Demonstration of a Ni-Like Kr Optical-Field-Ionization Collisional Soft X-Ray Laser at 32.8 nm

S. Sebban,¹ T. Mocek,² D. Ros,³ L. Upcraft,¹ Ph. Balcou,¹ R. Haroutunian,¹ G. Grillon,¹ B. Rus,² A. Klisnick,³

A. Carillon,³ G. Jamelot,³ C. Valentin,¹ A. Rousse,¹ J. P. Rousseau,¹ L. Notebaert,¹ M. Pittman,¹ and D. Hulin¹

¹Laboratoire d'Optique Appliquée, CNRS UMR 7639, F-91761 Palaiseau cedex, France
²Institute of Physics, Academy of Sciences of the Czech Benyhlic, Praque 8, Czech Benyh

Institute of Physics, Academy of Sciences of the Czech Republic, Prague 8, Czech Republic ³

Laboratoire de Spectroscopie Atomique et Ionique, CNRS UMR 8624, F-91405 Orsay cedex, France

(Received 19 February 2002; published 27 November 2002)

We report the first experimental demonstration of a Ni-like optical-field ionization collisional soft x-ray laser. The amplifying medium is generated by focusing a circularly polarized 760 mJ, 30 fs, 10-Hz Ti:sapphire laser beam in a few mm cell filled with krypton. We have measured a gain coefficient of 78 cm⁻¹ on the $3d^94d^1S_0-3d^94p^1P_1$ transition at 32.8 nm, which is here amplified for the first time. This radiation source represents the shortest wavelength optical-field ionization collisional soft x-ray laser ever produced. The influence of the gas pressure and the pumping energy on the lasing output are also presented.

DOI: 10.1103/PhysRevLett.89.253901 PACS numbers: 42.55.Vc, 32.30.Rj, 52.50.Jm

Using ultrashort laser pulses to collisionally pump soft x-ray laser transitions has been proposed since 1988 [1]. In this scheme the ion stage is produced by optical-field ionization (OFI) and is controlled by the laser intensity while the energy distribution of the resulting electrons is controlled by varying the polarization of the pumping laser. Linearly polarized lasers generate relatively cold free electrons which are suitable for recombination-type soft x-ray lasers $[2-4]$. On the other hand, the interaction of circularly polarized lasers with a gas medium produces higher energy electrons which can be used to form a population inversion in the extreme ultraviolet range by collisional processes.

In 1994, Lemoff *et al.* modeled [5] three specific collisional x-ray laser schemes in the 30–50 nm range that can be driven by an ultrashort pulse. One year later the same group demonstrated lasing in palladium-like xenon at 41.8 nm [6]. However, it has taken five years to reproduce and improve the original results; saturated amplification on the $4d^95d^1S_0-4d^95p^1P_1$ transition in Xe⁸⁺ at 41.8 nm has recently been achieved by focusing a 330 mJ, 35 fs circularly polarized laser pulse into a gas cell filled with 15 Torr of xenon [7]. In parallel another challenge is to reach shorter wavelength. Ni-like schemes offer a more efficient approach to shorter wavelengths; lasing on the 4*d*-4*p* transition in Ni-like krypton at 32 nm can be anticipated [5]. Ni-like collisional x-ray lasers has been successfully demonstrated using sequential pumping techniques [8–12] and more recently the transient collisional scheme [13–15]. However, the extension of the OFI collisional x-ray laser sources to the Ni-like scheme has not been realized to date. Moreover, an amplified 4*d*-4*p* x-ray laser transition in Ni-like krypton has never been observed using any operating x-ray laser techniques.

In this Letter we report the first experimental demonstration of a 10-Hz Ni-like OFI soft x-ray laser. We have observed a strong amplification at 32.8 nm on the $3d^9 4d^1S_0 - 3d^9 4p^1P_1$ transition in Kr⁸⁺ by focusing a \sim 760 mJ, 30 fs circularly polarized laser pulse into a gas cell filled with krypton. We have measured a gain coefficient of 78 cm⁻¹, and the lasing output has been optimized by adapting the gas pressure, the position of the spot focus, the length of the gas cell, the driving energy, and the ellipticity of the polarization. This source represents the shortest wavelength high repetition rate OFI collisional x-ray laser ever produced.

