PHYSICAL REVIEW E VOLUME 62, NUMBER 3 SEPTEMBER 2000

Soft-x-ray emission from small-sized Ne clusters heated by intense, femtosecond laser pulses

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(Received 16 February 2000)

Soft-x-ray emission from a cryogenically cooled Ne jet irradiated by intense, 25-fs laser pulses was measured. The Ne spectrum started to drastically change in emitting ions from Ne^{5+} to Ne^{7+} below the preexpansion temperature of $-120\,^{\circ}\mathrm{C}$. The significant change in the spectrum is attributed to the collisional heating of small-sized Ne clusters formed in the cooled jet. The increase of the laser pulse length from 25 fs to 100 fs resulted in further increase of x-ray emission from Ne^{7+} states.

PACS number(s): 52.50.Jm, 52.25.Nr, 36.40.Gk, 36.40.Mr

The interaction of high-intensity, ultrashort laser pulses with atomic clusters has been of interest in the past few years [1–3]. The main characteristic of a gaseous cluster is the near-solid-density inside the cluster, which results in an enhanced absorption of laser energy via collisional and resonance absorptions [2,4] and consequently in an increase of x-ray conversion efficiency. Experimentally, strong x-ray radiation from anomalously high charge states has been observed by irradiating clusters made of noble gases (Ar,Kr, Xe) with subpicosecond laser pulses [1,2]. A recent result obtained by irradiating 25-fs laser pulses on Ar clusters also showed strong enhancement of x-ray emission [5]. To date, however, there has been no experimental result on the interaction with Ne clusters since Ne is characteristized by its poor propensity to undergo clustering. In this paper, we report the observation of x-ray emission from small-sized Ne clusters irradiated by a high-intensity, femtosecond laser. The appearance of spectral lines from Ne^{7+} with the cooling of Ne gas below $-120\,^{\circ}\mathrm{C}$ clearly showed the cluster formation from Ne atoms, since the spectral lines could not be produced from the optical field ionization of Ne atoms by the laser intensity used. The effect of laser pulse duration was also investigated by extending the laser pulse length from 25 fs to 100 fs.

A common way to achieve cluster formation is via an adiabatic expansion of gas from high-pressure valve into a vacuum. The onset of clustering in an expanding gas jet can be estimated with the help of the Hagena parameter [6,7],

$$\Gamma^* = k\,\frac{(d/\tan\alpha)^{0.85} p_0}{T_0^{2.29}}, \tag{1}$$

where p_0 is the backing pressure (mbar), T_0 is the preexpansion gas temperature (K), d is the nozzle diameter (μm), α is the expansion angle, and k is the condensation constant depending on the particular gas (for example, $k=185$ for Ne, 1650 for Ar, and 5500 for Xe [7]). The number of atoms per cluster scales as $\Gamma^{*2.0-2.5}$ [8] and published data indicate that clusters composed of about 100 atoms can be formed when $\Gamma^* > 10^3$ depending on the particular valve and experimental condition [7,9]. Due to the small value of k for Ne, clustering can be achieved only by applying very high backing pressures and/or by lowering the preexpansion gas temperature.

The experiment was performed with a Ti:sapphire laser delivering 25-fs, 35-mJ pulses at a fundamental wavelength of 810 nm [10]. We were able to vary the laser pulse length from 25 fs up to 100 fs by changing the distance between gratings in the pulse compressor. Great care was taken to eliminate undesirable prepulses that could result in the fragmentation of clusters and creation of an underdense plasma before arrival of the main heating pulse. The Ti:sapphire oscillator was operated in a cavity-dumped mode by employing a Bragg cell, and the prepulse to main pulse ratio lower than 10^{-5} has been measured. The linearly polarized laser beam was focused with a $f=45$-cm spherical mirror, yielding the maximum peak intensity of about 7×10^{16} W/cm^2. Figure 1 shows the schematic of the used cryogenically cooled gas jet. The gas jet was produced by a solenoid-driven pulsed valve fitted with a 200-μm-diameter circular nozzle. The gas was cooled by passing through a reservoir filled with cold nitrogen gas or liquid nitrogen that was forced under pressure from a separate liquid nitrogen bath. The nozzle was directly attached to the reservoir as shown in Fig. 1, providing additional cooling. The temperature just before the solenoid valve was measured with a thermocouple inserted into the copper pipe carrying Ne gas. Using this

