

# Possibility of Recombination Gain Increase in CV Ions at 4.0 nm Via Coherence

Y. Luo, A. Morozov, D. Gordon, P. Sprangle, A. Svidzinsky, H. Xia, M. Scully and S. Suckewer

**Abstract** This paper is about the recent experimental results on amplification of the CV line in the “water window” at 4.03 nm from resonance transition to the ground level of He-like ions in recombination scheme. The indication of the amplification of the CV line has been observed when an elongated narrow plasma channel was created, where high intensity 100 fs beams, optimal for creating CV ions in high density plasma, was propagated up to 0.5–0.6 mm. Without channeling the effective plasma length was much shorter and there was no indication of amplification. The large interest in gain generation in He-like ions in the transition to ground state is due to the possibility of applying a recently developed theory of Lasing Without Inversion (LWI) in XUV and X-ray regions to largely increase the gain for such transitions. The presented results of the indication of CV line amplifications are being discussed from the point of view of using LWI as a superradiance gain increase, hence to construct a very compact soft X-ray laser in the “water window”. The last part of the paper is related to the application of the ultra-intensive fs plasma laser, which is currently in the process of development by using stimulated Raman backscattering (SRBS) to create a plasma amplifier and compressor, as the pump for compact laser operating in the “water window” and also at shorter wavelengths.

## 1 Introduction

Tremendous progress has been made in beam quality, compactness, and especially in the application of soft X-ray lasers (SXLs), since the first large gain-length products (gl) were demonstrated in the soft X-ray region nearly three decades ago. The qualities of SXL beams, reaching 10 nm and slightly below, have been improved in their coherence, divergence and stability (i.e. very good reproducibility from shot-to-shot) is presented in a number of recent and past reviews [1–9], including presentations at this conference (for example [10–12]).

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S. Suckewer (✉) · Y. Luo · A. Morozov · A. Svidzinsky · H. Xia · M. Scully  
Princeton University, Princeton, NJ 08544, USA  
e-mail: suckewer@princeton.edu

A. Svidzinsky · H. Xia · M. Scully  
Texas A&M University, College Station, TX, USA

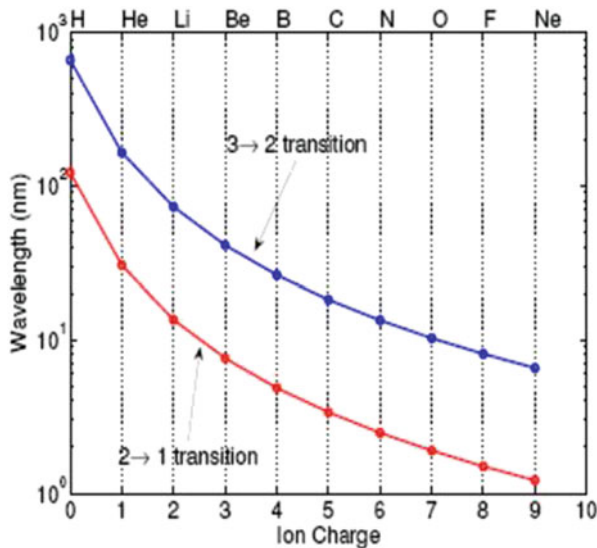
D. Gordon · P. Sprangle  
Naval Research Lab, Washington DC, USA

Special attention has been directed toward SXL compactness. The development of the table top SXLs, which will have a number of practical applications in science and technology, has become a high priority for many researchers in the field. Of course, the compactness of SXL depends on the efficiency of the lasing system (especially quantum efficiency) and the pumping efficiency. For the maximum quantum efficiency, systems in which lasing is in transition to ground state are very attractive. As for pumping efficiency, longitudinal pumping usually is more efficient than transverse pumping.

To realize lasing to the ground states of ions in the soft- and x-ray regions, ultra-short laser pump pulses have to be used in order to provide population inversion between the excited and ground states in ions. With the development of very high intensity fsec laser pulses it becomes possible to realize recombination lasing to the ground states in ions, which are first stripped of electrons by optical field ionization, OFI.

