Demonstration of X-Ray Amplifiers near the Carbon *K* Edge

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The Ni-like $4d$-$4p$ laser scheme has been extended to wavelengths near the K absorption edge of carbon. A gain of 2.3 cm^{-1} with a duration of 250 psec was observed in Ni-like Ta at 44.83 Å (a wavelength close to optimal for holographic imaging of live cells). Ni-like W produced a gain of 2.6 cm^{-1} with a total of 7 gainlengths of amplification at 43.18 Å. This is the first demonstration of an x-ray amplifier on the short-wavelength side of the carbon K edge, within the "water window." Both lasers should be scalable to coherent power sufficient for holographic imaging and other applications.

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Since 1984 when extreme ultraviolet (XUV) amplification was demonstrated[1,2] near 200 Å, there has been much effort to develop an amplifier to produce significant power at wavelengths below the carbon K absorption ege at 43.76 Å.[3] One motivation was to produce a coherent, high brightness source suitable for holography of biological specimens within the "water window" between the K edges of carbon and oxygen.[4] More recently, a study of the power requirements specific to x-ray holographic imaging by London, Rosen, and Trebes[5] has shown that while for x-ray microscopy it can be advantageous to be inside the water window, for x-ray holography[4,6] the dosage received by a cell is minimized for a given image resolution if the illumination wavelength is slightly to the long-wavelength side of the carbon K edge.

Nickel-like x-ray lasers[7–11] were first demonstrated in a laser-produced plasma of Eu in 1987.[8] Four gainlengths of amplification were observed at 71.0 Å in Ni-like Eu. Subsequently, the scheme was isoelectronically extrapolated to 50.26 Å in Ni-like Yb.[10] The result reported here extrapolates the system further to the edge of the water window, at 44.83 Å, and provides the basis for the design of an x-ray laser source that should fulfill some of the requirements for holography of living cells.[5,6] Also reported here is the further extrapolation of the scheme to 43.18 Å, within the water window, a wavelength more suited perhaps, to short-pulse x-ray microscopy.

Collisionally pumped Ni-like x-ray lasers are $4d$-$4p$ transitions in high-Z Ni-like ions. The $4d$ levels are populated through a combination of direct collisional excitation from the ground state and cascading from upper levels. The $4d$-$4p$ population inversion is maintained by fast radiative decay from the $4p$ levels to the $3d^{10}$ Ni-like ground state while the $4d$ levels are metastable to radiative decay to the ground state. The largest gain is expected on the $J=0$-1 transition $(\underline{3d}_{3/2}4d_{3/2})_0$-$(\underline{3d}_{3/2}4p_{1/2})_1$, at 44.83 Å in Ta^{45+}, where $\underline{3d}_{3/2}$ denotes a vacancy in an otherwise full $\underline{3d}$ shell. The upper state of this transition is populated mainly by collisional excitation from the ground state. The same upper state has another $J=0$-1 lasing transition, to the $(\underline{3d}_{5/2}4p_{3/2})_1$ level (at 50.97 Å in Ta^{45+}). This second, longer-wavelength transition has more gain in lower-Z ions[8] but in higher-Z ions its stimulated emission cross section is smaller than that of its shorter-wavelength partner.[12] For Ta the stimulated emission cross sections of the two transitions are in the ratio 1.3:1.

The Ni-like amplifier is produced by irradiating a thin foil of Ta with a high-intensity optical laser such as the Nova laser at LLNL. The heated foil then becomes a plasma that expands to form a large, uniform gain medium.[2] The plasma needs to be uniform as steep transverse electron-density gradients will cause the x rays to refract out of the plasma before traveling its length.[2] At a density of 10^{21}, transverse scale lengths larger than 50 μm are needed for a 44.83-Å x ray to propagate the length of a 3-cm amplifier. The plasma, at an electron temperature of order 1 keV, and density of order 10^{21} cm^{-3}, should have a significant Ni-like population. The gain in the amplifier may be influenced by trapping, i.e., self-absorption of the $4p$-$3p$ dump lines. The typical line-center optical depth of these lines is $\sim$3–6 with the $(\underline{3d}_{5/2}4p_{3/2})_1$-$3d^{10}$ having almost twice the opacity of the other dump line; hence the 50.97-Å $J=0$-1 line should be more sensitive to trapping. Doppler shifts due to velocity gradients in the explosion of the foil lead to a reduction of the opacity of both dump lines in the direction perpendicular to the original foil surface; hence radiation should escape in this direction. Calculations utilizing the LASNEX, XRASER, and SPECTRE, hydrodynamics, time-dependent kinetics, and line-transfer models[11] predict that the two $J=0$-1 lines should have similar gains (see Table I).

