PHYSICAL REVIEW A **71**, 061804(R) (2005)

Collisional excitation soft x-ray laser pumped by optical field ionization in a cluster jet

H.-H. Chu,[1,2] H.-E. Tsai,[1,3] M.-C. Chou,[1,4] L.-S. Yang,[1] J.-Y. Lin,[4] C.-H. Lee,[5] J. Wang,[1,3] and S.-Y. Chen[1]
[1]*Institute of Atomic and Molecular Sciences, Academia Sinica, Taipei 106, Taiwan*
[2]*Department of Physics, National Taiwan University, Taipei 106, Taiwan*
[3]*Graduate Institute of Electro-Optical Engineering, National Taiwan University, Taipei 106, Taiwan*
[4]*Department of Physics, National Chung Cheng University, Chia-Yi 621, Taiwan*
[5]*Research Center for Applied Sciences, Academia Sinica, Taipei 115, Taiwan*
(Received 23 December 2004; published 20 June 2005)

An optical-field-ionization soft x-ray laser using a clustered gas jet was demonstrated. Pd-like xenon lasing at 41.8 nm with 95-nJ pulse energy and 5.2-mrad divergence was achieved, indicating near-saturation amplification. By using a prepulse to control the expansion of ionized clusters, it was found that the microscopic uniformity of the plasma is essential for efficient lasing. In addition, the optimal atom density for maximum lasing reported previously is verified to result from the tradeoff between large gain coefficient and short gain length due to ionization-induced refraction.

DOI: 10.1103/PhysRevA.71.061804 PACS number(s): 42.55.Vc, 52.50.Jm

Since the first demonstration of soft x-ray lasing in 1985 [1,2], researchers have made much effort to reduce the size of x-ray laser and to increase its repetition rate. A promising scheme is the optical-field-ionization (OFI) collisional-excitation x-ray laser [3], in which a gas target is optical-field ionized by a longitudinally incident laser pulse to form a non-equilibrium plasma channel. Electrons produced from ionization gain energy from the laser pulse via above-threshold-ionization (ATI) heating and collide with the ions to produce population inversion for the lasing levels of specific ion species. The ionization stage can be controlled by laser intensity, and the electron energy distribution and thus the lasing intensity strongly depends on laser polarization [4].

The first OFI collisional-excitation x-ray laser was demonstrated in 1995 for the $5d$-$5p$ line emission at 41.8 nm of Pd-like xenon by using a 70-mJ, 40-fs, circularly polarized laser pulse focused into a xenon gas cell [5]. Saturated amplification of the same spectral line was achieved in 2001 [6], by using a 330-mJ, 35-fs pump pulse. The output reaches 5×10^9 photons/pulse. Strong amplification at 32.8 nm for the $4d$-$4p$ transition of Ni-like krypton was also demonstrated later [7], with a 760-mJ, 30-fs, circularly polarized laser pulse. Recently, two breakthroughs in enhancing lasing efficiency of OFI laser were reported. One is the amplification in a preformed plasma waveguide driven by discharge in a capillary tube filled with xenon and buffer gas [8], resulting in a gain length up to 30 mm and a lasing intensity of 4 times that using a 4-mm-length gas cell. The other breakthrough is seeding the amplifier with pulses from high-harmonic generation [9], which led to a 20-fold increase in the x-ray output and sub-picosecond x-ray pulse duration.

In this Rapid Communication, we report the demonstration of an OFI soft x-ray laser using a clustered gas jet. Comparing to gas cells and gas-filled capillary tubes, gas-jet targets have additional advantages such as much higher density can be reached and damage to the entrance pin hole of the gas cell or the capillary tube and the resulting contamination can be avoided. These features make gas jets a favorable choice for high-efficiency long-term high-repetition-rate operation of OFI soft x-ray laser, particularly when used in conjunction with a plasma waveguide produced by optical method. In addition, for experimental investigation gas-jet targets allow the use of various diagnostics in the transverse direction. This is important for resolving the detail of the propagation and amplification process. However, the production of high-density gas jet of high-Z gases leads to formation of clusters [10], especially so for a supersonic jet which is used for producing a flat-top gas jet with sharp boundaries. Therefore, it is crucial to explore soft x-ray amplification in cluster jets. To make a uniform plasma out of gas clusters, we use a prepulse to ionize and heat up the clusters. By controlling the expansion of the ionized clusters with the prepulse intensity and the delay between the prepulse and the pump pulse, it is shown that the microscopic uniformity of the plasma is essential for lasing, and the optimal atom density for maximum lasing reported previously [6,7] is a result of tradeoff between a larger gain coefficient and a shorter gain length due to ionization-induced refraction.

