Gain Saturation Regime for Laser-Driven Tabletop, Transient Ni-Like Ion X-Ray Lasers

J. Dunn, ¹ Y. Li, ^{2,*} A. L. Osterheld, ¹ J. Nilsen, ¹ J. R. Hunter, ¹ and V. N. Shlyaptsev ³

¹Lawrence Livermore National Laboratory, Livermore, California 94550

²Institute for Laser Science and Applications, Lawrence Livermore National Laboratory, Livermore, California 94550

³Department of Applied Science, University of California Davis-Livermore, Livermore, California 94550

(Received 3 December 1999)

We have demonstrated small signal gain saturation on several transient-gain Ni-like ion x-ray lasers by using a high-power, chirped-pulse amplification, tabletop laser. These results have been achieved at wavelengths from 139–203 Å using a total of 5–7 J energy in a traveling-wave excitation scheme. Strong amplification is also observed for Ni-like Sn at 119 Å. Gain of 62 cm⁻¹ and gL product of 18 are determined on the $4d \rightarrow 4p$ transition for Ni-like Pd at 147 Å with an output energy of 12 μ J. A systematic evaluation of the laser driver parameters yields optimum beam divergence and small deflection angles of 2–5 mrads, in good agreement with simulations.

PACS numbers: 42.55.Vc, 32.30.Rj, 42.60.By, 52.50.Jm

Gain saturation is the desired operating regime for x-ray lasers if the maximum extraction efficiency of energy from the plasma gain medium is to be attained. This has been demonstrated for Ne-like ion collisional excitation schemes for $3p \rightarrow 3s$ $J = 2 \rightarrow 1$ lasers of Se at 204 Å and Ge at 232 Å with 500 J to kilojoules of drive energy [1]. The main challenge to improving x-ray lasers has been taking a tabletop scheme and successfully driving it into saturation. There has been considerable difficulty in meeting this challenge by many promising schemes including optical field ionization and recombination [2]. Towards this goal, substantial improvement has been made in recent years in the reduction of the laser drive energy required to generate the population inversion by collisional excitation. This has allowed further progress towards high output, shorter wavelength x-ray lasers. The prepulse technique [3] when used in conjunction with 75 ps pulses with 75 to 150 J energy has achieved gain saturation on various Ni-like $4d \rightarrow 4p$ $J = 0 \rightarrow 1$ transitions from 140 to 59 Å for Ag and Dy ions [4]. Recently, the laser drive energy required for saturated operation has been reduced further to 30 J for Ni-like Pd $4d \rightarrow 4p \ J = 0 \rightarrow 1 \text{ x-ray lasers at } 147 \ \text{Å} [5].$

Progress towards a tabletop x-ray laser has advanced in parallel because the advantages of reduced size, low cost, and high repetition rate are important for the future development of applications in this field. The fast capillary discharge plasma operating at 469 Å for Ne-like Ar has shown gain saturation by single and double passing with a half cavity, and recently this has been extended to high repetition rate, high average power operation [6]. The transient collisional excitation scheme has been proposed to achieve tabletop operation for laser-driven schemes [7]. This utilizes two laser pulses where a long nanosecond pulse generates the plasma and creates the required closed shell Ne-like or Ni-like ionization conditions. After a delay to allow for plasma cooling and expansion which is desirable for both optimum pumping and ray propagation along the plasma column, a 1 ps laser pulse generates a transient population inversion. This fast heating time scale allows efficient pumping without perturbing the ionization. Very high x-ray laser gains greater than 100 cm⁻¹ are predicted and so saturation for target lengths of less than 1 cm is possible. The advantage with this scheme is that less than 5-10 J of laser energy from a chirped-pulse amplification (CPA) tabletop laser is sufficient to drive the inversion. The initial demonstration of the transient scheme was shown for Ne-like Ti $3p \rightarrow 3s$ transition at 326 Å [8] and was extended to the Ni-like ion sequence for the Pd $4d \rightarrow 4p$ line at 146.8 Å [9]. Transient gain saturation was demonstrated on Ne-like Ti at 326 Å and Ge at 196 Å using the larger Vulcan-CPA laser at the Rutherford Appleton Laboratory [10]. However, the laser drive energy to achieve saturation was reported to be 32 and 60 J, respectively, which is currently beyond the output of present tabletop lasers.