The target used in the Ta experiment was a foil of 127 $\mu g\,cm^{-2}$ Ta on a 24-$\mu g\,cm^{-2}$ Lexan substrate irradiated simultaneously with two of Nova's beams, superposed with a total irradiance of 4.6×10^{14} $W\,cm^{-2}$ in a 120-μm-wide line focus up to 2 cm in length. A total of 5.5

TABLE I. Wavelengths (in angstroms) and gains of the Ni-like Ta and W lines. The estimated uncertainties are given in parentheses. Wavelengths are calibrated against known wavelengths for the Cu-like Ta (Ref. 14) and W (Ref. 19) resonance lines.

		Wavelength (Å)		Gain (cm^{-1})			
	Transition	Theory[a]	Observed	SFFS (Peak)	MCPIGS	SFFS (Int.)	XRASER
Ta^{45+}	$(\underline{3d}_{3/2}4d_{3/2})_0$-$(\underline{3d}_{3/2}4p_{1/2})_1$	44.76	44.83(0.02)	2.3(0.2)	1.4(0.3)	1.6(0.2)	2.6
	$(\underline{3d}_{3/2}4d_{3/2})_0$-$(\underline{3d}_{5/2}4p_{3/2})_1$	50.84	50.97(0.02)	0.5(0.4)	−1(1)	−0.9(1)	2.0
	$(\underline{3d}_{5/2}4d_{5/2})_2$-$(\underline{3d}_{5/2}4p_{3/2})_1$	74.31	74.42(0.02)		−1(1)		3.9
	$(\underline{3d}_{5/2}4d_{5/2})_1$-$(\underline{3d}_{5/2}4p_{3/2})_1$	77.40	77.47(0.02)		1.3(0.4)		1.8
W^{46+}	$(\underline{3d}_{3/2}4d_{3/2})_0$-$(\underline{3d}_{3/2}4p_{1/2})_1$	43.08	43.185(0.01)		2.6(0.2)		5.5
	$(\underline{3d}_{3/2}4d_{3/2})_0$-$(\underline{3d}_{5/2}4p_{3/2})_1$	49.24	Not observed				4.2
	$(\underline{3d}_{5/2}4d_{5/2})_2$-$(\underline{3d}_{5/2}4p_{3/2})_1$	72.25	72.40(0.015)		−0.6(1)		4.9
	$(\underline{3d}_{5/2}4d_{5/2})_1$-$(\underline{3d}_{5/2}4p_{3/2})_1$	75.23	75.35(0.015)		0.8(0.3)		2.2

[a]Reference 15.

kJ of 2ω was used in a 500-psec (full width at half power) Gaussian pulse. The foil was set up in a target chamber vacuum vessel with a large number of XUV and x-ray spectrometers and imaging diagnostics. In particular, the foil was viewed from one end of its axis (the preferred axis for stimulated emission) by a gated grazing incidence XUV spectrometer with a microchannel plate detector (the MCPIGS). Another MCPIGS spectrometer viewed the plasma from an off-axis direction while various time-resolved and time-integrated x-ray spectrometers recorded the x-ray transitions near 2 keV as a signature of the ionization state of the plasma.

Time-integrated spectra from the off-axis MCPIGS spectrometer showed optically thick Cu-like 4-4 resonance lines that have been previously observed and their wavelengths well documented.[13,14] The on-axis spectra were dominated by strong line emission at 44.83 Å; weaker Ni-like emission was visible at 50.97, 74.42, and 77.47 Å. These last two lines are $J=2$-1 and 1-1 transitions which are expected to have gain and share the same lower state as the 50.97-Å $J=0$-1 line. The wavelengths both calculated and measured (using the Cu-like wavelengths as fiducials from Ref. 14) are given in Table I. The calculations are Grant's multiconfigurational Dirac-Fock code with an optimized level scheme.[15]

Figure 1 shows spectra from the other on-axis diagnostic, the streaked flat-field spectrometer (SFFS) which is a grating spectrometer coupled to an x-ray streak camera with 70-psec continuous time resolution. Spectra at one instant in time are shown from two different length foils. Figure 1 illustrates the nonlinear growth of the line intensity with foil length. The spectrometer viewed second, third, and fourth order to obtain good dispersion. The aluminized Mylar filter bandpass restricted its spectral range to the region between the carbon K edge and slightly to the long-wavelength side of the 50.97-Å $J=0$-1 line.