To realize a Ni-like krypton OFI collisional soft x-ray laser, appropriate laser irradiation and polarization are required. According to the barrier suppression ionization theory [16], the Kr IX species are generated for a threshold laser intensity of 2.3×10^{16} W cm⁻². Note that laser intensities larger than 2.1×10^{17} W cm⁻² will generate Kr X and thus overionize the amplifying medium. To pump the laser transition from the Ni-like ground state $(1s²2s²2p⁶3d¹⁰)$ to the upper level of the lasing transition $4d$ (${}^{1}S_{0}$) electron energies larger than 145 eV should be created [4]. Using a circularly polarized laser beam; a non-Maxwellian electron distribution is generated with electron energies ranging from about 20 eV for the first ionization stage to about 2 keV for the eighth released electron [17]. Finally, a circularly polarized laser delivering about \sim 1 \times 10¹⁷ W cm⁻² at the focus over a few millimeters should be suitable to generate a plasma column with appropriate conditions for the amplification of the $3d^9 4d^1S_0-3d^9 4p^1P_1$ transition in Kr IX.

The experiment has been performed at the Laboratoire d'Optique Appliquée, Palaiseau (France). The experimental setup is shown in Fig. 1. It is similar to the one described in [6] except for some important modifications of the energy (up to 1 J on target), the duration (30 fs instead of 35 fs), and the diameter (55 mm instead of 40 mm) of the driving laser [18]. The laser contrast has

FIG. 1. Schematic description of the experimental setup.

been measured to be about 6×10^7 . Taking into account the total efficiency of the optical system used, the net intensity in the focus spot was estimated to be up to $5 \times$ 10^{17} W cm⁻². The principal diagnostic was an on-axis soft x-ray transmission grating spectrometer. The spectrograph utilizes a grazing incidence gold coated spherical mirror, transmission grating with 2000 lines/mm, and a back-illuminated x-ray charge-coupled device (CCD). Since the CCD detector has not been absolutely calibrated, the measured signal is given in arbitrary units. Each data point presented in this paper corresponds to signal accumulated over 20 laser shots.

Figure 2 shows the on-axis time integrated spectrum of a 4 mm long krypton plasma column produced by a 600 mJ circularly polarized laser pulse. The spectrum is dominated by emission on the $3d^9 4d^1 S_0 - 3d^9 4p^1 P_1$ transition of Kr^{8+} at 32.8 nm, which suggests that high gain is present. The bright satellite lines that can be observed on both sides of the lasing line result from diffraction of the lasing signal on the supporting grid of the transmission grating. Several incoherent emission lines belonging to Kr^{7+} and Kr^{8+} were also observed in the spectrum, however significantly (350 times) weaker than the 4*d*-4*p* lasing line. To find optimum conditions for lasing we have systematically studied the dependency of the x-ray laser output on major experimental parameters: pressure, pumping energy, ellipticity of polarization, and

FIG. 2. Time integrated spectrum of a 4-mm-long Kr plasma column irradiated by a 30 fs, 600 mJ laser pulses (20 shots).

position of focus with respect to the entrance pinhole. Note that the lasing action of the 32.8 nm line has shown to be sensitive to the contrast of the driving laser and has been effective only for a laser contrast better to about 10⁶. The detrimental effect of the preionization of the plasma by decreasing the on-axis ion density is largely responsible for the reduction of the small-signal gain coefficient, i.e., the amplification of the lasing line [19]. This is qualitatively clearly observed in our experiment. In Fig. 3 the output intensity of the 32.8 nm lasing line is shown as a function of the gas pressure. The length of the gas cell was 4.5 mm and the net driving energy on the target was 600 mJ. Lasing appears to be efficient in the range 15–25 Torr with a maximum at 20 Torr. A normalized pressure dependence of the 5*p*-5*d* Pd-like xenon lasing line at 41.8 nm is included for comparison in Fig. 2. It shows that the xenon laser is amplified over a narrow range of gas densities, while the efficient pressure range is broader for Ni-like krypton. This finding is consistent with the fact that Ni-like ions are more stable against further collisional ionization than Pd-like ions. Nevertheless, the pressure dependencies of both laser sources are very similar in the sense that the lasing output first increases with the gas density being, however, maximized for quite low gas pressure. This can be qualitatively explained by considering the ionization-induced refraction [20] of the driving laser beam which does not permit one to create a sufficiently long medium for higher pressures. The refraction of the laser beam depends on the electron density profiles which are expected to be very similar for both Ni-like krypton and Pd-like xenon plasmas; eight electrons have to be removed from the neutral atoms. Therefore our experimental results suggest that the measured optimum pressure is the best compromise between high plasma density and laser propagation obtainable in this geometry.

Figure 4 compares intensity of the lasing line at 32.8 nm with one of the incoherent lines observed in

FIG. 3. Pressure dependence of the 32.8 nm 4*d*-4*p* line of Nilike Kr (solid dots) and the 41.8 nm 5*d*-5*p* line of Pd-like Xe (open dots).