In this Letter, we report significant reduction in the laser energy to $\sim 5-7$ J to drive a number of Ni-like ion transient gain lines operating between 139 and 203 Å into the saturation regime. The output energy up to 12 μ J has been extracted by using traveling-wave excitation. This is the first time that gain saturation with gL of 18 has been achieved on a laser-driven tabletop x-ray laser. This also represents the shortest wavelength saturated tabletop x-ray laser. Reduction in the x-ray laser beam divergence and deflection angles to a few mrads is observed for increasing target lengths. In addition, we observe strong output on the Ni-like Sn $4d \rightarrow 4p$ 119 Å line with an estimated gL product of ~ 10 which is the shortest wavelength x-ray laser reported on a tabletop facility.

The experiments were performed on the Compact Multipulse Terawatt laser system at LLNL [11]. This laser, operating at 1054 nm wavelength, utilizes the technique of CPA to produce two beams of nominally 500 fs and 600 ps (FWHM) pulse duration with a repetition rate of 1 shot every 4 min. The short pulse was lengthened to 1.0–1.5 ps with energy of 4.5–5.5 J while the long pulse energy was typically 0.5 to 3 J delivered in the line focus

at the target chamber. The peak-to-peak delay between the laser pulses was found to be optimal at 700 ps with the short pulse arriving after the long pulse. The line focus length of 1.1 cm was achieved with a cylindrical lens and an on-axis paraboloid.

The on-axis x-ray laser output was observed with a 1200 line mm⁻¹ variable-spaced flat-field grating spectrometer with a back-thinned 1024 × 1024 chargecoupled device (CCD). Filters giving attenuation between $10 \times -30 \times$ were used to prevent the x-ray laser saturating the CCD. Fiducial wires were placed close to the spectrometer entrance slit and aligned to the target surface with a telescope to calibrate the angular deflection and beam divergence of the x-ray laser in the horizontal direction. Flat polished target slabs were used in the experiment and tilted back by ~5 mrad in the horizontal direction to compensate for refraction of the x-ray laser in the plasma column. A CCD x-ray slit camera with 25 µm spatial resolution monitored the line focus plasma uniformity and overlap of the laser pulses. An on-axis soft x-ray multilayer-coated imaging system spatially resolved the two-dimensional x-ray laser beam exit profile and output energy for the 189 Å Ni-like Mo $4d \rightarrow 4p$ laser [12].

Several changes in the experiment were introduced in comparison with our previous x-ray laser campaigns [9,11]. The long pulse was defocused to a width of \sim 150 μ m (FWHM) while the short pulse beam was focused to 80 μ m. The main idea was to produce a more uniform lateral plasma medium and increase absorption prior to the excitation process driven by the picosecond laser. More importantly, a traveling-wave scheme was introduced to mitigate against the reduced amplification at longer target lengths resulting from the short-lived transient gain lifetime and increase the laser output. A simple reflection echelon technique was adopted to produce the traveling wave as described in previously related x-ray laser work [13]. The traveling-wave optic consisted of five flat mirror segments placed before the focusing optics. Each segment was offset by 0.12 cm to introduce a traveling wave towards the spectrometer with a delay of 7.7 ps per step. This corresponded to a phase velocity of c along the line focus length, matched to the propagation of the x-ray laser in the gain region. For nontraveling-wave excitation, the reflection echelon was replaced with a flat mirror. Optical streak camera measurements at the line focus with the short pulse beam confirmed that the flat mirror setup produced simultaneous excitation along the line focus.

Figure 1 shows the spectra of Ni-like ion sequence for 0.9 cm targets of Mo (Z=42), Pd (Z=46), Ag (Z=47), Cd (Z=48), and Sn (Z=50) irradiated with a 1.2 \pm 0.5 J, 600 ps laser pulse followed by a 5.0 \pm 0.3 J, 1 ps pulse after a delay of 700 ps. The spectra were obtained with the five-segment traveling-wave reflection echelon optic. The $4d \rightarrow 4p$ x-ray laser transitions are dominant in each spectrum with the highest intensity ob-

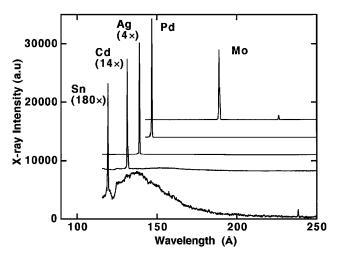


FIG. 1. Single shot spectra from the Ni-like ion sequence from Mo to Sn. In each case, the strong $4d \rightarrow 4p$ x-ray laser line from 189 to 119 Å dominates the spectrum. Note that the baseline of all spectra except Sn are offset for clarity. Ag, Cd, and Sn have intensity multipliers of $4\times$, $14\times$, and $180\times$, respectively, to normalize the line intensity.

