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Emission of electrons from rare gas clusters after irradiation with intense VUV pulses of wavelength 100 nm and 32 nm

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\textbf{Abstract.} Kinetic Boltzmann equations are used to describe electron emission spectra obtained after irradiation of noble-gas clusters with intense vacuum ultraviolet (VUV) radiation from a free-electron-laser (FEL). The experimental photoelectron spectra give a complementary and more detailed view of nonlinear processes within atoms and clusters in an intense laser field compared to mass spectroscopy data. Results from our model obtained in this study confirm the experimental and theoretical findings on the differing ionization scenarios at longer (100 nm) and shorter (32 nm) VUV radiation wavelengths. At the wavelength of 100 nm the thermoelectronic electron emission dominates the emission spectra. This indicates the plasma formation and the inverse bremsstrahlung (IB) heating of electrons inside the plasma. This effect is clearly visible for xenon (with the fitted temperature of 6–7 eV), and less visible for argon (with the fitted temperature of 2–3 eV). The two-photon-ionization rate for argon that initiates the cluster ionization, is much lower than the single-photoionization rate for xenon. Also, more of the photoelectrons created within an argon cluster are able to leave it, as they are more energetic than those released from a xenon cluster. Therefore, the IB heating of plasma electrons in argon is

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less efficient than in xenon, as the density of the electrons remaining within the cluster is lower.

At a wavelength of 32 nm the dominant ionization mechanism identified from the electron spectra of argon clusters is the direct multistep ionization. The signature of the thermalization of electrons is also observed. However, as the heating of electrons due to the inverse bremsstrahlung process is weak at these radiation wavelengths and pulse fluences, the increase of the electron temperature with the pulse intensity is mainly due to the increasing photoionization rate within the irradiated sample.

Contents

1. Introduction 2
2. Experimental scheme and simulations 3
3. Ionization dynamics at 100 nm 4
4. Ionization dynamics at 32 nm 5
Acknowledgments 9
References 9

1. Introduction

Studies performed with atomic clusters exposed to intense short wavelength radiation from free-electron-lasers (FELs) [1]–[7] and to higher harmonic radiation [8] reveal interesting information on the ionization dynamics of the irradiated samples. These studies are important for planned experiments with FELs in solid state physics, materials science and for studies of the extreme states of matter [9]–[11]. Accurate predictions on the ionization, thermalization and expansion timescales that can be obtained with cluster experiments are also needed for exploring the limits of experiments on single particle diffraction imaging [12]–[17].

Time-of-flight (TOF) mass spectroscopic studies performed at 100 nm radiation wavelength demonstrated a strong energy absorption within clusters, compared to absorption by single atoms. Coulomb explosion of Xe and Ar clusters resulting in emission of multiply charged ions with keV kinetic energies was recorded, using mass spectroscopic methods [3, 4, 18]. In contrast, the TOF ion spectra of Ar recorded at 32 nm wavelength exhibited rather narrow peaks, showing that Ar fragments do not carry significant kinetic energies [2]. This indicated a less efficient ionization mechanism.

The photoemission spectroscopy studies performed in [1, 2] gave a complementary and more detailed view on ionization of atoms and clusters in an intense laser field, compared to the results from mass spectroscopy. At a peak intensity of $\sim 4.4 \times 10^{12}$ W cm$^{-2}$ and average pulse duration of 100 fs, the kinetic energy distribution of emitted electrons strongly decreased at kinetic energies of 30–40 eV. A characteristic electron temperature below 10 eV was obtained. Simulations with a time-dependent Thomas–Fermi model supported the description of electron spectra as resulting from the thermoelectronic emission from the plasma.

Photoelectron studies performed at 32 nm wavelength [2] suggested a differing ionization dynamics within the irradiated clusters that is driven by a direct multistep photo- and collisional ionization. The inverse bremsstrahlung (IB) heating of the nanoplasma is of minor importance.
at these short radiation wavelengths. Non-dynamic Monte Carlo simulations were performed for 32 nm wavelength in order to support this hypothesis [2]. Here we obtain predictions for the photoemission spectra, using the comprehensive theoretical framework of the kinetic Boltzmann model [19]–[22]. Analysis of ionization dynamics at similar photon energies was also performed in [23, 24]. So far, our model has been successful in reproducing the mass spectroscopic results obtained from the first cluster experiment at FLASH [3, 4] performed at 100 nm wavelength. Below we describe our results on the electron emission spectra in detail.

