Rare-gas clusters in intense VUV, XUV and soft x-ray pulses: signatures of the transition from nanoplasma-driven cluster expansion to Coulomb explosion in ion and electron spectra

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Abstract. We investigate the wavelength-dependent ionization, heating, and expansion dynamics of medium-sized rare-gas clusters (Ar$_{923}$) under intense femtosecond short-wavelength free-electron laser pulses by quasi-classical molecular dynamics simulations. A comparison of the interaction dynamics for pulses with $\hbar\omega = 20$, 38 and 90 eV photon energy at fixed total excitation energy indicates a smooth transition from plasma-driven cluster expansion, where predominantly surface ions are expelled by hydrodynamic forces, to quasi-electrostatic behavior with almost pure Coulomb explosion. Corresponding signatures in the time-dependent cluster dynamics, as well as in the final ion and electron spectra, corroborate that this transition is linked to a crossover in the electron emission processes. The resulting signatures in the electron spectra are shown to be even more reliable for identifying the cluster expansion mechanisms than ion energy spectra. It is shown that the prevailing ionization mechanism and the dominant expansion process can be roughly estimated by a simple frustration parameter.
1. Introduction

Within the last decade, the rapid development of intense laser–matter science at short wavelength has been fueled by tremendous progress in free-electron laser (FEL) technology (DESY’s free-electron laser (http://flash.desy.de/); European XFEL project (http://www.xfel.eu/de/); SPring-8 Compact SASE Source (SCSS), Japan (http://www.spring8.or.jp/end/); Stanford Linac Coherent Light Source (LCLS), USA (http://slacportal.slac.stanford.edu)). The availability of high-intensity pulses from the vacuum ultraviolet (VUV) to the extreme ultraviolet (XUV) up to the x-ray domain opens up new horizons for studying nonlinear laser–matter processes in various kinds of systems, including atoms and molecules [1], clusters [2, 3] and dense plasmas [4]. In particular, the study of clusters in intense FEL pulses offers a route to explore ultrafast laser-driven excitation and decay dynamics of many-particle systems [5, 6]. Moreover, corresponding knowledge about related radiation damage processes and timescales of target destruction is of central importance for novel applications, such as single-shot diffractive imaging of biological samples or time-resolved x-ray holography of highly excited nanosystems [7]–[12].

For studying the nonlinear response processes of finite systems in intense short-wavelength laser radiation, atomic clusters have been proved to be versatile since the early days of VUV-FEL at DESY [13]. A key result of the first VUV experiments and a considerable number of successive theoretical works is that collisional plasma heating through inverse bremsstrahlung (IBS) is a major (typically the leading) heating process in the $\lambda \approx 100$ nm range, and electron emission proceeds mostly via thermal evaporation; see e.g. [14]–[17]. With increasing photon energy, however, collisional plasma heating processes diminish rapidly and different heating and electron emission processes take the lead.

A key process at higher photon energy is sequential direct emission of photo- or Auger electrons into the continuum [18]–[21]. This so-called multistep ionization mechanism produces characteristic plateau features in the electron emission spectra and leaves behind an on-average homogeneously charged cluster, as schematically sketched in figures 1(a) and (b). It should be noted that the width of the multistep plateau has even been considered as a measure of the pulse fluence [22]. Multistep ionization becomes suppressed as soon as the cluster Coulomb potential produces an energy downshift larger than the atomic excess energy of the direct photo- or Auger electrons; see figure 1(c). This suppression effect begins at the cluster center (partial frustration) and then gradually spreads to the surface (full frustration) [18, 24]. If electron impact ionization can be neglected in early stages of the interaction, frustration of direct photoemission is required.
Figure 1. Schematics of the cluster ionization dynamics in intense short-wavelength laser pulses based on the effective cluster potential. After direct photoemission of the first electron (a), subsequently emitted photoelectrons experience a continuous Coulomb downshift with increasing cluster charge (b). This multistep ionization becomes frustrated at a certain ionization stage and nanoplasma formation sets in (c). Collisions between trapped electrons induce evaporation electron emission (d). Finally, the cluster expands due to charging and hydrodynamic forces (e). Note that these key stages have strong conceptual similarities with the physics of ultracold plasmas [23].

