Observation of a Short-Wavelength Laser Pumped by Auger Decay

H. C. Kapteyn,(1) R. W. Lee,(2) and R. W. Falcone(1)
(1)*Department of Physics, University of California, Berkeley, California 94720*
(2)*Lawrence Livermore National Laboratory, University of California, Livermore, California 94550*

(Received 15 September 1986)

We report the observation of gain in a new type of short-wavelength laser system at 108.9 nm. Soft x rays from a laser-produced plasma photoeject an inner-shell electron from xenon, and a population inversion is created by subsequent Auger decay to particular excited states. A model for this system is described, spectroscopic data are presented, and gain limitations due to parasitic amplified spontaneous emission are discussed. A maximum gain of 0.8 cm^{-1} over a length of 9 cm is reported.

PACS numbers: 42.55.−f, 32.80.Hd, 52.25.Nr

Interest in the development of new short-wavelength lasers has resulted in recent demonstrations[1,2] of stimulated emission in the region of 10–20 nm. The techniques used to produce population inversions in those experiments involved collisional ionization and excitation by energetic electrons in dense laser-produced plasmas,[1] and recombination of ionized species in magnetically confined laser-produced plasmas.[2]

In this Letter we report the demonstration of a new class of short-wavelength lasers pumped by photoionization followed by Auger decay. As shown in Fig. 1, soft x-ray emission from a high-temperature laser-produced plasma causes photoejection of an inner-shell 4*d* electron from neutral xenon. The resulting ion, Xe II $4d^9 5s^2 5p^6\,{}^2D_{3/2,5/2}$, rapidly undergoes Auger decay, producing various excited states of Xe III. The branching of the Auger decay results in a population inversion between the levels Xe III $5s^0 5p^6\,{}^1S_0$ and Xe III $5s^1 5p^5\,{}^1P_1$. Gain is observed at the transition wavelength of 108.9 nm.

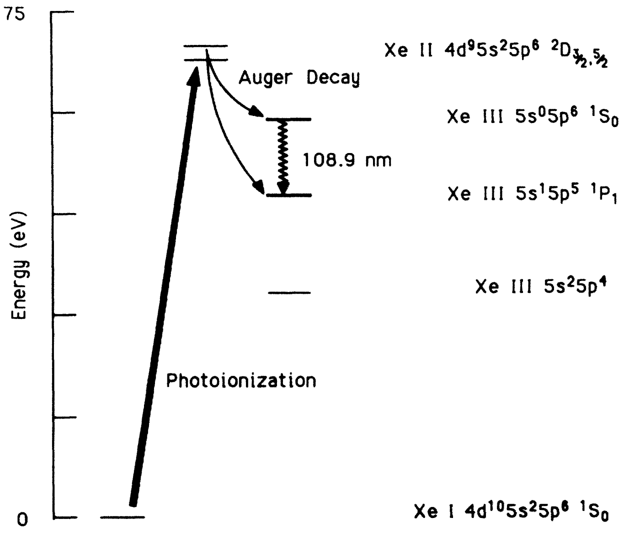

FIG. 1. Energy levels of xenon showing pumping of Xe III levels by photoionization followed by Auger decay. The lasing transition is at 108.9 nm.

The production of a population inversion by core-electron photoejection followed by selective Auger decay was originally proposed by McGuire[3] and recently discussed by Mendelsohn and Harris.[4] An advantage of this type of short-wavelength laser scheme over others[1,2] is that the population inversion is produced by a flux of photons, which results in low ion and electron densities in the lasing region, thus circumventing complex collisional pumping kinetics. This allows simpler modeling of population dynamics and results in smaller plasma refractive-index gradients. Large excited-state populations can be produced in this scheme since laser-produced plasmas are intense and efficient soft x-ray sources, and inner-shell photoionization cross sections and Auger branching ratios can be favorable.[4] Importantly, this scheme should be scalable to shorter wavelengths with existing technology.

The original proposal for photoionization pumping of short-wavelength lasers was made by Duguay and Rentzepis.[5] More recently, several related pumping schemes produced population inversions on longer-wavelength transitions; tunable lasers have been used to populate states which were selectively photoionized[6] or which selectively autoionized[7] into particular final states. Caro *et al.* have shown that soft x rays from laser-produced plasmas can create high densities of excited-state species by photoionization pumping.[8] Plasma-produced soft x rays have also been used to pump lasing states directly.[9]

A diagram of the experiment is shown in Fig. 2. A Nd-doped glass laser pulse with an energy of 55 J and a pulse width of 1 nsec is directed into a target cell containing 1 Torr of xenon gas. The laser is focused to a 50-μm × 9-cm line on a solid tantalum target at a power density of 10^{12} W/cm^2. Broad-band soft x rays radiate into a large solid angle from the resulting plasma. A 3×3-mm^2 × 9-cm-long channel faces the plasma and is 2 cm away from and parallel to it. The channel defines a long and narrow region which is viewed by the detection system. The xenon-filled target chamber is isolated from