The line intensities from the SFFS are plotted in Fig. 2(a) as a function of foil length. They are fitted with the formula for the axial emission from a distributed source of amplified spontaneous emission from Linford *et al.*[16] The data shown are for the time of peak gain. The time-resolved values of the gains on the two Ni-like $J=0$-1 lines are shown in Fig. 2(b) illustrating the 250-psec duration of the 44.83-Å gain. The time-integrated MCPIGS line intensities were fitted in the same way; the results are summarized in Table I. The SFFS data for the 44.83-Å line peak at a gain of 2.3 ± 0.2 cm^{-1} while the MCPIGS gain is 1.4 ± 0.3 cm^{-1}. The value of the gain obtained by integrating the SFFS data in time is 1.6 ± 0.2 cm^{-1} which is consistent with the MCPIGS value and, as expected, shows that our time-integrated gain measurements may underestimate the peak gain.

To extrapolate this scheme to shorter wavelengths and try and demonstrate a higher gainlength product, tungsten foils of thickness 89 $\mu g\,cm^{-2}$ on 20-$\mu g\,cm^{-2}$ Lexan up to 3 cm in length were irradiated with a total of 3.1×10^{14} $W\,cm^{-2}$ of 2ω light. The Ta experiment

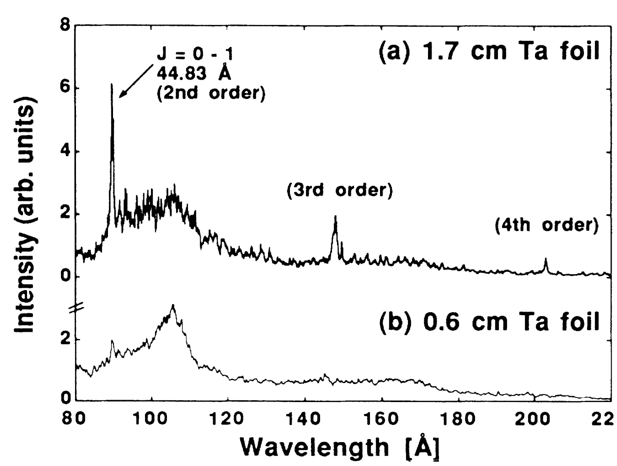

FIG. 1. On-axis spectra from the SFFS streaked spectrometer from (a) 1.7- and (b) 0.6-cm-long Ta foils. Data obtained at the peak of the 44.83-Å gain.

