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Frustration of direct photoionization of Ar clusters in intense extreme ultraviolet pulses from a free electron laser

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Abstract

We have measured the kinetic energies of fragment ions from Ar clusters (average cluster size $\langle N\rangle \sim 10$–600) exposed to intense extreme ultraviolet free electron laser pulses ($\lambda \sim 61$ nm, $I \sim 1.3\times 10^{11}$ W cm^{-2}). For small clusters ($\langle N\rangle \lesssim 200$), the average kinetic energy of ions strongly increases with increasing the cluster size, indicating a promotion of the multiple ionization, whereas the average kinetic energy is observed to be saturated for $\langle N\rangle \gtrsim 200$. Considering how many photoelectrons can escape from the cluster, it was found that the size dependence of the ion kinetic energy exhibited the frustration of direct photoionization, which resulted from the strong Coulomb potential of the highly ionized cluster.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The interaction of rare-gas clusters with intense femtosecond laser pulses has been of considerable interest. In the infrared region, one of the most remarkable features is that clusters very efficiently absorb laser energy, compared to isolated atoms and molecules [1–5]. Many experimental and theoretical investigations have revealed that intense laser irradiations cause a transient nanoplasma within the cluster, and the resonant collective modes of the nanoplasma strongly enhances absorption by inverse bremsstrahlung [4, 5]. In the short wavelength region ($\lambda < 100$ nm), however, it was expected that the energy absorption of clusters would be much weaker because the light frequency is too high to reach the plasma resonant frequency, and the quiver energy of an initially generated electron in the radiation field, scaled with the inverse frequency square, is very small.

Recent developments of self-amplified spontaneous emission free electron laser (SASE FEL) allow one to utilize intense laser pulses in the extreme ultraviolet (EUV) region. First experiments on large Xe clusters performed by Wabnitz *et al* [6] have revealed that energy absorption at 98 nm is still very efficient. This surprising result triggered many experimental and theoretical works, and it is suggested that this efficient energy absorption of clusters is due to plasma-screening effects on the ionic scattering potential and barrier suppression for an inner ionization [7–11]. Thus, the efficient energy absorption of a nanoplasma has become one of the interesting topics even at shorter wavelength.

A specific feature of the short wavelength region is that the photon energy is high enough to completely eject electrons from the cluster in the early stage of photoabsorption. However, when the charge state of the cluster significantly increases, kinetic energies of photoelectrons decreases and some electrons cannot escape from the cluster due to the strong Coulomb potential [12]. As a result, many electrons are trapped in the highly charged cluster. In the short wavelength region, this frustration of direct photoelectron emissions plays an important role in the formation of the nanoplasma, which is required for the plasma heating process.

In this paper, we report on the size dependence of kinetic energies of fragment ions from Ar clusters (average size $\langle N\rangle \sim$ 10–600) exposed to intense EUV-FEL pulse with a wavelength of 61 nm. The size dependence of the kinetic energy is discussed in relation with the multiple ionization process of Ar clusters and frustration of direct photoionization.

2. Experiment

Our experimental procedures were similar to our previous report [13]. Briefly, the experiments were performed at the SPring-8 Compact SASE Source (SCSS) test accelerator in Japan [14]. In the present experiments, the wavelength was 61 nm, its polarization axis was in the horizontal plane and the laser pulse repetition rate was 10 Hz. The laser power was calculated to be $\sim 1.3 \times 10^{11}$ W cm^{-2} with an estimated uncertainty of a factor of 2 on the basis of the laser pulse energy ($\sim 30\,\mu$J), pulse duration ($\sim$100 fs), effective focus size ($\sim 15\,\mu$m), total reflectivity of three mirrors ($\sim$13%), the size of the skimmer and partial light stopper and a laser power reduction of 10% by the variable horizontal slits upstream the beam stopper.

