12.8-eV Laser in Neutral Cesium

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We report the operation of a saturated 96.9-nm laser in Cs vapor that has an extrapolated small-signal gain of exp(83) in a total length of 17 cm. We believe that lasing occurs from a core-excited level embedded in the continuum of the valence electron. The laser is pumped by soft x rays from a synchronous, traveling-wave, laser-produced (2.5 J, 20 ps, 1064 nm) plasma.

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In this Letter we report the operation of what we believe is the first laser with its upper level embedded in the continuum of the valence electron.[1] The laser employs a grating-assisted traveling-wave geometry[2,3] that creates a $\sim$20-ps-long pulse of laser-produced soft x rays traveling synchronously with the generated 96.9-nm (12.8-eV) radiation. The gain coefficient is 4.9 cm^{-1} over a 17-cm length, which results in a total extrapolated small-signal gain of exp(83). After about 4 cm, the output energy grows linearly with length, indicating that the laser transition is fully saturated.

Core-excited levels that are embedded within a continuum usually autoionize rapidly, making the accumulation of population difficult. But this need not be the case; recent work by Spong *et al.*[4,5] has shown, for example, that there are many levels in neutral Rb that have autoionizing lifetimes exceeding 10 ps, and several that exceed 100 ps. Such long lifetimes can result either from angular momentum selection rules that to first order prohibit autoionization, or from fortuitous radial matrix element cancellations. The possibility of using such levels to make extreme ultraviolet and soft x-ray lasers has been noted by several workers.[6-8] The existence of an inversion from an upper level embedded within a continuum has been inferred from fluorescence intensity measurements by Silfvast and Wood.[9]

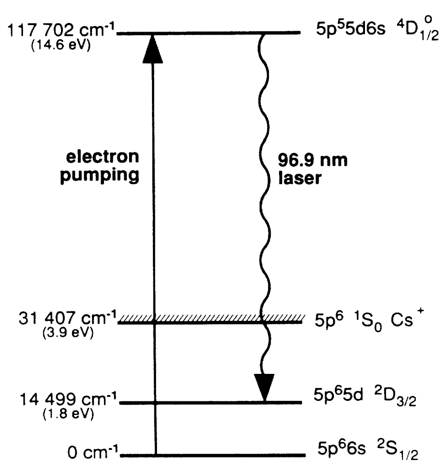

FIG. 1. Partial energy-level diagram for neutral Cs showing the laser transition.

Figure 1 is a partial energy-level diagram for the neutral Cs system showing the 96.9-nm laser transition. The 117702-cm^{-1} energy of the upper level has been measured by vacuum-ultraviolet absorption spectroscopy,[10,11] and the energy of the $5p^6 5d\,^2D_{3/2}$ lower level is well known.[11] The difference (96.897 nm) agrees with our measured emission wavelength of 96.86 $\pm$ 0.05 nm. Our identification of the upper level is based on a comparison of its characteristics as predicted by the multiconfigurational *RCN/RCG* atomic physics code of Cowan[12] with experimental measurements. Code-calculated oscillator strengths from the ground level are in good agreement with the absorption data of Connerade[10] and the ejected-electron data of Pejčev and Ross.[13] For simplicity, we have labeled the upper level $5p^5 5d6s\,^4D_{1/2}$ although it contains a large admixture of the $5d^2$ configuration. The code calculates a transition Einstein A rate of 2.3×10^7 s^{-1}, and an autoionizing rate of 1.6×10^{10} s^{-1}, yielding a radiative branching ratio of 0.0014. We calculate a Doppler-broadened stimulated emission cross section of 1.7×10^{-14} cm^2. Thus, it should be very difficult to observe spontaneous emission from this transition; significant outputs will occur only if the upper level is excited very rapidly and the stimulated emission rate exceeds the autoionizing rate. In our experiments rapid excitation is provided by the combination of a $\sim$20-ps-long pumping pulse and a synchronous traveling-wave geometry.