served for the 188.9, 146.8, and 138.9 Å lines of Mo, Pd, and Ag, respectively. Recent gain measurements for Mo together with spatial imaging of the gain region indicate that the gain length product is 16.8 ± 0.6 and that the laser intensity is several times above the saturation regime [12]. Overall, the Pd x-ray laser has the highest intensity, approximately $2\times-3\times$ higher than Mo and $4\times$ higher than the Ag output. The general trend is decreasing intensity of the $4d \rightarrow 4p$ laser line for the highest atomic numbers Cd and Sn. This is primarily a plasma temperature effect where the short pulse heating is insufficient to strongly collisionally pump the higher excitation energy from the ground state to the upper excited level and drive into saturation.

Figure 2 shows the intensity versus length of the Pd x-ray laser with and without the traveling wave. Nominal energy in the line focus is $2.0 \pm 0.2 \,\mathrm{J}$ for the long pulse and 4.8 ± 0.5 J for the short pulse. The small signal gain is determined to be 41 cm⁻¹ using the Linford formula [14]. The laser output smoothly increases to achieve an overall gain length product of 18.1. With no traveling wave, the x-ray laser output flattens out for targets above 0.4 cm as observed qualitatively previously [8]. In contrast, the traveling-wave data shows a continued increase in output to between 20 to 100 times higher at 0.9 cm. One of the main issues regarding the interpretation of experimental data from the transient scheme is being able to identify the characteristics of gain saturation, transient gain lifetime, and refraction of the x-ray laser beam out of the high gain region. All of these effects cause roll-off in the x-ray laser intensity as a function of target length. Our analysis using the RADEX simulation code [7-9] concludes that the roll-off in the x-ray laser output without the traveling-wave excitation is determined primarily by

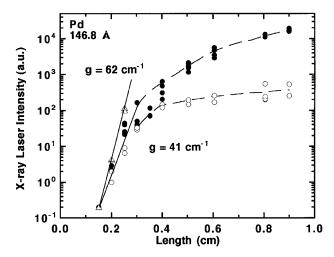


FIG. 2. Intensity versus length plot for Ni-like Pd $4d \rightarrow 4p$ x-ray laser line. Nominal experimental conditions are 2 J, 600 ps and 4.8 J, 1 ps of energy in the line focus for traveling wave (full circles) and no traveling wave (open circles). The dashed curve is to guide the eye. A gain of 41 cm⁻¹ (solid line) is determined at short target lengths for traveling wave and approximately 35 cm⁻¹ for no traveling wave. Traveling wave with 0.5 J energy in 600 ps (triangles) has higher gain 62 cm⁻¹ (solid line).

the short transient gain lifetime in the plasma column and, second, by refraction issues related to density gradients. The transient gain lifetime was estimated to have an exponential decay constant of \sim 7 ps [15] which corresponds to ~0.2 cm photon transit path. The traveling-wave excitation scheme substantially reduces the transient gain effect, hence, leaving plasma refraction as the most important effect. In spite of the transient inversion x-ray lasers operating at shorter target lengths than quasisteady state schemes, refraction has a significant influence as a result of the higher gains produced at high densities. Included in Fig. 2 are lower energy 0.5 J long pulse driver laser shots which generate consistently higher x-ray laser output and inferred gain \sim 62 cm⁻¹ for short plasmas up to 0.25 cm. Calculations show that a lower starting temperature aids the transient inversion by reducing the initial excited level population. It also revealed that instantaneous values of the gain are even larger 90–100 cm⁻¹ but are masked at these small lengths with long-lasting spontaneous emission and refraction.

