Femtosecond-pulse-driven 10-Hz 41.8-nm laser in Xe IX

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We report the observation of extreme UV lasing at 41.81 nm on the $4d^9 5d\ ^1S_0 - 4d^9 5p\ ^1P_1$ transition in Xe IX, as proposed by Lemoff *et al.* [Opt. Lett. **19**, 569 (1994)]. A 10-Hz circularly polarized 800-nm laser pulse with an energy of ~ 70 mJ and a duration of ~ 40 fs is longitudinally focused to a peak intensity of $> 3 \times 10^{16}$ W/cm² over a length of 8.4 nm in a differentially pumped cell containing 12 Torr of Xe gas. Laser amplification was observed with an estimated gain coefficient of 13 cm⁻¹ and a total gain of exp(11). © 1996 Optical Society of America

1. INTRODUCTION

Approximately 10 years ago Matthews and colleagues,¹ working at the Lawrence Livermore National Laboratory, reported laser amplification at wavelengths of 20.6 and 21.0 nm in Se XXV. This neonlike Se laser operates by sequential ionization of the target species until a closed shell is reached, followed by electron collisional excitation from the ionic ground state to the upper laser level. At the same time, Suckewer and colleagues,² working at Princeton University, reported laser amplification at 18.2 nm in C VI. The carbon laser operates by fully ionizing the target species by sequential ionization, followed by recombination into the upper laser level. Both schemes required solid-density targets and pump laser pulse energies of several hundred joules. Since that time, many neonlike³ and nickellike⁴ ions have been made to lase by the collisional excitation scheme first demonstrated at the Lawrence Livermore Lab. There also has been substantial effort and progress in reducing the necessary pump energy.⁵⁻⁷ In a related study, Rocca *et al.*,⁸ using a capillary discharge, demonstrated lasing at 46.9 nm in Ar IX.

We recently demonstrated a new class of extreme UV (XUV) laser in which an intense circularly polarized femtosecond laser pulse is used to tunnel ionize a gaseous target species while simultaneously producing the hot electrons necessary for collisional excitation of the species.⁹ Corkum and Burnett first mentioned the possibility of pumping a collisionally excited laser in this way,¹⁰ although much more attention has been paid to their prediction of XUV lasing with a cold plasma produced by linearly polarized optical-field-induced ionization.^{10,11} The first specific systems that used circularly polarized field-induced ionization were proposed by Lemoff *et al.*,¹² who gave calculations for lasing in Ar IX (neonlike), Kr IX (nickellike), and Xe IX (palladiumlike). We have observed lasing at 41.81 nm on the $4d^9 5d\ ^1S_0 - 4d^9 5p\ ^1P_1$ transition in Xe IX. A 10-Hz, 800-nm laser pulse with an energy of ~ 70 mJ and a time duration of ~ 40 fs is longitudinally focused in a differentially pumped cell containing 5–12 Torr of Xe gas. Laser amplification was observed with an estimated gain coefficient of 13 cm⁻¹ and a total gain of exp(11). To our knowledge, this is the highest gain ever observed in a tabletop laser system

operating below 90 nm. We believe that this is also the first XUV laser to operate at a 10-Hz repetition rate and the first that does not require a solid target.

2. INVERSION SCHEME

Figure 1 is the energy-level diagram for the Xe IX laser, showing important radiative and collisional transitions. This Pd-like system, directly analogous to the Ni-like system,⁴ has a full subshell $4d^{10}$ ground state, and lasing occurs on the $J = 0$ to $J = 1\ 5d - 5p$ transition. Both Pd-like and Ni-like systems are direct extensions of the original Ne-like system to closed shells of higher principal quantum number.¹³ In contrast to those for earlier XUV lasers operating by collisional excitation as mentioned above, our gain species, Xe⁸⁺ ions, are created by the optical pulse through tunneling ionization. The tunneling ionization rate is determined by the instantaneous value of the laser field strength.¹⁴ In the tunneling limit, the field-induced ionization rate varies exponentially with an exponent, $-k(E_i^3/I_L)^{1/2}$, where E_i is the ionization potential, I_L is the laser intensity, and k is a constant.¹⁰ With tunneling ionization formulas and an estimated focal intensity of 3×10^{16} W/cm², the tunneling rate from neutral Xe to Xe IX is 1.5×10^{17} s⁻¹. At this laser intensity the ionization rate from Xe IX to Xe X is only 5×10^5 s⁻¹ and is thus insignificant. This vast difference in ionization rates results from the increased energy necessary to ionize the filled $4d$ subshell. We thus expect the 40-fs pulse to produce, over some volume, a nearly uniform plasma of Xe IX in the focal region. Following the ideas of Corkum and Burnett¹⁰ and Corkum *et al.*,¹⁵ we expect that the electrons, released by circularly polarized laser field ionization, will retain a kinetic energy that is equal to the quiver energy, $\epsilon = e^2 E^2 / 4m\omega^2$, at the time of ionization. This is necessary to satisfy conservation of angular momentum or, equivalently, zero velocity at the instant of ionization. Calculations combining the driving laser pulse shape with the tunneling rate formulas yield eight energy groups in the electron energy distribution immediately following the end of the laser pulse. These energy groups, varying from ~ 9 eV for the first electron ionized to ~ 550 eV for the eighth electron ionized,¹² correspond to the different stages of ionization. The last four electrons

