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Compact X-ray laser amplifier in the "Water Window"

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ABSTRACT

Spectroscopy and microscopy in the so-called "water-window" is a holy grail of modern molecular biology. A pulsed source of coherent X-rays within this spectral window, falling between 2.3 nm and 4.4 nm, provides a unique tool for time-resolved imaging of bio-systems in their naturally water-rich state. Within this spectral range, water is mostly transparent, while proteins are mostly opaque. This results in a high-contrast image on the sub-cellular level. Here we present, for the first time, generation of a very high gain of G≈ 60/cm in He-like CV ions via transitions to the ground state at 4.03 nm in a table-top device.

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1. Introduction

Water is a key ingredient in living systems. Indeed, it is properly said that blood is essentially water but "thicker than". Thus, it is important to develop and perfect spectroscopic and microscopic techniques which are not overwhelmed by water.

For example, infrared (IR) vibrational spectroscopy is often not useful in living – bio systems since the IR radiation is strongly absorbed by the water background. However, Raman spectroscopy is especially useful in probing living matter since the optical (pump and Stokes) probes are not strongly absorbed by water.

Prof. Wolfgang Kiefer, to whom we dedicate this paper, has made pioneering contributions to much of femtosecond chemical physics and in particular to problems involving Raman spectroscopy using femtosecond pulses.

In the present paper, we note that X-rays, which are of course ubiquitous in the study of bio-systems, are often strongly absorbed by water. Thus, it is of interest to develop probes involving lasers in the water window at 4.03 nm. This is the subject of the present paper and indeed a subject of research at our Princeton group over the last several decades [1–3]. It is a pleasure to note that this amplification of X-ray radiation uses femtosecond lasers to advantage even as Prof. Kiefer has pioneered their use in Raman spectroscopy.

In this paper, we present the development of very high gain in a compact X-ray laser (XRL) amplifier (Fig. 1) in the "water window" (WW) at 4.03 nm. This is achieved on the transition from excited state of n = 2 to ground state of n = l in He-like CV ions as shown in Fig. 2. A high-resolution compact microscope operating in the WW is a particularly promising application for the table-top XRL. This microscope enables time-resolved imaging of bio-systems in their natural environment (*i.e.* water), with spatial resolution down to nanometers. Additional experimental details will be presented elsewhere [4]. XRL application to the study of live cells was discussed in the past [2] and is a subject of active present work.

Fig. 1 illustrates the experimental setup for our XRL amplifier operating in the "water window" at 4.03 nm via singlet CV ions lasing to ground state of $2^1P – 1^1S$. With a 100 fs, 150 mJ/pulse drive laser beam, focused to 10 µm, we were able to obtain high peak intensity of ~2×10^{18} W/cm^2 in a 0.5–0.7 mm long, 10 µm diameter plasma channel, as required for a maximum gain of 60–70 cm^{-1}. As shown in Fig. 1, the initial plasma channel is created by using about 2 orders of magnitude lower beam intensities, with the same energy and pulse duration as for the high intensity beam but a correspondingly larger focal spot. Such a large-diameter initial plasma channel works to guide a much higher-intensity pumping beam with lower energy losses, as we have observed in multiple experiments on beam propagation in plasma waveguides.

The first demonstration of lasing to a ground state was in H-like LiIII in a rapidly-recombining plasma [3] using a 250 fs, 50 mJ pumping KrF laser with a pulse intensity of ~3×10^{17} W/cm^2. This intensity is required for high gain generation in LiIII at 13.5 nm for the n = 2 to n = 1 transition [5–8]. The initial plasma was created by a line focus of 1 J, 12 ns Nd-glass laser beam on a LiF slab. The pumping laser beams were focused on the slab by cylindrical lenses of various line-focal lengths for LiIII(2–1) line intensity measurements vs length yielding the gain value.

