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Charge and energy transfer in argon-core–neon-shell clusters irradiated by free-electron-laser pulses at 62 nm

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The multiple ionization of Ar-core–Ne-shell clusters in intense extreme-ultraviolet laser pulses ($\lambda \sim 62$ nm) from the free-electron laser in Japan was investigated utilizing a momentum imaging technique. The Ar composition dependence of the kinetic energies and the yields of the fragment ions give evidence for charge transfer from the Ar core to the Ne shell. We have extended the uniformly charged sphere model originally applied to pristine clusters [Islam *et al.*, Phys. Rev. A **73**, 041201(R) (2006)] to the core-shell heterogeneous clusters to estimate the amounts of charge and energy transfers.

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I. INTRODUCTION

The dynamics of matter in intense photon fields is of fundamental interest [1]. Atomic clusters are ideal objects for such studies, not only because their size can be varied from a single atom to a bulklike macroscopic object, but also because there is no energy dissipation into a surrounding medium.

Since the invention of the laser in the last century, the ionization dynamics of clusters in intense near-infrared (NIR) laser fields has been intensively investigated [2]. In the NIR spectral regime, the electric field induced by the laser at a high intensity ($\geqslant 10^{15}$ W/cm^2) is strong enough to distort the Coulomb potentials of the atomic nuclei, thus ionizing the cluster via tunneling ionization. The stripped electrons then become quasifree inside the whole cluster and the resulting transient nanoplasma is efficiently heated by the laser fields via an inverse bremsstrahlung (IBS) process, leading finally to a Coulomb explosion of the clusters [3–8].

The recent advent of free-electron-laser (FEL) light sources has made it possible to investigate intense photon-matter interaction at shorter wavelengths [9–11]. Because the ponderomotive energy is proportional to the inverse square of the laser frequency, ω^{-2}, the direct effect of the laser field on the electron movement is much smaller at short wavelengths than at NIR energies. However, the experiment in the vacuum-ultraviolet (VUV) regime at 98 nm and intensities up to 7×10^{13} W/cm^2 reported that Xe clusters absorbed an unexpectedly large amount of energy and, as a result, experienced a complete Coulomb explosion [12]. This finding sparked significant theoretical efforts. Several new concepts for the description of the absorption and cluster ionization, such as enhanced inverse bremsstrahlung (IBS) [13], charge recombination [14], and lowering the threshold for ionization [15], have been developed. However, the discussion about the significantly enhanced energy absorption at short wavelength is still controversial. While IBS is predicted theoretically to provide strong contributions down to 62 nm [16], an electron spectroscopy experiment for Ar clusters at 32 nm [17] and an ion spectroscopy study for Xe clusters at 61 nm [18] found no evidence for IBS. Recent experimental results also revealed clear significance for other processes occurring during cluster ionization, such as sequential photoabsorption [17], charge recombination [19,20], and inhomogeneous charge redistribution [20,21].

A deeper insight into the spatial origin of the charge states can be obtained from core-shell heterogeneous systems. So far, Xe-core–Ar-shell systems irradiated at $\lambda = 13.7$ nm have been investigated using a time-of-flight (TOF) mass spectrometry [19]. By using the Xe 4*d* resonance absorption, a charge transfer from the Xe core to the Ar shell was observed, but its quantitative estimation was not reported.

In this paper, we adopted Ar-Ne mixed clusters which are known to be self-assembled to a core-shell structure [22]. We present experimental studies on pristine Ne clusters and heterogeneous Ar-core–Ne-shell clusters irradiated by intense 62 nm (20 eV) pulses from the free-electron laser. At this photon energy, one photon suffices for the ionization of an Ar atom (whose ionization potential is 15.8 eV), whereas at least two-photon absorption is needed for the ionization of a Ne atom (whose ionization potential is 21.6 eV). This results in preferential energy injection into the Ar core, and thus the ionization dynamics followed by a charge-transfer process can be studied by varying the Ar core size. The excitation process is expected to be much simpler than in the previously reported Xe-core–Ar-shell systems because the valence electrons are predominantly excited in the ionization. The momentum of each fragment ion along with the mass-to-charge ratio is recorded with our momentum imaging spectrometer, and we find evidence for efficient charge transfer from the Ar core to the Ne shell.