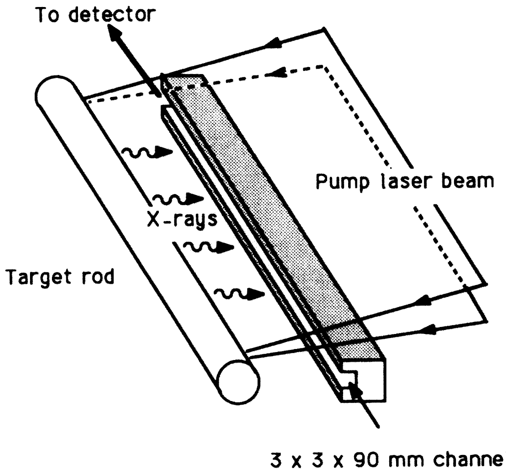

FIG. 2. Target cell design showing the line focus of the Nd-doped glass laser on the tantalum target rod and emission of x rays from the resulting plasma. The channel confines the observed region of excited xenon.

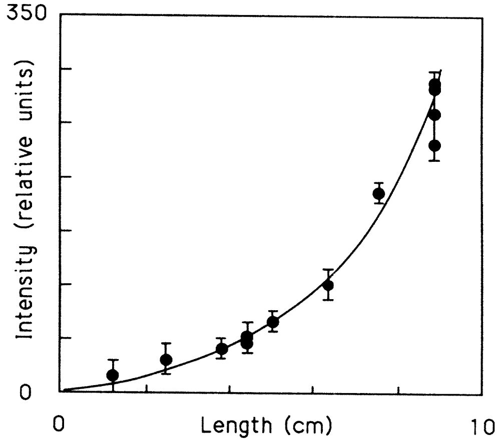

FIG. 3. 108.9-nm emission intensity as a function of illuminated channel length with the use of a normal mixture of xenon isotopes. The data are fitted by a function describing emission from a Doppler-broadened transition, yielding the gain coefficient $\alpha = 0.4$ cm^{-1}.

a 0.5-m vacuum spectrometer by a 1-mm-thick lithium fluoride window. Fluorescence from the ionized xenon is dispersed by the spectrometer and detected with the use of an x-ray streak camera, which records the emission intensity as a function of time and wavelength for each laser shot. The streak camera was modified[10] for high soft x-ray sensitivity by replacement of the normal photocathode with a microchannel plate with adjustable gain. In our experiments data were taken with a wavelength resolution of 1.5 nm and a time resolution of 150 psec. The information was recorded on calibrated film and analyzed by use of an optical scanning digitizer.

The fluorescence intensity at 108.9 nm is determined as a function of length of the pumped region by the shadowing of varying portions of the channel from the x-ray source. This technique maintains constant plasma-light-source characteristics and constant electron and ion densities along the length of the channel, except at the ends. Apertures limit the collection solid angle of the detector so that the field of view is confined inside the channel. The detection efficiency is approximately constant along the length of the channel. This was verified by the observation of approximately equal signal intensity from each half-length of the channel. Most data were taken with the illuminated lengths centered along the slot to minimize end effects.

As shown in Figs. 3 and 4, we found that the emission intensity increased nonlinearly with illuminated length. Each datum point indicates the peak intensity of the 108.9-nm emission for one laser shot, and error bars indicate the noise level of the digitized film data. The Nd-doped glass pumping-laser energy was within ± 10% of the nominal value for all points. The data were fitted by a spectrally integrated brightness function (shown as the solid line) generated by the consideration of the narrow angular and spectrally unresolved emission from an inhomogeneously broadened line source, with the assumption of uniform gain along its length. The two-parameter fit involves an overall intensity constant and the gain coefficient. Although the indicated curve is a numerical integration, the function reduces to $\exp(\alpha l)/(\alpha l)^{1/2}$ when $\alpha l > 2$, where α is the gain coefficient per unit length at line center and l is the length.[1]

The results shown in Fig. 3 are from an experiment in which normal xenon (an isotopic mixture with the largest component 27% ^{132}Xe) was used. No significant difference was observed with use of isotopically enriched xenon (84% ^{136}Xe). Figure 4 shows the results with use of the isotopically enriched xenon, when a 150-nm-thick parylene filter was placed over the open side of the channel facing the plasma. Parylene with this thickness will