for efficient nanoplasma generation in the cluster (figure 1(d)). Still, even if electron trapping sets in, collisional heating of the cluster-bound (quasifree) electrons remains negligible over a wide intensity range [25] and the thermal energy of the nanoplasma electrons is determined by inner ionization heating, i.e. by the excess energy from inner photoionization [24], and dissipation through additional impact ionization. From this stage on, energy equilibration through electron–electron collisions and evaporative electron emission yields an additional nearly thermal distribution to the multistep component of the electron spectrum [19, 21], cf figure 1(d). Hence, the cluster charging and the formation and energetics of the nanoplasma depend on the particular ionization regime, which, in turn, determines the cluster expansion dynamics (figure 1(e)); see e.g. [26]–[28]. In the limit of pure multistep ionization, the quasi-homogeneous charging of the system can be shown to induce a pure Coulomb explosion of the cluster. In contrast to that, ions in the inner region of the cluster can be efficiently screened in the presence of a dense nanoplasma, leading to substantial charge recombination in the cluster center [26] and hydrodynamic expansion behavior [27]. However, as will be shown, the identification of the expansion mechanism from ion energy spectra is often ambiguous. Alternative methods would thus be desirable for identifying the relative impact of the different explosion mechanisms.

The key motivation for the present study is the question of whether electron spectra can be used for an easier identification of the leading expansion processes. Therefore, we investigate the transition from hydrodynamic expansion to Coulomb explosion behavior as a function of wavelength and examine the relations between the ionization regime and the electron and ion spectra. As a numerical example, we analyze the cluster response for excitations with VUV, XUV and soft x-ray pulses using Ar_{23} as a medium-sized model system. The applied molecular dynamics approach is similar to that of a previous study on the intensity dependence of the electron emission from small rare-gas clusters at fixed wavelength [24]. For comparability of
Table 1. Photoionization cross-sections ($\sigma^\alpha$) and ionization energies ($I_p^\alpha$) for atomic Ar. Superscript $\alpha$ indicates electron removal from 3s and 3p shells (as indicated) with initial shell occupation $occ^\alpha$. The remaining ionization potentials are calculated with an atomic all-electron Dirac-LDA code [32]. The remaining cross-sections have been extrapolated from the next known values assuming linear scaling with shell occupation.

| $occ^{3\text{s}}$ | $occ^{3\text{p}}$ | $I_p^{3\text{s}}$ (eV) | $\sigma^{3\text{s}}$ (Mb) (20/38/90 eV) | $I_p^{3\text{p}}$ (eV) | $\sigma^{3\text{p}}$ (Mb) (20/38/90 eV) |
|------------------|------------------|---------------------|--------------------------------|---------------------|--------------------------------|
| Ar               | 2                | 6                   | 29.3                           | – / / 0.2           | 15.76b                          | 30a / 5.0a / 1.2a |
| Ar$^{+}$         | 2                | 5                   | 43.67                          | – / – / 0.2         | 27.63b                          | – / 2.6c / 1.0  |
| Ar$^{2+}$        | 2                | 4                   | 58.09                          | – / – / 0.2         | 42.54                           | – / – / 0.8     |
| Ar$^{3+}$        | 2                | 3                   | 73.60                          | – / – / 0.2         | 57.60                           | – / – / 0.6     |
| Ar$^{4+}$        | 2                | 2                   | 90.07                          | – / – / –           | 74.96                           | – / – / 0.4     |

$^a$ Values taken from [33].

$^b$ Values taken from National Institute of Standards and Technology (NIST).

$^c$ Values taken from [34].

The simulations and to highlight the fundamental differences between the excitation regimes, the laser intensity is chosen such that a fixed total energy absorption is achieved. We further define a simple estimate for the prevailing ionization regime by means of a frustration parameter that can be used for the classification of experiments over a wide range of cluster sizes and laser intensities. For extracting characteristic signatures from the ion spectra that can be used for comparison with experimental spectra, ion energy distributions are analyzed in two ways, i.e. shell resolved as well as charge-state resolved. Therein, electron–ion recombination in the expanding cluster is taken into account, which is of particular interest for the investigation of core-shell effects [26, 27].

The remaining text is structured as follows. The theoretical approach and its numerical implementation are described in section 2. Simulation results are presented in section 3, containing an analysis from the time-dependent perspective (section 3.1) in relation to the final emission spectra (section 3.2) followed by a projection of our results on recent experimental data (section 3.3). Conclusions and an outlook are presented in section 4.

