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Frustration of direct photoionizations of rare gas clusters in intense extreme ultraviolet free-electron laser pulses

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Abstract. We performed a momentum imaging measurements of Ar clusters irradiated by extreme ultraviolet free electron laser pulses (61nm 1.3×10^{11}W/cm^2) for various cluster sizes. Dominant fragment ions were Ar^+ and Ar_2^+ ions with significant kinetic energy (up to 40eV). This indicates that Ar clusters absorbed many photons, become highly ionized and finally dissociated into many fragments through Coulomb explosion. The size dependence of the ion kinetic energy distribution indicates that larger clusters significantly suffer from the frustration of direct photoelectron emissions and multiple ionizations.

1. Introduction

The interaction of clusters with intense femtosecond laser radiation has been of considerable interests.

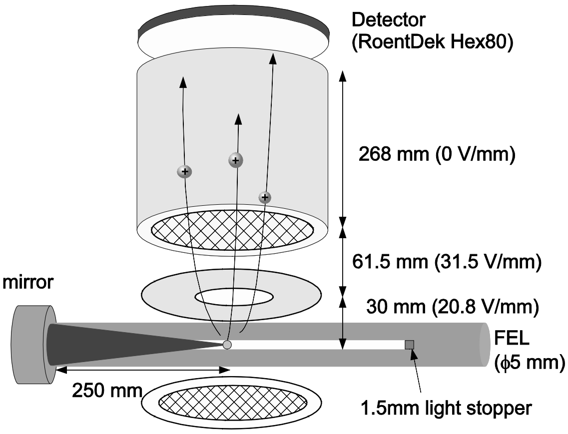

Figure 1. Schematic view of our experimental apparatus.

When the clusters are irradiated with strong visible or near-infrared laser pulses, it is known that high energy ions and electrons are generated from the clusters [1-3]. Many experimental and theoretical works revealed that a cluster absorbs a large amount of photon energy by the creation of transient nanoplasma and the inverse Bremsstrahlung absorption [4]. This efficient energy absorption results in Coulomb explosion of highly charged clusters, which dissociates into many high energy ions. The ion energy distributions give us valuable information on the energy absorption process of clusters in intense laser fields.

Recent progress in free electron lasers (FEL) based on self-amplified spontaneous emission (SASE) allows to utilize intense laser pulses in the short wavelength region. At the TESLA test facility (TTF) at Deutsches Elektronen-Synchrotron (DESY) , Wabnitz *et al.* investigated multiple ionization of Xe clusters by 98-nm FEL radiation, and found complete Coulomb explosion and ions with charge states up to 8+. This result shows extremely strong absorptions of the laser radiation by Xe clusters [5]. The efficient energy absorption of clusters is interesting topics even in the short wavelength laser.

Very recently, the SPring-8 Compact SASE Source (SCSS) test accelerator, has started operation in Japan [6]. It provides linearly polarized EUV-FEL pulses with ~30 μJ per pulse, ~100 fs pulse width, and 10–20 Hz repetition rate in the wavelength region of 51–61 nm. This energy regime is of particular interest because all atoms in any forms of matter can be ionized by just a single photon owing to huge photoionization cross sections.

Recently we reported experimental results of rare gas clusters irradiated by extreme ultraviolet free electron laser (EUV-FEL) pulses from the new FEL facility [7-10]. In this paper, we describe the details on experimental results of Ar clusters irradiated by 61nm 1.3×10^{11}W/cm^2 EUV-FEL pulses.

2. Experiment

The experiments were performed at the SPring-8 Compact SASE Source (SCSS) test accelerator in Japan [6]. 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 $\sim1.3\times10^{11}$ W cm^{-2} with an estimated uncertainty of a factor of 2 on the basis of the laser pulse energy (~30μJ), pulse duration (~100 fs), effective focus size (~15μm), total reflectivity of three mirrors (~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.