The deflection and beam divergence angles for the Pd laser line are shown as a function of length in Fig. 3. For the longest targets, the average deflection is 3 mrad away from the target surface with some variation from 1.5 to 5 mrad. At lengths below 0.25 cm, beam deflection angles are greater than 10 mrad. These rays originate in a very high gain, but dense and highly refractive region. With mitigation of refraction, one can even achieve saturation here that indicates further potential for the transient scheme approach that is just starting to be experimentally realized. This deflection angle decreases with increasing target length also due to refraction that tends to select the

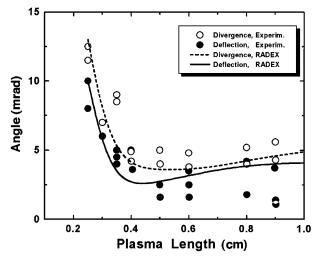


FIG. 3. Beam deflection (full circles) and divergence (open circles) of the Pd x-ray laser with plasma length in comparison with RADEX calculations (solid and dash lines, respectively).

more optimal but less dense areas. Then, it slightly increases with length reaching an asymptotic value while these rays propagate through the whole active medium. The maximum deflection angle $(n_e/n_c)^{0.5}$, where n_c is critical density for this x-ray wavelength, is a reliable tool which reveals the optimal amplification density for Pd ions of $n_e \sim 0.9 \times 10^{20}$ cm⁻³. It should be noted that only 4%-5% of the ps-laser energy is absorbed at this density with this flat target. The divergence also initially decreases with length and finally rebroadens at larger lengths as a consequence of intensity saturation. Figure 4 shows typical angular profiles of the x-ray laser beams for 0.9 cm Ag and 0.9 cm Cd targets: The measured beam divergence is 2.3 and 3.6 mrad (FWHM) with deflection angles of 2.0

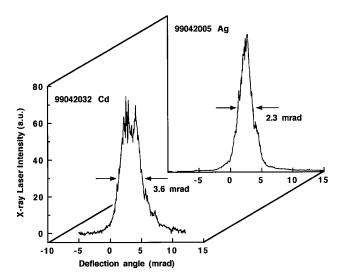


FIG. 4. Angular profile of the x-ray laser beams for the Ag 138.9 Å and Cd 131.5 Å $4d \rightarrow 4p$ lines from 0.9 cm targets, where increasing deflection is defined as away from the target surface. The intensities have been normalized.

and 3.3 mrad, respectively. This is similar to the Pd x-ray laser before the strong saturation rebroadens the beam divergence.

Saturation intensity can be written as $I_{\text{sat}} = (A_u E_v)/$ σ_{stim} , where A_u is the total upper laser level destruction rate, E_{ν} is the x-ray laser energy, and $\sigma_{\rm stim}$ is the stimulated emission cross section [16]. The Ni-like Pd x-ray laser saturation intensity can be calculated from numerical simulations of the plasma conditions, where the density, electron, and ion temperatures at the time of lasing are $n_e \sim 0.9 \times 10^{20}$ cm⁻³, $T_e \sim 400$ eV, and $T_i \sim$ 80 eV, respectively. Values for the stimulated emission cross section $\sigma_{\rm stim} = 5.2 \times 10^{-15} \ {\rm cm^{-2}}$ and upper level destruction rate of $A_u = 1.4 \times 10^{12} \text{ s}^{-1}$ are used. The corresponding saturation intensity is $3.7 \times 10^9 \text{ W cm}^{-2}$. The estimated total uncertainty in this analysis is a factor of 2. The experimental 147 Å Pd x-ray laser intensity, $I_{\rm exp}$, can be also estimated from the studies of the 188.9 Å $4d \rightarrow 4p$ Mo laser where the laser energy output and gain region dimensions have been measured using a multilayer imaging system [17]. Output energy of $3-5 \mu J$ per shot was measured for the Mo x-ray laser after factoring in the recorded signal, mirror reflectivity, filter transmission, and CCD quantum efficiency of the system. The relative sensitivity of the spectrometer at different wavelengths was used to convert the integrated x-ray laser signal at the spectrometer detector into x-ray laser output energy. The Pd x-ray laser output was determined to be $12 \pm 4 \mu J$. The gain region of 80 μ m \times 50 μ m and x-ray laser pulse duration of 5-10 ps, estimated from the analysis in [15], yield an experimental Pd x-ray laser intensity of $I_{\rm exp} = (4 \pm 2) \times 10^{10} \ {\rm W \, cm^{-2}}$ which is ~10 times higher than $I_{\rm sat}$. Because of high small-signal gain, the laser output is still increasing nonlinearly with length at the longest targets while the laser is operating in the saturation regime. The output is smoothly increasing at a continually decreasing rate and it should be possible to extract \sim 5-10× more energy with a few additional millimeters of target under the present irradiation conditions or by driving the plasma harder. More significantly, this demonstrates the strong robustness of this concept where driving tabletop x-ray lasers into sub-100 Å wavelength range will be of primary importance in the future. These experiments and simulations show one of the reliable routes towards this end by creating and utilizing very high gain.

