Photoionized Xeon Cluster Plasmas Generated by a Soft X-Ray Laser Pulse

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Abstract. The xenon clusters were irradiated with a soft x-ray laser pulse having a wavelength of 13.9 nm (h\nu=89.2 eV) and intensity of up to 2\times10^{10} W/cm^2 for the first time. The photon energy was high enough to photoionize inner-shell electrons (4d) of the Xe atom. In contrast to the results obtained in synchrotron radiation experiments, the enhancement of double Auger decay probability with increasing cluster size and x-ray laser intensity was observed.

1. Introduction
The interaction of rare gas clusters with ultrashort, high-intensity laser pulses from IR to UV wavelength regime has attracted a great deal of interest for fundamental condensed matter physics and various applications [1]. On the other hand, Wabnitz et al. performed the first experiment to investigate the interaction of xenon (Xe) clusters and a vacuum ultraviolet free electron laser (VUV-FEL, wavelength: 98 nm, pulse width: 100 fs, intensity: 2\times10^{13} W/cm^2) and found that each atom in the large Xe clusters absorbed 30 photons (400 eV) and highly charged ions up to 8+ were produced [2]. This result was very surprising, since for short wavelength laser pulses the collisional heating is significantly suppressed due to their small ponderomotive energy and quiver amplitude [3], which are the most decisive differences between the conventional optical lasers and short wavelength lasers. In order to identify the photoabsorption and heating mechanisms in the clusters, various numerical approaches have been attempted so far [4-6]. However, little is known about the ionization process and expansion dynamics of the clusters irradiated with an intense x-ray laser pulse, in which the photoionization of inner-shell electrons, followed by Auger decays, plays an important role in the generation of highly-charged ions.

We have tried to clarify the ionization dynamics in Xe clusters irradiated with much shorter wavelength laser (13.9 nm) than that of VUV-FEL. The photon energy of the x-ray laser (89.2 eV) is sufficiently high to photoionize Xe 4d inner-shell electrons (threshold energy: \sim 70eV) and its cross section is by ten times larger than that of the valence electron. Subsequently, this inner-hole is filled by the valence electron with a decay time of less than or equal to a few femtoseconds, resulting in the
productions of an Auger electron and a Xe$^{3+}$ ion, which has been extensively investigated by synchrotron radiation experiments involving free Xe atoms [7]. On the other hand, Xe$^{3+}$ ions can also be generated by double Auger (DA) decay of a 4$d$ vacancy, although the transition probability for this process is small.

In this study, the charge state distribution was examined using a time-of-flight ion mass spectrometer (TOF-MS). It was found that Xe$^{3+}$ ions arising from double Auger transition became the dominant final ionic product with increasing cluster size and laser intensity, which was in contrast to the results of synchrotron radiation experiments. The results obtained in the present study suggest that the strongly coupled cluster plasmas were generated by x-ray irradiations, whereby the ionization potential lowering took place due to plasma screening effect.

2. Experimental Setup
The experiment was performed using the x-ray laser facility at JAEA. The x-ray laser with a wavelength of 13.9 nm, a pulse duration $\sim 7$ ps and a resolution of $\lambda/\Delta \lambda < 1 \times 10^{-4}$ was generated by transient collisional excitation scheme for nickel-like silver [8]. The maximum laser intensity focused using a Mo/Si multilayer spherical mirror was $\approx 2 \times 10^{10}$ W/cm$^2$ on target.

The Xe cluster targets were prepared by injecting the high stagnation pressure gas into vacuum using a supersonic conical nozzle with a throat diameter of 200 $\mu$m. The details of the nozzle shape were described in Ref. 9. The average cluster size was estimated by Hagena scaling law and its range was estimated to be $1 \times 10^2$ to $3 \times 10^5$ atoms/cluster [10]. Moreover, a seeding technique (Xe 30%-He 70%) was also employed to promote the formation of larger clusters.

The Xe$^{q+}$ ion stages and their energies were measured using a TOF-MS. The observation was achieved in the direction perpendicular to the laser polarization and the propagation axes [11]. To measure the x-ray laser intensity, a soft x-ray CCD camera was also installed 50 cm from the intersection of the laser and cluster beam.

3. Results and Discussion
In order to confirm the presence of large Xe clusters in the gas jet, a Ti:S laser (800 nm, 60 fs, $\sim 1 \times 10^{15}$ W/cm$^2$) was focused onto the gas targets. Figure 1 shows the TOF spectra and the ion kinetic energies (inset) obtained under a stagnation pressure of 5.6 atm. It was found that the Xe$^{10+}$ ions with the energy of $\sim 10$ keV were generated due to the efficient electron heating within the clusters.