The kinetic energy of fragment ions was recorded by momentum imaging measurements with a position sensitive delay-line detector (HEX80 RoentDek [15]). The three-dimensional momentum vector for each ion was extracted from times-of-flight of ions and their positions on the detector. The ion momentum imaging spectrometer is described elsewhere [16, 17]. We recorded the signal waveforms from the detector with two 4-channel digitizers (Acqiris DC282), which can substantially reduce dead times of signals compared to a conventional time-to-digital converter system. Details of the experimental apparatus will be described elsewhere.

Argon clusters were produced by a supersonic expansion through a pinhole (ϕ = 30 μm) kept at 130 K. Stagnation pressure was adjusted to control the average cluster size $\langle N\rangle$ according to well-known scaling laws [18, 19].

3. Results

Time-of-flight mass spectra for various cluster sizes ($\langle N\rangle \sim$ 10, 150 and 600) are shown in figure 1. Major fragment ions are singly charged ions Ar^+ and Ar_2^+, and relative abundances of Ar^+ ions are roughly 80% for each cluster size. We hardly observed multiply charged atomic ions Ar^{z+} ($z \geqslant 2$) under the present laser conditions even for an atomic beam. This may be due to the fact that a minimum energy of 43.4 eV is required to doubly ionize an Ar atom, which means that at least three photons have to be absorbed at the present wavelength. Therefore, the probability of multiphoton absorptions of an Ar atom is negligibly low in the present condition. The peak widths of Ar^+ and Ar_2^+ have sharp and broad components. The broad component indicates that the Ar fragment ions have considerable kinetic energy and that the parent clusters are multiply ionized and dissociated by the Coulomb explosion after intense FEL irradiation. With increasing cluster size, the peaks of the Ar fragment ions become broader. We note that peaks of Ar_2^+ have sharp (low-energetic) and broad (energetic) components. The former fragment ions come from singly ionized clusters, whereas the latter ions originate from multiply ionized ones. With increasing the cluster size, the relative abundance of the former components strongly decreases, reflecting the fact that the multiple ionization becomes dominant for larger clusters.

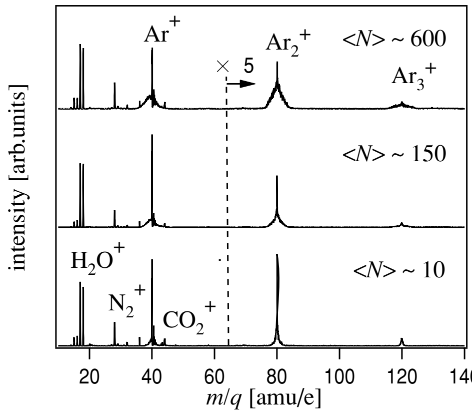

Figure 1. Time-of-flight mass spectra of Ar clusters for various cluster sizes ($\langle N\rangle \sim$ 10, 150 and 600). The ions such as H_2O^+ and N_2^+ come from residual gas in the ionization region.

Figure 2 shows the kinetic energy distributions of Ar^+ and Ar_2^+ for various cluster sizes. In our settings of the ion spectrometer, all ions ejected to the whole 4π sr solid angle with less than 20 eV were collected by the detector. The angular distribution of ions is isotropic below the kinetic energy of 20 eV. However, some of the Ar^+ ions with more than 20 eV miss the detector. To achieve the same collection angle for up to 50 eV energetic ions, we restricted the analysis of Ar^+ ions to a 1.5 sr detection cone towards the ion detector.

The kinetic energies of fragment ions are a few tens eV and much less than those measured at longer wavelength [4, 6]. As inferred from the peak widths of the TOF spectra, the kinetic energies of Ar^+ and Ar_2^+ increase as the cluster size increases. This indicates that the clusters are more strongly ionized when increasing the cluster size. In the small cluster region ($\langle N\rangle <$ 150), the kinetic energy of Ar^+ ions rapidly increases as the cluster size increases, while the size dependence of the kinetic energy seems to be saturated for larger clusters.

