Surface plasma resonance in Xe clusters studied by EUV pump-NIR probe experiments

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Abstract

The ionization dynamics of Xe cluster was analyzed by extreme ultraviolet (EUV) pump-near infrared (NIR) probe experiments. The experiments were carried out for Xe clusters containing around 5000 atoms on average at the SPring-8 Compact SASE Source (SCSS) test accelerator in Japan. We recorded ion spectra emitted from Xe clusters as a function of the time delay between the EUV and NIR pulses. The ion yields up to highly-charged states were substantially enhanced when Xe clusters excited by the EUV pulses were illuminated by the NIR laser pulses at 1 to 2 ps after the pump pulse, providing a compelling evidence for nano-plasma formation and subsequent NIR absorption via surface plasma resonance. We also observed emergence of low kinetic energy Xe+ at long time delay, which can be attributed to the ionization of excited neutral atoms resulting from the electron-ion recombination in the expanding nano-plasma.

Recent developments of laser technology give us opportunities to study ultra-fast processes during the laser-matter interaction. Atomic clusters produced in vacuum are ideal objects to study electronic and ionic dynamics triggered by an intense laser pulse, because energy dissipation into a surrounding medium is negligibly small [1]. When a rare-gas cluster is exposed to an intense infrared (IR) laser pulse, the strong electric field strips electrons from atoms by tunneling ionization and subsequent collisional ionization, and as a result, the cluster transforms to a nano-meter scale plasma, called nano-plasma, in which the quasi-free electrons are trapped by the Coulombic potential of the cluster ion [2, 3]. The nano-plasma formation was intensively studied in the IR spectral regime where various interesting phenomena such as the ejection of keV electrons [4], MeV ions [5] as well as x-ray photons in the keV energy range [6, 7] were observed when clusters were exposed to an intense laser pulse. Pioneering work on intense IR laser - cluster interactions [8] revealed that the hydrodynamic expansion model gives good insight for the dynamics of nano-plasma. After the nano-plasma formation, it starts to expand/disintegrate rapidly due to the internal pressure of nano-plasma, therefore the information on the temporal evolution of nano-plasma is important to understand the dynamics induced by irradiating intense laser pulses. Zweiback et al. [9] carried out an IR pump-IR probe experiment for Xe clusters, in which they measured the absorption of the probe radiation as a function of the pump-probe time delay and found a maximum absorption at 1 to 2 ps after the pump laser illuminated to clusters. They interpreted the data with the help of the Mie resonance scattering theory [10], and found that the cluster can obtain enormous energy from the radiation fields of the IR laser when the created nano-plasma meets the condition of Mie-resonance on the way of their expansion.

When a rare-gas cluster is irradiated by an extreme ultraviolet (EUV) free electron laser (FEL) pulse, the cluster becomes highly ionized by sequential photoionizations of constituent atoms [11, 12], and photoelectrons...
are trapped by the strong Coulombic potential of the cluster ion. As a result, nano-plasma can be formed by the irradiation of the intense EUV-FEL pulses. The first experiment in the vacuum-ultraviolet (VUV) regime (12.6 eV) revealed that unexpectedly large amounts of energy were absorbed in Xe clusters [13]. Ultrafast ionization dynamics and electron emission were studied in the extreme ultraviolet regime (90 eV) [14]. The formation of Xe nano-plasma was also confirmed in the soft x-ray regime (850 eV) [15]. An experiment at 24 eV utilizing the EUV-FEL facility showed that inhomogeneous charge redistribution takes place in a self-organized manner before Coulomb explosion [16].

Although the ionization mechanism depends crucially on the laser wavelength, nano-plasma is generated in common when some of electrons that are ionized from individual atoms (inner ionization) remain within the cluster as quasi-free electrons [17]. For the deeper insight to the dynamics of EUV-FEL induced nano-plasma, theorists [18] recommended a EUV pump-IR probe experiment as an elegant way to study the dynamics of collectively excited electrons. When the frequency of collective motion of quasi-free electrons in the cluster coincide with the IR laser frequency, resonant energy absorption due to surface plasma resonance, which is mathematically equivalent to the Mie resonance, occurs. Because EUV and IR radiations would induce photoionization and surface plasma resonance, respectively, the EUV pump-IR probe scheme could provide opportunity to trace the dynamics of nano-plasma by varying the time delay between the pulses.

On the other hand, Schütte et al [19] performed the EUV pump-IR probe experiments on Ar and Ar-Xe clusters by using weak EUV pulses with the intensity of $2 \times 10^{12}$ W cm$^{-2}$, in which they mainly traced electron-ion recombination in nano-plasma. They pointed out the importance of recombination in expanding nano-plasma and revealed that the expanding nano-plasma can absorb energy from IR laser pulse even when the condition of surface plasma resonance is not satisfied. Here the IR pulse ionize the Rydberg atoms generated via the recombination.