2. Experimental scheme and simulations

First, we describe briefly the experimental scheme [1, 2]. The rare gas clusters of Xe, Ar were prepared in a supersonic gas expansion. The average cluster size was tuned by varying the stagnation pressure. The full-width at half-maximum of the size distribution is then approximately equal to the average cluster size. FEL radiation was focused on the cluster beam, using an elliptical mirror at grazing incidence. Pulse duration was estimated indirectly with statistical and spectral methods. In case of irradiation with 100 nm radiation, the estimated pulse length was $\sim 100$ fs. In the case of irradiation with 32 nm radiation, the pulse length was $\sim 25$ fs. In both cases the peak intensity was up to $\sim 10^{13}$ W cm$^{-2}$.

The electrons generated during the laser cluster interaction were detected with the microchannel plate of a TOF spectrometer. Kinetic energies of electrons ranging from a few electron volts up to a few tens of electron volts could be analysed. The measured electron distribution curve was converted into kinetic energy spectra [1, 2] and corrected with respect to the calculated transmission of the spectrometer. The electron spectra were obtained by averaging the single-shot spectra over many laser shots. Due to the strongly varying transmission of the spectrometer towards low kinetic energies, the line shapes could not be properly evaluated at energies below 5 eV.

Our theoretical simulations were performed for clusters exposed to single rectangular shaped vacuum ultraviolet (VUV) pulses of fixed fluence. The fluence was a product of the experimentally recorded average pulse intensity and of the pulse length. We obtained predictions for the position of the cluster integrated over the approximately estimated Gaussian spatial profile of the pulse, along the lines of the experimental conditions.

For the theoretical simulations we have used a unified Boltzmann model [20, 21] based on kinetic equations. It included the following interactions: photo- and collisional ionization, three-body recombination, elastic electron–ion and electron–atom scattering, IB heating of quasi-free electrons, shifts of energy levels within atomic potentials due to the plasma environment, and shielded electron–electron interactions. IB rates were calculated as in [25, 26], using the effective atomic potentials. The calculated IB rates were included explicitly in the kinetic equations in the form of the source terms. These interactions and further details of the model were discussed in [20].

Our model follows the full dynamics of irradiated clusters starting from the non-equilibrium ionization phase up to the semi-equilibrium expansion phase. It treats all relevant interactions within the irradiated sample. It gives a complete, consistent description of the cluster evolution. This is the advantage of this model when compared to the simplified models used for the simulations in [1, 2]. The hydrodynamic Thomas–Fermi model [27] used in [1] described only the dynamics of small argon clusters Ar$_{55}$ irradiated with 100 nm radiation, whereas the experimental data were obtained for Ar$_{300}$ and Xe$_{70}$ clusters. This model assumed
the full single ionization of the sample at the initial state, so photoionization was not treated there. In the framework of this hydrodynamic model the non-equilibrium phase of sample evolution was neglected. Also, the frozen core approximation was used, i.e. the positions of ions were fixed during the simulations.

Dedicated simulations performed in order to describe experimental results of [2] were nondynamic Monte Carlo simulations. They were assuming: (i) statistical photoionization of the sample constituents; (ii) instantaneous escape of electrons with an asymptotic kinetic energy (only direct ionization events with positive kinetic energy were accepted) and (iii) no motion of atoms/ions during the pulse (no other processes were included). This simulation scheme just tested the hypothesis that the direct multistep ionization dominates the electron spectra.

3. Ionization dynamics at 100 nm

Predictions obtained with other theoretical models [25, 26], [28]–[32] and with our model [19]–[22] demonstrated that a heating mechanism within a many-body system irradiated with VUV radiation leads to the fast formation of an electron–ion plasma. Enhanced IB [25, 26] was among the potential mechanisms of the plasma heating. As in [25, 26], we apply this plasma heating mechanism in our model.

Shortly after the pulse is off, the plasma evolves towards a local thermodynamic equilibrium (LTE). The electron emission spectrum for xenon gives a clear indication that the heating of electrons was efficient and fast electron thermalization took place: the full emission spectrum can be well fitted by the Maxwell–Boltzmann (M–B) distribution (figure 1(a)). The experimentally estimated temperature of $T = \text{7.1 eV}$ is in good agreement with the prediction from our model, $T = \text{6.5 eV}$ (figure 1(a)). The photoelectron peak due to single photoionization of neutral xenon atoms ($<1\text{ eV}$) cannot be properly identified as it shows up below the spectrometer transmission limit of 5 eV (experimental data) and overlaps with the peak of the M–B distribution (model predictions).

