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Formation of the energetic doubly charged Ne ion by irradiation of large neon clusters using intense EUV-FEL pulses at 52 nm

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Abstract. The interaction of clusters with intense EUV-FEL pulses was investigated using the SPring-8 Compact SASE Source (SCSS) test facility in Japan. Neon clusters of mean sizes $<N>$=1000 and 4000 were irradiated by intense FEL pulses at 52 nm and emitted ions were detected by a momentum imaging spectrometer. The production of energetic doubly charged ions was not found for Ne_{1000}, but it was observed for Ne_{4000} clusters, which suggests that an inhomogeneous charge distribution is generated for the larger clusters.

1. Introduction

The response of matter to intense pulses of laser light in the short wavelength region is an issue of fundamental interest. The recent development of Free Electron Lasers (FEL) based on Self-Amplified Spontaneous-Emission (SASE) in the Extreme UltraViolet (EUV) spectral region below λ=100 nm has enabled us to explore the interaction of intense short wavelength laser pulses with atoms [1–3], molecules [4, 5], and clusters [6–9].

Atomic clusters are ideal objects to study the dynamics driven by the FEL radiation because their size can be controlled from an atom to the bulk-like regime and there is no energy dissipation into surrounding media like in solids. In the present study, we investigated the interaction of neon clusters with intense FEL light. Especially multiple ionization processes were studied at a wavelength of 52 nm by using a dead-time-free multi-particle momentum spectroscopy technique.

2. Experiment

Intense EUV-FEL pulses were provided by the SPring-8 Compact SASE Source (SCSS) test accelerator in Japan [10] with linear polarization in the wavelength region between 51nm and 62nm. A Mg/Si multi-layer mirror fabricated at Lawrence Berkeley National Laboratory (LBNL) was used to focus the FEL pulses into the interaction of a spectrometer set-up. The maximum laser power at the focusing point was estimated to be at most 2×10^{14} W/cm^2 using the estimated (diffraction limited) focal spot size of 3 μm; the experimental conditions considered for this estimation included the laser pulse energy (~30 μJ), the pulse duration (~100 fs), the total reflectivity of three mirrors, the size of skimmers and of a partial light stopper for preventing the incidence of unfocused FEL radiation on the cluster beam [11].

We used a pulsed cluster beam apparatus equipped with a solenoid type pulsed valve (Series 99 valve, Parker Instrumentation Corp.) and a liquid circulating cryostat [12]. The neon cluster beam was produced by supersonic expansion of neat neon gas through a nozzle having an inner diameter of 250 μm. The neon gas was cooled to 85 K using liquid nitrogen and the stagnation pressure was adjusted to control the cluster size. The average number of atoms in the cluster, $<N>$, was estimated to be 1000 and 4000 when the stagnation pressure, P_0, was 4.0 atm and 8.6 atm, respectively, by using well-known scaling laws [13, 14].

The momentum imaging spectrometer equipped with a position sensitive delay-line detector [15] was utilized to record the ions emitted from the cluster by FEL irradiation. The produced ions were extracted into the spectrometer using a constant electric field, traveled a second acceleration region and a field free region, and were finally detected by 120 mm diameter microchannel plates (MCPs) with delay-line anode (Roentdek HEX120). The three dimensional momentum vectors for each ion were evaluated from the time-of-flight (TOF) of those ions and their hit positions on the detector. We recorded the signal waveforms from the detector using two 4-channel digitizers (Acqiris DC282), which can substantially reduce the dead times of signals, as compared to a conventional time-to-digital converter system [16].