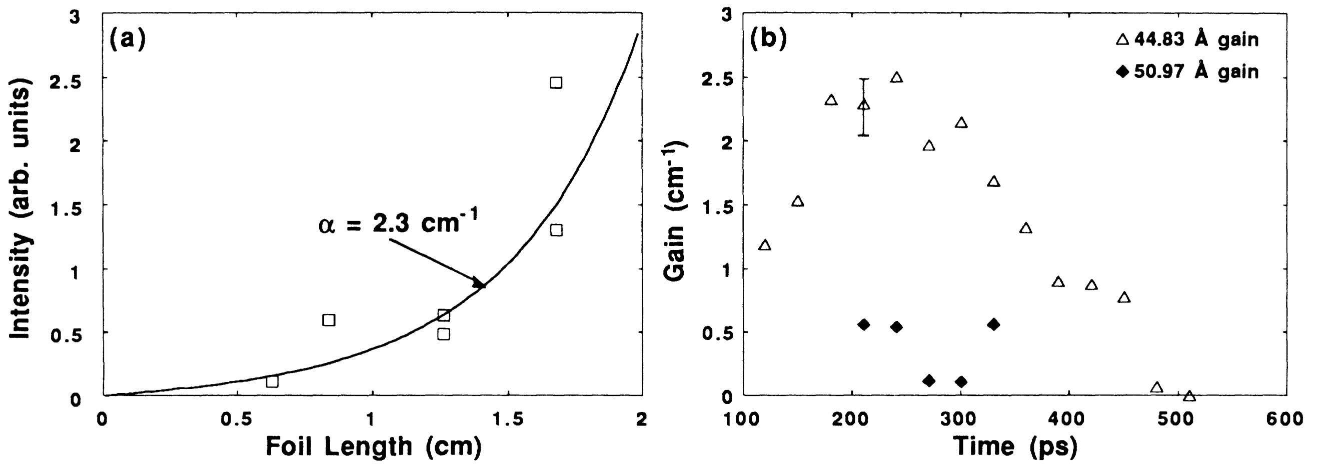

FIG. 2. (a) Line intensity as a function of foil length from SFFS at the peak of the gain. The fit is with the equation $I=(\varepsilon/\alpha)(e^{\alpha l}-1)^{1.5}/(\alpha l e^{\alpha l})^{1/2}$ for a distributed source with emissivity ε, small signal gain α, and length l (Ref. 16). (b) The time history of the gain deduced from the SFFS data.

had shown significant amounts of (Co-like) Ta^{46+} 4-3 and 5-3 x-ray emission indicating that we could reduce the irradiance and still ionize to Ni-like. Figure 3(a) shows on-axis MCPIGS spectra from 2.5- and 1.7-cm-long W foils with the $J=0$-1 laser line at 43.185 Å in first and second order and weaker $J=2$-1 and $J=1$-1 Ni-like lines at 72.40 and 75.35 Å, respectively. In Fig. 3(b) the intensity of the 43.185-Å line is plotted as a function of foil length from the MCPIGS spectrograph. The time-integrated gain from the fit with the same formula as before is 2.6 cm^{-1} with a maximum gainlength of 7 recorded. Time-resolved XUV spectra were not obtained from the W shots but the peak gain may be higher than the time-averaged MCPIGS value. The other $J=0$-1 line (predicted to be at 49.24 Å) was not observed. The gains on the $J=1$-1 and 2-1 lines were qualitatively the same as for Ta and are summarized in Table I.

Table I also shows calculated (time-integrated) gains from simulations similar to those of Ref. 11. Note that there is qualitative agreement (a factor of 2) between the theoretical and measured (time-integrated) gains on the 44.83- and 43.185-Å lines and also their respective $J=1$-1 lines (at 77.47 and 75.35 Å). The calculated gains on the other lines terminating on the $(\underline{3d}_{5/2}4p_{3/2})_1$ level are inconsistent. In particular, the model predicts more gain on the $J=2$-1 line at 74.42 Å (72.40) than the $J=1$-1 line at 77.47 Å (75.35) while the experiment shows no gain on the $J=2$-1. (In Ref. 8 a low gain was seen on the 2-1 and 1-1 lines of Eu^{35+}, in disagreement with calculation.) The ratio of the gains on the two $J=0$-1 lines is predicted to be the same as the ratio of