New Journal of Physics 11 (2009) 103012 (http://www.njp.org/)
In the argon spectra the distinct two-photon-ionization peak at energies $\sim 10$ eV is clearly visible (figure 1(b)). This two-photon-ionization mechanism initiates the (single) ionization of the neutral Ar cluster, as the photon energy (13 eV) is below the threshold for the single photoionization (15.8 eV). The two-photon single ionization rate is about 40–80 times smaller than the rate for the single photoionization of xenon at the considered pulse intensity $\sim 4 \times 10^{12}$ W cm$^{-2}$. Also, at this pulse intensity the resonant excitation does not contribute significantly to the total heating rate, and thus can be neglected [18]. Therefore, the ionization in argon progresses much slower than in case of xenon. Also, more energetic photoelectrons in argon ($E_{\text{kin}} = 10$ eV), compared to xenon ($E_{\text{kin}} < 1$ eV) are more likely to escape from the irradiated cluster. As a result, a dilute plasma of quasi-free electrons is created inside the Ar cluster, and therefore the IB heating within argon is much less efficient than that in xenon.

Due to the presence of the large photoelectron peak at 10 eV and also to the additional small peak at 23 eV, which is presumably due to single-photon ionization of neutral argon atoms by third harmonic of FEL radiation [1], fitting the full M–B distribution to the argon data is difficult (figure 1(b)). The final M–B fit to the experimental data reveals the photoelectron temperature, $T = 3.5$ eV, and is in good agreement with the predictions on photoelectron spectra from our model, $T = 2.8$ eV. Our photoelectron temperature estimates are lower than those obtained in [1], where the data from the region below 5 eV have been also used for fitting, and an asymptotic exponential fit was applied, instead of the full M–B distribution [1] that is now fitted to the data.

Finally, we mention that the additional small peaks for argon which are presumably due to the ionization of neutral atoms by third harmonic of FEL radiation [1] cannot be obtained within our model, as we did not include the irradiation by the third harmonic there.

We show also the results from the model on: (i) the total electron temperature in the whole simulation box (measured after the thermalization of quasi-free electrons within the cluster and after the escape of the photoelectrons from the simulation box), (ii) the average charge state formed inside the cluster, and (iii) the estimated average energy per ion, as functions of fluence in the range 0.05–0.5 J cm$^{-2}$ (figure 3). For the considered pulse length of 100 fs this corresponds to pulse intensities in the range $5 \times 10^{11} - 5 \times 10^{12}$ W cm$^{-2}$. These predictions confirm the above dynamics scenarios for xenon and argon. The total electron temperature for argon clusters is lower than the temperature for xenon clusters due to the less efficient IB heating within the argon cluster. The density of the photoelectron plasma in the argon cluster is lower, due to the slow two-photon-ionization rate of argon and the higher escape rate of energetic photoelectrons from argon clusters. The lower electron temperature results in a lower average charge state generated within the argon cluster, compared to case of xenon clusters, and finally in a lower average ion energy. These quantities increase sublinearly with the pulse fluence, and saturate at higher pulse fluences.

The results obtained with Boltzmann model were cross checked with our molecular dynamics (MD) simulations, showing that they are not biased by the choice of the particular simulation method.

4. Ionization dynamics at 32 nm

Analysis of the photoelectron spectra at 32 nm performed in [2] indicated that the direct multistep ionization may be the dominant ionization mechanism at this radiation wavelength. Our predictions confirm this hypothesis. After performing dedicated simulations at the highest...
pulse intensities, we found out that the IB heating contributes to the total energy absorption by argon clusters only by the order of 2% at most at this particular wavelength. These findings are in compliance with recent independent MD-calculations [33].

The direct multistep ionization process is initiated by single photoionization of Ar atoms and indicated in the emission spectra by a pronounced 3p photoionization peak at ∼22 eV. Other processes follow the photoionization, i.e. (i) photoionization of Ar⁺, (ii) the escape of photoelectrons from the cluster, (iii) thermalization of quasi-free photoelectrons within the plasma, (iv) creation of secondary electrons during collisions of photoelectrons with ions/atoms within the cluster [20, 21] and (v) collisional (three-body) recombination [22].

With the increasing number of photoelectrons emitted, the attractive Coulomb field inside the cluster grows. It reduces the kinetic energy of subsequently emitted electrons and eventually prevents further electron emission, leading to the generation of a thermally equilibrated nanoplasma. The contribution of thermalized electrons observed in photoemission spectra increases with time, and, as expected, electron temperature increases with increasing pulse intensity (figure 2). However, as the heating of electrons due to the IB process is negligible (2% effect), the increase of the electron temperature with the pulse intensity is mainly due to the increasing photoionization rate within the irradiated sample.