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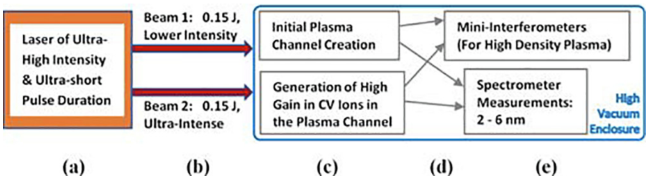

Fig. 1. Block Diagram of the experimental setup for generating high gain in CV ions at 4.03 nm, in a narrow plasma channel, by high intensity femtosecond-type laser beam. The laser beam energy of 150 mJ, pulse duration of 100 fs, focused into a 10 µm diameter channel, provides pumping intensity of 2×10^{18} W/cm^2 for generating the maximum gain of ~60–70 cm^{-1}. (a) A compact laser with ~0.5 J output pulse energy, providing pulses of ~100 fs duration, focusable to a maximum intensity of ~5×10^{18} W/cm^2; (b) Beam 1, at a relatively low intensity, serving to create the initial plasma channel; Beam 2, at an ultra-high intensity, for generating high gain in a small-diameter plasma channel; (c) The initial plasma channel + Gain generation in the narrow plasma channel; (d) Beam lines connecting to Applied Diagnostics; (e) Spectroscopic and Interferometric Diagnostics (All in a High Vacuum Enclosure).

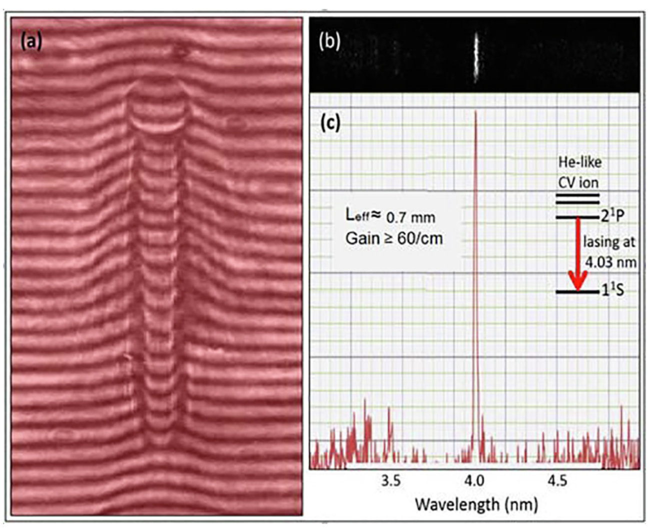

Fig. 2. Example of X-ray laser (XRL) high gain in "water window" at 4.03 nm; (a) Laser interferogram showing a phase profile of the plasma channel; (b) and (c): X-Ray Laser output spectrum with a strong laser line at 4.03 nm, in the "water window" [recorded by X-ray spectrometer on film (b), which scan is shown in (c)]. The inset in part (c) shows the level schematics in CV ions.

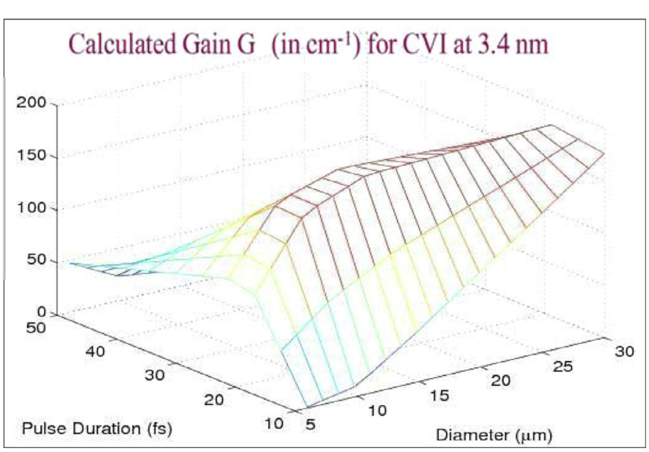

Fig. 3. Gain modeling in H-like CVI ions for 2-1 transitions at 3.37 nm; details of the gain modeling versus pump beam diameter and duration of pumping pulses can be found in [9].

However, generating gain in He-like CV at 4.03 nm requires two orders of magnitude higher pumping intensities than for LiIII, and one order of magnitude higher plasma densities [8,9]. Achieving such conditions experimentally with high gain is a difficult task, especially in creating a sufficiently long and reasonably uniform plasma waveguide ("plasma channel"). Such a plasma waveguide has been used to obtain gain-length product GL $\geq$ 4.5 for sufficiently large intensities of XRL pulses [2,9]. The corresponding X-ray pulse intensity would then be sufficient for generating multi pulses for high-resolution imaging of live cells.

In addition to the quest for ever shorter wavelength lasers, a major motivation for the present work is developing an XRL as the source of radiation for high resolution X-ray microscopy, allowing us to obtain images of live cells in their natural environment.