In this work we carried out a series of EUV pump-IR probe experiments for Xe clusters with an average size of 5000. The plasma density in the parent clusters was controlled by varying the EUV intensity. We adopted the FEL intensity of $\sim 10^{15}$ W cm$^{-2}$, where we expect the creation of dense nano-plasma sufficient to cause surface plasma resonance. TOF spectra of daughter ions were measured at various time delays from $-50$ ps to $+100$ ps. From the TOF spectra not only the total ion yield (TII) but also the partial ion yields (PIYs) of the ions with the charge $q$ from 1 to 12 are deduced as a function of the time delay. We found that the experimental results provide the evidence of surface plasma resonance in EUV pump-IR probe measurements proposed by the theoretical work [18]. The experimental results are interpreted by the combination of surface plasma resonance and electron-ion recombination in expanding nano-plasma.

### 1. Experiment

The EUV pump-IR probe experiments on Xe clusters were performed at the SPring-8 Compact SASE Source (SCSS) test accelerator in Japan [20]. EUV-FEL with the photon energy of 24.3 eV and NIR laser with 1.55 eV were injected, nearly parallel to each other, into a spectrometer chamber [21], where the lasers met the cluster beam at a right angle and the time-of-flight (TOF) of Xe ions from photo-excited clusters were measured using TOF mass spectrometer equipped with a microchannel plates and a phosphor screen. The pulse widths for both lasers were 30 fs. The focal sizes were 13 μm for EUV and 500 μm for NIR. The intensity of EUV pulses was measured on a shot by shot basis, giving the mode FEL intensity of $1.1 \times 10^{14}$ W cm$^{-2}$. The intensity of NIR laser was adjusted to about $1.0 \times 10^{13}$ W cm$^{-2}$ so that Xe clusters could not be ionized solely by the NIR pulses. The maximum time jitter between the two pulses was ±0.5 ps [22]. Further experimental details about the FEL beam line was described elsewhere [23].

Before the pump-probe experiments, spatial and temporal overlap between the EUV and IR laser pulses was achieved. For the spatial overlapping, a trace amount of Xe gas was introduced into the chamber, where Xe ions generated along EUV and non-attenuated NIR laser beams were detected as images on a phosphor screen using the spatial focusing mode of the spectrometer. Then the orientation of NIR laser beam was adjusted to the best focusing position. For the temporal overlapping helium gas was used because He atoms with the ionization potential $I_p$ of 24.6 eV cannot be ionized unless irradiated by both EUV pulses with 24.3 eV and NIR pulses with 1.55 eV [24]. We measured He ion yield as a function of the time delay of NIR pulse relative to the EUV pulse, and identified the time-zero as a step-wise increase of the yield.

Xe clusters were produced by pulsed supersonic expansion through a pinhole, 250 μm in diameter, kept at room temperature [25]. The stagnation pressure was 7 bar so that the average cluster size $\langle N \rangle$ was adjusted to be about 5000 atoms according to well-known scaling laws [26, 27].
2. Results and discussions

Figure 1(a) (the bottom panel) shows the ion TOF spectrum for the Xe clusters irradiated only by EUV pulses. Sharp peaks of atomic Xe ions coming from uncondensed atoms in gas jet are visible up to Xe$^{6+}$. Fine structures are assigned to Xe isotopes. Broad peaks presumably due to the clusters are seen up to Xe$^{4+}$. The broad peaks indicate that the Xe fragment ions obtained considerable kinetic energy through the Coulomb explosion or hydrodynamic expansion [8]. Each peak is substantially broadened and splits into two limbs depending on the initial ejection direction. Energetic ions ejected toward the direction opposite to the spectrometer were not fully detected owing to the limited acceptance angles determined by the spectrometer geometry and the voltage settings. For example, the longer time limb of energetic Xe$^{2+}$ suffers from this problem, resulting in a cut-off near 14.5 μs. Decomposition of TOF spectra into each charge state will be discussed later. Other peaks are due to residual gases such as H$_2$O.

In figure 1(b) TOF spectra taken in the EUV pump-NIR probe measurements are depicted at various NIR delays. Dramatic increases in the ion yields and the maximum ion charges are observed at the delays from 2 to 10 ps, which indicates tremendous energy conversion from the laser fields to the clusters presumably due to the surface plasma resonance.