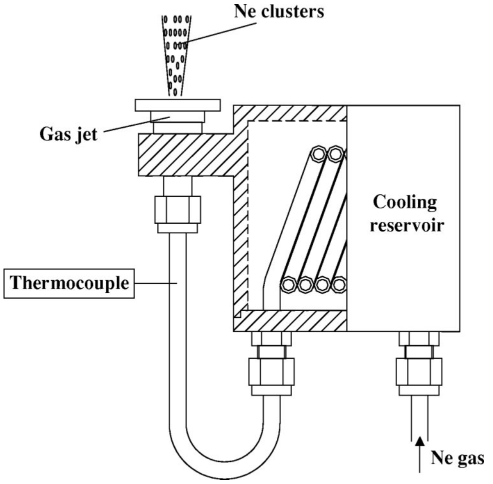

FIG. 1. Schematic of a cryogenically cooled gas jet.

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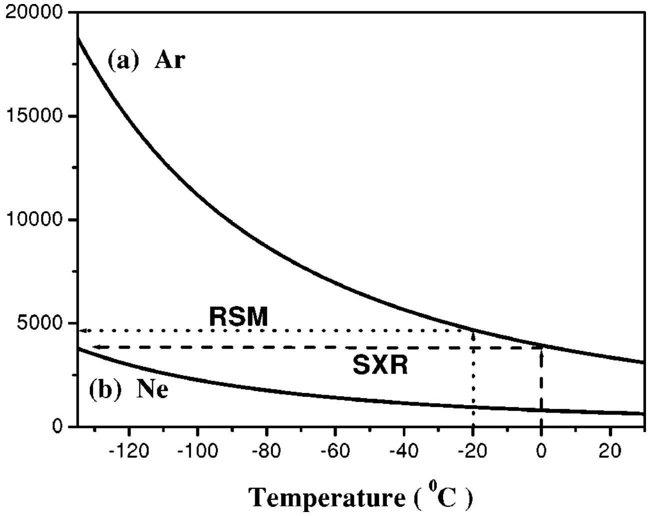

FIG. 2. Scaling of Hagena parameter for (a) Ar at 10 bars and (b) Ne at 18 bars with the preexpansion gas temperature. The onset in the Rayleigh-scattering measurement (RSM) and the transition of soft-x-ray (SXR) spectrum to higher charged ions are indicated by dotted and dashed lines, respectively.

arrangement, gas could be cooled from the room temperature down to about −130 °C. We have not observed any significant drop of backing pressure in the valve that could indicate the formation of liquid droplets instead of clusters. The laser illuminated the gas jet perpendicularly with respect to the flow of the gas and the laser focus was placed about 0.5 mm above the nozzle tip. The use of reservoir for precooling of the gas has made it possible to operate the valve at a repetition rate of 10 Hz without substantial change in the gas temperature. The experiments were repeated with 1 Hz and 0.1 Hz, and the same results were obtained.

The presence of clusters in the cooled jet was first investigated by employing the Rayleigh-scattering technique; however, no significant scattered signal above the noise level was observed over the whole range of pressures (up to 20 bar) and temperatures (down to −130 °C) studied. Figure 2 compares the Hagena parameter for Ne at 18 bars with that for Ar at 10 bars as a function of preexpansion temperature. Our previous study [5] on x-ray emission from laser-irradiated Ar clusters revealed that the first change in the soft-x-ray (SXR) spectrum was observed at a somewhat higher temperature than that corresponding to the onset of signal detection in the Rayleigh-scattering measurement (RSM), as indicated in Fig. 2. The spectroscopic result clearly showed that x-ray spectroscopy serves as a more sensitive indicator of the presence of small-sized clusters than the RSM that presumably starts to show appreciable signal for clusters with typically more than 100 atoms. This suggests that the collisional heating is important even for small-sized Ar clusters composed of a few tens of atoms. Consequently, we may expect that such small-sized Ne clusters can start to be formed in an Ne jet for the comparable value of Γ^*, i.e., around the temperature of −130 °C.

The time-integrated soft-x-ray emission in the wavelength region 4–18 nm was measured with a space-resolving, flat-field extreme ultraviolet spectrometer [11]. The spectrograph utilizes a varied line-spacing concave grating with 1200 lines/mm in combination with a gold-coated toroidal mirror and a back-illuminated x-ray charge-coupled device (CCD; Princeton Instruments). The spectrograph was positioned to view the plasma transversely to the driving laser. The obtained spectra were corrected for the diffraction efficiency of the grating, reflectivity of the toroidal mirror and quantum efficiency (QE) of the CCD [12].

