Enhancement of soft x-ray emission from a cryogenically cooled Ar gas jet irradiated by 25 fs laser pulse

Department of Physics, Korea Advanced Institute of Science and Technology (KAIST), 373-1 Kusong-dong, Yusong-gu, Taejon 305-701, Korea

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Soft x-ray spectra (40–180 Å) produced by the interaction of 25 fs laser pulses at an intensity of \( \sim 7 \times 10^{16} \) W/cm\(^2\) with a cryogenically cooled Ar gas jet have been measured. New spectral lines from Ar\(^{8+}\) and Ar\(^{9+}\) charge states appeared with decreasing gas temperature. Nonlinear increase of x-ray line emission from Ar\(^{7+}\), Ar\(^{8+}\), and Ar\(^{9+}\) was observed with cooling, which saturated below certain temperature. The drastic change in the spectrum is attributed to efficient collisional heating and collisional ionization of growing (\(10^2–10^3\) atoms) Ar clusters from the cooled jet.

In the past few years there has been much interest in understanding the interaction of femtosecond, high intensity laser pulses with van der Waals bonded atomic clusters.\(^1\)–\(^5\) The main difference between a gaseous cluster composed of a large number of atoms and the other forms of matter is the combination of high local density inside the cluster and low average density. Several theoretical models have been proposed to describe the laser-cluster interaction.\(^1\)–\(^3\) Recently, the hydrodynamic model developed by Ditmire \(et al.\)\(^2\) is being recognized due to the good agreement with experimental results.\(^4\),\(^6\) In this approximation each cluster was considered as a small ball of high density plasma and the main processes involved in the interaction were optical field ionization (OFI), heating through inverse bremsstrahlung, collisional ionization by thermal and laser-driven electrons, and cluster expansion. Here the short (subpicosecond) pulse duration of a driving laser was essential for an efficient transfer of laser energy into plasma electrons on a time scale shorter than the disassembly time of cluster. Previous studies\(^4\),\(^6\) also pointed out the importance of resonance absorption for efficient deposition of laser energy into clusters. If the laser pulse is longer than the time for the cluster to expand to the resonance condition \((n_e = 3 n_c)\), where \(n_e\) is the electron density and \(n_c\) is the critical density) where the oscillating laser field resonantly drives the free electrons in the cluster, collisional heating can be greatly enhanced. When using an extremely short pulse, however, resonance absorption is expected to be marginal since by the time it takes to reach the resonance condition, the laser pulse is past. Even in this case the interaction of an ultrashort laser pulse with a near-solid-density cluster can still be efficient due to collisional processes. To date, laser pulses typically longer than 100 fs have been used to irradiate clusters of noble gases while no experimental attempt with pulses shorter than 50 fs has been reported yet.

Experimental studies have shown that laser-irradiated clusters could be a source of strong x-ray radiation ranging from extreme ultraviolet (XUV) up to hard x-ray emission.\(^1\),\(^2\),\(^7\) Though the x-ray emission from clusters has been investigated by several groups, most experiments have been performed at room temperature using high pressure valves and just a few data exist for cooled targets.\(^7\),\(^8\) No experimental study of argon cluster plasma using cryogenic gas target has been done to date.

In this letter we examine experimentally the soft x-ray emission from a cryogenically cooled Ar gas jet irradiated by intense femtosecond laser pulses and report significant enhancement of x-ray line emission with increasing cluster size. Our data show that collisional heating via inverse bremsstrahlung can still play a crucial role in the interaction of laser pulses as short as 25 fs with clusters.

The experiment was performed with a Ti:sapphire laser delivering 25 fs, 35 mJ pulses at a fundamental wavelength of 810 nm. The laser is based on the chirped pulse amplification technique coupled with the long wavelength injection method for broad amplified spectrum.\(^9\) To achieve a high contrast ratio, a Bragg cell was installed and the Ti:sapphire oscillator was operated in a cavity-dumped mode. The prepulse to main pulse ratio less than 10\(^{-3}\) has been measured. The linearly polarized laser beam was focused with a \(f = 45\) cm spherical mirror, yielding a peak intensity of up to about \(7 \times 10^{16}\) W/cm\(^2\). The laser illuminated the gas jet transversely with respect to the flow of the gas and the laser focus was placed \(\sim 0.7\) mm above the nozzle tip. The gas jet was produced by a solenoid-driven pulsed valve fitted with a 200 \(\mu\)m diameter circular nozzle. The gas was cooled by passing through a reservoir filled with cold nitrogen gas which was forced under pressure from a liquid nitrogen bath. The nozzle was directly attached to the reservoir providing additional cooling. The temperature just before the solenoid valve was measured with a thermocouple inserted into the copper pipe carrying Ar gas.