Here we give rough estimate of the conditions for surface plasma resonance. We expect the surface plasma resonance when the IR laser frequency, $\omega_{\text{laser}} = 800$ nm, coincide with the surface plasma frequency $\Omega_{\text{pl}}$. $\Omega_{\text{pl}}$ is given by the $\Omega_{\text{pl}} = \sqrt{e^2 n Z / \varepsilon_0 m_e} = \omega_{\text{pl}} / \sqrt{3}$, where $e$ is the elementary charge, $n$ is the number density of atoms or ions in the cluster, $Z$ is their average charge, $\varepsilon_0$ is the vacuum permittivity, $m_e$ is electron mass and $\omega_{\text{pl}}$ is bulk plasma frequency [18]. When we adopt the $1.4 \times 10^{22}$ cm$^{-3}$ for the atomic number density in Xe cluster, surface plasma resonance at $\omega_{\text{laser}} = 800$ nm is satisfied for $Z \sim 0.4$. Considering photoabsorption cross section of $\sim 20$ Mb at $h\nu = 24$ eV [28], the FEL intensity that satisfies the surface plasma resonance is estimated about $\sim 1 \times 10^{12}$ W cm$^{-2}$. The current FEL intensities are well above this intensity. As a result, the electron density is reduced via the nano-plasma expansion, it eventually meet the resonant condition. Therefore, we safely conclude that the observed enhancement of ion yield can be attributed to the influence of surface plasma resonance.

Total ion yield (TIY) estimated by integrating each TOF spectrum is plotted by red dots as a function of the NIR time delay $t$ in the inset of figure 2. For comparison TIY without NIR pulses is indicated by the green dashed line. The TIY with NIR pulses rises sharply at $t = 0$ followed by a gradual decrease at later times. The TIY decay is appreciably slower than that in the previous study where NIR lasers were used both for pump and probe pulses [9]. Figure 2 (the main panel) shows TIY against $t$ at various FEL intensities, in which shot-to-shot fluctuations of FEL intensity were used to derive FEL intensity dependence. The TIY peak position slightly shifts toward larger $t$. 
with decreasing $I$. The peak positions of TIY correspond to the time of the resonance where $\Omega_{pl} = \omega_{laser}$. The shifts of the peak position can be interpreted by the difference of the expansion time for surface plasma resonance condition. The present results revealed that the time becomes shorter as the FEL intensity increases. It is noted that the time for the resonance depends on the initial plasma density and expansion speed. One can expect that weak FEL intensity results in the shorter time for plasma resonance via low initial plasma density, if the initial plasma density mainly plays a role for the resonance conditions. Therefore, the expansion speed of nano-plasma is more important for interpretation of the temporal dependence of ion yields. Here we focus the hydrodynamic expansion of nano-plasma triggered by the intense EUV-FEL pulses, whose speed depends on the FEL intensity. From the hydrodynamic expansion model \cite{8}, the expansion speed, $v_{ex}$, of nano-plasma can be expressed as 

$$v_{ex} = \frac{m_{q} v_{q}}{2 \sigma_{q}^2} \times f_q(v).$$

(1) 

Here $m_q$ is the ion mass, $v_q$ is the mean velocity and $\sigma_q$ is the Gaussian width for a charge state $q$. The function $f_q(v)$ is needed to incorporate the geometrical limitation of accepting ion signals, as follows:

$$f_q(v) = \frac{1}{2} \left[ 1 + \text{erf} \left( \frac{m_{q} v}{2 \sigma_{q}^2} \frac{v^2 - v_{q,\text{cut}}^2}{2\sigma_{q,\text{cut}}^2} \right) \right].$$

(2) 

Here $v_{q,\text{cut}}$ is the cut-off velocity and $\sigma_{\text{cut}}$ represents the sharpness of the error function. Then, by using a usual linear relation between the time-of-flight $T$ and $v$ (i.e. $T = a_q + b_q v$), equation (1) was converted to a function of $T$ for each $q$. Note that simple kinematics leads to $a_q \propto q$ and $b_q \propto q^{-1}$. The applicability of equations (1) and (2) was confirmed by using a simulation code SIMION \cite{29}. In figure 3(a), we illustrate how to decompose a TOF spectrum into partial $q$-contributions. The experimental TOF spectrum recorded at $t = +2$ ps was excellently reproduced by a summation of the velocity distributions up to $q = 18$, where we have made further

![Figure 2. (Inset) Total ion yield (TIY) estimated by integrating each TOF spectrum is plotted by red dots as a function of the NIR time delay $t$. (Main panel) TIY versus $t$ at various FEL intensities. The lines are the guide for the eyes.](image-url)
assumptions: the velocity perpendicular to the spectrometer axis was ignored because of the small acceptance angle, and the peak positions of highly charged ions \((q \geq 5)\) were evaluated by extrapolating those for \(q = 1\) to 4.

Figure 3(b) displays the most probable kinetic energy (MPKE) of daughter ions derived from fitting of TOF spectrum for \(t = +2\) ps.