2. Computer simulation of the gain for the XRL in transitions to ground states

Recombination XRL schemes in transitions to the ground state (here the n = 2 to n = 1 transition), characterized by short lasing wavelength and high quantum efficiency, are particularly attractive. For these schemes the WW lasing is attainable for He-like CV and H-like CVI ions using pumping lasers with relatively low pulse energy but high peak intensity. The low pulse energy allows to, quite easily, operate such lasers at high repetition rates. Our computer simulations have shown the feasibility of achieving high gain with the recombination scheme in the WW with peak pump intensity of 2×10^{18} W/cm^2 for He-like CV ions and intensity of 6×10^{18} W/cm^2 for H-like CVI ions [2,9].

In the present experiment, the length of the high gain region is quite short, no longer than 0.7 mm for the XRL wavelength of 4.0 nm, as can be seen from the interferogram of Figure 2. This is due to the limit of propagation length of pumping laser pulses of ultra-high intensity and ultra-short duration. Moreover, in order to operate the pumping laser at high repetition rates with low pulse energy but of very high intensity, the plasma waveguide ("plasma channel") has to be of very small diameter, ~10 µm for 150 mJ pulse energy to reach intensity of 2x10^{18} W/cm^2 at the exit of the waveguide. In order to create a plasma channel of 0.7 mm length, with the above-mentioned parameters, we have used an axicon lens. However, because of the limitation of this approach to rather short channel length, we are considering using a relatively small Z-pinch (about 10 cm long), with a compressed magnetic field, creating an on-axis elongated and very narrow waveguide for propagation of the high intensity pumping pulses [10].

In relation to gain generation for XRL in WW, we should notice that free electrons, produced during ionization of atoms and ions, consist of two, energetically different, parts of the initial highly non-Maxwellian distribution function. In particular, high density of low-energy electrons, mostly in the range below 10 eV and up to 20 eV, play a crucial role in 3- body recombination, generating gain in H-like and He-like ions like CVI and CV respectively, at 3.37 nm and 4.03 nm transitions from n = 2 to the ground level n = 1. The other, much more energetic part of the electron distribution, significantly above 20 eV, practically does not contribute to the recombination process, hence does not contribute to the gain increase in CVI and CV ions. The role of these high-energy electrons for XRL generation is mostly negative, due to accelerating Maxwellization processes, which are shortening the "window of opportunity" for efficient XRL development.

Fortunately, these more energetic electrons quickly escape the narrow plasma channel shortly after ionization, due to their small electron-ion interaction cross section. After a short time of about 15–20 ps, the bulk of the electrons in the plasma channel "decay" to a Maxwellian electron distribution with a peak energy at about 250–300 eV. Therefore, very short and intense laser pulses are required in order to create a high electron density in the plasma with fully ionized carbon ions and with a highly non-Maxwellian electron energy distribution. The Optical Field Ionization (OFI) processes such as tunneling and multiphoton ionizations are capable of providing suitable conditions for generating high gain in H-like CVI and singlet He-like CV ions [5–9].

The OFI, due to short pumping pulses, does not create significant plasma heating. The main source of plasma heating during the OFI process is the above-threshold ionization (ATI) heating. This heating arises from the variation in the oscillation phase between the free electrons (resulting from the ionization of atoms and ions) and the phase of the laser electric field.

Further to the issue of recombination and gain generation: the recombination occurs here primarily, as was mentioned already, by a three-body process (electron–ion + electron), while recombination rates are proportional to the square of the electron density, N_e^2, and to the main quantum number n to power 4, n^4. Therefore, transitions from fully ionized carbon C^{6+} to H-like CVI (C^{5+}) occur primarily to the states with high n, while collisional and radiative transitions to level n = 2 are faster than to the ground level n = 1, which can lead to population inversion between excited states with $n \geq 2$ and the ground state, n = 1. Similarly, for creating population inversion in singlet He-like CV ions, C^{5+} primarily recombines to C^{4+} (CV), although some contribution to higher population inversion may occur due to collisional (not radiative) transitions between triplet and singlet states. Here we are primarily interested in the population inversion between states n = 2 and the ground state n = 1.

