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

The recent advent of self-amplified spontaneous emission (SASE) free electron laser (FEL) light sources has opened new research fields on the interaction of intense short-wavelength laser pulses with matter [1-3]. Atomic clusters are ideal objects for such studies, not only because their size can be varied from a single atom to a bulk-like macroscopic object, but also because there is no energy dissipation into the surrounding medium. Pioneering works on clusters have revealed characteristic ionization dynamics induced by intense FEL pulses, such as sequential photoabsorption [4], charge recombination [5,6], and inhomogeneous charge redistribution [6,7].

A deeper insight into the spatial origin of the charge distributions can be obtained from core-shell heterogeneous systems. An Ar-Ne cluster is known to be self-assembled to a core-shell structure [8]. We have carried out experimental studies on Ar core-Ne shell clusters irradiated by intense 62 nm (20 eV) pulses from a free electron laser [9]. 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. This type of study may be of great importance with respect to suggestions to delay and reduce the Coulomb explosion of bio-molecules by embedding them in a tamper [10,11] for future X-ray diffraction imaging experiments using X-ray lasers.

Experiments were performed at the SPring-8 compact SASE source (SCSS) test accelerator in Japan [2]. Figure 1 is a schematic of the experimental setup employed in this experiment [12]. Briefly, the FEL beam was back-focused onto the cluster beam by a multi-layer focusing mirror. The FEL beam was partially blocked by a beam stopper before the ionization region so that the unfocused beam did not irradiate the cluster beam directly. The estimated power density at the focus was ~10^{14} W/cm^2 at the full power of the FEL in the present experiment. The Ar core-Ne shell clusters were prepared by adiabatic expansion of Ne-Ar premixed gas (1% and 3% Ar in Ne) through a pulsed 250 μm nozzle. The stagnation pressures and the nozzle temperature were adjusted to control cluster size. We measured the time-of-flight mass spectra and the kinetic energy distributions (KED) of ions with our momentum imaging spectrometer [12].

The most representative results of the present experiments were the Ar concentration dependence of KED. As shown in Fig. 2, the fragment Ne^+ from the 1% Ar-Ne cluster has a kinetic energy up to ~60 eV, whereas those from the 3% Ar-Ne cluster have much higher energies (up to ~90 eV). In addition, we found a clear tendency that the number of fragment Ne^+ ions increases with Ar concentration (i.e., Ar core size). These results indicate that a portion of the charges produced in the Ar core are transferred to the Ne shell before the fragmentation. The kinetic energy of Ar^+ ions was much smaller than that of Ne^+ ions. This indicates that the prepared Ne-Ar premixed cluster surely has the Ar core-Ne shell structure.

We have estimated the number of transferred charges and energies from Ar to Ne within a core-shell cluster by assuming a uniformly charged sphere model [9]. Here, we simply assume the Ar core-Ne shell cluster as an ideal concentric sphere, and that its core is uniformly charged with the total charge Q_{Ar} and its shell with Q_{Ne}. The kinetic energies of Ne^+ ions and Ar^+ ions produced via Coulomb explosion can be easily calculated within classical mechanics.

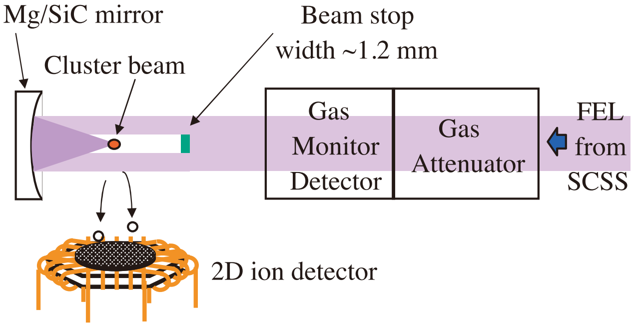

Fig. 1. Schematic illustration of the experimental setup.

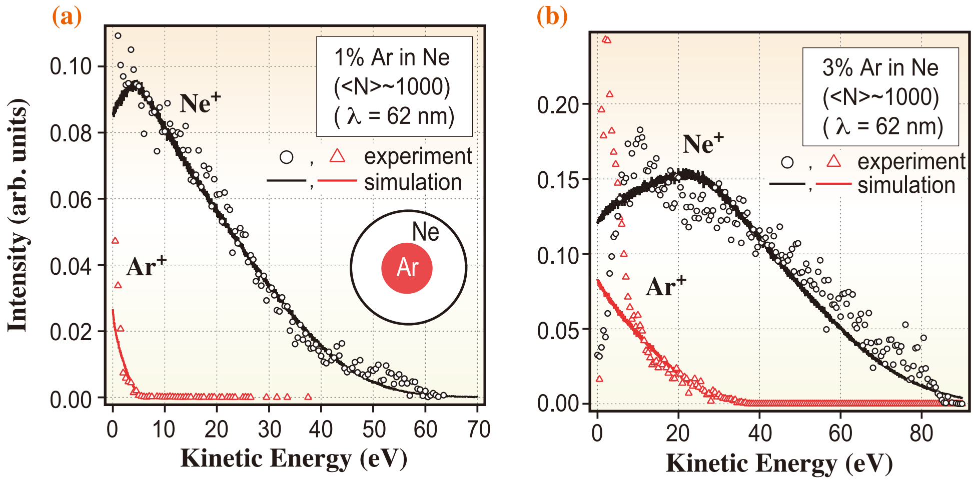

Fig. 2. Kinetic energy distributions of Ne^+ and Ar^+ ions from **(a)** core-shell clusters from 1% Ar-Ne and **(b)** core-shell clusters from 3% Ar-Ne. The observed kinetic energy spectra are shown by symbols (open circles for Ne^+ ions and open red triangles for Ar^+ ions) and the simulated kinetic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

The estimated numbers of charges created in the Ne and Ar core-Ne shell clusters, are listed in Table 1, in addition to the energies stored in the Ar core and Ne shell. The number of charges transferred from the Ar core to the Ne shell is estimated by the difference between the number of charges obtained by the Ne shell and that obtained by the pure Ne cluster. This is ~10 for 1% Ar-Ne and ~39 for 3% Ar-Ne. For the estimation of transferred energy, we assume that some part of the energy initially created in the Ar core migrates to the Ne core and the rest remains in the core. The latter is directly obtained from the experiments, and the former is estimated as the difference between the average energy stored in the Ne core of a core-shell cluster and that in a pristine Ne cluster. Hence we determine 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. These results indicate that more than 90% of the energy absorbed by the Ar core is transmitted to the Ne shell. The present results strongly support the usefulness of a tamper [10,11]. Embedding biomolecules in a tamper would make it possible to reduce the influence of Coulomb explosion during the X-ray diffraction experiments using X-ray lasers.

	Total Charge	Charge in Ne Shell (stored energy)	Charge in Ar Core (stored energy)
Ne cluster	+48	+48 (2.1 keV)	
1% Ar in Ne	+60	+58 (2.9 keV)	+1.6 (0.029 keV)
3% Ar in Ne	+100	+87 (6.5 keV)	+13 (0.44 keV)

Table 1. 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 from a cluster with **<Ni>** = 1000 at the focus point. The energy stored in the Ar core or the Ne shell is listed in parentheses.

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