2. Simulation method

The laser–cluster interaction dynamics are modeled by a quasi-classical molecular dynamics approach. Therein, atomic ionization processes are described quantum mechanically via appropriate rates, whereas the resulting ions and electrons are treated classically. This general strategy has been widely applied to laser–cluster interactions from the near infrared up to the x-ray domain; see [5, 6] and references therein.

As optical field ionization is negligible in the wavelength range from the VUV to the soft x-ray range, single-photon ionization is considered to be the main inner ionization process. Electron impact ionization and atomic Auger processes are neglected in the present study. Departing from an initial Ar$_{923}$ cluster in relaxed icosahedral structure, photoionization of the cluster constituents is evaluated stochastically using single-photon ionization cross-sections taken from free atomic Ar; see table 1. It should be noted that medium-induced atomic ionization threshold lowering due to screening and local plasma field effects is explicitly taken into account.
account [29] and enables the charging of cluster constituents beyond the maximum charge state of the atomic species; for details see [16, 30, 31]. The photoionization probability is determined from the instantaneous laser intensity $I(t) = I_0 f^2(t)$, where $I_0$ is the peak intensity and $f(t) = \exp(-2 \ln 2 t^2 / \tau^2)$ is the normalized temporal field envelope of a Gaussian pulse with a pulse duration $\tau$ (full-width at half-maximum). The resulting ions and electrons are propagated classically in the linearly polarized laser field (dipole approximation) and under the influence of a regularized Coulomb interaction of the form

$$V_{ij}(r_{ij}, q_1, q_2) = \frac{e^2}{4\pi \varepsilon_0} \frac{q_i q_j}{r_{ij}} \text{erf}\left(\frac{r_{ij}}{s}\right),$$

(1)

where $e$ is the elementary charge, $r_{ij}$ is the distance between the interacting particles with charge states $q_i$ and $q_j$ and $s = 1.128 \, \text{Å}$ is a numerical smoothing parameter. The latter prevents classical electron–ion recombination below the lowest quantum energy level. The classical trajectories are integrated using a standard velocity–Verlet algorithm. For the classification of active electrons, we define a single-particle energy

$$E_{sp}^i = \frac{m_i}{2} v_i^2 + \sum_{i \neq j} V_{ij},$$

(2)

where $m_i$ and $v_i$ are the mass and velocity of the $i$th particle. Electrons are called quasi-free (delocalized, still bound to the cluster) if $E_{sp} < 0$ and free if $E_{sp} \geq 0$ (continuum energy). Note that only the latter contribute to the cluster charge state.

In order to calculate final ion charge states, e.g. to analyze charge-state-dependent energy spectra, recombination of quasi-free cluster electrons with ions is treated by an approximate scheme developed in [29]. As radiative recombination rates are negligibly small for rapidly expanding clusters, only collisional recombination, with dominantly three-body recombination (TBR), is resolved in our model. Since TBR proceeds mainly to highly excited, Rydberg-like atomic levels, a classical description within the molecular dynamics model is justified. To evaluate recombination, we continue the classical propagation after the laser pulse. Electrons are treated as recombined when localized in an ionic cell after a sufficiently long propagation time (here we use $t_{\text{recomb}} = 3 \, \text{ps}$). The charge state of the corresponding ion is reduced by the number of recombined electrons. The remaining quasi-free electrons persist and are assumed to be removed in an experiment by the extraction fields of the ion detector.

3. Results and discussion

For examining the wavelength-dependent cluster response, we compare excitations of $\text{Ar}_{923}$ with $\tau = 30 \, \text{fs}$ laser pulses at $\hbar \omega = 20, 38$ and $90 \, \text{eV}$, representing typical parameters, at present available at FEL light sources. For brevity these cases will henceforth be denoted as VUV, XUV and soft x-ray. To achieve a fixed total energy absorption (13 keV), the pulse intensities are chosen as $I_0 = 2.5 \times 10^{12}, 1.5 \times 10^{13}$ and $5 \times 10^{13} \, \text{W cm}^{-2}$, respectively. The increase of intensity with photon energy reflects the reduced photoionization cross-sections at shorter wavelengths.