The predictions obtained from our model for: (i) the total electron temperature inside the whole simulation box (measured after the thermalization of electrons within the cluster and after the escape of the photoelectrons from the simulation box), (ii) the average charge state created and (iii) the estimated average energy per ion, in the argon clusters Ar₈₀ and Ar₁₅₀ support the above scenario (figure 3). These predictions were obtained for fluences in the range 0.005–1.25 J cm⁻². For the considered pulse length of 25 fs this corresponds to the pulse intensities in the range 2 × 10¹ⁱ–5 × 10¹³ W cm⁻².

At a low pulse fluence electrons are produced almost solely during the photoionization process. These photoelectrons can then escape from the cluster. The escape of photoelectrons increases the potential energy of the system. The total electron temperature recorded after the time longer than the electron thermalization time and photoelectron escape time is low, as almost all photoelectrons have left the cluster and the simulation box by that time.

With the increasing pulse fluence more photoelectrons are released. They can escape from the cluster until the attractive Coulomb field of the cluster becomes so large that it prevents further electron emission. This leads to the generation of a thermally equilibrated nanoplasma, as has been observed also in recent MD calculations [2, 33]. More electrons can escape from the larger clusters, creating higher potential energy within the system. This is reflected by a stronger increase of the estimated average energy per ion and of the total electron temperature with the pulse fluence for Ar₁₅₀ clusters than for Ar₈₀ clusters. In contrast, the average charge state created is not cluster-size dependent (figure 3), as it was created mainly during photoionization. As in case of irradiation with 100 nm wavelength, the dependence of the total electron temperature, the average charge state and the average energy per ion on the pulse fluence is sublinear, and saturates at higher pulse fluences.

Performing our studies at 100 nm and 32 nm, we restricted ourselves only to the cluster sizes and FEL pulse parameters that were investigated experimentally. This was done in order to test the predictivity and accuracy of our model. As differing energy absorption processes are predominant at different wavelengths, and the ionization dynamics is a nonlinear function of pulse fluence (figure 3), the comparison of the results obtained at 100 nm and 32 nm is not straightforward. The photoionization rate for argon at 100 nm is five times smaller than
Figure 2. Electron emission spectra for: (a)–(d) Ar$_{80}$, and (e) Ar$_{150}$ clusters. Ar$_{80}$ clusters were irradiated with single FEL pulses ($\lambda_{\text{FEL}} = 32 \text{ nm}$) of intensities in the range of $0.02-5 \times 10^{13} \text{ W cm}^{-2}$ and the average pulse duration of 25 fs. The Ar$_{150}$ cluster was irradiated with a FEL pulse of intensity, $3 \times 10^{13} \text{ W cm}^{-2}$. We show also the results from our MD simulations. The vertical line at 5 eV shows the data cut off.

at 32 nm but the IB heating, which is negligible at 32 nm, contributes significantly to the energy absorption at 100 nm. In contrast, more photoelectrons can escape from argon clusters at 32 nm, as they are more energetic than those released from clusters at 100 nm. This greatly increases the potential energy within the clusters at 32 nm. Due to these competing processes, the net energy
Figure 3. (a) Temperature of electrons inside the whole simulation box after the electron thermalization and after the escape of photoelectrons from the simulation box, (c) average charge state created within a cluster, and (e) estimated average energy per ion as functions of fluence at 100 nm wavelength for Xe$_{70}$ and Ar$_{300}$ clusters. Plots (b), (d) and (f) show the same parameters estimated at 32 nm for Ar$_{150}$ and Ar$_{80}$ clusters. We show both the results obtained with the Boltzmann model (BM) and our MD code (MD).

absorption within the investigated clusters that is reflected by the estimated average energy per ion (figure 3(e) and (f)), does not differ much for the case of irradiation with 100 nm and 32 nm wavelength.

New Journal of Physics 11 (2009) 103012 (http://www.njp.org/)
In summary, our simulations performed within the framework of the Boltzmann model that included all predominant interactions and followed the non-equilibrium and equilibrium dynamics of irradiated samples, confirm the differing ionization scenarios at irradiation of atomic clusters with short- and long-wavelength VUV radiation as posed in [1, 2]. The physical mechanisms contributing are now understood and explained quantitatively. The results obtained have been successfully cross-checked with MD simulations by the Rostock group [33] and our own results showing that they are not biased by the choice of the particular simulation method. The MD results [33] are prepared for publication. All of the experimental electron emission spectra recorded at 100 nm and at 32 nm are now quantitatively reproduced and comprehensively understood within the framework of our kinetic model.

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