II. EXPERIMENT

Experiments were performed at the SPring-8 Compact SASE Source (SCSS) test accelerator in Japan [10]. Our experimental setup employed in this experiment was almost the same as the one reported in [23,24]. Briefly, the cluster beam crossed the FEL beam at 45° in the horizontal plane. The photon energy was tuned to 20 eV (62 nm). The FEL beam was partially blocked by a 1.5-mm-wide horizontal beam stopper before the ionization region so that the unfocused beam did not irradiate the cluster beam directly. The FEL beam was reflected and focused onto the cluster beam by a multilayer focusing mirror fabricated at Lawrence Berkeley National Laboratory, which was the same one as used in [23]. Taking all of the optical elements (steering and focusing mirrors, etc.) between the radiation source and the ionization point into account, we estimated the power density at the focus to be at most $\sim 10^{14}$ W/cm^2 at the full power of the FEL. We consider this number to be an upper limit because it relies on the design values of the focusing mirror given above and thus we estimate that the uncertainty may be a factor of four, mostly due to the uncertainty of the spot size.

The Ne clusters and the Ar-core–Ne-shell clusters were prepared by adiabatic expansion of Ne pure gas and Ne-Ar premixed gas (1% or 3% Ar in Ne) through a pulsed 250 μm nozzle, respectively. The stagnation pressures in both cases were 4.6 bar, and the nozzle was cooled to 80 K using liquid nitrogen [25]. The average cluster sizes $\langle N \rangle$ are estimated to be $\sim$1000 atoms according to the well-known scaling law [26,27]. The pulsed gas jet was cut to 0.6 mm width and 0.4 mm height with knife-edge slits and traveled to the focus spot, which was located 1.7 m downstream from the nozzle. The base pressure of the interaction chamber was 8×10^{-11} torr.

We measured the time-of-flight (TOF) mass spectra (MS) and the kinetic-energy distributions (KED) of ions with our momentum imaging spectrometer [23,24]. The fragment ions were vertically extracted by a uniform electrostatic field. They traveled through an extraction field (75 V/cm in strength, 40 mm in length), followed by an acceleration region (110 V/cm in strength, 52 mm in length), and a field free drift path (308 mm), and were finally detected by a set of microchannel plate detectors equipped with a three-layer-type delay-line anode (Roentdek HEX120) [28]. The signals from the detector and an ion chamber for a laser-intensity monitor were recorded by an eight-channel digitizer (Acqiris DC282 × 2), and the timing signals were extracted by a software constant fraction discriminator (CFD) [29]. From the TOF and the detected positions, KEDs were obtained according to the method reported in [30] in which the angular-dependent detection efficiency of our spectrometer was taken into account.

In addition, TOF-MS and KED were measured for clusters ($\langle N \rangle = 1000$) produced from 3% Ar and enriched ^{22}Ne premixed gas by using weak ($\sim 10^{11}$ W/cm^2) FEL pulses with $\lambda = 51$ nm (i.e., 2.4 eV above the Ne ionization potential of 21.6 eV) to determine the Ar content within the clusters.

III. RESULTS

Figure 1(a) shows a TOF mass spectrum for Ne clusters ($\langle N \rangle = 1000$) as a function of the mass-to-charge ratio, m/q. From the Ne clusters, only singly charged Ne$^+$ ions are observed. The peak of Ne$^+$ contains sharp components centered at $m/q = 20$ and 22, as well as a broad, slightly asymmetrical peak centered at $m/q = 20$. Other peaks are due to imperfect subtraction of background signals. The sharp peaks clearly represent negligibly small kinetic energies of ions stemming from uncondensed atoms within the cluster gas jet. On the other hand, the broad peak indicates a non-negligible amount of kinetic energy. The KED of the fragment Ne$^+$ is presented in Fig. 2(a). Ne$^+$ ions have kinetic energy up to $\sim$30 eV. This implies that irradiated Ne clusters absorb a significant number of 20 eV photons, become highly charged, and subsequently dissociate into many Ne$^+$ ions, even though a single photon does not have enough energy to ionize a Ne atom.