The experimental geometry is a modification of that used by Sher *et al.*[2] for the Xe 108.9-nm Auger laser. As shown in Fig. 2, a 2.5-J, 15- to 20-ps-long 1064-nm pulse is incident upon a cylindrical lens at 65° from normal and is focused onto a target parallel to the lens. The width of the line focus is $\sim$100 μm. The large angle of incidence expands the length of the line focus by $1/\cos 65 = 2.4$, producing a 17-cm-long plasma. By itself, this geometry would produce a plasma sweeping along the target at a speed of $c/\sin 65 = 1.1c$, resulting in a synchronism mismatch of 3.1 ps/cm of target length. In this experiment, however, the 20-ps-long pulse is formed

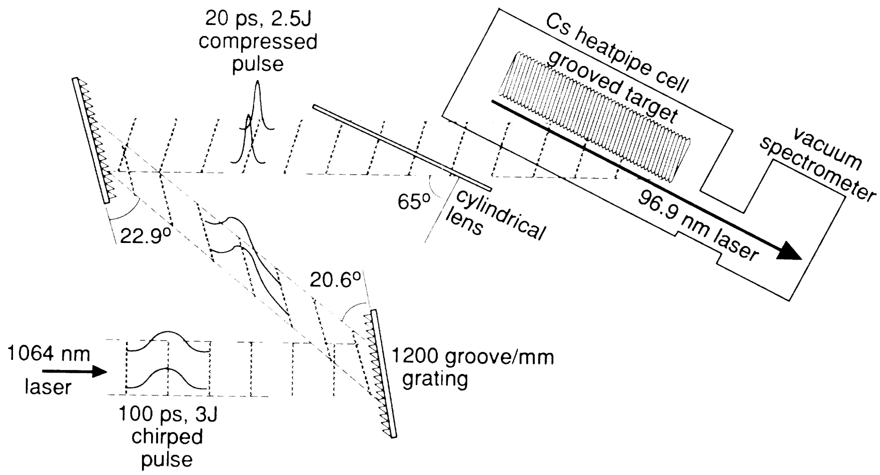

FIG. 2. Grating-assisted, traveling-wave geometry.

by chirping a mode-locked 1064-nm pulse in a fiber, amplifying it in Nd-doped yttrium-aluminum-garnet and Nd:glass stages, and compressing the resulting 120-ps pulse with a parallel grating pair.[14] The second grating of this pair is tilted off true parallelism by 2.3° so as to produce a tilted wave front[3] that exactly compensates for the group-velocity lead of the oblique geometry. The result is a plasma, and its associated pulse of soft x rays, which travels along the target at the speed of light. This correction is essential to produce the largest observed gains.

The target chamber is a Cs heat pipe operating at a density of 6.3×10^{16} cm^{-3}. The surface of the stainless-steel target rod is grooved[2] at a pitch of 43 cm^{-1} and during experiments is wet with liquid Cs. Heating of the target to remove the Cs reduced the observed signals by about 100. Radiation from a $\sim$1-cm region in front of the target was collected by a 1-m normal-incidence spectrometer and detected by a microchannel plate having a 600-ps time resolution. Thin films of In and Al and LiF filters were used to check for possible grating second-order and ghost signals. For the wavelength measurements, the 96.9-nm laser beam was scattered from two ground glass plates before entering the spectrometer.

Gain coefficients were determined by our measuring the relative 96.9-nm energy as a function of plasma length for short sections of the target and fitting the data with the functional form for frequency-integrated superfluorescence output.[15] The length was varied by our masking the input plasma-producing beam. The gain measured at several sections along the 17-cm target was uniform, averaging 4.9 cm^{-1}, thus yielding a total extrapolated small-signal gain of exp(83). Figure 3 shows the dependence of the 96.9-nm output energy on plasma length. The linear increase after about 4 cm clearly indicates saturation. The absolute output energy, measured with an Al vacuum photodiode and a calibrated In filter, was 1.5 μJ.

The predicted poor radiative yield of this transition means that it should be very difficult to observe 96.9-nm spontaneous emission. Using a very short (0.6 cm) plasma in the same cell, we were unable to observe 96.9-nm radiation, and estimate that its intensity was at least a factor of 40 smaller than the 90.1-nm Cs II resonance line, a factor of 60 smaller than the 63.8-nm Cs III resonance line, and a factor of 40 smaller than the 87.5-nm Cs IV resonance line, all of which we observed. Emission at 96.9 nm may have been observed from a discharge in earlier work[16] at a signal level $\sim$200 below the Cs II 90.1-nm resonance line.