Fig. 1. Partial energy-level diagram of Xe IX, showing observed lines at 16.53 and 40.02 nm and the laser line at 41.81 nm. The dashed lines indicate the dominant collisional excitation and deexcitation channels of the upper laser level.

ionized for each Xe^{8+} ion created are expected to have energies greater than 104 eV, thus contributing to collisional excitation of the upper laser level. Because electron ion relaxation time is of the order of 10 ns, the ions remain very near room temperature during the period in which we are interested. The laser line is therefore believed to be predominantly Stark broadened. At 12 Torr, we estimate the laser transition to have a linewidth of $\sim 0.001\text{ nm}$.¹⁶ The plasma is calculated to be optically thick at 16.53 nm, with an absorption length at 12 Torr of $0.06\text{ }\mu\text{m}$. Thus the lower laser level will be trapped, forcing stimulated-emission gain to self-terminate¹² after one upper-state lifetime ($\sim 30\text{ ps}$ for pressures $< 1\text{ Torr}$, $\sim 3.4\text{ ps}$ at 12 Torr). It should also be noted that a 100-eV electron traverses a distance of $50\text{ }\mu\text{m}$ in a time of $\sim 8\text{ ps}$. This is also a limiting factor in determining the pulse duration.

3. EXPERIMENTAL APPARATUS

The laser system used to create the plasma in this experiment is a Ti:sapphire chirped pulse amplification system capable of producing 35-fs, 125-mJ pulses.¹⁷ The recompressed pulse is passed through a 4-cm-diameter, 5-mm thick MgF_2 window into an evacuated chamber as shown in Fig. 2. Inside the vacuum chamber, a 5.08-cm-diameter, 41- μm thick mica quarter-wave plate produces a circularly polarized beam, which is focused by a 50-cm focal-length concave mirror into the differentially pumped Xe target cell, shown in Fig. 3. The pulse energy reaching the target is $\sim 70\text{ mJ}$.

The Xe-gas target cell, together with the gas supply line, has a volume of approximately 7 L and is held at a constant, uniform pressure. Two replaceable pinholes of diameter $< 500\text{ }\mu\text{m}$ provide the entrance orifice for the pump laser and the exit orifice for both the pump and the XUV lasers. These pinholes are drilled *in situ* in 0.1-mm-thick brass stock by the high-power femtosecond laser pulse. The pinholes must be replaced after $\sim 10^6$ shots because their diameter increases progressively. At a cell pressure of 12 Torr of Xe, the background pressure in the continuously pumped vacuum chamber is less than 2 mTorr. By means of translation stages, the pinhole separation can be varied from zero (typically, nonparallelism and bowing of the pinholes put at a minimum of $\sim 1\text{ mm}$ on the separation) to greater

than 4 cm. In this geometry, the femtosecond laser pulse propagates in a nearly gas-free region until it reaches the region immediately outside the entrance pinhole.

Although the pinhole translation stages permit accurate measurement of changes in pinhole separation, the determination of the effective lasing length is more complicated and depends on the pressure distribution outside the pinholes as well as on the focusing properties of the pumping laser. To estimate the effective length, we measure the 41.8-nm signal versus pinhole separation at a sufficiently low pressure (0 to 3 Torr) that the signal varies linearly with length and extrapolate to zero signal. Throughout this paper we assume that the effective length is independent of pressure; however, because of beam-propagation effects, this may not be a valid assumption at higher pressure.