Figure 1(b) shows an m/q spectrum for Ar-core–Ne-shell clusters with an average size $\langle N \rangle$ of 1000 produced from 1% Ar-Ne premixed gas (hereafter referred to as 1% Ar-Ne). Other mixed clusters are also denoted by the Ar concentration of premixed gas. When 1% Ar is mixed with Ne, Ar$^+$ ions are observed along with Ne$^+$ ions. The sharpness of the Ar$^+$ peak again indicates that their kinetic energy is very small. Since the Ar$^+$ peak contains significant contributions from clusters, its sharpness indicates that Ar atoms were located in the central part of the cluster, where the repulsive Coulomb forces are most efficiently canceled out and the ions hardly gain kinetic energy [31]. The Ne$^+$ peak shape of Ar-core–Ne-shell clusters from 1% Ar-Ne is similar to the one from pure Ne clusters. The KEDs of Ne$^+$ ions and Ar$^+$ ions from Ar-core–Ne-shell clusters are presented in Fig. 2(b). The KED of Ne$^+$ from core-shell clusters is slightly broader than the one from pure Ne clusters. The Ne$^+$ yields for Ne clusters and core-shell clusters (1% Ar-Ne and 3% Ar-Ne) are presented in Table I. The yield of Ne$^+$ from 1% Ar-Ne is also slightly larger than the one from pure Ne clusters.

Figure 1(c) shows an m/q spectrum and Fig. 2(c) shows the KEDs of Ne$^+$ and Ar$^+$ from core-shell clusters produced from

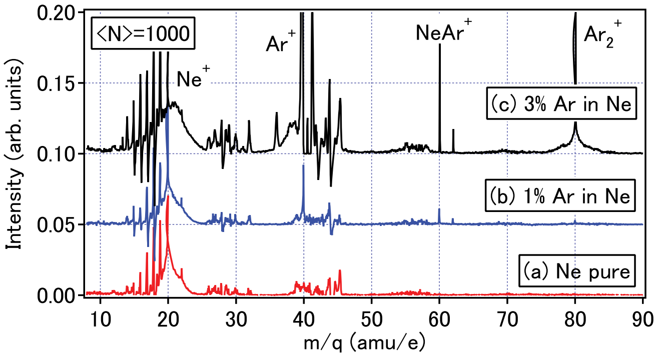

FIG. 1. (Color online) m/q spectra for $\langle N \rangle = 1000$ clusters. (a) Ne pure clusters, (b) Ar-core–Ne-shell clusters produced from 1% Ar-Ne premixed gas, and (c) Ar-core–Ne-shell clusters from 3% Ar-Ne gas. The respective backgrounds are subtracted.