On the other hand, TOF spectra for the Xe clusters irradiated with the x-ray laser pulse are shown

![Figure 1. TOF spectra obtained for Xe clusters irradiated with a Ti:S laser pulse (intensity: $\sim 1 \times 10^{15}$ W/cm$^2$). The ion kinetic energy is also shown in the inset.](image-url)
in figure 2 for various Xe-He gas backing pressures. In contrast to the results of Ti:S laser experiment, the ion mass spectra up to Xe^{3+} were observed. It should be noted that the cross section for the inner-shell ionization of 4d electron is ~16 Mb, whereas for the outermost valence electron ~1.5 Mb at a photon energy of ~90 eV [12]. The similar experiments with synchrotron radiations involving isolated atoms revealed that the dominant decay process following the inner vacancy is the normal Auger process and the ratio of Xe^{2+} to Xe^{3+} ion yield was estimated to be ~4, which was independent of the incidence photon energy in the 4d shape resonance region [13]. Nevertheless, in the present study the Xe^{3+} ions became the dominant ionic fragment for higher backing pressures (Xe^{2+}/Xe^{3+} ratio: ~0.37 at 5.6 atm). Moreover, a similar trend with the laser intensity was also obtained.

![Figure 2. TOF spectra for various sizes of Xe clusters subjected to a 13.9 nm x-ray laser pulse having an intensity of ~1×10^{10} W/cm².](image)

The reason why the Xe^{3+} ion yield increased with the cluster size and laser intensity might be explained by the collisional ionization of the Xe^{2+} ion by the photo- and Auger electrons. In order to estimate the ionization events during the laser irradiation, a set of coupled rate equations involving a Xe atom and a Xe^{q+} ion were solved. The cross sections of the collisional ionizations from the representative levels of Xe^{q+} ion and atom were calculated using the empirical Lotz formula [14]. Moreover, the photoionization for Xe\(^+\) and Xe\(^{2+}\) was also incorporated in the program. Figure 3 shows the ion yield ratio after the passage of laser pulse. Neither simple photoionization (a) nor collisional ionization (b) provided a reasonable explanation for the enhancement of the Xe^{3+} ion yield.

The most probable explanation for the increase in the Xe^{3+} yield with the cluster size or laser intensity could be a reduction in the ionization threshold within the strongly coupled cluster plasmas. The rate equations for Xe\(^+\) and Xe\(^{2+}\) ions show that ~10% of atoms inside the clusters are photoionized at a laser intensity of ~1×10^{10} W/cm². Thus, the plasma screening effect in the cluster is likely to reduce the ionization potential significantly [15]. If the energies of the Xe^{3+} state are reduced within a cluster, the number of possible final Xe^{3+} states will increase and consequently the probability of 4d\(^1\) decaying to the Xe^{3+} states via the DA decay will also increase. Actually, for a free atom the energetically possible final Xe^{3+} states from the 4d\(^1\)\(_{3/2}\) state are \(^4S_{3/2}, ^2D_{3/2, 5/2}\) and \(^2P_{1/2, 3/2}\), whereas for the 4d\(^1\)\(_{5/2}\) state only \(^2D_{5/2}\) and \(^2P_{1/2, 3/2}\) are allowed final states. This difference in the number of the allowed final states drastically changes the ratio of Xe^{2+} to Xe^{3+} products; the yield ratio is estimated to be ~3.5 for 4d\(^1\)\(_{3/2}\) and ~5.0 for 4d\(^1\)\(_{5/2}\) [13]. Although the mechanism underlying this increase in the
DA probability has not been clearly identified, a further investigation using electron TOF spectrometer will make it possible to clarify the ionization and decay processes of the inner-shell vacancy in the clusters.

4. Summary
The interaction of Xe clusters with an x-ray laser pulse with a wavelength of 13.9 nm was investigated using ion TOF mass spectrometry for the first time. In contrast to the results observed in synchrotron radiation experiments involving free atoms, an enhancement of Xe$^{3+}$ yield was observed with increasing cluster size and laser intensity. The most probable explanation for the phenomena could be a reduction in the ionization threshold within the cluster plasmas. Although the mechanism underlying the enhancement of DA probability with the cluster size and x-ray laser intensity has not been clarified, the further investigation using electron TOF spectrometer will make it possible to identify the ionization and decay processes of inner-hole in the clusters.

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