3.1. Time-dependent analysis

A time-dependent analysis of characteristic quantities as extracted from the three simulation scenarios is presented in figure 2. For sufficient statistics, we performed ensemble averaging.
over a sufficiently large number of simulations for each case, i.e. all observables reflect statistical mean values. Figures 2(a)–(c) show the time evolutions of the number of inner ionized, quasi-free and emitted electrons (black/red/blue). The indicated critical cluster charge states for partial ($q_{\text{par}}$) and full frustration ($q_{\text{full}}$) of direct photoionization (top panels) are estimated for a spherical, homogeneously charged cluster with radius $R = r_s N^{1/3}$ ($r_s$ is the Wigner–Seitz radius for Ar) and 3p photoemission ($I_p = 15.76\text{ eV}$) from neutral Ar atoms [24] as $q_{\text{full}} = (\hbar \omega - I_p) r_s N^{1/3}/14.4\text{ eV} \AA$ and $q_{\text{par}} = 2 q_{\text{full}}/3$. We further define a frustration parameter

$$ \alpha = \frac{\tilde{N}_{\text{tot}}}{q_{\text{full}}} $$

as the estimated ratio of photo-activated electrons $\tilde{N}_{\text{tot}} = I_0 \tau \sigma N / \hbar \omega$, where $\sigma$ is the atomic cross-section for the dominant ionization channel, to possible direct photoelectrons prior to full frustration. While $\alpha \lesssim 1$ indicates mostly direct photoemission, $\alpha \gg 1$ results in strong frustration and dense nanoplasma formation.
Focusing on the VUV case ($\hbar \omega = 20 \text{ eV}$), direct photoionization is operational only in a very early period of the laser pulse before the critical charge state for full frustration of $q \approx 6$ is reached; see figure 2(a). As indicated by the large frustration parameter of $\alpha_{\text{VUV}} \approx 100$, additional inner ionization leads to the formation of a dense nanoplasma (red curve). Therein, collisional equilibration of quasi-free electrons leads to thermal electron evaporation, which is the dominant emission process in this case. However, the vast majority of activated electrons remain trapped in the cluster potential. The electron spill-out produced by hydrodynamic electron pressure and the concentration of positive charge at the cluster surface result in a cluster potential that expels ions predominantly from the outer shells; see figure 2(d). Ions in inner shells hardly move within the displayed time interval, indicating efficient screening of the corresponding ions by the remaining quasi-free electrons.

The ionization and expansion behavior gradually changes with increasing photon energy. In the intermediate XUV case ($\hbar \omega = 38 \text{ eV}$), a notable period of multistep ionization occurs in the leading edge of the pulse, where only free electrons are created (figure 2(b)). This direct emission stops abruptly as soon as the full frustration threshold is reached at about $t \approx -15 \text{ fs}$; see figure 2(b). The subsequent generation of quasi-free electrons again induces evaporative electron emission, which roughly sets in at time zero and produces a number of free electrons similar to the previous multistep ionization phase. In contrast to the ion dynamics in the VUV case, a much more pronounced expansion is observed for the second outer shell, and also the inner shells show a considerable expansion effect; see figure 2(e). Comparison to the VUV case indicates the reduced importance of electron trapping over direct photoionization, as is reflected by the lower frustration parameter of $\alpha_{\text{XUV}} \approx 10$.

In the soft x-ray case ($\hbar \omega = 90 \text{ eV}$), multistep ionization is even more pronounced and frustration of direct emission begins only after the pulse peak; see figure 2(c). The subsequently produced quasi-free electrons are less abundant than free electrons and only very weak evaporative emission is observed. This dominance of direct photoemission is reflected by a frustration parameter of $\alpha_{\text{x-ray}} \approx 1$. It should be noted that the direct photoemission stops on a level that is slightly below the predicted threshold for full frustration (dashed line). This effect is attributed to the additional trapping field resulting from quasi-free electrons that are produced in the inner region of the cluster as soon as the partial frustration level is reached. Because of their high excess energy (about 74 eV for 3p electron detachment from Ar), these electrons induce a significant charge separation at the surface (spill-out) that enhances electron trapping. Further, all ionic shells expand similarly to a pure electrostatic Coulomb explosion of the cluster (figure 2(f)).

Deeper insights into the importance of the different expansion mechanisms, i.e. the contributions from hydrodynamic versus electrostatic Coulomb forces to the final total ion kinetic energy, can be gained from a time-resolved analysis of the energy distribution in the system; see figures 2(g)–(i). The evolution of the potential Coulomb energy and the kinetic energy contributions from ions and active electrons can be used to reconstruct the energy exchange processes. Note that the total energy (dashed lines) is the sum of kinetic and potential energies of all plasma particles plus the accumulated atomic ionization potentials for the generated ions. After the laser pulse, i.e. without further inner ionization, the sum of the kinetic and potential energy terms remains constant because of energy conservation (not shown). It is further assumed that the change in the kinetic energy of free electrons is negligible, which is fulfilled in good approximation in all scenarios discussed here. Under these circumstances, any gain of ion kinetic energy is either due to the release of thermal energy by expansion.
cooling of quasi-free electrons (hydrodynamic) or due to the release of potential Coulomb energy (Coulomb explosion).

