $10^{14}$ W cm\textsuperscript{-2}, we enter the ideal regime after 0.8 fs of exposure, during the irradiation with the intensity of $3 \times 10^{13}$ W cm\textsuperscript{-2}, we enter the ideal regime after 3.1 fs of exposure, and during the irradiation with the intensity $10^{13}$ W cm\textsuperscript{-2}, we enter the ideal regime after 11 fs of exposure.

Therefore it is to be expected that many-body effects can be significant for clusters irradiated with low radiation fluxes. This is an interesting point as we see that our predictions obtained at the lowest radiation flux, $F = 0.05$ J cm\textsuperscript{-2}, cannot reproduce the experimental data. This may be the regime where many-body-recombination effects as proposed in [34] become important.

6. Summary

We performed simulations of ionization dynamics within the Xe\textsubscript{2500} clusters irradiated with flat VUV pulses of intensity $\leq 10^{14}$ W cm\textsuperscript{-2} and duration $\leq 50$ fs. Our model includes the following interactions: photoionization, collisional ionization, elastic scattering of electrons on ions, IB heating, electrostatic interaction of charges, interaction of charges with laser field, shifts of energy levels within atomic potentials due to the plasma environment and shielded electron-electron interactions. Limitations and possible improvements of the model were discussed in section 5.

Within the theoretical framework defined above, we studied the impact of various IB rates and the effect of the plasma environment on the overall ionization dynamics. The results obtained were compared to experimental data. We arrived at the following conclusions:

1. All physical mechanisms that were included in the model contributed to the ionization dynamics. The total ionization rate within the sample was affected most by the heating rate applied, then less strongly by the charge interactions (also the shielded electron–electron interactions) and the plasma environment effects.

2. The heating rate estimated with Coulomb atomic potentials [38, 39] was too low to enable sequential electron impact ionization leading to the production of charges higher than +2. Our analysis included the shifts of the ionization potentials due to the electron screening and to the vicinity of other ions.

3. High charges up to +8 were created with the enhanced IB rate that was estimated with an effective atomic potential [29]. These high-charge states were also observed when shifts of ionization potentials due to the plasma environment were neglected, i.e. when atomic potentials were approximated with those of isolated atoms and ions. In both cases, the total distribution of ion charges obtained with the enhanced IB rate overestimated that obtained with the experimental data. This effect was especially pronounced at high-energy fluxes.

In analogy with [31], the charge distribution within the cluster observed at the end of the ionization phase was inhomogeneous. The cluster consisted of a neutral ion–electron core of net charge equal to 0 and a positively charged outer shell of ions. The highest ion charge was concentrated within the core. The ions of lowest charges could be found only within the surface layer of the cluster (figure 9). The distribution of ions within this surface layer followed the experimental data recorded by the TOF detector. Therefore we expect that the recombination
of the core during the expansion phase (not considered here) should significantly reduce the ion charge within the core. This hypothesis should be quantitatively verified with an expansion code. With the present model we could also obtain the upper and lower limits of the average kinetic energy per ion. These limits were compared to the available experimental data on the average ejection energy per atom. The experimental data lay within the model estimates.

As we showed above, various processes influence the dynamics of samples irradiated by VUV photons. As there are no full ab initio calculations within the strongly coupled systems, the estimated contribution of these processes can be model-dependent. Dedicated experiments could be helpful in sorting out the relevant models. Experimental estimates of the electron temperature within the irradiated clusters at the end of the ionization phase could verify the theoretical estimates for the electron heating rate. It is expected that such estimates of the electron temperature could be obtained with cluster experiments similar to the recent holographic experiment that measured the temperature-dependent expansion rate of irradiated polystyrene spheres [64].

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