In conclusion, we have observed gain saturation on various transient collisional Ni-like ion schemes operating at 203 to 139 Å, Nb to Ag, at a repetition rate of 1 shot/4 min. Gains of 41-62 cm⁻¹ have been determined for Pd at 147 Å with a gL product of \sim 18.1 with 5 J. This corresponds to 12 μ J output energy. An estimated brightness of 10^{24} photons mm⁻² mrad⁻² s⁻¹ in 0.01% bandwidth is comparable with other x-ray laser sources driven by larger lasers and represents the highest brightness tabletop source operating at this wavelength. The estimated Ni-like Cd intensity at 131.5 Å wavelength

is close to saturation. The shortest wavelength x-ray laser observed in this work at 119.1 Å, $E_v = 104$ eV, is robust and it is estimated that a 50% increase in laser intensity would be sufficient to drive this line into saturation. A final note is that these results were achieved with a simple target design with substantial refraction effects and ps-laser absorption of only a few percent.

The continued support of this research by M. Eckart and A. Hazi are greatly appreciated. We acknowledge the technical contributions from B. Sellick and A. Ellis as well as discussions with A. Faenov and T. Pikuz of VNIIFTRI, Russia, in an earlier experiment. Y. L. and V. N. S. acknowledge support from L. Da Silva and H. Baldis of ILSA. This work was performed under the auspices of the U.S. Department of Energy by the University of California Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

- *Current address: Argonne National Laboratory, Argonne, IL 60561.
- B. J. MacGowan *et al.*, Phys. Fluids B 4, 2326 (1992);
 A. Carillon *et al.*, Phys. Rev. Lett. 68, 2917 (1992).
- [2] X-ray Lasers—1998, edited by Y. Kato, H. Takuma, and H. Daido, IOP Conf. Proc. No. 159 (IOP, Bristol, 1999), p. 720.
- [3] J. Nilsen et al., Phys. Rev. A 48, 4682 (1993).
- [4] J. Zhang *et al.*, Phys. Rev. Lett. **78**, 3856 (1997); S. Sebhan *et al.*, in Ref. [2], p. 91; J. Y. Lin *et al.*, Opt. Commun. **158**, 55 (1998); R. Smith *et al.*, Phys. Rev. A **59**, R47 (1999).
- [5] R. Tommasini, F. Löwenthal, and J. E. Balmer, Phys. Rev. A 59, 1577 (1999).
- [6] J.J. Rocca et al., Phys. Rev. Lett. 77, 1476 (1996); B.R. Benware et al., ibid. 81, 5804 (1998).
- [7] Yu. V. Afanasiev and V. N. Shlyaptsev, Sov. J. Quantum Electron. 19, 1606 (1989); V. N. Shlyaptsev *et al.*, Proc. SPIE—Int. Soc. Opt. Eng. 2012, 111 (1993).
- [8] P. V. Nickles et al., Phys. Rev. Lett. 78, 2748 (1997).
- [9] J. Dunn et al., Phys. Rev. Lett. 80, 2825 (1998).
- [10] M. P. Kalachnikov et al., Phys. Rev. A 57, 4778 (1998);
 P. J. Warwick et al., J. Opt. Soc. Am. B 15, 1808 (1998).
- [11] J. Dunn, J. Nilsen, A.L. Osterheld, Y. Li, and V.N. Shlyaptsev, Opt. Lett. 24, 101 (1999).
- [12] Y. Li, J. Dunn, J. Nilsen, A.L. Osterheld, and V.N. Shlyaptsev, "A Saturated Tabletop X-Ray Laser System at 19 nm," J. Opt. Soc. Am. B (to be published).
- [13] J. R. Crespo et al., Proc. SPIE—Int. Soc. Opt. Eng. 2012, 258 (1993).
- [14] G. J. Linford, E. R. Peressini, W. R. Sooy, and M. L. Spaeth, Appl. Opt. 13, 379 (1974).
- [15] A. L. Osterheld, J. Dunn, and V. N. Shlyaptsev, in Ref. [2], p. 131.
- [16] R. C. Elton, *X-ray Lasers* (Academic Press, San Diego, 1990), p. 23.
- [17] Y. Li et al., Proc. SPIE—Int. Soc. Opt. Eng. 3776, 45 (1999).