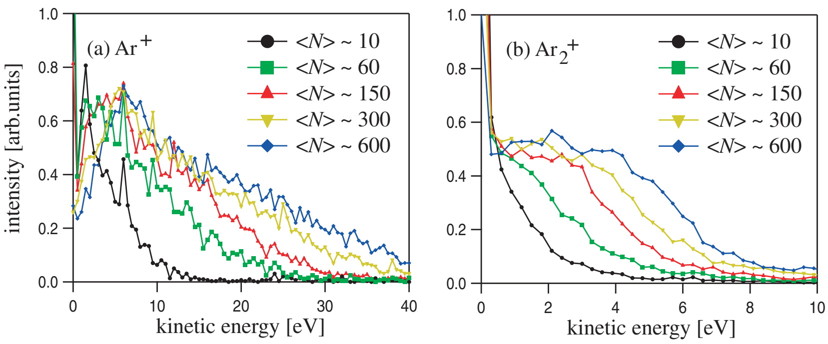

Figure 2. Kinetic energy distribution of (a) Ar^+ and (b) Ar_2^+ ions from irradiated Ar clusters ($\langle N \rangle \sim$ 10, 60, 150, 300 and 600).

4. Discussion

In the infrared region, intense laser irradiation of a rare-gas cluster creates a transient nanoplasma through the inner ionization process [5], which means an excitation from localized electrons to quasifree electrons moving inside the cluster. This nanoplasma formation is required for the extremely high energy absorption. In the EUV region, a photoelectron can escape from the cluster in the early stage of photoabsorption since the photon energy is significantly larger than the ionization energy. When the number of ejected electrons from the cluster significantly increases, some electrons cannot escape from the cluster due to the strong Coulomb potential of the retaining cluster ion [12]. This frustration of direct photoelectron emissions causes the inner ionization and the subsequent nanoplasma formation.

To interpret the size dependence of kinetic energy distributions, we discuss the number of photons a cluster absorbs and the number of removed electrons from the cluster. In our experimental conditions of laser power (1.3×10^{11} W cm^{-2}) and wavelength (61 nm), the ponderomotive energy, which is the cycle-averaged quiver energy of a free electron in the radiation field, is 4×10^{-5} eV, and the Keldysh parameter is much higher than 1 [20, 21]. This means that an optical field ionization such as tunnelling ionization does not play an important role. The fragment ions were only singly charged, and their average kinetic energy is much lower than that for longer wavelength. Considering these results, the laser field driven plasma heating process is negligible. Therefore, the main photon absorption process is the sequence of single photon absorptions of individual Ar atoms in the cluster [12]. From the atomic photoabsorption cross section of an Ar atom at 61 nm and our photon intensity, we estimate a photon absorption probability to be 0.14 photons per atom with an estimated uncertainty of a factor of 2.

The kinetic energies of fragment ions are estimated by using a classical molecular dynamics calculation. At first, we do not take into account the frustration of photoelectron emissions and assume that all photoelectrons completely and instantaneously escape from a cluster. In this case, the number of removed electrons from the cluster is equal to the number of absorbed photons. Here, the cluster size distribution is assumed as the log-normal function, and the distribution of absorbed photon numbers is evaluated by using the laser power of 1.3×10^{11} W cm^{-2} and a Monte Carlo method for each calculation. Single charges are randomly distributed to atoms within a cluster.