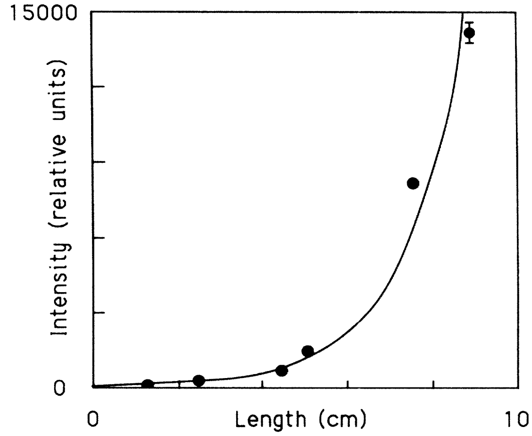

FIG. 4. Same as Fig. 3 except with the use of isotopically enriched xenon and 150-nm-thick parylene to filter the pump soft x rays. Note that the relative intensity units are the same as in Fig. 3. The fit to the data yields a gain coefficient of $\alpha = 0.8$ cm^{-1}.

transmit 60% of the pumping soft x rays, but is opaque to photons below 50 eV energy. We expect that the filtering action of the parylene will increase the observed gain in the experiment by eliminating two gain-limiting effects. First, intense parasitic amplified spontaneous emission (ASE) at the lasing transition (11.4 eV), generated in the xenon-filled region between the plasma light source and the channel, results in a photon flux which could radiatively quench the population inversion in the channel. Second, intense pump radiation from the laser-produced plasma at 12–67 eV, capable of ejecting a Xe 5*s* electron but not a 4*d* electron, results in a higher density of electrons which could collisionally quench the population inversion. We observed a gain coefficient of 0.4 cm^{-1} with normal xenon and without the parylene filter, and 0.8 cm^{-1} with the isotopically enriched xenon and the filter. This corresponds to an output 50 times larger with the 9-cm length in the latter case and is perhaps the most dramatic indication of stimulated emission.

A calculation of the expected gain was made with the assumption that the laser plasma source radiated with a spectral distribution corresponding to a blackbody temperature of 30 eV, with an assumed conversion efficiency of 7% from laser energy to total radiated energy.[11] Photoionization cross sections are known[12] and estimates were made of the Auger branching rates from electron-spectroscopy data,[13-15] a multiconfiguration Dirac-Fock code,[16] and fluorescence measurements.[17] The inversion density is thus calculated to be approximately 2% of the total Xe II ions initially produced by 4*d* electron photoionziation. We measured[17] the lifetime of the upper laser level to be 4.75 ± 0.15 nsec, in good agreement with calculations[16] which also indicate that the primary decay of Xe III $5s^0 5p^5\,{}^1S_0$ is the 108.9-nm transition. The resultant gain cross section, calculated with the assumption of Doppler broadening of a single line, is 3×10^{-13} cm^2. At a pressure of 1 Torr and 2 cm from the plasma target, the peak gain coefficient is thus estimated to be 2 cm^{-1} in the absence of quenching. The effects of isotope shifts and hyperfine splittings on the gain are difficult to predict since their magnitudes are calculated[18] to be on the order of the Doppler linewidth. However, we expect the gain for the normal xenon isotopic mixture to be 2–3 times smaller than the single-line gain predicted above for isotopically enriched ^{136}Xe.

In the absence of the parylene filter, parasitic ASE could act to limit the gain to a maximum value. However, the actual effect of the filter is difficult to predict. In addition to eliminating the inversion quenching effects of parasitic ASE, the filter is expected to reduce the pumping flux, and, thus, the inversion density, by 40% while decreasing the total electron density by a factor of 2. The reduction in the electron density could be significant in determination of the observable gain if collisional quenching (by recombination or deexcitation) limits the population inversion. The experimental observation is that both the isotopically enriched xenon and the parylene filter are required to observe an increase in the gain over the normal-xenon, unfiltered case. Neither improvement alone was sufficient to increase the gain significantly. For the data in Figs. 3 and 4, the emission pulse width was about 600 psec. These results imply that both ASE (in the unfiltered case) and electron collisional quenching act to limit the gain in our geometry. Collisional quenching by ground-state xenon atoms was determined[17] to be unimportant at a pressure of 1 Torr.

Evidence that photoionization followed by rapid Auger decay is the primary pumping mechanism in this system was provided by a separate experiment,[17] which used a plasma produced by a lower-energy, high-repetition-rate, short-pulse laser as a soft x-ray light source to excite the xenon. We observed that the ratio of fluorescence intensities from the upper and lower laser levels was in good agreement with predictions of the Auger branching ratio from both the electron spectroscopy data[13-15] and the code calculation.[16] We also observed that the start of the fluorescence decay from the upper laser level was simultaneous with the several-hundred-picosecond–long soft x-ray pulse from the plasma source, indicating a rapid pumping process.