FIG. 4. Driving energy dependence of the 32.8 nm 4*d*-4*p* lasing line of Ni-like Kr (solid dots) and the 29.6 nm incoherent line (open dots).

the spectrum, namely, the transition Kr^{8+} 3 $d^{9}5f^{(3}D_1)$ - $3d^94d(^1S_0)$ at 29.6 nm as a function of the driving energy. The data in Fig. 4 have been obtained using the 4.5 mm long gas cell, and a gas pressure of 20 Torr. While the incoherent line was always present in the spectrum, the lasing line completely disappeared for pumping energies below 360 mJ. For both emission lines the output signal monotonically increases with the driving energy up to 760 mJ with no sign of optimum. As expected, the 29.6 nm incoherent signal grows linearly due to volume effects. By contrast, the signal from lasing transition exhibits rapid nonlinear increase with the driving energy and thus clearly demonstrates that an effective prolongation of the krypton plasma column results in a larger amplification.

The absolute photon number has been estimated by comparing the relative intensities of the 41.8 and 32.8 nm lasing lines obtained during the same experiment, using a noncalibrated but identical x-ray camera, taking into account the filter transmission and the grating efficiency, and considering that the 41.8 nm laser emits up to 5×10^9 photons per shot as previously measured [7], we can reasonably estimate that the 32.8 nm laser source delivers up to $(2-3) \times 10^9$ photons per shot for a pumping energy of 760 mJ, a cell length of 4.5 mm, and gas pressure of 20 Torr.

Figure 5 shows the intensity of the 4*d*-4*p* line as a function of the length of gas medium. Note that we took into account the effect of gas streaming out of the gas cell which makes the effective interaction length longer than the nominal distance between the pinholes. No lasing has been observed for length shorter than 3 mm. By increasing the gas length from 3 to 3.5 mm, the lasing signal increased by a factor of \sim 40. Further increase of the cell length resulted in an enhancement of the output signal by one more order of magnitude before a saturation-like behavior has been observed. The experimental data have been fitted using the set of formulas presented in Ref. [7]

FIG. 5. Measured (squares) and calculated (line) intensities of the 32.8 lasing line as a function of the gas length.

in which saturation effects are taken into account. Good agreement between experimental results and calculations has been obtained for a gain coefficient of 78 ± 2 cm⁻¹ and a saturation intensity being reached for a length of 3.5 mm. This corresponds to a gain-length product at saturation (Gl) _s of about 27.

While the above gain coefficient is similar to the gain measured for the 5*d*-5*p* Pd-like xenon line [7], the gainlength product inferred from Fig. 5 seems to be overestimated for several reasons. First, comparing to the Pd-like xenon laser the detected output signal for Nilike krypton is too weak to correspond to such a high gain-length product. Second, the fact that no signal has been observed for lengths shorter than $3 \text{ mm } [(Gl)_s \approx 23]$ is not consistent with an expected output from so highly an amplified x-ray laser source. The last point clearly indicates that only an unknown fraction of the gas cell length participates in the process of amplification of the 4*d*-4*p* line and that a realistic estimation of the gainlength product is not possible at the moment.

For the modeling of our experiment, we have used a time dependent collisional radiative method [21] which considers collisional excitation and radiative decays between 207 LSJ coupled states with principal quantum numbers $n = 3$, 4, and 5. The atomic data necessary (level energies, radiative and collisional rates) has been calculated using the Cowan code [22]. The computational model determines the initial state of a 1D slice of plasma in a radial direction at the focal position of the driving laser by use of the tunneling rates of Delone *et al.* [23].We have used the Fokker-Planck code of Pert [24] to examine the production of the electron distribution through OFI and its subsequent relaxation towards the Maxwellian state through electron-electron collisions. The code also calculates inverse-bremsstrahlung heating of the electrons although the effect is found to be insignificant in this case. For the calculation of the collisional excitation rates, the thermal energy distribution is then described by a Maxwellian distribution, with $T_e = \frac{2}{3} \bar{\varepsilon}_e$ where $\bar{\varepsilon}_e$ is the mean electron energy resulting from the OFI process, of

the equivalent temperature. The consideration of all ion stage populations throughout the driving pulse results in an electron distribution with $\bar{\varepsilon}_e = 680$ eV. These calculations predict a peak gain of around 500 cm^{-1} at 8 ps following the driving laser. This large gain figure is predominantly due to the very narrow linewidth which results from the ions being left effectively at room temperature. These calculations are in reasonable agreement with those made by Lemoff *et al.* for lower gas densities [5]. To compare to our experimental results, it is more appropriate to consider the gain coefficient averaged over the total gain duration; the average gain is here close to 220 cm^{-1} , which is 3 times larger than the measured one. We can identify two main reasons for this difference: First, the experimental uncertainties in measuring the effective length and homogeneity of the plasma in the case of longitudinal pumping and, second, in the use of thermal distributions in the simulation which overestimate the pumping rates. Both of these issues will be addressed in future work. The same atomic code also allows calculations of the saturation intensity and predicts values of 9×10^6 W cm⁻² for Kr at the experimentally optimal pressures 20 Torr. This saturation intensity has to be compared with the calculated value of $2 \times$ 10^7 W cm⁻² for the 41.8 nm Xe laser for the optimal gas pressure of 15 Torr. The relative intensities of the Kr and Xe lasers measured in our experiments are thus in good agreement with the theoretical expectation at saturation.