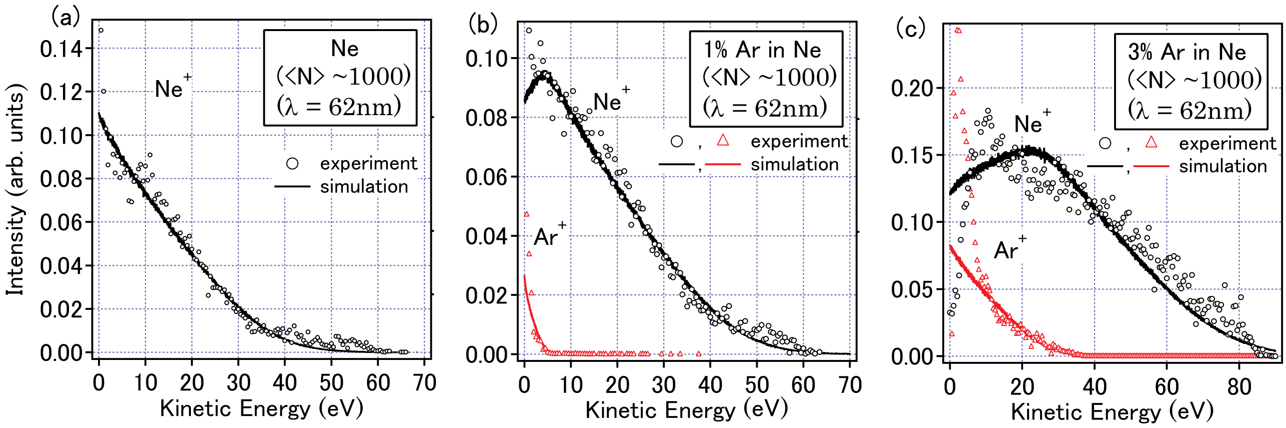

FIG. 2. (Color online) Kinetic-energy distributions of Ne^+ and Ar^+ from (a) Ne clusters, (b) core-shell clusters from 1% Ar-Ne, and (c) core-shell clusters from 3% Ar-Ne. The observed kinetic-energy spectra are shown by symbols (open circles for Ne^+ ions and red open triangles for Ar^+ ions) and the simulated kinetic-energy spectra are plotted with lines (black lines for Ne^+ ions and red lines for Ar^+).

3% Ar-Ne. Here the m/q spectrum changes dramatically. As in the case of 1% Ar-Ne, the peaks of Ne^+ and Ar^+ appear in the m/q spectrum, as shown in Fig. 1(c), but both their heights and widths become much larger. It should be noted that the Ar^+ peak due to uncondensed gas is so intense that the side toward larger m/q of the Ar^+ peak is rather distorted. Thus, only the smaller m/q was employed for further analysis. The present results indicate that both the yield and kinetic energy of Ne^+ emitted from 3% Ar-Ne are much larger than those from 1% Ar-Ne. The tendency of the number of fragment Ne^+ ions to increase with the Ar concentration (i.e., the Ar core size) indicates that a part of the charges produced in the Ar core is transferred to the Ne shell before the fragmentation. As in the case of 1% Ar-Ne, the kinetic energy of Ar^+ ions from 3% Ar-Ne is much smaller than that of Ne^+. This again indicates that the prepared Ne-Ar premixed cluster surely has the Ar-core–Ne-shell structure.

IV. DISCUSSION

The present observation that appreciable amounts of Ne^+ ions were produced by FEL photons of 20 eV, which is lower than the ionization potential of Ne, is a nontrivial result, which will be analyzed in more detail in a separate paper [32].

In this paper, we restrict ourselves to discuss the effects of adding Ar atoms to Ne clusters. Having confirmed experimentally that the charge transfer from the Ar core to the Ne shell occurs before fragmentation, we now discuss how many charges are generated in the cluster and how many charges are transferred from the Ar core to the Ne shell. For this purpose, we employed a uniformly charged sphere model [8] and extended it to core-shell clusters.

TABLE I. Ne^+ yields for Ne clusters and Ar-core–Ne-shell clusters (1% Ar-Ne and 3% Ar-Ne).

	Ne^+ yield (counts/shot)
Ne cluster ($\langle N\rangle = 1000$)	6.1
1% Ar-core–Ne-shell cluster ($\langle N\rangle = 1000$)	7.6
3% Ar-core–Ne-shell cluster ($\langle N\rangle = 1000$)	17.4

For the sake of simplicity, we treat the Ar-core–Ne-shell cluster as an ideal concentric sphere with a core radius of R_{Ar} and a total radius of R. We consider that the cluster is located at the focus point (laser power I) and that its core is uniformly charged with the total charge Q_{Ar} and its shell is charged with Q_{Ne}. The kinetic energies of Ne^+ ions and Ar^+ ions produced via Coulomb explosion on the basis of this configuration can be easily calculated within classical mechanics.