Figures 3(a)–3(c) show a series of Ne spectra obtained with a 25-fs laser pulse at temperatures −115 °C, −120 °C, and −128 °C, respectively. The gas jet backing pressure was 18 bars and the signal was integrated over 100 shots. The spectrum in Fig. 3(a) at −115 °C is very similar to that obtained at room temperature showing transitions in Ne^{3+}, Ne^{4+}, and Ne^{5+} ions [13]. In this case, the applied laser intensity of about 7×10^{16} W/cm^2 is sufficient to strip neutral Ne atoms up to Ne^{6+} ions by optical-field ionization (OFI) [14]. It is then followed by recombination to populate excited levels of Ne^{5+}, the next lower ionization states, and to generate spectral lines of Ne^{5+}. When the temperature reached −120 °C [Fig. 3(b)], the spectrum started to exhibit new features. First, lines belonging to transitions in Ne^{6+} and Ne^{7+} appeared in the spectrum. Second, the spectral lines became more intense and well resolved. Spatially resolved spectrum also showed that the size of plasma increased with cooling. Note that the threshold laser intensity to produce Ne^{7+} ions by OFI is about 1.5×10^{17} W/cm^2, which is more than 2 times higher than the peak intensity used. The Ne^{7+} lines were rapidly enhanced with further cooling and finally became dominant in the spectrum at −128 °C [Fig. 3(c)].

The change in x-ray spectrum indicated transformation of the target medium from atomic gas to small clusters that grow in size with decreasing temperature. Though the first electrons are always produced by OFI due to the high local density within the cluster, the collisional heating via inverse bremsstrahlung becomes quickly dominant and leads to the generation of high-charge states. The theoretical simulation by Ditmire [15] predicted that collisional processes could be important even for small-sized clusters composed of a few tens of atoms. As the average cluster size increases with decreasing temperature, the higher charge state formed in a larger cluster confines the free electrons inside the cluster for a longer time and they can be heated more efficiently. The resulting highly ionized plasma with electron temperature fairly exceeding the value given by above-threshold ionization undergoes expansion while being adiabatically cooled. From the spectral data in Figs. 3(b) and 3(c), we deduce that the average charge state increases with cluster size and the corresponding longer decay of an underdense plasma results in stronger x-ray emission.

A very interesting feature observed in Fig. 3(c) is that the spectral lines below 7.5 nm (denoted A through E), which belong to transitions from highly placed levels in Ne^{7+} ions, are anomalously strong. On the other hand, lines belonging to resonance lines of Ne^{7+}, namely, Ne^{7+} $2s$-$3p$ at 8.8 nm (denoted as F) and Ne^{7+} $2p$-$3d$ at 9.8 nm (denoted as G), were extremely weak even though they have considerably larger oscillator strengths [16]. The sensitivity change of a back-side illuminated CCD detector below the silicon L edge (~12 nm) was already taken into account in the spectra, using the data provided by the manufacturer [12]. It was, however, about 20% only in this region due to a very thin