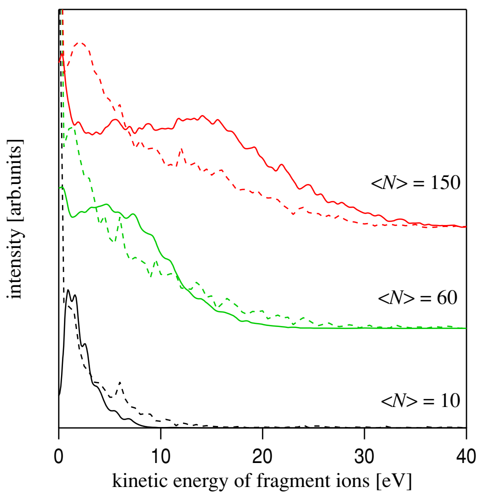

Figure 3. Simulated (solid line) and experimental (dashed line) kinetic energy of fragment ions from multiply charged clusters. For experimental kinetic energy distribution, we take account of relative abundance of Ar^+ and Ar_2^+ ions.

Figure 3 shows simulated kinetic energy distributions. For small clusters, the simulation can reproduce the experimental distribution well. For large clusters, however, the simulation overestimates the ion kinetic energy. We believe that this is because a considerable number of photoelectrons could not escape and inner ionization processes become significant due to the strong Coulomb potential of the cluster ion. It is suggested that the larger clusters significantly suffer from the frustration of direct photoelectron emission and multiple ionization.

Then we consider the energy absorbed by the cluster and the energies of fragments after the Coulomb explosion. We can derive the following equations from the energy conservation:

$$N_{\rm ph} h\nu = N_{\rm ion}(\langle E_{\rm ion}\rangle + I_{\rm p}) + N_{\rm ele}\langle E_{\rm ele}\rangle + N_{\rm neu}\langle E_{\rm neu}\rangle, \quad (1)$$

$$\langle E_{\rm ion}\rangle = \frac{N_{\rm ph}}{N_{\rm ion}} h\nu - I_{\rm p} - \langle E_{\rm ele}\rangle - \frac{N_{\rm neu}}{N_{\rm ion}}\langle E_{\rm neu}\rangle, \quad (2)$$

$$\leqslant \frac{N_{\rm ph}}{N_{\rm ion}} h\nu - I_{\rm p}. \quad (3)$$

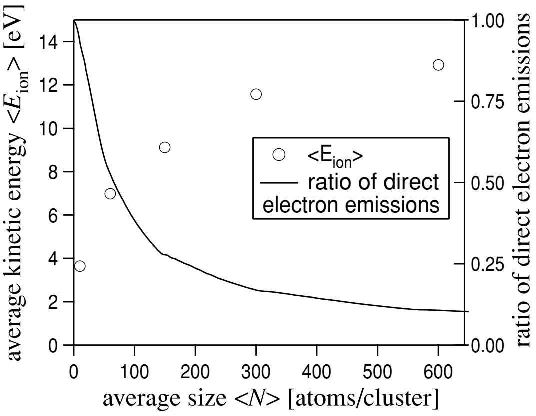

Figure 4. Size dependence of experimental average kinetic energy (circles) and calculation of ratio of photoelectrons directly emitted from the cluster to the sum of all primary electrons, including those which do not escape from the cluster (solid line).

Here $h\nu$ is the photon energy (20.3 eV), I_p is the ionization potential (15.8 eV), $\langle E_{ion(ele,neu)}\rangle$ is the average kinetic energy of fragment ions (ejected electrons, neutral fragments) after the Coulomb explosion, N_{ph} is the number of absorbed photons, $N_{ion(ele,neu)}$ is the number of fragment ions (ejected electron and neutral fragments) generated by the Coulomb explosion. It is noted that the number of ejected electrons from the cluster was equal to that of fragment ions because we observed only singly charged ions. If all photoelectrons would be ejected from the cluster, N_{ion} would be equal to N_{ph}, and this means that $\langle E_{ion}\rangle$ is less than 4.5 eV. This means that in a photoabsorption event always 15.8 eV of the total photon energy (20.3 eV) is spent for overcoming the ionization potential if N_{ion} is equal to N_{ph}. As a result, the kinetic energies of ions, electrons and neutral fragments share 4.5 eV. Therefore $\langle E_{ion}\rangle$ is less than 4.5 eV in this case.