Islam *et al.* [8] proposed a method to deduce the KED of fragment ions ejected from a spherical cluster irradiated with laser power I. Assuming that the potential energy is converted to kinetic energy during the Coulomb explosion, a fragment ion located at a radius r within a cluster having the total radius R gets the kinetic energy $K(r) = \frac{Q^2e^2r^2}{NR^3}$, where Q is the total charge and N is the number of atoms within the cluster. This can be easily extended to the Ar-core–Ne-shell system: the kinetic energy of an Ar ion located at r_{Ar} within the core of radius R_{Ar}, $K_{\mathrm{Ar}}(r_{\mathrm{Ar}})$, and that of a Ne ion located at r_{Ne} in a shell extending from R_{Ar} to R, $K_{\mathrm{Ne}}(r_{\mathrm{Ne}})$, are given by

$$K_{\mathrm{Ar}}(r_{\mathrm{Ar}}) = \frac{Q_{\mathrm{Ar}}^2e^2r_{\mathrm{Ar}}^2}{N_{\mathrm{Ar}}R_{\mathrm{Ar}}^3}, \tag{1}$$

$$K_{\mathrm{Ne}}(r_{\mathrm{Ne}}) = \left(Q_{\mathrm{Ar}} + \frac{r_{\mathrm{Ne}}^3 - R_{\mathrm{Ar}}^3}{R^3 - R_{\mathrm{Ar}}^3}Q_{\mathrm{Ne}}\right)\frac{Q_{\mathrm{Ne}}e^2}{N_{\mathrm{Ne}}r_{\mathrm{Ne}}}, \tag{2}$$

where Q_{Ar} and Q_{Ne} are charges stored within the core and shell, respectively, and N_{Ar} (N_{Ne}) is the number of Ar (Ne) atoms within a cluster. By summing up $K_{\mathrm{Ar}}(r_{\mathrm{Ar}})$ in the core and $K_{\mathrm{Ne}}(r_{\mathrm{Ne}})$ in the shell, then adding the ionization potentials $E_{\mathrm{Ne}}^{\mathrm{I.P}}$ and $E_{\mathrm{Ar}}^{\mathrm{I.P}}$ of Ne and Ar atoms, the energies stored in the Ar core, E_{Ar}, and in the Ne shell, E_{Ne}, are described by

$$E_{\mathrm{Ar}}(Q_{\mathrm{Ar}},R_{\mathrm{Ar}}) = \frac{3Q_{\mathrm{Ar}}^2e^2}{5R_{\mathrm{Ar}}} + Q_{\mathrm{Ar}}E_{\mathrm{Ar}}^{\mathrm{I.P.}}, \tag{3}$$

$$\begin{aligned} &E_{\mathrm{Ne}}(Q_{\mathrm{Ne}},Q_{\mathrm{Ar}},R,R_{\mathrm{Ar}}) \\ &= \left\{\frac{3}{2}\frac{R^2 - R_{\mathrm{Ar}}^2}{R^3 - R_{\mathrm{Ar}}^3}Q_{\mathrm{Ar}}Q_{\mathrm{Ne}}e^2 + \left[\frac{3}{5}\frac{R^5 - R_{\mathrm{Ar}}^5}{(R^3 - R_{\mathrm{Ar}}^3)^2} \right.\right. \\ &\quad \left.\left. - \frac{3}{2}\frac{R^2 - R_{\mathrm{Ar}}^2}{(R^3 - R_{\mathrm{Ar}}^3)^2}R_{\mathrm{Ar}}^3\right]Q_{\mathrm{Ne}}^2e^2\right\} + Q_{\mathrm{Ne}}E_{\mathrm{Ne}}^{\mathrm{I.P.}}. \end{aligned} \tag{4}$$

TABLE II. The estimated number of charges created in the Ne clusters and the Ar-core–Ne-shell clusters (1% and 3% Ar-Ne). The listed value represents the charge number, which is obtained by a cluster with $\langle N \rangle = 1000$ at the focus point. The energy stored in the Ar core or the Ne shell is listed in parentheses. EF is the assumed enrichment factor of Ar atoms in the core.