3. Results and discussion

Figure 1 shows the TOF spectrum of a neon cluster beam ($<N>$ = 1000 and 4000) irradiated with intense EUV-FEL pulses at 52 nm with the maximum laser power density. Background signals of residual gases have been subtracted in the spectra of Fig. 1. For a cluster beam with $<N>$ = 1000, the peaks of singly charged ions (Ne^+ and Ne_2^+) have broad components due to energetic fragment ions in addition to sharp contributions, whereas the peak of Ne^{2+} has only a sharp component. These sharp peaks of Ne^+ and Ne^{2+} stem from the atomic component within the cluster gas jet. The broad peak of Ne_n^+ (n = 1, 2) implies that irradiated Ne clusters become highly charged and subsequently dissociated into many singly charged ions. When P_0 was increased to 8.6 atm, two peaks appear around the atomic Ne^{2+} peak, corresponding to energetic Ne^{2+} ions from the clusters. The striking contrast in peak shape between singly and doubly charged ions suggests the difference to be due to the kinetic energy distribution (KED) among these ions.

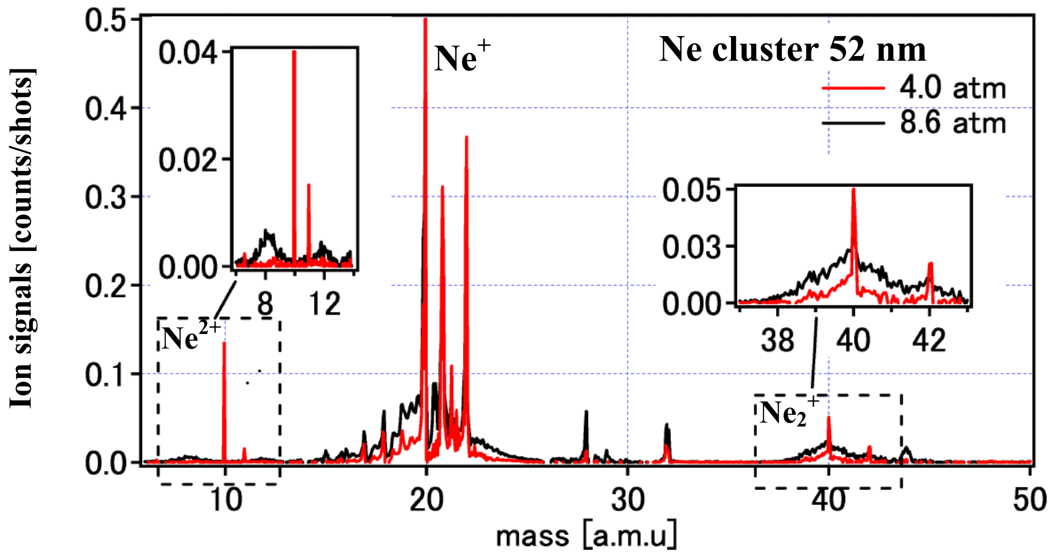

Figure 1. TOF spectrum for Ne clusters irradiated with intense FEL pulses at 52nm. Estimated average cluster size is 1000 and 4000 for the stagnation pressure of 4.0 atm and 8.6 atm, respectively.