A 10-TW, 55-fs, 810-nm, 10-Hz Ti:sapphire laser system [11] was used for this experiment. The laser temporal contrast was $>10^7$ at -10 ns, $>10^6$ at -50 ps, and $>10^4$ at -1 ps. A prepulse and a pump pulse both with 55-fs duration were focused with a concave mirror of 1-m focal length onto a xenon cluster jet. The two pulses were set to propagate colinearly with an adjustable temporal separation. The focal spot size of the pump pulse was 25 μm in full width at half maximum (FWHM), with 85% energy enclosed in a Gaussian-fit profile. The peak intensity reached 7×10^{17} W/cm^2 for a pump-pulse energy of 350 mJ. The beam size of the prepulse in the jet was tuned to 180 μm FWHM by installing a telescope in the prepulse beam path before combining with the pump pulse. A motorized quarter-wave plate was inserted before the concave mirror to control the polarization ellipticity of the two laser pulses.

The cluster jet was produced by using a nozzle attached to a pulsed valve with 1-ms open time. There are two different nozzles used for the experiments. One is a conical nozzle with 6-mm outlet diameter, 1.2-mm throat diameter, and 5.4° opening angle, producing a jet profile with a flat-top region of 4.5 mm in length and a sharp boundary of 1 mm at both edges. Clusters are formed inside the jet as a result of rapid adiabatic cooling due to expansion. Since the conditions of the cluster jet are not within the applicable range of the Hagena's or modified Hagena's formula [12], the cluster size cannot be calculated with it. However, a rough estimation can be obtained by using the pump-probe Rayleigh scattering method [13,14]. By measuring the time it takes for the ionized clusters to expand to the resonant density and comparing the result with that of the clusters produced from another supersonic nozzle [14] for which the modified Hagena's formula is applicable, the average cluster size was estimated to be 15–50 nm for a backing pressure of 0.7–4 MPa. The corresponding average atom density was measured to be $1.1\times10^{17}-6.9\times10^{17}$ cm^{-3}. The second nozzle is a slit nozzle with an outlet of 5 mm$\times$0.5 mm dimension and a round throat of 1.2-mm diameter. In the major axis of the outlet, the inner opening angle of the nozzle is 5.4°, and in the minor axis the inner width shrunk from 1.2 mm at the throat to 0.5 mm at the outlet. The jet profile had a flat-top region of 3.5 mm in length and a sharp boundary of 0.75 mm at both edges along the major axis. The average atom density was measured to be $1.1\times10^{17}-3.3\times10^{19}$ cm^{-3} for a backing pressure of 0.013–4 MPa.

A flat-field spectrometer consisting of an aperiodic grazing-incident cylindrical grating and a 16-bit x-ray charge-coupled device (CCD) camera was used to measure the x-ray emission spectrum and the divergence angle in the direction of laser propagation. The spectral range was 30–60 nm and the resolution was 0.05 nm. Aluminum filters were used to block transmitted laser pulses and attenuate x-ray emission. A dipole magnet was used in front of the spectrometer to deflect the electrons emitted from the interaction region, guarding against false signal in the soft x-ray CCD camera. By calibrating the grating reflectivity, the filter transmittance, and the CCD response, we obtained the absolute emission yield. Mach-Zehnder interferometry with a probe pulse passing transversely through the cluster jet was used to measure the evolution of the density distribution of plasma electrons. Side imaging of Rayleigh scattering from nanoplasmas, bandpass filtered at 810 nm, was used to time-resolve the expansion of the nanoplasmas [14]. In this paper, a nanoplasma means an ionized and heated cluster, which is a spherical plasma with a diameter ranging from 1 nm to 1000 nm, and a nanoplasma gas means a cloud of nanoplasmas, which is produced from ionization of clustered gas.

The inset in Fig. 1 shows the soft x-ray emission spectrum for an average atom density of 5×10^{17} cm^{-3} and a pump-pulse delay of 3.8 ns with respect to the prepulse. The energy of the pump pulse was 350 mJ, and its focal point was put at the rear boundary of the cluster jet. The peak intensity of the prepulse was 8.6×10^{14} W/cm^2. Strong lasing signal appeared at 41.8 nm when both pulses were circularly polarized. When the polarizations were changed to linear, the lasing signal vanished. Such a strong dependence of lasing intensity on laser polarization identifies that the x-ray laser was pumped by optical-field ionization and ATI heating. The output energy reached 60 nJ/pulse (1.3×10^{10} photons), larger than the saturated output energy reported previously with similar pump energy, atom density, and gain medium length [6]. The divergence of the lasing beam was 8 mrad, smaller than the aspect ratio of the plasma column. These results indicate that the amplification was close to saturation. When the prepulse was turned off, no lasing signal was observed for either polarization.