Figure 1 shows a schematic view of our experimental apparatus. We performed momentum imaging measurements with a position sensitive delay-line detector (HEX80 RoentDek [11]). To focus the FEL beam onto the cluster beam of 1 mm in diameter, we used a concave mirror at normal incidence. This mirror was fabricated at Tohoku University, using a tungsten-vanadium coating on a superpolished quartz substrate with a focal length of 250 mm. The FEL beam was partially blocked by a 1.5-mm wide vertical beam stopper. We calibrated the position of this vertical beam stopper to avoid

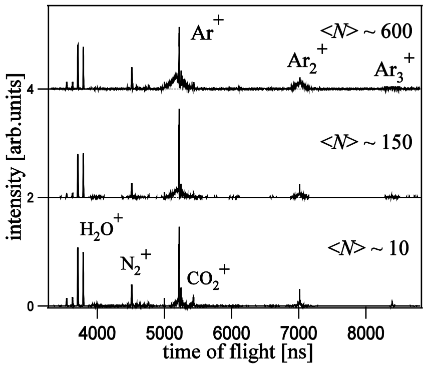

Figure 2. Time-of-flight spectra for various cluster size (<N>=10,150 and 600).The ions such as H_2O^+ and N_2^+ come from residual gas on the FEL beam.

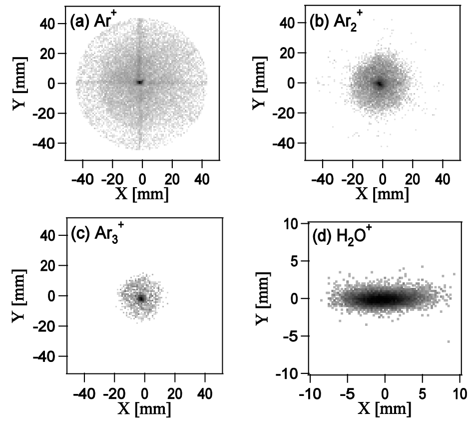

Figure 3. Ion image on the detector for (a) Ar^+, (b) Ar_2^+, and (c) Ar_3^+ ions for the cluster size <N>=150. The ion image of H_2O^+ ions is also shown in (d).

undesirable irradiation of unfocused FEL beam on the cluster beam. Argon clusters were produced by a supersonic expansion through a pin hole (φ = 30 μm) kept at 130 K. The stagnation pressures of Argon gas were 1.5, 3, 4.5, 6 and 8bar. We estimated the average cluster size <N> to be 10, 60, 150, 300 and 600 for each gas pressure using the Hagena's scaling law [12].

The kinetic energy of fragment ions was recorded by momentum imaging measurements with a position sensitive delay-line detector (HEX80 RoentDek). The three dimensional momentum vector for each ion was extracted from times of flight of ions and their positions on the detector [13]. 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 [9].

3. Results

Time-of-flight spectra for <N> = 10, 150 and 600 are shown in Fig. 2. The spectra reveal that dominant fragment ions were Ar^+, Ar_2^+ and Ar_3^+ ions, and relative abundances of Ar^+ ions are roughly 80% for each cluster size. The peaks of Ar^+ ions have sharp and broad components. While the ions in the sharp peak come from atomic Argon in the cluster beam, the broad component corresponds to Ar fragment ions which have a considerable kinetic energy. These energetic fragments indicate that the parent clusters were multiply ionized and dissociated through Coulomb explosion after intense FEL irradiation. With increasing cluster size, the peaks of Ar fragment ions become broader. This indicates that the kinetic energy of fragment ions become larger with the increase of the cluster size.

Figure 3 shows the ion images on the position sensitive detector. The ion images of residual gas ions such as H_2O^+ are nearly lines because they were mainly ionized by incident and unfocus FEL pulses (see Fig. 1). Ion images of Ar_n^+ (n=1, 2 and 3) are circular and broad. This means that only focused FEL beam irradiated the cluster beam. These broad images show that fragment ions come from Coulomb explosion of highly ionized clusters. The ion image of Ar^+ ions seems to be larger than the detector (φ=86mm), and some Ar^+ ions should have missed the detector.

Using the time of flight and the detection position, we calculate the ion kinetic energy for each ion. Figure 4 represents the ion kinetic energy of Ar^+, Ar_2^+ and Ar_3^+ ions. The kinetic energies of Ar^+ ions are several tens eV. In Fig 4, only ions are selected within a cone of 45 deg relative to the spectrometer axis. In this cone, the spectrometer has a energy independent solid angle up t a maximum energy of 50eV. The kinetic energy distributions of Ar^+, Ar_2^+ and Ar_3^+ ions increase with the increase of the cluster size. This result directly shows that larger clusters absorb more photons and dissociate more violently than smaller ones.