Based on these assumptions, the following picture can be deduced from the above scenarios. For the VUV case, the ion energy gain results almost completely from the release of thermal energy of quasi-free electrons, indicating the hydrodynamic expansion regime, cf figure 2(g). The small decrease of potential energy indicates that electrostatic Coulomb explosion is only a minor effect. Note that in a nearly neutral plasma the potential energy is negative, while a strongly charged system has positive potential energy. As IBS heating is negligible for all runs presented here (well below 1%), the available thermal energy of quasi-free electrons is determined by ionization heating. Note that significant IBS heating may occur for VUV excitation, but only at much higher pulse fluence \[31\].

An intermediate situation is found with XUV excitation, cf figure 2(h), where nearly equal release of thermal electron energy and potential energy takes place. This scenario thus represents a dynamical mixture of hydrodynamic expansion and Coulomb explosion with comparable contributions. Finally, nearly pure Coulomb explosion is observed in the soft x-ray case (figure 2(i)), where potential energy release yields the main contribution to the ion recoil energy. The three different scenarios thus reflect the transition from hydrodynamic to Coulomb-driven cluster expansion, which is accompanied by a change in the frustration parameter from \(\alpha_{\text{VUV}} \approx 100\) down to \(\alpha_{\text{x-ray}} \approx 1\).

### 3.2. Electron and ion spectra

In the next step, the final electron and ion spectra are examined in detail in order to extract characteristic features of the different ionization and expansion regimes. The spectra are sampled after 3 ps of propagation and include ensemble averaging.

The energy distributions of emitted electrons in figures 3(a)–(c) show a transition from an exponential to a plateau-like structure. In the VUV scenario (figure 3(a)), only a small feature from multistep ionization below 4 eV (see arrow) is observed within the otherwise smooth exponential spectrum, reflecting the dominant contribution from electron evaporation. For XUV excitation, multistep ionization produces a pronounced plateau with the characteristic sharp cutoff at the atomic photoline (at 22 eV); see figure 3(b). Nevertheless, the spectrum still contains a sizable exponential component in the low energy range and beyond the atomic photoline. In contrast to that, the electron spectrum is completely dominated by the multistep plateau for the soft x-ray scenario in figure 3(c), where the evaporative part is negligible. It should be emphasized that the plateau contains a small step at about 60 eV in this case. This feature results from the fact that 3s and 3p ionizations of the argon atoms produce separate multistep plateaus that are shifted by the difference of the corresponding ionization potentials (about 14 eV) against each other. Such a step in the spectrum is not possible for VUV excitation \(I_{3s} > \hbar \omega\) and is less pronounced for the XUV case, where 3p ionization is much more dominant over the 3s channel as in the soft x-ray case.

The electron spectra are thus found to be very sensitive to the ionization processes and the excitation regime being probed and thus represent a useful sensor of the cluster expansion dynamics. Strongly exponential electron spectra occurring for frustration parameters \(\alpha \gg 1\) indicate a strong contribution of hydrodynamic expansion, while plateau structures typical for multistep ionization at \(\alpha \lesssim 1\) are a clear marker for Coulomb explosion.

We now turn to the analysis of the spectra of emitted ions, where the signatures are much more complex. The final ion energy spectrum for the VUV case (black dotted curve
Figure 3. Calculated electron and ion spectra for the same simulation parameters as in figure 2; (a–c) electron energy spectra; (d–f): shell-resolved ion energy spectra (as indicated); (g–i): charge-state-resolved ion energy spectra with and without recombination (as indicated) and corresponding ion charge distributions (insets).