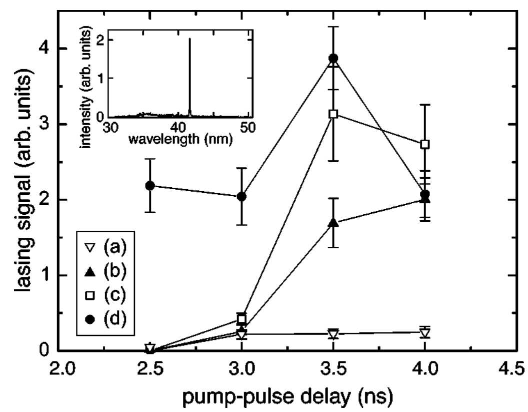

FIG. 1. X-ray lasing intensity as a function of pump-pulse delay with respect to the prepulse for various prepulse intensities: (a) 4.3×10^{14}, (b) 6.7×10^{14}, (c) 8.6×10^{14}, and (d) 1.7×10^{15} W/cm^2. The cluster jet is produced from the conical nozzle with an average atom density of 5×10^{17} cm^{-3}. The energy of the circularly-polarized pump pulse is 350 mJ. Inset shows the soft x-ray emission spectrum for case (c) with a pump-pulse delay of 3.8 ns.

If the pump pulse interacts with a nanoplasma gas instead of a uniform plasma, the collisional ionization and heating may dominate over optical-field ionization and ATI heating because of the high local density. This leads to a smaller population of lasing ion species and/or lower electron temperature, and thus results in a much lower lasing intensity. In addition, the nanoplasma structure may disrupt soft x-ray or pump pulse propagation and consequently hinder the amplification process. To clarify the influence of the nanoplasma structure on x-ray lasing, the dependence of lasing intensity on the pump-pulse delay was measured for various prepulse intensities. Figure 1 shows the results for a pump pulse energy of 350 mJ. The backing pressure of the gas jet was 3.2 MPa, resulting in a cluster size of 40 nm and an average atom density of 5×10^{17} cm^{-3}. When the prepulse intensity was 4.3×10^{14} W/cm^2, relatively weak lasing was observed even when the pump pulse was delayed by up to 4 ns. When the prepulse intensity was increased to 6.7×10^{14} W/cm^2, six-fold increase in lasing intensity was achieved at a pump-pulse delay exceeding 3.5 ns. When the prepulse intensity was further increased to 1.7×10^{15} W/cm^2, strong lasing was obtained at a pump-pulse delay of larger than 2.5 ns. Such a six-fold or higher rise of lasing intensity with just a change of pump-pulse delay by 0.5 ns can be understood from its exponential dependence on the gain coefficient.

To clarify the underlying mechanism for such dependence, the expansion time for the ionized clusters from the initial solid density to the resonant density was measured by using the pump–probe Rayleigh scattering method [13,14]. In this measurement prepulses of various intensities were used to ignite the nanoplasma expansion and the 18-mJ pump pulse served as a probe pulse for Rayleigh scattering. Figure 2 shows the integrated Rayleigh scattering intensity

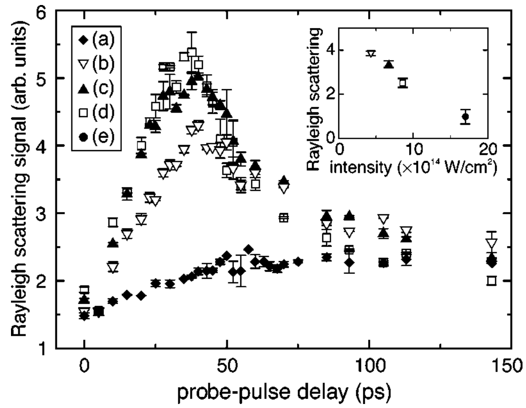

FIG. 2. Integrated Rayleigh scattering intensity as a function of probe-pulse delay for various prepulse intensities: (a) 2.2×10^{14}, (b) 4.3×10^{14}, (c) 6.7×10^{14}, (d) 8.6×10^{14}, and (e) 1.7×10^{15} W/cm^2. The cluster jet condition is the same as in Fig. 1. The energy of the probe pulse is 18 mJ. Inset shows the integrated intensity of Rayleigh scattering of the 350-mJ pump pulse as a function of prepulse intensity for a pump-pulse delay of 3.5 ns (note the vertical scale is in a different unit).