	EF	Total charge	Charge in Ne shell	Charge in Ar core
			(stored energy)	(stored energy)
Ne cluster, $\langle N \rangle = 1000$		+48	+48 (2.1 keV)	
1% Ar in Ne, $\langle N \rangle = 1000$	1	+60	+58 (2.9 keV)	+1.6 (0.029 keV)
	2	+60	+58 (2.9 keV)	+1.9 (0.035 keV)
	3	+60	+58 (2.9 keV)	+1.9 (0.034 keV)
	4	+60	+58 (2.8 keV)	+2.0 (0.036 keV)
3% Ar in Ne, $\langle N \rangle = 1000$	1	+100	+87 (6.5 keV)	+13 (0.44 keV)
	2	+102	+89 (6.7 keV)	+13 (0.38 keV)
	3	+104	+89 (6.8 keV)	+15 (0.43 keV)
	4	+107	+91 (6.9 keV)	+16 (0.44 keV)

So far we have assumed that the cluster is located at the focus point; however, to reproduce the experimentally measured KED, we need to take the spatial distribution of the laser power and the cluster size distribution into account. Assuming that the absorbed energy and hence the stored energy is proportional to the laser power and the cluster size, the energies stored in the Ar core and in the Ne shell for a different laser power I' and cluster size $R', R'_{\rm Ar}$ conditions scale as follows:

$$E_{\rm Ar}(Q'_{\rm Ar}, R'_{\rm Ar}) = E_{\rm Ar}(Q_{\rm Ar}, R_{\rm Ar}) \frac{I' R'^3}{I R^3}, \tag{5}$$

$$E_{\rm Ne}(Q'_{\rm Ne}, Q'_{\rm Ar}, R', R'_{\rm Ar}) = E_{\rm Ne}(Q_{\rm Ne}, Q_{\rm Ar}, R, R_{\rm Ar}) \frac{I' R'^3}{I R^3}. \tag{6}$$

With Eqs. (3)–(6), $Q'_{\rm Ar}$ and $Q'_{\rm Ne}$ can be derived for the different laser powers and cluster sizes. Namely, assuming a Gaussian shape for the spatial laser power profile and a log normal distribution for the cluster size [8], the KEDs are simulated by a Monte Carlo method. We then fit the model curves to the experimental KEDs, regarding Q_{Ar} and $Q_{\rm Ne}$ as fitting parameters.

The results are presented in Fig. 2 by the lines. The KEDs of $\rm Ne^+$ and $\rm Ar^+$ emitted from Ne clusters and the 1% Ar-Ne clusters are well described by our model. For the 3% Ar-Ne clusters, however, the experimental results are not perfectly reproduced by the uniformly charged model. The excess yield of $\rm Ne^+$ at high energies may be a precursor of the inhomogeneous charge redistribution to minimize the total potential energy that was observed for pristine Xe [21] and Ar clusters [33] under intensive EUV-FEL irradiation. In addition, an oscillatory behavior at lower energies might be associated with the existence of Ar-Ne interface (i.e., interfacial charges due to the discontinuity of dielectric permittivity). Considering these uncertainties, we treat only the total charges in the following discussion.

The estimated numbers of charges created in the Ne and Ar-core–Ne-shell clusters, respectively, are listed in Table II, in addition to the energies stored in the Ar core and Ne shell. The amount of charge transfer from the Ar core to the Ne shell is estimated by the difference between the number of the charge obtained by the Ne shell and the one obtained by the pure Ne cluster. It is ~10 for 1% Ar-Ne and ~39 for 3% Ar-Ne.