In figure 3(d) is weakly structured and exhibits a negative slope over almost the full energy range. This latter feature is typical for hydrodynamic expansion [35]. The shell-resolved analysis reveals that only ions from the outermost shell and a small portion of the second outer shell achieve high recoil energy, whereas ions from inner shells acquire very little energy. The weak modulation of the full spectrum results from different charge states of ions originating from the outermost shell; cf figure 3(d) in comparison with the charge-state-resolved spectrum in figure 3(g). In addition, the charge-state-resolved spectra for higher final ionization levels show isolated peak structures (green curve for $q = 2$ and blue curve for $q = 3$ in figure 3(g)), which can be explained by electron–ion recombination. While highly charged ions from outer shells can escape quickly, the ionization levels of transiently produced high-$z$ ions from inner shells are reduced by recombination (compare ion spectra before and after recombination in figure 3(g)). Note that ions with a transient charge state $q = 4$ are removed from the final spectra almost completely due to recombination. Isolated features in charge-state-resolved energy spectra thus indicate the presence of a dense nanoplasma (electrons are required to recombine) and are thus typical in the regime of hydrodynamic expansion. This picture is substantiated by recent experiments where such isolated features have been observed in charge-state-resolved ion spectra from xenon clusters in 62 nm pulses [36].
For the XUV case, the total ion energy spectrum is much more structured and exhibits a strong oscillatory behavior. This structure can be traced back to different ion charge states originating mostly from the two outermost shells, see figures 3(e) and (h). The charge-state-resolved contributions before recombination show nearly the same profiles (after rescaling the respective energy axis by the ion charge state). These features indicate that ions from outermost shells already expand similar to a regular Coulomb explosion. Ions from inner shells, on the other hand, still exhibit smooth energy spectra with negative slopes and much lower energies. Comparing the spectra before and after recombination, the signal reduction at low energies for \( q = 2 \) and \( q = 3 \) results from recombination to the next lower charge states \( (q \rightarrow q - 1) \), which transfers the contributions to the energy spectra of charge state \( q - 1 \). Similarly, the signal gain at high energy in the final spectra for \( q = 1 \) and \( q = 2 \) can be traced back to recombination from the next higher ionization stage \( (q + 1 \rightarrow q) \), leading to an additional step on the high-energy side (see figure 3(h)). However, in the XUV scenario recombination is less efficient than for the VUV case, as the lower fraction of quasi-free electrons and the higher temperature of the nanoplasma reduce the corresponding rates (compare spectra before and after recombination in figures 3(g) and (h)). Nonetheless, the signatures from recombination reflect that a dense nanoplasma is still present in the XUV case, although hydrodynamic expansion and Coulomb explosion contribute with comparable strength.

The strongest oscillatory patterns in the ion energy spectra are found for soft x-ray excitation together with a clearly positive slope of the total energy spectrum at low energy, cf figure 3(f). This feature reflects that Coulomb explosion is the dominant mechanism. The maxima of the shell-resolved distributions occur at nonzero energy values that gradually increase with shell number. Note that the peaks at higher ion energy (at about 160 and 260 eV) contain contributions from the four outermost shells, representing a clear signature from overrun effects in the exploding cluster [37]. Inspection of the charge-state-resolved spectra shows that recombination is negligible for this scenario. The latter two effects, however, can be resolved only with the full microscopic information available from the simulation.

From the above analysis it can be concluded that the ion spectra also contain useful features for identifying the type of expansion and thus the ionization regime. The most important features that may be analyzed in corresponding experiments are the slope of the full ion energy spectrum at low energies and the presence of well-isolated peaks in the energy spectra of high-\( z \) ions. The presence of a negative slope hints at hydrodynamic expansion effects, which are closely connected with extensive nanoplasma generation and ionization heating. A sizable contribution from Coulomb explosion, on the other hand, is indicated by a positive slope of the ion energy spectra at low energy, a strong oscillatory pattern and self-similar structures (after rescaling with charge state) of charge-state-resolved ion energy spectra. These features are typical of the multistep ionization regime. Intermediate cases show step structures at the high energy side with respect to the maximum in the charge-state-resolved ion spectra that are produced from recombination of the respective higher charge states. The identification of the strengths of hydrodynamic and Coulomb explosion effects from the ion spectra, however, is expected to be difficult in experiments, as focus averaging and a finite size distribution of the clusters induce additional blurring of the signatures.

3.3. Comparison to experimental data

The above scenarios demonstrate that the ionization regime in intense short-wavelength pulses, i.e. direct emission versus strong electron trapping with thermal evaporation, can be extracted.
from the structure of the electron energy spectra and be roughly estimated by the magnitude of the frustration parameter. Further, because of the direct links between ionization and expansion dynamics, electron spectra or the frustration parameter allow the identification of the dominant cluster expansion mechanism.