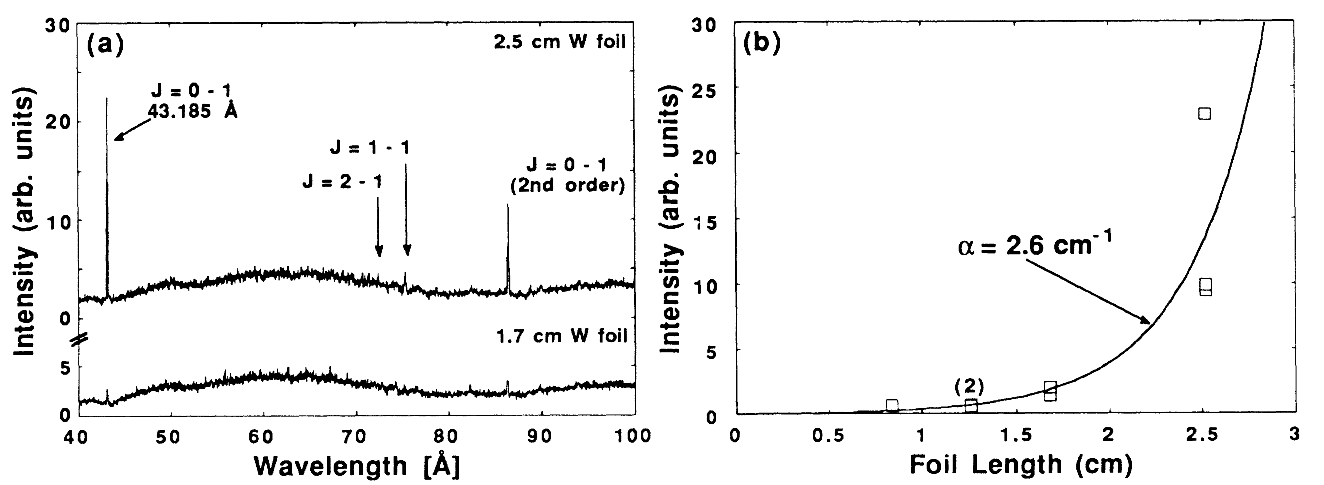

FIG. 3. (a) On-axis spectra (MCPIGS) from 2.5- and 1.7-cm-long W foils. The 2.5-cm foil had a gainlength product of 7 on the 43.18-Å $J=0$-1 laser line. (b) Intensity of the 43.18-Å transition of Ni-like W as a function of foil length, from the MCPIGS spectrometer.

their stimulated emission cross sections, while the Ta $J=0$-1 at 50.97 Å has little or no gain and the analogous line in W was not even observed. These observations may be explained by errors in the population mechanisms for the $(\underline{3d}_{5/2}4p_{3/2})_1$ level, either through an underestimate of the significance of trapping on the $4p$-$3d$ dump lines, or through an error in the coupling of the $4p$ levels to other levels such as the $4s$ states. Postulating a reduced electron temperature in the plasma (which might explain the lower than expected short-wavelength $J=0$-1 gains) does not account for the ratio of the $J=0$-1 gains. This inconsistency of the gains of two $J=0$-1 lines, which share the same upper state, points to a shortcoming in our modeling of Ni-like lasers.

Although we have not yet measured the energy output of these amplifiers, we estimate (using the expected divergence and source size of the amplifier) that the 7 gainlengths of the brightest W laser gave an energy of order 20 μJ at 43.18 Å. The gain duration of order 250 psec is long enough to allow the use of normal incidence multilayer x-ray mirrors to double pass the gain medium.[17] Using a 5% reflectivity mirror a net amplification equivalent to 11 gainlengths should be possible by double passing a 3-cm foil. This amplifier would have an energy output of order 1 mJ. Optimization of the gain in the target through varying its thickness could increase this output further. The saturation flux of a Ni-like Ta x-ray laser (XRL) is estimated to be of order 10^{11} W cm^{-2} which would occur at 12 to 16 gainlengths (depending on the amplifier's divergence) and an energy of 4 mJ. If the divergence is restricted due to the geometry of the target or through use of apertures in combination with the mirror, the amplifier will saturate at a similar energy but higher gainlength product. Estimates of the x-ray laser intensity needed for holography vary. For single-frame holography of a cell with resolution of 300 Å an energy of 300 μJ has been estimated in Ref. 5 while more sophisticated experiments have been proposed using as little as 10 μJ.[18] This energy should be in one transverse mode of the laser. The number of modes emitted by the 44.83-Å x-ray laser will be studied experimentally now that we have passed the initial hurdle of generating gain and significant energy. A worst-case estimate for a 3-cm Ta amplifier is of order 10^4 modes, but this may be overly pessimistic as it ignores any coherence in the beam beyond that expected from its geometric divergence and source size. Now that we have produced gains of order 2.5 cm^{-1} at 43.18 and 44.83 Å, the optimization of that gain may allow the saturation of a Ta laser with gainlength greater than 12, using two beams of Nova, and provide enough energy per mode to perform holography experiments. Possibilities exist to increase the efficiency of the Ta XRL such that pump sources smaller than Nova may be usable. The use of 1ω light would save energy currently lost in the conversion process to 2ω. It may also be possible[20] to use a low-energy, long-pulse ($\sim$1 nsec) laser, to produce a uniform plasma amplifier, then a short pulse ($\sim$20 psec) to heat and ionize it to Ni-like, hence saving the energy that is conducted or radiated away from the hot plasma while it is expanding.

In conclusion, we have demonstrated a working x-ray laser operating at an optimum wavelength for producing holographic images of living cellular material. In addition, we observed seven gainlengths at 43.185 Å in Ni-like tungsten in the first demonstration of an x-ray amplifier operating within the "water window." It remains to enhance output power and characterize and improve coherence in order to have an x-ray laser source, at 44.83 Å, suitable for holography.

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