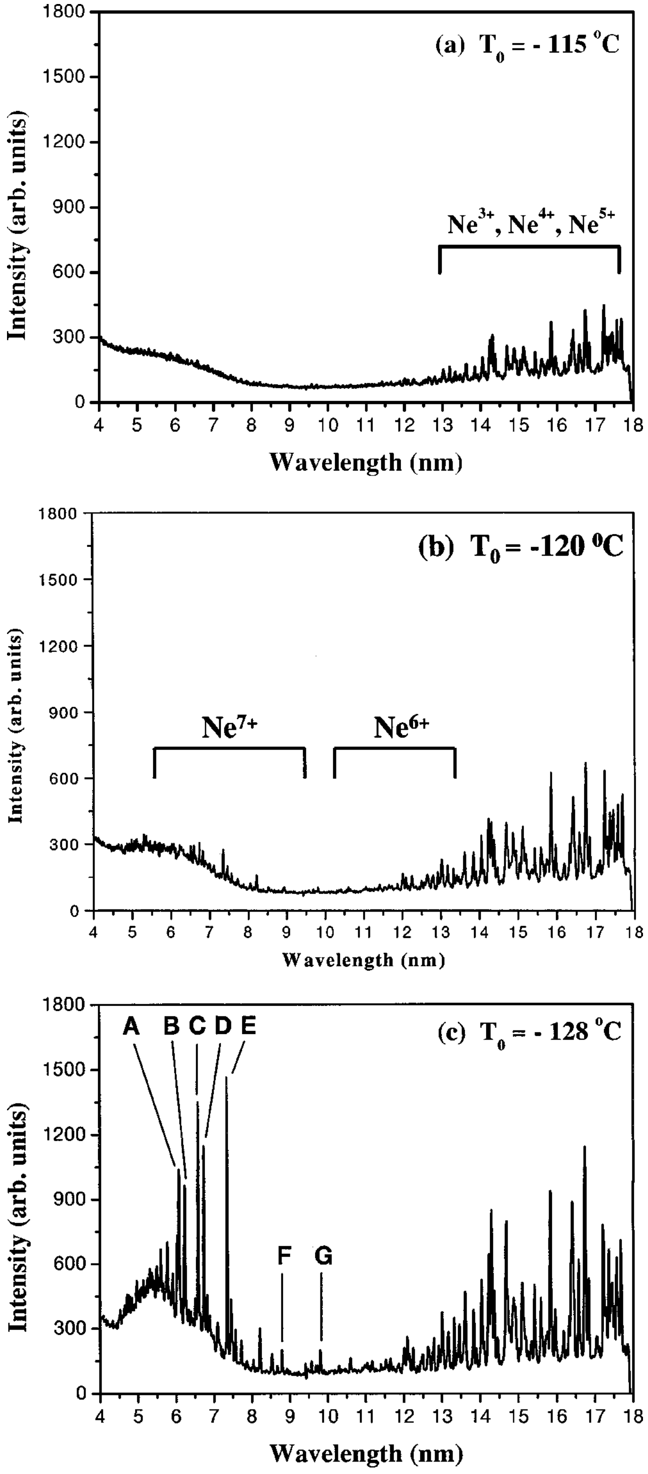

FIG. 3. X-ray spectra from gas jet of Ne ($p_0 = 18$ bars) at preexpansion temperature of (a) $-115\,°C$, (b) $-120\,°C$, and (c) $-128\,°C$, respectively, irradiated by 25-fs laser pulse. In (c), the marked lines from Ne^{7+} ions belong to the transitions: (*A*) $2s$-$5p$, (*B*) $2p$-$6d$, (*C*) $2p$-$5d$, (*D*) $2s$-$4p$, (*E*) $2p$-$4d$, (*F*) $2s$-$3p$, and (*G*) $2p$-$3d$.

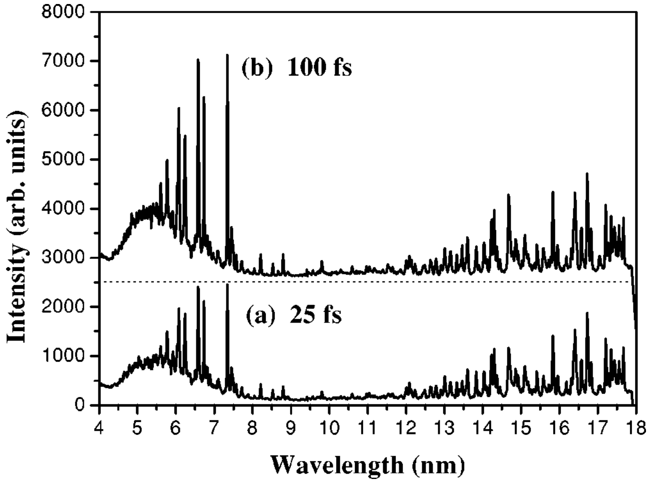

FIG. 4. Comparison of Ne spectra ($p_0 = 16$ bars, $T_0 = -128\,°C$) obtained with (a) 25-fs and (b) 100-fs laser pulse. The upper spectrum is offset by 2500 arbitrary units for clarity.

dead layer, which was further confirmed through the comparison with a theoretical model [17] developed to calculate QE of a thinned CCD. Another consideration on the anomaly is the absorption of radiation by the surrounding gas or plasma that was not taken into account yet in Fig. 3. The absorption by Ne gas, estimated using the database [18], at 10-nm region is larger by a factor of about 2 than that at 7-nm region when the gas density of $1.4 \times 10^{19} cm^{-3}$ and the medium length of 0.35 mm is assumed. This still is not large enough to resolve the anomaly; there must be another reason for this anomaly.