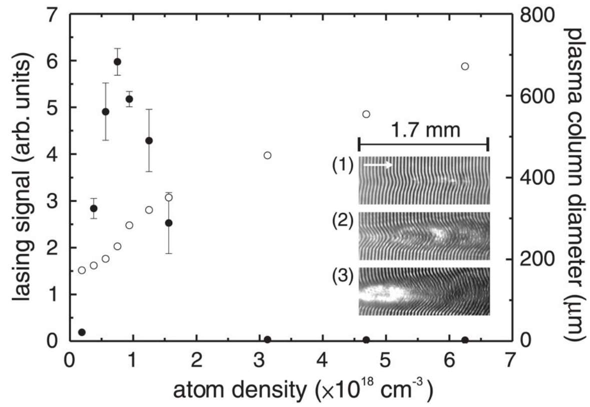

FIG. 3. Lasing intensity (solid circles) and the diameter of the plasma column produced by the pump pulse at 4 mm away from the entrance (open circles) as functions of average atom density. Inset shows interferograms of the plasma column taken at 50 ps after the pump pulse for (1) 7.5×10^{17}, (2) 3.1×10^{18}, and (3) 6.2×10^{18}–cm^{-3} atom densities. The pump pulse energy is 350 mJ, with no prepulse used.

of the probe pulse as a function of probe-pulse delay. The gas jet was operated under the same condition as in Fig. 1. As shown, when the prepulse intensity was 2.2×10^{14} W/cm^2, the Rayleigh scattering intensity reached a maximum after 50-ps probe-pulse delay, indicating that the nanoplasmas ignited by the prepulse expanded to the resonant density in this time scale. As the prepulse intensity was increased, the time scale decreased and reduced to 35 ps for a prepulse intensity of 8.6×10^{14} W/cm^2. This agrees with the expectation that a more intense prepulse results in a higher electron temperature of the ionized clusters. From the measured expansion rates (expand from 40 nm radius to 100 nm radius in about 50 ps) and the average inter-cluster distance (about 2 μm) the time for the mixing of adjacent nanoplasmas to form a uniform plasma was estimated to be a few nanoseconds, which is in good agreement with the time scale of the required pump-pulse delay for obtaining intense x-ray lasing. The correlation between Fig. 1 and Fig. 2 reveals that the microscopic uniformization of the plasma is essential for x-ray lasing. With higher prepulse intensity the time for expansion of ionized clusters decreases, therefore intense x-ray lasing can be reached for a shorter pump-pulse delay.

More evidence can be obtained from the simultaneous measurement of Rayleigh scattering of the pump pulse under the condition of Fig. 1. It is known [13,14] that after the expansion of nanoplasmas has passed the resonant condition, the intensity of Rayleigh scattering of the pump pulse should decrease with increasing delay. When the nanoplasmas merge with each other to form a uniform plasma, the Rayleigh scattering reaches a steady-state level determined by incoherent Thomson scattering from free electrons. In other words, the Rayleigh-scattering intensity can serve as an indicator for the local uniformity of the plasma. The results for 3.5-ns pump-pulse delay are shown in the inset of Fig. 2. At the same delay stronger prepulse resulted in a lower Rayleigh-scattering intensity while the transverse interferometry showed no significant variation of average on-axis electron density and laser beam propagation, indicating a more uniform plasma. This corresponds well to the increasing x-ray lasing intensity.

To increase the efficiency of x-ray lasing, in principle the atom density should be increased to produce higher population-inversion density and thus higher gain coefficient. However, previous studies on the longitudinally-pumped optical-field-ionization soft x-ray laser showed that there exists an optimal atom density for most efficient x-ray lasing [6,7]. The presence of an optimal atom density was ascribed to the effect of ionization-induced refraction. To give a clear experimental proof of this effect, we replaced the 6-mm-diameter conical nozzle by the 5-mm-length slit nozzle, which produced a gas jet with much higher atom density than that from the conical nozzle under the same backing pressure. This provided a tunable range of the average atom density that can adequately cover the optimal density.