Several experiments were performed to test the importance of synchronous traveling-wave pumping and of short-pulse excitation. The grating angle and target angle of incidence were changed to produce a group-velocity lead for the traveling excitation of 10 ps/cm of target length. For this condition, the output signal for a 2.4-cm plasma length was reduced by a factor of 525, and the gain was reduced to 1.8 cm^{-1} by the lack of synchronism. We also compared the 96.9-nm output from a 5.9-cm length of target using the normal 20-ps pumping pulse and an unchirped 220-ps pulse of the same energy. The output signal was $\sim$2000 times weaker with the long pulse. The signal levels of the 90.1-nm Cs II resonance line and the 63.8-nm Cs III resonance line were unchanged for short- and long-pulse excitations, indicating that the laser-signal reduction with longer-pulse pumping should not be attributed to reduced x-ray conversion efficiency. Taken together, the above results strongly support the hypothesis that the 96.9-nm laser emission originates from a level with a very poor radiative yield and a short lifetime.

We believe that the upper laser level is pumped primarily by electrons produced by incoherent soft x rays emitted from the plasma. The expected gain can be es-

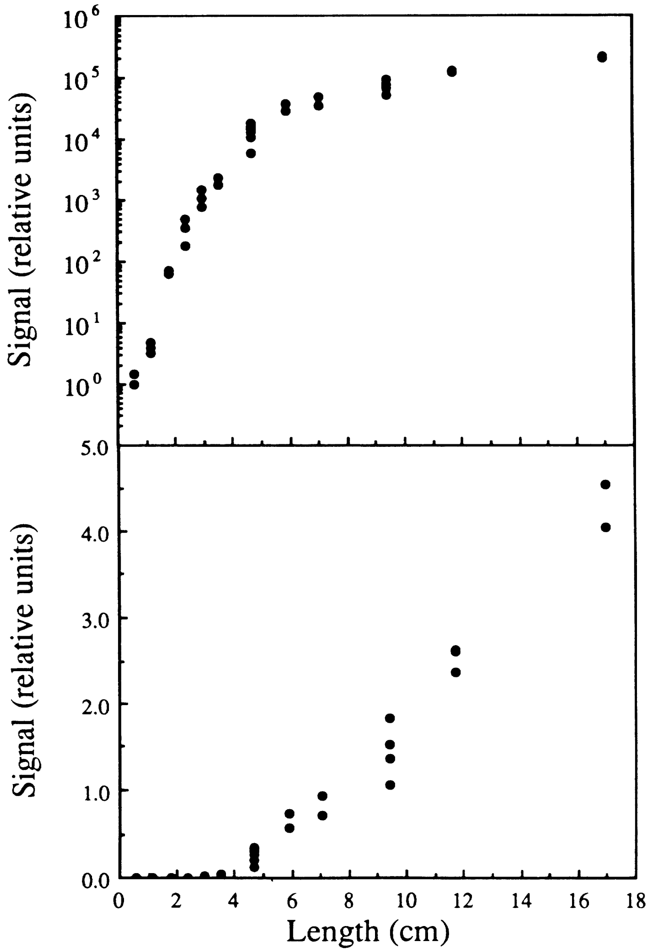

FIG. 3. Output energy at 96.9 nm as a function of plasma length; the same data are presented on both linear and logarithmic scales.

timated as follows. For our 1064-nm power density on target of 1.5×10^{12} W cm^{-2}, and with the assumption of a 25-eV effective blackbody plasma temperature, ~3% of the 1064-nm laser energy will be converted to soft x rays. At a distance of 1 mm from target this flux will create an electron density of about 10^{16} cm^{-3} with a temperature of ~30 eV. The *RCN/RCG* code calculates a temperature-averaged electron excitation cross section times velocity product for the upper laser level of 3.5×10^{-9} cm^3 s^{-1}. Use of the calculated upper level autoionizing lifetime of 62 ps as the effective pumping time yields an upper level population density of 1.4×10^{14} cm^{-3}. This value is consistent with experiments[17] demonstrating that laser-produced plasmas can produce populations in excess of 10^{14} cm^{-3} in metastable levels embedded within a continuum. This upper level population times the calculated gain cross section of 1.7×10^{-14} cm^2 gives a gain coefficient of 2.4 cm^{-1}, if we assume that the lower level is empty, as compared to our measured value of 4.9 cm^{-1}. The autoionizing lifetime and oscillator strength calculated by the *RCN/RCG* code can vary by about a factor of 2 depending on the relative energy spacing used between the $5d6s$, $5d^2$, and $6p^2$ configurations. Direct excitation by the soft x rays may also play a role in the production of upper level population.