The onset of clustering in an expanding gas jet can be estimated with the help of an empirical parameter \(\Gamma^*\) scaling as \(p_0/T_0^{2.29}\), where \(p_0\) is the backing pressure (mbar) and \(T_0\) the pre-expansion gas temperature (K).\(^{10,11}\) The number of atoms per cluster \(N_c\) scales as \(\Gamma^*^{2.0–2.5}\) and published data indicate that clusters composed of about 100 atoms can be formed when \(\Gamma^* > 10^3\) depending on particular valve and experimental conditions.\(^1,11,12\) Therefore the onset of clustering for a given valve has to be determined experimentally, for example by means of Rayleigh scattering where \(N_c \approx 100\) is generally considered as a threshold for observing
optical scattering.\textsuperscript{4,13} In our experiment, $\Gamma^*$ varied from $\sim 3 \times 10^7$ up to $\sim 1 \times 10^9$. The presence of clusters in the cooled jet was confirmed with the help of the Rayleigh scattering technique. The gas jet was irradiated by a beam of a frequency-doubled Nd:yttrium–aluminium–garnet laser (10 ns) focused with $f = 40$ cm lens. The side-scattered light was imaged on a photomultiplier with $f = 2.5$ cm lens. For 10 bar of Ar we observed a weak signal above the noise level around $-15 ^\circ C$, and with decreasing temperature the signal showed a non-linear increase. By performing pressure scan at low temperatures we found that the scattered signal does vary as $p^3$, which is in good agreement with published data.\textsuperscript{13} From our measurements we inferred that the average cluster radius ranges from $9 \pm 2 \text{ Å}$ ($N_c \sim 90 \text{ at } -13 ^\circ C$) up to $20 \pm 4 \text{ Å}$ ($N_c \sim 930 \text{ at } -100 ^\circ C$). The error in the estimate of $N_c$ is about $\pm 50\%$.

Assuming adiabatic expansion of cluster plasma the lower bound for the time necessary to expand from near solid density to $3n_c$ may be assessed. Considering that the initial temperature for our experimental conditions is to the order of hundreds of electron volts,\textsuperscript{2} an initial electron temperature of 1 keV implies the expansion time of about 50 fs for an initial cluster radius of 20 Å, while it amounts to 25 fs for a 10 Å cluster. This indicates that the electron resonance density may be reached late only after the pulse peak, and thus that the resonance absorption is not a dominant heating mechanism in our experiment.

The time-integrated soft x-ray emission was measured by a space-resolving, flat field XUV spectrometer.\textsuperscript{14} The spectrograph utilizes a varied line-spacing concave grating with 1200 lines/mm in combination with gold-coated toroidal spectrograph utilizes a varied line-spacing concave grating.

Figure 1 compares the spectra from Ar between 40 and 180 Å obtained at room temperature and at $-77 ^\circ C$. The gas jet backing pressure was 10 bar and the signal was integrated over 100 shots. In Fig. 1(a) at room temperature transitions in $\text{Ar}^{8+}$ and $\text{Ar}^{7+}$ contribute to the majority of the observed lines.\textsuperscript{16} Considering the peak laser intensity of $\sim 7 \times 10^{16}$ W/cm$^2$, charge states of up to $\text{Ar}^{8+}$ should be readily generated by OFI since the threshold intensity$^{17}$ to produce $\text{Ar}^{8+}$ ions is about $3 \times 10^{16}$ W/cm$^2$. As the driving laser is linearly polarized, relativistically low electron temperature ($\lesssim 29$ eV) is expected. Such a cold OFI plasma is dominated by three-body recombination processes feeding electrons into the upper excited levels of $\text{Ar}^{7+}$. For that reason, the x-ray spectrum in Fig. 1(a) has an abundance of lines belonging to $\text{Ar}^{7+}$ and also $\text{Ar}^{5+}$ due to the collisional radiative cascade processes. By contrast, Fig. 1(b) at $-77 ^\circ C$ shows a completely different spectrum. First, the x-ray line emission significantly increased. Second, the spectral distribution has changed in such a way that the lines belonging to $\text{Ar}^{8+}$ and $\text{Ar}^{9+}$ appeared as dominant in Fig. 1(b). Note that the threshold intensity to generate $\text{Ar}^{9+}$ ions by OFI is about 1.6 $\times 10^{19}$ W/cm$^2$, approximately 20 times higher than used in our experiments. Additionally, the presence of strong resonance lines of $\text{Ar}^{8+} (2p-3s$ at 48.7 Å and 49.2 Å$)$ indicates that high electron temperature was achieved and the plasma parameters are fairly different from the previous case of cold recombing OFI plasma. From this it follows that another mechanism(s) became dominant in the interaction and we attribute the drastic changes in the spectrum to inverse bremsstrahlung heating and collisional ionization of near-solid-density Ar clusters.