The results of PIY are shown in figure 4(a) for \(q = 1\) to 5, and in figure 4(b) for \(q = 6\) to 12 as a function of the NIR time delay. The intensity of the irradiated FEL pulses is \(1.1 \times 10^{14} \text{W cm}^{-2}\). The lines are guides for the eye.

The results reveal that not only the peak height but also the width (i.e. decay time \(\tau_q\)) decrease rapidly with increasing \(q\). Typically, singly charged Xe\(^+\) ions survive beyond 30 ps, while highly charged Xe\(^{12+}\) ions are generated only within a few picoseconds. NIR delay dependence of PIY suggests the creation of highly charged ions as a result of surface plasma resonance in expanding nano-plasma. It is well known that the surface plasma resonance donates huge energy from the laser field to nano-plasma when the surface plasma frequency is equal to the laser frequency [8, 18]. As a result of the resonant heating, we can expect creation of highly charged ions, as well as the emission of energetic ions by subsequent explosion of the heated nano-plasma. Emission of energetic highly charged ions will be quickly suppressed at the long time delay where the nano-plasma is no more resonant conditions.

On the other hand, we observed apparent enhancement of Xe\(^+\) ion at long delay (\(\geq 50\) ps). Figure 5 shows the comparison of the TOF spectra displayed in enlarged scale. It revealed that the low energy part (i.e. the central part of Xe\(^+\) peak in the TOF spectrum, kinetic energy is less than 20 eV) is enhanced in the TOF spectrum at \(+50\) ps, whereas the energetic ions (estimated MPKE is \(\sim 40\) eV) are emitted at the shorter time delays (\(t = 10\) ps and...
The singly charged Xe ions observed at the long time delay can be attributed to the reionization of excited atoms created by recombination (REAR) in nano-plasma, as pointed out by Schütte et al [19], who carried out the XUV pump-NIR probe experiments on Ar and Ar-Xe clusters. The recombination in nano-plasma occurs in the slowly expanding plasma and therefore REAR results in the emission of low kinetic energy Xe$^+$. In the time delay of 10 ps and 20 ps, we see the enhancement of energetic Xe$^+$ as well as the low energy Xe$^+$. Therefore we cannot exclude the possibility that the surface plasma resonance also enhances the low energy Xe$^+$. It is difficult to analyze the present data using a simple model such as expanding uniform nano-plasma model, etc., because both the surface plasma resonance and REAR may contribute to the creation of low-energy Xe$^+$ ions. Here, we restrict ourselves to point out that our results may be understood by the combination of surface plasma resonance and the electron-ion recombination, and the former is dominant at the short time delay whereas the latter is dominant at long time delay.

The observed phenomena, surface plasma resonance [8, 18] and electron-ion recombination [19], have been known by the preceding studies and are expected to occur regardless of the wavelength of the pump laser. We observed that during dynamics in nano-plasma created by intense EUV laser pulse the surface plasma resonance has the main contribution to the enhancement of ion yields via the energy deposition to nano-plasmas from the IR probe laser at an early time, and the creation of highly excited ions and neutral atoms via electron-ion recombination has the main contribution to the creation of ions via photoionization by IR probe at a late time, although the two overlap temporally.

The experimental fact that the electron-ion recombination in nano-plasma results in the formation of excited atoms even a few tens ps after nano-plasma formation, it is worth noting also that ion signals coming from the recombination are easy to detect even though the spatial and temporal overlap between the pump and probe lasers is insufficient. Considering that the recombination effect emerges in nano-plasma created by the pump laser with weak intensity [19], enhancement of ion signals from nano-plasma by the IR probe can be expected to be observed widely in both spatial and temporal regimes in terms of the overlap between the two lasers. In addition, the enhancement of highly excited ion signals becomes stronger as the spatial and temporal overlap gets closer to the perfect condition of the surface plasma resonance. A combination of these points allows us to achieve on-the-fly adjustment of the spatial and temporal overlap between the pump and probe lasers by maximizing the signals of highly charged ions as monitoring recombination signals. From the technical point of view, therefore, ion enhancement signals in these phenomena are useful to make spatial and temporal overlap between two lasers during experiments.

3. Summary

EUV pump-NIR probe experiments were carried out on Xe clusters with the average cluster size around 5000 atoms. In the short time delay region, the ion yields up to Xe$^{18+}$ were substantially enhanced when Xe clusters excited by EUV pulses were illuminated by NIR laser pulses. We also observed enhancement of low-kinetic energy Xe$^+$ at the long time delay. The present results are understood well by the combination of surface plasma resonance and electron-ion recombination, in which the former mainly contributes at an early time delay whereas the latter contributes the creation of ions at a late time.
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