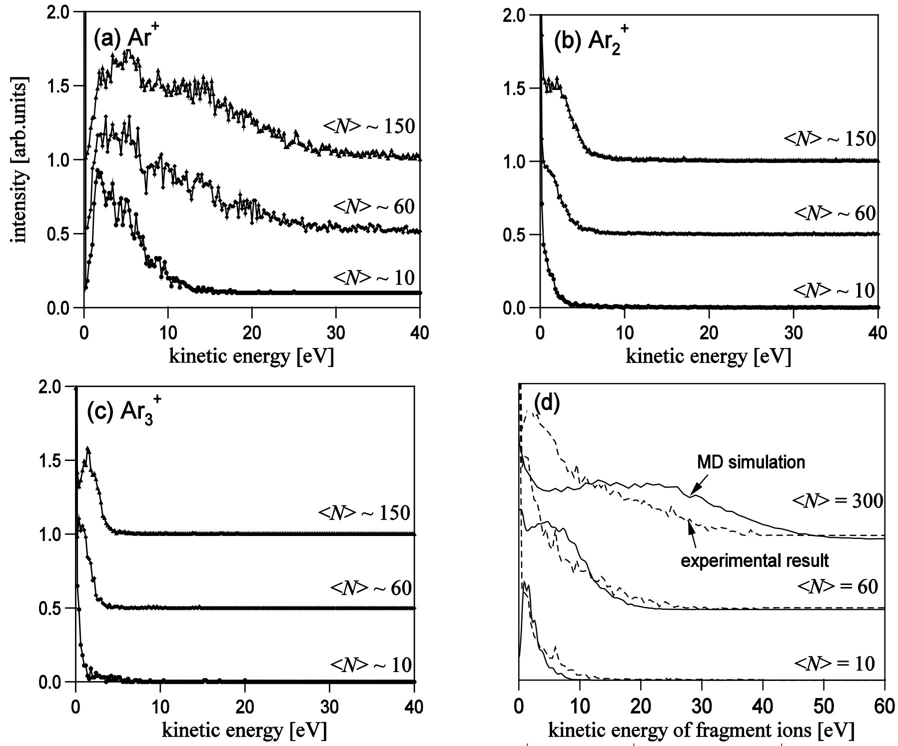

Figure 4. Kinetic energy distributions of (a) Ar^+, (b) Ar_2^+ and (c) Ar_3^+ ions. The kinetic energy distribution of fragment ions in (d) is the sum of Ar_n^+ (n=1, 2 and 3) ions. The simulated results are also shown in (d).

In our laser condition, the ponderomotive energy is 4×10^{-5} eV, and the Keldysh parameter is much higher than one [14]. This means that an optical field ionization does not play an important role. Instead the main photon absorption process is the sequence of single photon absorptions by individual Ar atoms in the cluster. The kinetic energies of fragment ions are estimated by using a classical molecular dynamics calculation. At first, we assume that all photoelectrons completely and instantaneously escape from a cluster. In this case, the charge state of cluster ions 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 4(d) shows simulated and experimental kinetic energy distributions. The experimental distribution is the sum of Ar_n^+ (n=1, 2 and 3) kinetic energy distributions based on their relative abundances. For small clusters, the simulation can reproduce the experimental distribution well. For large clusters, however, the size dependence of the kinetic energy for experimental results seems to be saturated, while kinetic energies of the simulation rapidly become higher with increasing the cluster size. We believe that this is because a considerable number of photoelectrons could not escape and were trapped in the strong Coulomb potential of the highly charged cluster. It is suggested that the larger clusters significantly suffer from the frustration of direct photoelectron emission and multiple ionization.

4. Summary

We have measured ion kinetic energy distributions of Argon clusters irradiated by EUV-FEL pulses for various cluster size. Main fragment ions were singly charged Ar^+, Ar_2^+ and Ar_3^+ ions. With increasing the cluster size, the kinetic energy of fragment ions becomes larger. We observed Ar^+ ions with up to 40 eV. Molecular dynamics simulation results suggest the clusters significantly suffer from the frustration of multiple ionizations.

5. Acknowledgements

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