To explore the role of cluster size in the interaction, we systematically studied the dependency of x-ray line emission on temperature. In Fig. 2 the relative peak intensity values for three selected lines belonging to different charge states of Ar ($\text{Ar}^{7+} 3s-4p$ near 159 Å, $\text{Ar}^{8+} 2p-3s$ at 48.7 Å, and $\text{Ar}^{8+} 2s^22p^5-2s2p^6$ at 165.5 Å) are shown as a function of gas temperature at fixed backing pressure of 10 bar. In all cases the x-ray yield rapidly increases as the gas is being cooled down, and finally saturates at a temperature which exhibits dependency on the ionization state. The growth is approximately exponential until saturation for all three lines. From the data we inferred that the emission on the $\text{Ar}^{7+}$ line saturates around $-40 ^\circ C$ being $\sim 45$ times stronger than that at the room temperature while both $\text{Ar}^{8+}$ and $\text{Ar}^{9+}$ lines reach saturation around $-80 ^\circ C$. Another interesting feature in Fig. 2 is the stepwise appearance of subsequent Ar charge states. At room temperature the highest charge state in the spectrum is $\text{Ar}^{7+}$. As the temperature is lowered down to $-0 ^\circ C$, $\text{Ar}^{8+}$ lines first appear and start to increase. When

![FIG. 1. Time-integrated soft x-ray spectrum from Ar (10 bar) irradiated by 25 fs pulses (100 shots) at room temperature of +26 °C (a) compared with the spectrum at −77 °C (b).](attachment:image.png)
the temperature reaches about $-20^\circ\text{C}$, the Ar$^{9+}$ lines finally appear. It should be noted that due to the existence of a large number of lines between 160 and 180 Å we were not able to distinguish exactly at what temperature the Ar$^{9+}$ lines appeared above the background level and it could be possible that the lines were present at somewhat higher temperatures just being overlapped with the neighboring lines. Similar data have been obtained for 5 bar of Ar, only the saturation points were shifted towards lower temperatures which confirms that the change in cluster size is responsible for the result in Fig. 2.

The experimental results can be explained in terms of a simplified physical picture. At the room temperature there are no clusters (or just negligible amount) in the Ar jet and the radiation on Ar$^{7+}$ lines is caused mainly due to the recombination in cold OFI plasma. While cooling, Ar clusters are formed in the jet and gradually grow in size with decreasing temperature. Though the first electrons are produced by OFI, due to the high local density within the clusters collisional ionization becomes quickly dominant and results in the generation of high ionization stages. As the cluster size increases, the higher positive charge of larger cluster confines ionized electrons inside the cluster, and they can be heated more efficiently via inverse bremsstrahlung longer. The resulting highly ionized hot plasma undergoes expansion associated with cooling. Although the time-integrated data do not provide information about the dynamics, it is realistic to assume that the emission on lines from Ar$^{9+}$ is due to collisional excitation. We have observed signal on the Ar$^{9+}$ $2s^22p^5 - 2s2p^43s$ line near 44 Å growing with decreasing temperature, however, the intensity was quite low when compared with the other spectral lines, Ar$^{9+}$ $2s^22p^5 - 2s2p^6$. This may indicate that the desirable high temperature for collisional excitation into the upper level was attained over a short time before it dropped due to the conductive cooling and hydrodynamic expansion. The emission on resonance lines of Ar$^{9+}$ is due to the combination of collisional excitation in Ar$^{8+}$ and recombination from Ar$^{9+}$. The same holds in case of Ar$^{7+}$ until they saturate. Since the signal on lines from Ar$^{8+}$ and Ar$^{9+}$ continues to grow with decreasing temperature, significant portion of the emission on Ar$^{7+}$ line can be attributed to the recombination from Ar$^{8+}$ in later time of plasma evolution. From the spectral data we deduce that the nonlinear growth of x-ray line emission with increasing cluster size is caused by more efficient collisional heating and slower hydrodynamic expansion of larger clusters. The average charge state increases with cluster size and the corresponding longer decay of an underdense plasma results in stronger x-ray emission due to recombination.

The effect of laser pulse duration on the x-ray spectrum was examined by varying the pulse length from 25 up to 100 fs while the laser energy was kept constant. At room temperature we observed gradual decrease of entire x-ray emission with increasing laser pulse length. In contrast, while cooling we found that the longer pulse resulted in stronger emission on lines from high charge states, such as Ar$^{8+}$, Ar$^{9+}$, and even Ar$^{10+}$. These data for different pulse lengths are consistent with the hydrodynamic model, suggesting that the resonant absorption condition could be reached for the 100 fs pulse.

In conclusion, we have shown that the soft x-ray emission from high intensity, femtosecond laser-irradiated Ar gas jet can be significantly enhanced by lowering the gas temperature. We found rapid nonlinear increase of soft x-ray emission on lines from Ar$^{7+}$, Ar$^{8+}$, and Ar$^{9+}$ charge states and attributed these phenomena to efficient collisional heating of growing Ar clusters. Even though the resonance absorption is no longer a dominant heating mechanism for 25 fs laser pulses, the experimental result showed that the collisional heating itself could provide sufficient heating of the cluster, and by varying the cluster size soft x-ray yield on certain transitions could be maximized.

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