In order to minimize plasma heating, the pump pulses have to be ultrashort due to the exceptionally short radiative lifetime τ of the first excited levels of higher Z ions, since τ decreases as $1/Z^4$, where Z is the atomic number of the ions. In fact, the pump pulse duration has to be much shorter than the radiative decay time, because it also needs to be shorter than the collisional decay time at high N_e. Computer simulation has shown (see Fig. 3) that the gain can be enhanced and also become less stringently dependent on exactly matching the required experimental parameters if hydrogen is added into the carbon plasma [9]. This is because the ionization potential of hydrogen is much lower than that of highly ionized carbon ions; by adding hydrogen, we can introduce free electrons into the plasma much more efficiently than by increasing carbon density.

3. Applications of the XRL in the WW to high resolution microscopy of live cells

An amplifier with very high gain at 4.03 nm opens the path toward applications to high resolution microscopy of living cells in a thin layer of water in the X-ray range between Oxygen and Carbon Kα edges of 2.3 nm and 4.4 nm, respectively. This WW is the optimal region for high resolution imaging microscopy of live cells, where contrast is high and resolution is very good. Better resolution is possible to obtain at shorter wavelength, however due to much worse contrast at such wavelength the overall imaging quality would be lower.

The microscopy in the WW offers a new way to look at unaltered cells in their natural state. The absorption edges in the X-ray spectra of naturally occurring cell constituents provide sharp contrast without the addition of heavy metals to the cell as in the case of electron microscopy. This work begun at Princeton by using our first recombination laser at 18.2 nm, pumped by 300 J CO_2 laser (system 1) and in following the second system (system 2) pumped by significantly smaller, very intense KrF excimer laser. X-ray lasers with these pumping systems (system 1 and 2), provided similar images of dehydrated normal and cancer cells. The ultimate goal of our X-ray laser microscopy program is to obtain images of live cells. Some details of this as well as our other work with X- ray laser microscopy are described in [8].

Imaging of cancer and normal cells in their natural environment relies on increasing the XRL pulse energy. Our work is an important step in this direction, as we have demonstrated high gain at 4.03 nm for the first time in He-like CV ions. This accomplishment holds promise for a new kind of high resolution X-ray microscopy.

In the past we obtained images of the cancer and the normal cells using a contact microscope and our first recombination soft X-ray laser as the illumination source [2,8]. Although at 18.2 nm the laser wavelength was far away from WW, we could already use this system to obtain high resolution images of dehydrated the HeLa cancer and the normal cells. The weak point of that research was the necessity of using dehydrated cells due to relatively long wavelength of 18.2 nm. We will be able to enhance this research by obtaining images of live cells in a thin layer of water using an XRL with high gain at 4.03 nm.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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It is our pleasure to dedicate this work to Prof. Wolfgang Kiefer who has been a pioneer in femtosecond Raman spectroscopy and much more.

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References

[1] S. Suckewer, C.H. Skinner, H. Milchberg, C. Keane, D. Voorhees, "Amplification of Stimulated Soft-X-Ray Emission in a Confined Plasma Column", Phys. Rev. Lett. 55 (1985) 1753.
[2] S. Suckewer, C.H. Skinner, "Soft X-ray Lasers and Their Applications", Science 247 (1990) 1553.
[3] D.V. Korobkin, C.H. Nam, S. Suckewer, A. Goltsov, "Demonstration of soft x-ray lasing to ground state in Li III", Phys. Rev. Lett. 77 (1996) 5206.
[4] S. Suckewer, A. Morozov, A. Goltsov, A. V Sokolov, M.O. Scully, "Development of a Compact X-Ray Laser in the 'Water Window' at 4.0 nm", to be published (2021).
[5] L.V. Keldysh, Ionization In The Field Of A Strong Electromagnetic Wave, Soviet Phys. JETP 20 (1965) 1307.
[6] N.H. Burnett, P.B. Corkum, Cold-plasma production for recombination extreme-ultraviolet lasers by optical-field-induced ionization, J. Opt. Soc. Am. B 6 (1989) 1195.
[7] D. Attwood, Soft X-rays and Extreme Ultraviolet Radiation: Principles and Applications, Cambridge Univ. Press, 1999.
[8] S. Suckewer, P. Jaeglé, X-Ray laser: past, present, and future, Laser Phys. Lett. 301–326 (2009).
[9] Y. Avitzour, S. Suckewer, Feasibility of achieving gain in transition to the ground state of C VI, J. Opt. Soc. Am. **B** 24 (2007) 819.
[10] J. Rocca, H. Kapteyn, D. Atwood, M. Murnane, C. Menoni, E. Anderson, "Tabletop Lasers in the Extreme Ultraviolet", Opt. Photon. News 17 (11) (2006) 30–38.