As a general trend, the importance of direct photoemission and Coulomb explosion increases over thermal evaporation and hydrodynamic explosion increases with photon energy if the clusters are driven away from strong atomic absorption edges and resonances. However, as the response is in general strongly dependent on pulse energy, ionization cross-section, cluster size and material, the frustration parameter is key to estimate the prevailing processes under specific experimental conditions. For example, in the soft x-ray study at 90 eV photon energy on Xe clusters reported by Thomas et al [27], frustration parameters are in the range \( \alpha \approx 40–1500 \) (larger values correspond to larger clusters) and thus indicate strong frustration and dense nanoplasma generation\(^2\). This picture is consistent with the reported smooth ion energy spectra that have been ascribed to hydrodynamic expansion and is in accordance with our results for the highest frustration parameters. Further, in the experiment of [27], oscillatory patterns of the ion energy spectra become stronger with decreasing size of clusters, where the frustration parameters are reduced. Also this trend is in agreement with our present analysis and supports a noticeable contribution from Coulomb explosion for the smallest clusters.

Another process that is highly dependent on the ionization regime is recombination of quasi-free electrons. Efficient recombination is operational only in the presence of a dense nanoplasma (high frustration parameter) and for not too high electron temperatures. Recent experiments at moderate frustration parameters (\( \alpha = 10–100 \)) on core-shell cluster systems by Hoener et al [26] found strong recombination in the cluster center. Our analysis predicts substantial recombination at comparable frustration parameters, which is consistent with this observation. For low frustration parameters (like in the simulated soft x-ray case), recombination is only weak due to the low nanoplasma population.

Although Auger processes and impact ionization have been neglected in the simulations for clarity, the main findings remain valid at least qualitatively even with their inclusion. The main arguments are that (i) Auger processes effectively only enhance the ionization probability and (ii) the frustration effects that lead to complete suppression of direct photoionization are mainly determined by the cluster charge state. Impact ionization effectively reduces the escape probability of photoelectrons prior to full frustration and introduces more efficient energy sharing within the system, without changing the energy balance. Details of the ion and electron spectra, including the relative abundance of certain ion species or slopes of the electron spectra, may change by the additional ionization channels. The net effect of impact ionization becomes increasingly important with increasing size of clusters at high photon energies (e.g. in the keV range) where ionization cascades induced by photoelectrons produce a large number of secondary electrons.

4. Summary and conclusions

To summarize, we have investigated the transition from nanoplasma-driven hydrodynamic cluster expansion to mostly electrostatic Coulomb explosion for medium-sized Ar\( \text{N}_N \) in intense

\(^2\) For the estimate we used \( I = 5 \times 10^{14} \text{ W cm}^{-2}, \quad \tau = 10 \text{ fs}, \quad \hbar \omega = 90 \text{ eV}, \quad \sigma_{4d} = 24 \text{ Mbarn}, \quad N = 50–10000, \quad r_{Xe} = 2.56 \text{ Å} \) (liquid xenon at boiling point) under the assumption that each photoionization event of a 4d electron is followed by an Auger electron. The excess energy has been taken for the Auger electron (33 eV).

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VUV, XUV and soft x-ray pulses from FEL. By analyzing the time-dependent cluster dynamics as well as final electron and ion spectra with quasi-classical simulations, we have shown that this transition is closely linked to a crossover in the ionization dynamics and electron emission processes. This link makes it possible to identify the dominant cluster expansion mechanism just from the evaluation of electron spectra, as exemplarily shown for three simulation scenarios. While plateau-shaped structures arising from multistep ionization indicate Coulomb explosion, exponential electron spectra from thermal electron evaporation are a typical sign of hydrodynamic cluster expansion. As the transition point measured in terms of photon energy is also strongly dependent on the specific cluster and laser parameters, we have defined a simple frustration parameter that can be used as a rough estimate to determine the ionization and expansion regimes.

Our findings are expected to simplify the interpretation of future strong-field FEL studies with clusters. In particular for the simultaneous measurement of electron and charge-state-resolved ion energy spectra, possibly even together with angular-resolved light scattering for avoiding focus and size averaging, our analysis offers a promising route to a more detailed reconstruction of cluster expansion, overrun effects and recombination processes from laser–cluster interactions in the short wavelength regime. Corresponding insights into the interrelation between ionization and expansion dynamics are of interest for far reaching applications, such as single-shot diffractive imaging or time-resolved x-ray holography on nanosystems.

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