The small-sized cluster could be a promising medium for a recombination x-ray laser. The rapid expansion of a cluster plasma may result in an enhanced recombination of Ne^{8+} ions into the high-lying excited levels of Ne^{7+}. To qualitatively justify our claim, we have performed a simple calculation. Assuming adiabatic expansion of cluster plasma the time-dependent profiles of electron density and temperature were calculated and used to evaluate the collisional excitation and deexcitation rates for the transitions observed in the spectrum. The initial conditions ranged from small-sized hot cluster (radius 0.35 nm, temperature 1 keV) to medium-sized cold cluster (radius 0.5 nm, temperature 100 eV). The collisional deexcitation rates integrated over the corresponding radiation decay times were all smaller than unity, which implies that spontaneous radiative decay is the governing process in the downward collisional-radiative cascade. The adiabatic cooling of cluster plasma, particularly fast-expanding small-sized clusters, appears to result in strong three-body recombination followed by radiative-decay dominated deexcitation cascade, leading to population inversions between the levels lying on the cascade. This picture qualitatively explains the anomaly in Fig. 3(c) that indicates population inversions between the levels $n = 4$ and 3 and also between $n = 5$ and 3 of Li-like Ne ions occur; however, it remains to be confirmed by generating an appreciable gain from Li-like Ne ions in future investigation.

The effect of laser pulse duration on the x-ray spectrum was examined by varying the pulse length from 25 fs up to 100 fs while the input laser energy was kept constant. At the room temperature, we observed a gradual decrease of the x-ray emission with increasing laser pulse length. Such behavior is consistent with the estimation that OFI, which strongly depends on laser intensity, can be considered as the main ionization mechanism at room temperature, i.e., when the laser pulse interacts with a purely gaseous medium. By contrast, completely opposite behavior was observed at low temperatures when the formation of small-sized Ne clusters is expected. Figure 4 compares spectra of Ne ($p_0 = 16$ bars, $T_0 = -128\,^\circ$C) irradiated with 25-fs and 100-fs laser pulses. Apparently, the application of a 100-fs pulse results in considerably stronger emission on Ne^{7+} lines, while the radiation from low ionization stages remained similar. The spectra in Fig. 4 indicate that the longer pulse yielded more efficient heating of the cluster plasma. As the collisional heating by long laser pulse is not efficient since the electron density drops rapidly in time, another absorption mechanism must contribute to the more efficient heating.

Of particular importance is the possible presence of spherical resonance absorption [2,9,19]. If the laser pulse is longer than the time for the cluster plasma to expand to the condition $n_e = 3n_c$ (n_e is the electron density and n_c is the critical density), where the oscillating laser field resonantly drives the electron cloud in the cluster, the absorption of laser energy can be greatly enhanced. We have performed a simple numerical simulation, having for its goal to assess the decay time of the cluster plasma from the solid to $3n_c$ density via adiabatic expansion. Considering an initial cluster radius of 0.5 nm and assuming initial electron temperatures of 100 eV and 1 keV, respectively, we have obtained 30 and 10 fs. Though these values are comparable with the shortest pulse we used (25 fs), the real time of reaching the resonance condition is largely underestimated since we have neglected the time necessary for the buildup of the initial temperature. This indicates that the spherical resonance absorption is marginal for the 25-fs pulse because the electron resonance density may be reached only late after the laser peak. Though we could not quantitatively resolve the interplay between collisional heating and spherical resonance absorption, it is realistic to consider that when the pulse was extended to 100 fs, the expanding overdense plasma could reach the resonance condition before the end of laser pulse, resulting in more efficient deposition of laser energy and higher x-ray yield. Nonetheless, the data in Fig. 4(a) show that collisional heating via inverse bremsstrahlung by laser pulse as short as 25 fs can still be considerable due to the high density inside the cluster.

In summary, we have shown that soft-x-ray emission from a cryogenically cooled Ne jet irradiated by an intense, femtosecond laser pulse exhibits features characteristic for highly ionized, dense cluster plasma dominated by collisional processes, which is fairly different from the plasma produced by OFI in a low-density atomic gas of Ne. The size of Ne clusters in our experiment is estimated to be quite small, i.e., a few tens of atoms per cluster. These clusters were undetectable by the standard Rayleigh-scattering technique, however, the appearance of spectral lines of Ne^{7+} ions presented clear evidence of collisional heating that occurred in near-solid-density cluster. The anomaly of strong lines from $n=4$ and 5 levels of Ne^{7+}, compared to the lines from $n=3$, indicated that population inversions between the levels $n=4$ and 3 and between $n=5$ and 3 might occur during the recombination from Ne^{8+}; however, it remains to be confirmed as yet. By extending the laser pulse length from 25 fs to 100 fs, considerably higher x-ray yield from the highest charge state (Ne^{7+}) was observed in the covered spectral region, showing more efficient absorption of an 100-fs laser pulse.

The authors would like to acknowledge helpful advice concerning the design of the cryogenically cooled valve from H. Fiedorowicz and Seok Ho Kim, and the technical assistance of V. V. Gavrilov during experiments. This research was supported by the Ministry of Science and Technology of Korea through the Creative Research Initiative Program.

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