Figure 4 shows the size dependence of the average kinetic energy of the fragment ions. For $\langle N\rangle > 300$, the average kinetic energies are more than 10 eV. This result also shows that the inner ionization and the frustration of direct photoelectron emissions should take place and some electrons of inner ionizations recombine with the ion.

We estimate the degree of inner ionization by calculating the energy of photoelectrons. Considering the ionization potential and the increase of stored Coulomb energy within the charged cluster, we estimate the kinetic energy of an electron released from the *j*th ion from the following equation [12]:

$$E_{ele} = h\nu - I_p - \frac{e^2}{4\pi\epsilon_0}\sum_{i\neq j}\frac{1}{r_{ij}}. \quad (4)$$

Here i runs over all other ions at the distance r_{ij}. Using this equation, we calculate the relative abundance of photoelectrons with $E_{ele} > 0$ eV as the ratio of direct electron emissions, whereas $E_{ele} < 0$ eV means inner ionization. For the cluster size $\langle N\rangle < 200$, the calculation shows that the ratio of direct electron emissions rapidly decreases due to the formation of the strong Coulomb potential. We note that $\langle E_{ele}\rangle$ also decreases due to the Coulomb potential, which was reported in [12]. Thus, the formation of a strong Coulomb potential causes the frustration of the photoionization and reduces the ratio of ions to absorbed photons, N_{ion}/N_{ph}. As seen in figure 4, this size dependence is correlated with the ion kinetic energy. Thus, the present results suggest that the strong increase of $\langle E_{ion}\rangle$ at $\langle N\rangle < 200$ results from the frustration of the direct photoelectron emission, which reduces N_{ion}/N_{ph} and $\langle E_{ele}\rangle$.

Then we consider the number of ejected electrons from the cluster during the Coulomb explosion. For $\langle N\rangle \sim 600$, N_{ele}/N_{ph} ($=N_{ion}/N_{ph}$) is estimated to be ~0.71 from $\langle E_{ion}\rangle$ of 12.9 eV and equation (3), when $\langle E_{ele}\rangle$ and $\langle E_{neu}\rangle$ are assumed to be far less than $\langle E_{ion}\rangle$ and, thus, negligible. On the other hand, the ratio of direct photoelectron emission is estimated to be only ~0.11 from equation (4) and figure 4, whereas the ratio of inner ionization is ~0.89. This indicates that $\sim 0.6N_{ph}$ ($=0.71N_{ph} - 0.11N_{ph}$) electrons of inner ionization are additionally released during the Coulomb explosion, and $\sim 0.29N_{ph}$ ($=0.89N_{ph} - 0.6N_{ph}$) electrons from inner ionizations should recombine with the ion. This indicates that the emission of an inner ionization electron requires 1.5 ($= 0.89\ N_{ph}/0.6N_{ph}$) photons in total. This energy corresponds well to the energy needed to generate an ion ($\langle E_{ion}\rangle + I_P = 28$ eV). Thus, we conclude that recombination of some inner electrons cause additional electron ejections.

5. Conclusion

In conclusion, we have studied the size dependence of the fragment ion kinetic energy emitted from Ar clusters in intense EUV-FEL ($\lambda \sim 61$ nm, $I \sim 1.3 \times 10^{11}$ W cm^{-2}) pulses, measured with an ion momentum imaging apparatus. We found Ar^+ and Ar_2^+ ions to dominate, whereas multiply charged ions were hardly observed. The average kinetic energies of the fragment ions are roughly 10 eV or less, indicating that the plasma heating process was negligible at the present wavelength and laser power. We observed that the kinetic energies of fragment ions strongly increase with the cluster size for relatively small clusters, while the size dependence of ion kinetic energies become saturated for large cluster sizes. Considering energy conservation, this size dependence was shown to result from the frustration of the direct photoelectron emission and photoionization, strongly lowering the ratio of ions to absorbed photons as well as the average electron kinetic energy.

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