Before drawing conclusions, one should examine Ar enrichment effects in the core [22]. For this purpose, the Ar enrichment factor for 3% Ar-Ne was directly determined: We produced Ar-core–Ne-shell clusters ($\langle N \rangle = 1000$) from 3% Ar and enriched (more than 99%) ^{22}Ne premixed gas, and measured TOF-MS and KED with 24.3 eV FEL pulses, which ionize not only Ar but also Ne atoms by one-photon absorption. Nonlinear optical phenomena were avoided by keeping the FEL intensity weak ($\sim 10^{11}$ W/cm^2). The reason for adopting the enriched ^{22}Ne isotope is to eliminate the effect of detecting $(\rm H_2O)^+$ ions, which is unavoidable when natural abundant Ne containing more than 90% ^{20}Ne is used. As seen in Fig. 3, the ^{22}Ne ion signals are well separated from a large $(\rm H_2O)^+$ peak. The Ar enrichment factor η is estimated to be 2.5 ± 0.2, which was deduced from the integrated intensity ratio of Ar to ^{22}Ne atoms and their ionization cross sections [34]. This result is smaller than the one ($\eta = 4$) determined by Lundwall *et al.* [22] from the Ne 2s and the Ar 2$p_{3/2}$ x-ray photoelectron spectroscopy (XPS) spectra. In Table II, the estimated results assuming the enrichment factors between 1 and 4 are also listed. Both the charges and stored energies do not significantly depend on the enrichment factor.

Finally, we estimate how much energy is transferred from the Ar core to the Ne shell. We assume that some part of the energy initially created in the Ar core migrates to the Ne

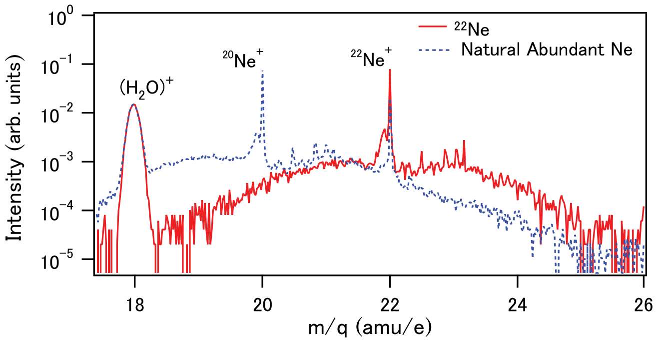

FIG. 3. (Color online) TOF spectrum for Ar-core–Ne-shell clusters ($\langle N \rangle = 1000$) produced from enriched ^{22}Ne (solid line) is compared with that produced from natural abundant Ne (dotted line). The Ar content in the premixed gas was 3% in both cases, and the spectra were recorded at 24.3 eV with weak FEL intensity ($\sim 10^{11}$ W/cm^2). The ^{22}Ne ion signals are well separated from a large peak due to $(\rm H_2O)^+$.

core and the rest remains in the core. The latter is directly obtained from the experiments, and the former is estimated as a difference between the average energy stored in the Ne core of a core-shell cluster and that in a pristine Ne cluster. Hence we obtain that about 0.03 keV is left in the Ar core and 0.8 keV (=2.9–2.1 keV) is transferred to the Ne shell in the 1% Ar-Ne clusters, whereas 0.44 keV remains in the Ar core and more than 4.4 keV is transferred to the Ne shell in the 3% Ar-Ne clusters. This result indicates that more than 90% of the energy absorbed by the Ar core is transmitted to the Ne shell.

V. CONCLUSION

We have investigated the fragmentation dynamics of pristine Ne clusters and heterogeneous Ar-core–Ne-shell clusters irradiated by intense EUV pulses at 62 nm. We found that the yield and the kinetic energy of fragment Ne^+ ions increase with the Ar core size. This result clearly indicates that the charges created in the Ar core are transferred to the Ne shell. The KEDs of Ne^+ and Ar^+ obtained in a uniformly charged sphere model show overall agreement with the experiment. Charge and energy transfer were estimated, and the results support the advantage of the core-shell structure for weakening the Coulomb explosion of laser-irradiated cluster cores. The current finding may be of great importance with respect to suggestions to delay and reduce the Coulomb explosion of biomolecules by embedding them in a tamper [35–40] for future x-ray-diffraction imaging experiments using x-ray lasers.

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