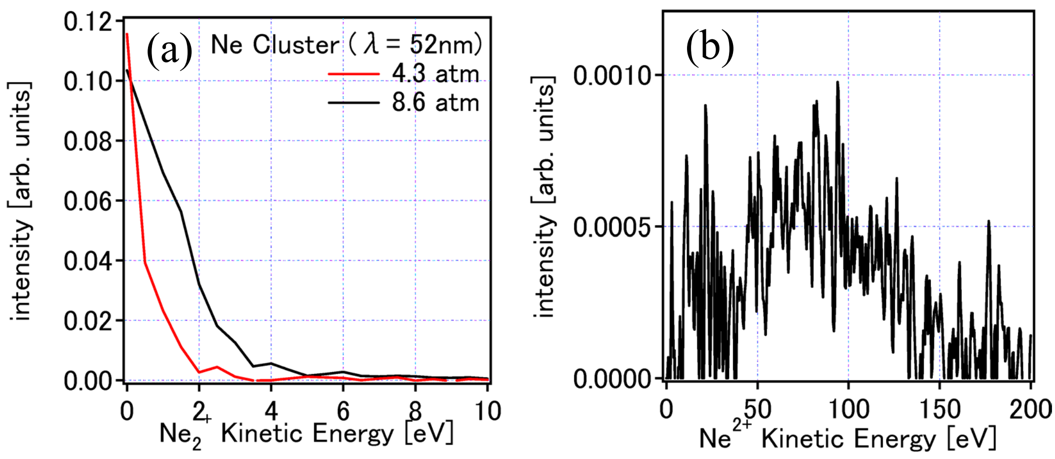

Figure 2. (a). Kinetic energy distribution of the singly charged dimmer, Ne_2^+, from Ne_{1000} (red line) and Ne_{4000} (black line). (b) Kinetic energy distribution of Ne^{2+} emitted from Ne_{4000} clusters irradiated by intense FEL pulses at 52nm.

We evaluated the Kinetic Energy Distribution (KED) of the fragment ions by using the time-of-flight, T, and the hit position, (X,Y), of each ion. To prevent unknown influences of the spectrometer acceptance, we restricted the emission angle of the ions within 25° for the KED analysis [17]. KEDs of Ne_2^+ and Ne^{2+} ions are plotted as a function of kinetic energy in Fig. 2. Here we do not discuss the KED of Ne^+ because we could not evaluate the kinetic energies of individual Ne^+ ions due to overlapping MCP signals from residual gases such as H_2O.

As shown in Fig 2(a), the KEDs of Ne_2^+ monotonically decrease with increasing kinetic energy for both sets of data (4.0 atm and 8.6 atm) displayed, The higher stagnation pressure P_0 makes the average kinetic energy of Ne_2^+ slightly larger. In contrast, the KED of Ne^{2+} has a clear peak around 80 eV, and shows a broad distribution over 100 eV, as shown in Fig. 2(b). Such a difference in the KED between singly and multiply charged ions were also found in the experiments on xenon clusters [12, 18], in which the increase of both, cluster size and laser power density causes the production of the energetic doubly charged ions. The difference in the KED between the singly and highly charged ions could be attributed to different positions of the ions before the dissociation starts by which they are emitted from within a cluster; that is, the highly charged ions are emitted from the cluster surface whereas the singly charged ones are predominantly emitted from the central part of the clusters.

To estimate the relation between the KED and the charge distribution, we have calculated the Coulomb energy stored in the cluster which loses Q electrons under two conditions [18]: One in which the cluster homogeneously contains Q singly charged ions, and another situation in which the cluster contains $(Q - 2)$ singly charged ions homogeneously and a doubly charged ion at the cluster surface. By considering the situation where the latter energy is smaller than the former energy, that is the doubly charged ion is stabilized, we obtained the inequality,

$$2(Q-2)e^2 / R > 5(I_{12} - I_{01}) - 12\, e^2/R \,.$$

Here, R is the radius of cluster, and I_{01} and I_{12} are the first- and second- ionization potentials of neon atom. The left-hand side of inequality corresponds to the potential energy for a doubly charged ion at the cluster surface, which is converted into its kinetic energy by the dissociation. We have evaluated a characteristic energy for the emergence of doubly charged neon from this inequality as 97 eV, which coincides well with the peak position of the experimental KED of Ne^{2+}. This agreement suggests that an inhomogeneous charge distribution within neon cluster is induced by irradiation with intense FEL pulses.

4. Summary

We investigated the interaction of neon clusters with intense EUV-FEL pulses at 52 nm using the SPring-8 Compact SASE Source (SCSS) test facility in Japan. The Neon clusters were irradiated with intense FEL pulses, and the resulting ions were detected by using a momentum imaging spectrometer. We observed the emission of energetic doubly charged Ne ions when large neon clusters ($\langle N \rangle$~4000) were irradiated by intense FEL pulses at a wavelength of 52 nm. Analyzing the kinetic energy distribution of the emitted ions we conclude that an inhomogeneous charge distribution is created within clusters.

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