The mechanism by which the population of the lower laser level is reduced below that of the upper level has not been determined and is critical to the understanding of this system. Calculations indicate that it is unlikely that electron collision alone can empty the level and produce the inversion. A 1064-nm two-photon transition to the continuum, with the $4f$ valence level as an intermediary, may play a role in this process. We also note that for levels embedded in a continuum, Fano-type interferences between autoionizing levels,[18,19] or in principle between a single level and the continuum,[20] may cause a cancellation of absorption and allow amplification without inversion.

In summary, we believe that this is the first observation of laser action on a transition having an upper level embedded within the continuum of an outer electron. Extremely large gains were produced with only 2.5 J of pumping energy. This fact bodes well for the extension of this concept to even shorter wavelengths.

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[1]S. E. Harris and J. F. Young, J. Opt. Soc. Am. B **4**, 547–562 (1987).

[2]M. H. Sher, J. J. Macklin, J. F. Young, and S. E. Harris, Opt. Lett. **12**, 891–893 (1987).

[3]Zs. Bor, S. Szatmári, and Alexander Müller, Appl. Phys. B **32**, 101–104 (1983).

[4]J. K. Spong, J. D. Kmetec, S. C. Wallace, J. F. Young, and S. E. Harris, Phys. Rev. Lett. **58**, 2631–2634 (1987).

[5]J. K. Spong, A. Imamoḡlu, R. Buffa, and S. E. Harris, Phys. Rev. A (to be published).

[6]E. J. McGuire and M. A. Duguay, Appl. Opt. **16**, 83 (1977).

[7]S. E. Harris, Opt. Lett. **5**, 1–3 (1980).

[8]H. Egger, T. S. Luk, W. Müller, H. Pummer, and C. K. Rhodes, in *Laser Techniques for Extreme Ultraviolet Spectroscopy—1984,* edited by S. E. Harris and T. B. Lucatorto, AIP Conference Proceedings No. 119 (American Institute of Physics, New York, 1984), pp. 64–78.

[9]W. T. Silfvast and O. R. Wood, II, Opt. Soc. Am. B **4**, 609–618 (1987).

[10]J. P. Connerade, Astrophys. J. **159**, 685–694 (1970).

[11]C. E. Moore, *Atomic Energy Levels,* U.S. National Bureau of Standards, National Standards Reference Data Series—3

(U.S. GPO, Washington, DC, 1971), p. 124.

[12]R. D. Cowan, *The Theory of Atomic Structure and Spectra* (Univ. of California, Berkeley, 1981), Secs. 8-1, 16-1, and 18-7.

[13]V. Pejčev and K. J. Ross, J. Phys. B **10**, 2935–2941 (1977).

[14]D. Strickland and G. Mourou, Opt. Commun. **56**, 219–221 (1985).

[15]G. J. Linford, E. R. Peressini, W. R. Sooy, and M. L. Spaeth, Appl. Opt. **13**, 379–390 (1974).

[16]A. J. Mendelsohn, C. P. J. Barty, M. H. Sher, J. F. Young, and S. E. Harris, Phys. Rev. A **35**, 2095–2101 (1987).

[17]J. C. Wang, R. G. Caro, and S. E. Harris, Phys. Rev. Lett. **51**, 767–770 (1983).

[18]J. E. Rothenberg, J. F. Young, and S. E. Harris, IEEE J. Quantum. Electron. **19**, 1795–1804 (1983).

[19]S. E. Harris, to be published.

[20]V. G. Arkhipkin and Yu. I. Heller, Phys. Lett. **98A**, 12–14 (1983).