The XUV radiation escaping from the target cell enters a 1.5-m grazing-incidence monochromator (Acton Research Corporation Model GIMS-551.5-M) whose entrance slit is located $\sim 62\text{ cm}$ from the center of the target cell. For the data presented here, entrance and exit slits are $20\text{ }\mu\text{m}$ wide and 1 cm high, and the concave grating has a groove density of 1200 lines/mm. The spectrometer was calibrated from the 25.632-nm line of He II (Ref. 18) and the 16.533-nm line of Xe IX.¹⁹ The spectrometer is accurate to 0.01 nm and in the vicinity of 40 nm has a resolution of $\sim 0.035\text{ nm}$, which is more than an order of magnitude wider than the predicted laser emission linewidth of $\sim 0.001\text{ nm}$. The signal is detected by a dc-biased, double microchannel plate (MCP) located behind the exit slit. The microchannel plate output is amplified by a rf preamplifier (Phillips Scientific Model 6954B-

Fig. 2. Experimental setup: NV, needle valve; GC, gas target cell; WP, quarter-wave plate; B, bellows; TS, translation stage.

Fig. 3. Schematic diagram of the differentially pumped target cell. Note that the cell sits inside a larger vacuum chamber.

Table 1. Comparison of Cowan Code Calculations with Experimentally Observed Energies for Several Levels of Xe IX

Level	Code (cm ⁻¹)	Energy Observed (cm ⁻¹)	Error (%)
$4d^9 5p \ ^3P_1$	599 840.9	598 400	+0.24
$4d^9 5p \ ^1P_1$	608 418.5	604 862	+0.58
$4d^9 5p \ ^3D_1$	620 719.2	618 284	+0.39
$4d^9 4f \ ^3D_1$	699 668.1	696 292	+0.49
$4d^9 5d \ ^1P_1$	792 898.7	789 600	+0.42
$4d^9 4f \ ^1P_1$	839 958.9	832 418	+0.90
$4d^9 5d \ ^1S_0$	848 060.6	844 000	+0.48
$4d^9 5f \ ^3D_1$	1 006 450.7	1 003 000	+0.34
$4d^9 5f \ ^1P_1$	1 043 186.1	1 040 000	+0.31

100) and integrated by a boxcar integrator (Stanford Research Systems Model 250) with a 5-ns time gate. Because the dynamic range of our measurements exceeds that of the detector at a fixed bias voltage, several voltages were used. Relative MCP gain as a function of bias was determined from the dependence of the MCP output on the bias voltage while other experimental conditions were kept the same. All signal voltage data have been normalized to be compared directly with a signal corresponding to a microchannel plate bias of 2000 V. The error involved in this normalization is small compared with the statistical scatter in the data.

4. SPECTROSCOPY OF Xe IX

We calculated the energy levels and the electric dipole spectrum of Xe IX by using the RCN/RCG atomic physics code of Cowan.²⁰ Using the experimental apparatus described above, we were able to measure the energies of these levels experimentally. All spectral lines observed with wavelengths below 16.5 nm can be assumed to be transitions to ground in Xe IX. Sugar and Kaufman, working at the National Bureau of Standards (now the National Institute of Standards and Technology), measured the energies of the 1P_1 and 3D_1 levels of the $4d^9 5p$ and $4d^9 4f$ configurations.¹⁹ We observed all four of these lines, and their positions agreed to within our experimental uncertainty with those reported by the National Bureau of Standards. In addition, we observed lines that we believe to be transitions to ground from the 3P_1 level of the $4d^9 5p$ configuration and the 1P_1 and 3D_1 levels of the $4d^9 5f$ configuration. Of these seven lines, all agree to better than 0.9% with the Cowan code predictions. The observed laser line at 41.81 nm is within 0.2% of the wavelength calculated by the code for the $4d^9 5d \ ^1S_0$ to $4d^9 5p \ ^1P_1$ transition. The prominent line that we observe at 40.02 nm is within 0.2% of the wavelength calculated by the code for the $4d^9 5f \ ^1P_1$ to $4d^9 5d \ ^1P_1$ transition. We therefore are quite confident in our identifications of these lines. Table 1 lists the nine energy levels calculated both from the Cowan code and from experimental data.

5. PRESSURE DEPENDENCE

Lasing at 41.81 nm is evident in the dependence of the signal on both pressure and length. In our experiment the most convenient parameter to adjust is pressure.