The gain-limiting effect of parasitic ASE must be considered in the design of photopumped laser schemes. In a first approximation, an atom at the center of a uniformly excited spherical region of radius r, with a radial gain-length product ar, will have an effective lifetime reduced by a factor of approximately $\exp(ar)$. In real systems this problem is manifested as a nonlocal, geometrically determined, gain-limiting depletion rate. A related observation is that high gains transverse to the observation axis of a laser make it difficult, if not impossible, to observe exponential increases in emission intensity with increasing length if the inversion lifetime is on the order of the transit time through the region. This problem is especially important for photon-pumped short-wavelength laser schemes involving plasma x-ray sources which radiate into large solid angles. The solution to this problem is to restrict gain in all but one direction. In our experiment this was accomplished with use of the narrow channel and the parylene filter to limit the gain transverse to the observed volume.

In conclusion, we have demonstrated gain in a new type of short-wavelength laser system. Excited-state ions are produced by Auger decay into selected levels following photoejection of an inner-shell electron by broadband soft x rays from a laser-produced plasma. We expect that this type of laser system will be scaled to shorter wavelengths with the use of more intense laser-produced–plasma x-ray sources and efficient x-ray focusing optics.

We thank M. H. Chen, M. J. Eckart, S. E. Harris, G. Kolbe, and J. H. Scofield for useful discussions. This work was supported by the National Science Foundation under Grant No. PHY 83-51689 and by the University of California Lawrence Livermore National Laboratory Institutional Research and Development Program. We also acknowledge the generous support of AT&T Bell Laboratories. One of us (R.W.F.) is a National Science Foundation Presidential Young Investigator.

---

[1] D. L. Mathews, P. L. Hagelstein, M. D. Rosen, M. J. Eckart, N. M. Ceglio, A. U. Hazi, H. Medecki, B. J. MacGowan, J. E. Trebes, B. L. Whitten, E. M. Campbell, C. W. Hatcher, A. M. Hawryluk, R. L. Kauffman, L. D. Pleasance, G. Rambach, J. H. Scofield, G. Stone, and T. A. Weaver, 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] E. J. McGuire, Phys. Rev. Lett. **35**, 844 (1975).

[4] A. J. Mendelsohn and S. E. Harris, Opt. Lett. **10**, 128 (1985).

[5] M. A. Duguay and P. M. Rentzepis, Appl. Phys. Lett. **10**, 350 (1967).

[6] W. R. Green and R. W. Falcone, Opt. Lett. **2**, 115 (1978).

[7] J. Boker, R. R. Freeman, and W. E. Cooke, Phys. Rev. Lett. **48**, 1242 (1982).

[8] R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, Phys. Rev. A **30**, 1407 (1984).

[9] W. T. Silfvast, J. J. Macklin, and O. R. Wood, Opt. Lett. **8**, 551 (19883).

[10] R. W. Falcone, H. C. Kapteyn, and R. W. Lee, to be published.

[11] R. G. Caro, J. C. Wang, R. W. Falcone, J. F. Young, and S. E. Harris, Appl. Phys. Lett. **42**, 9 (1983).

[12] J. Berkowitz, *Photoabsorption, Photoionization and Photoelectron Spectroscopy* (Academic, New York, 1979).

[13] L. O. Werme, T. Bergmark, and K. Siegbahn, Phys. Scr. **6**, 141 (1972).

[14] H. Aksela, S. Aksela, and H. Pulkkinen, Phys. Rev. A **30**, 865 (1984).

[15] S. Southworth, U. Becker, C. Truesdale, P. Kobrin, D. Lindle, S. Owaki, and D. A. Shirley, Phys. Rev. A **28**, 261 (1983).

[16] M. H. Chen, private communication.

[17] H. C. Kapteyn, M. M. Murnane, R. W. Falcone, G. Kolbe, and R. W. Lee, to be published.

[18] J. H. Scofield, private communication.

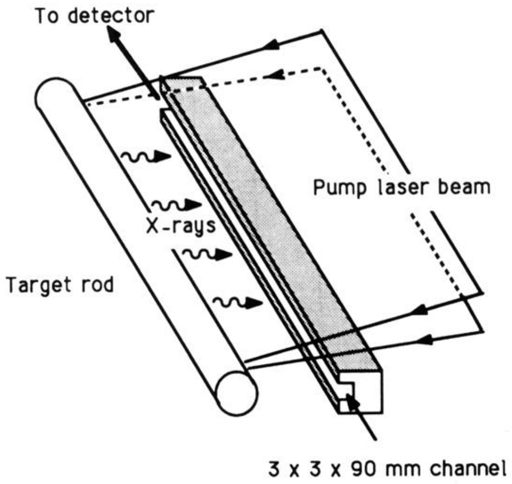

FIG. 2. Target cell design showing the line focus of the Nd-doped glass laser on the tantalum target rod and emission of x rays from the resulting plasma. The channel confines the observed region of excited xenon.