By focusing a multi-TW Ti:sapphire laser pulse into a gas cell filled with krypton, we have experimentally demonstrated the first amplification of the 4*d*-4*p* line of Ni-like krypton at 32.8 nm. The most intense signal has been obtained using a 760 mJ, 30 fs circularly polarized laser pulse and a 4.5 mm long static cell filled with 20 Torr of krypton gas. A gain coefficient of 78 ± 2 cm⁻¹ has been inferred; however, the experimental data strongly indicate that the lasing signal has not been amplified over the full length of the gas cell. While the question on saturated amplification of the $3d^9 4d^1S_0 - 3d^9 4p^1P_1$ transition of Ni-like krypton laser still remains opened, our result presents the first ever extension of the OFI collisional x-ray laser technique to shorter wavelengths using Ni-like ions.

We thank Professor G. J. Pert at the University of York for the computational codes used in the numerical investigations. The authors acknowledge support of the European community in the access to the LOA facility under the Contract No. HPRI 1999-00086.f.

- [1] P. B. Corkum and N. H. Burnett, in *OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications*, edited by R.W. Falcone and J. Kirz (Optical Society of America, Washington, DC, 1988), Vol. 2, p. 255.
- [2] Y. Nagata *et al.*, Phys. Rev. Lett. **71**, 3774 (1993).
- [3] D.V. Korobkin, C. H. Nam, S. Suckewer, and A. Goltsov, Phys. Rev. Lett. **77**, 5206 (1996).
- [4] B. N. Chichkov *et al.*, Phys. Rev. A **52**, 1629 (1995).
- [5] B. E. Lemoff, C. P. J. Barty, and S. E. Harris, Opt. Lett. **19**, 569 (1994).
- [6] B. E. Lemoff, C. L. Gordon III, C. P. J. Barty, and S. E. Harris, Phys. Rev. Lett. **74**, 1574 (1995).
- [7] S. Sebban *et al.*, Phys. Rev. Lett. **86**, 3004 (2001).
- [8] H. Daido *et al.*, Opt. Lett. **21**, 958 (1996).
- [9] J. Nilsen and J. C. Moreno, Opt. Lett. **20**, 1386 (1995).
- [10] J. Zhang *et al.*, Science **276**, 1097 (1997).
- [11] F. Löwenthal, R. Tommasini, and J. E. Balmer, Phys. Rev. A **59**, 1577 (1999).
- [12] S. Sebban *et al.*, Phys. Rev. A **61**, 43810 (2000).
- [13] A. L. Osterheld *et al.*, Phys. Rev. Lett. **80**, 2825 (1998).
- [14] A. Klisnick *et al.*, J. Opt. Soc. Am. B **17**, 1093 (2000).
- [15] J. Dunn *et al.*, Phys. Rev. Lett. **84**, 4834 (2000).
- [16] A. Augst, D. D. Meyerhofer, D. Strickland, and S. L. Chin, J. Opt. Soc. Am. B **8**, 858–867 (1991).
- [17] B. E. Lemoff, Ph.D. thesis, Stanford University, 1994.
- [18] M. Pitmann, J. P. Rousseau, L. Notebaert, S. Ferre, J. P. Chambaret, and G. Cheriaux, in *Proceedings of the CLEO 2001 Conference, Baltimore, 2001* (Optical Society of America, Washington, DC, 2001), p. 72.
- [19] S. M. Hooker, P.T. Epp, and G.Y. Yin, J. Opt. Soc. Am. B **14**, 2735 (1997).
- [20] C. Decker, D.C. Eder, and R.A. London, Phys. Plasmas **3**, 414 (1996).
- [21] G. J. Pert, J. Phys. B **23**, 619 (1990).
- [22] R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California Press, Berkeley, CA, 1981).
- [23] N. B. Delone, V. B. Krainov, and M.V. Ammosov, Sov. Phys. JETP **64**, 1191 (1986).
- [24] G. J. Pert, J. Phys. B **34**, 881 (2001).