Figures 4(a) and 4(b) show spectral scans in the vicinity of 41.8 nm and a length of 8.4 mm at Xe pressures of 3 and 12 Torr, respectively. Note the scale change of 100 between the two plots. The 3-Torr scan shows three prominent lines: the 40.02-nm line corresponds to the Xe IX $4d^9 5f \ ^1P_1$ to $4d^9 5d \ ^1P_1$ transition (see Fig. 1), the 41.45-nm line may be the $5s5p \ ^3P_2$ – $5s6s \ ^3S_1$ line of Xe VII,²¹ and the 41.81-nm line corresponds to the laser transition. The 12-Torr spectrum has a dynamic range of 40. At this pressure the laser line dominates, and no lines other than 41.81 nm are visible.

Figure 5 shows the 41.81-nm signal at a length of 8.4 mm as the pressure is continuously varied from 0 to 10 Torr. Doubling the pressure from 4 to 8 Torr increases the signal by a factor of 80. For comparison, Fig. 6 shows the (nearly linear) dependence of the emission at 40.02 and 16.53 nm. The extent of the dependence on pressure is somewhat obscured by the 5-ns time gate. Even at 1 Torr, stimulated emission is expected to terminate no later than 30 ps following ionization,¹² whereas spontaneous emission is observed to continue for longer than the 5-ns gate width. The observed ratio of

Fig. 4. Observed emission spectrum at an estimated length of 8.4 mm and a pressure of (a) 3 Torr and (b) 12 Torr. Note the factor-of-100 scale change between the two plots.

Fig. 5. Pressure dependence at an estimated length of 8.4 nm of the observed 41.81-nm line emission: (a) linear scale, (b) logarithmic scale.

Fig. 6. Pressure dependence at an estimated length of 8.4 mm of the observed line emission at (a) 40.02 nm and (b) 16.53 nm.

Fig. 7. Length dependence at a pressure of 12 Torr of the observed 41.81-nm line emission. Linford curves are shown fit to all the data (solid curves) and 3.9–7.44 mm only (dashed curves). (a) Linear scale, (b) logarithmic scale.

the 41.81-nm signal at 10 Torr to that at 1 Torr is ~ 700 . Assuming that the 1-Torr signal is entirely spontaneous emission, a 30-ps time-gated measurement would reveal a ratio of greater than 10^5 , which is consistent with a gain greater than $\exp(11)$. Note that we have not measured the XUV laser pulse duration.

6. LENGTH DEPENDENCE

Varying length, although experimentally less convenient, provides a more direct measurement of stimulated gain. Varying the length requires that the vacuum chamber be vented and opened and the pinhole separation changed. Figure 7 shows the 41.81-nm signal obtained at a pressure of 12 Torr and several lengths ranging from 3.9 to 8.4 mm. For this set of data, only the exit pinhole position was changed. Each data point represents an average over ~ 3000 shots. Also shown (solid curve) is the best fit to the Linford expression²²:

$$I \sim \frac{[\exp(gL) - 1]^{3/2}}{\sqrt{gL \exp(gL)}}.$$

From this fit the differential gain is estimated to be $13.3 \pm 0.9 \text{ cm}^{-1}$. At an effective length of 8.4 mm our best estimate of the total gain is $\exp(11.2)$. When a Linford curve is fitted only to the data from 3.9 to 7.4 mm (dashed curve), a better fit is obtained with $g = 16.8 \pm 0.5 \text{ cm}^{-1}$. Assuming a length of 7.4 mm, we obtain a gain of $\exp(12.4)$. The disagreement between the two fits has several possible explanations. The simplest explanation is simply statistical fluctuation. Another possible explanation is that the gain really is 16.8 cm^{-1} over the first 7.4 mm and that it is lower over the final millimeter as the result of a lower pump laser intensity. Finally, it may be that the gain actually is 16.8 cm^{-1} over the entire 8.4 mm, giving a total gain of $\exp(14.1)$, which may be high enough that gain saturation could explain the apparent lower gain of the final millimeter.

7. ADDITIONAL REMARKS

The experiment was repeated with linear polarization. In this case, no stimulated and very little spontaneous emission were observed at 41.8 nm. Under these conditions we looked for and did not observe high-order harmonic generation. When Xe is replaced with 5 Torr of Ar, bright harmonics are observed. The nearest harmonic to the laser wavelength is peaked at $\sim 41.1 \text{ nm}$, has a bandwidth of $\sim 1.0 \text{ nm}$, and has an integrated line fluence that is approximately equal to that of our laser line under optimal conditions. The harmonic has an observed linewidth of ~ 1000 times that of the predicted laser linewidth.

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