With the slit nozzle strong x-ray lasing was obtained even without the prepulse. To make sure this result is consistent with our model the occurrence of pre-ionization at the time of few nanoseconds before the pump pulse must be verified. This was done by adding a transverse prepulse focused to a line of 6-mm length and 10-μm width overlapping with the propagation axis of the pump pulse. The pulse duration of the transverse prepulse was 90 ps and the peak intensity was 5.2×10^{12} W/cm^2 with 250-mJ energy. The Rayleigh scattering image of only the prepulse shows that the prepulse was able to ionize xenon clusters. The images for the cases with both the prepulse and pump pulse show that when the transverse prepulse arrived at 3 ns prior to the pump pulse, the scattering intensity of the pump pulse from the region illuminated by the transverse prepulse was greatly reduced. This reveals that the transverse prepulse arrived before the ionization front of the pump pulse, and the Rayleigh scattering intensity for the case with only the pump pulse was actually dominated by the pre-ionizing pedestal. When the prepulse arrived before the pump pulse by less than 1.5 ns, the reduction of scattering intensity was small. These results show that the pre-ionization of clusters by the pedestal of the pump pulse due to finite laser temporal contrast occurred at between 1.5 ns and 3 ns. The time scale is consistent with our model. The measurements of interferogram and x-ray lasing also showed that when the prepulse intensity was too high or the delay was too long, the x-ray lasing intensity decreased as a result of reduction of on-axis ion density due to the expansion of the plasma column. The early initiation of the nanoplasma expansion for the slit nozzle compared to the conical nozzle is believed to be a result of different cluster size.

To examine the presence and cause of an optimal atom density, the dependence of the lasing intensity and the diameter of the pump-pulse created plasma column on atom density were measured, as shown in Fig. 3. A maximum of the lasing signal appears at an atom density of 7.5×10^{17} cm^{-3}, which is very close to that found in the experiments using a gas cell [6,7]. The simultaneous measurement of interferogram showed that the diameter of the plasma column created by the pump pulse increased with increasing average atom density. As the density was increased from 2×10^{17} cm^{-3} to 1.5×10^{18} cm^{-3}, the plasma column diameter increased by a factor of 2. The results prove that ionization-induced refraction indeed results in stronger defocusing of the pump pulse for higher average atom density and thus faster decrease of pump intensity with propagation. Therefore, an optimal atom density exists as a result of trade-off between a higher gain coefficient and a shorter gain length with increasing atom density. At a pump pulse energy of 350 mJ and an average atom density of 7.5×10^{17} cm^{-3}, the energy of the 41.8-nm lasing line reached 95 nJ (2×10^{10} photons), and the divergence angle of which was 5.2 mrad. Although the increase of average atom density can also lead to the decrease of lasing intensity through the dominance of collisional ionization and heating, even for a uniform atom gas, this limit should be at a much higher density.

In summary, near-saturated soft x-ray lasing for Xe^{8+} at 41.8 nm with a clustered gas jet was achieved by using a prepulse to prepare a suitable plasma condition. Maximum x-ray lasing intensity was obtained when the pump-pulse delay was optimized such that the nanoplasmas have expanded and merged with adjacent ones to form a uniform bulk plasma but before the average atom density drops significantly. The optimal atom density for maximum lasing was identified to be a result of trade-off between larger gain coefficient and shorter gain length caused by ionization-induced refraction with increasing density.

[1] D. L. Matthews *et al.*, Phys. Rev. Lett. **54**, 110 (1985).
[2] S. Suckewer, C. H. Skinner, H. Milchberg, C. Keane, and D. Voorhees, Phys. Rev. Lett. **55**, 1753 (1985).
[3] B. E. Lemoff, C. P. J. Barty, and S. E. Harris, Opt. Lett. **19**, 569 (1994), and references therein.
[4] P. B. Corkum, N. H. Burnett, and F. Brunel, Phys. Rev. Lett. **62**, 1259 (1989); N. H. Burnett and P. B. Corkum, J. Opt. Soc. Am. B **6**, 1195 (1989).
[5] B. E. Lemoff, G. Y. Yin, C. L. Gordan, III, C. P. J. Barty, and S. E. Harris, Phys. Rev. Lett. **74**, 1574 (1995).
[6] S. Sebban *et al.*, Phys. Rev. Lett. **86**, 3004 (2001).
[7] S. Sebban *et al.*, Phys. Rev. Lett. **89**, 253901 (2002).
[8] A. Butler *et al.*, Phys. Rev. Lett. **91**, 205001 (2003).
[9] P. Zeitoun *et al.*, Nature (London) **431**, 426 (2004).
[10] O. F. Hagena and W. Obert, J. Chem. Phys. **56**, 1793 (1972).
[11] H.-H. Chu *et al.*, Appl. Phys. B **79**, 193 (2004).
[12] F. Dorchies *et al.*, Phys. Rev. A **68**, 023201 (2003).
[13] K. Y. Kim, I. Alexeev, E. Parra, and H. M. Milchberg, Phys. Rev. Lett. **90**, 023401 (2003).
[14] H.-H. Chu *et al.*, Phys. Rev. E **69**, 035403(R) (2004).