Achieving gain using recombination scheme is highly desirable in the pursuit of x-ray lasers due to requiring relatively low pumping pulse energy. This, combined with the high quantum efficiency achieved by using the transition to the ground state, makes the creation of a practical tabletop x-ray laser (XRL) quite feasible. Furthermore, the highly favorable scaling of the required pumping energy with decreasing the wavelengths may enable reaching the “water window” (the wavelength range 2.3–4.4 nm, for which absorption in water is low) with a tabletop x-ray laser system. Hence, such XRL would be an excellent source for high resolution X-ray microscopy of biological cells in their natural environment, placed in a relatively small biological laboratory.

The highly efficient recombination system utilizing transition to ground states of ions was theoretically predicted [13, 14], and experimentally high gain was demonstrated in Hydrogen-like Li III in a transition to the ground state ( $2 \rightarrow 1$  transition) at 13.5 nm [15, 16] and explained theoretically [17] with added computer simulation of experimental results. This computer simulation takes into account the uniqueness of the OFI processes such as the non-Maxwellian nature of created plasma and its relatively low temperature, which is a very important feature for generating high gain in fast recombining ions in plasma. For example, the experiment on obtaining high gain in the  $2 \rightarrow 1$  transition in LiIII ions has been an important benchmark for computer simulations. Better efficiency of the gain generation using  $\sim 0.25 \mu\text{m}$  pumping wavelength in [15] in comparison to  $\sim 1.0 \mu\text{m}$  in [16], both experiments use pre-plasma in microcapillaries for waveguiding pumping beams at  $\sim 3 \times 10^{17} \text{ W/cm}^2$  intensities, was verified by computer modeling. Furthermore, by investigating the effects of different laser and plasma parameters on the gain generation at various lasing wavelengths, the optimal conditions required to achieve high gain in CVI ions in  $2 \rightarrow 1$  transition at 3.4 nm were predicted [18], providing a “prescription” for designing and conducting experiments.

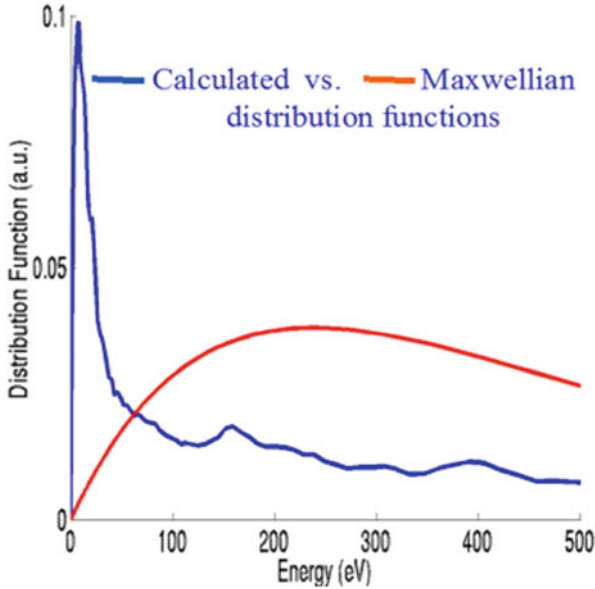


**Fig. 1** Wavelength scaling of recombination schemes for  $2 \rightarrow 1$  (in red) and  $3 \rightarrow 2$  transitions for H-like ions

## 2 Remarks About Recombination XRL: Rapid Convergence to Short Wavelengths

The recombination scheme is attractive due to its fast convergence to short wavelength of  $3 \rightarrow 2$  and  $2 \rightarrow 1$  transitions in H-like ions as illustrated by Fig. 1. Especially attractive are the transitions to ground state ( $2 \rightarrow 1$  transitions) with their high quantum efficiency and for which the “water window” is attainable for CV and CVI ions using pumping lasers with relatively low energy, but at high intensity and repetition rate. Our extensive computer calculations have shown the feasibility of achieving high gain with the transient recombination scheme in the “water window” with peak pump intensity in the order of  $I_p \sim 10^{18} - 10^{19}$  W/cm<sup>2</sup> and duration of pulses less than 100 fs for He-like ions CV (lower intensity range) and H-like ions CVI (higher intensity range). The crucial aspect of the generation of high gains for these transitions is a strong non-Maxwellian electron energy distribution function. Such distribution function, obtained by computer simulation of Optical Field Ionization, OFI, in the process of creating a fully ionized carbon, is shown in Fig. 2 from [18]. These computer calculations indicate that most of the electrons from the fully stripped carbon, have very low kinetic energies in the order of 10–20 eV, in comparison to the  $\sim 400 - 500$  eV ionization energies of the CV and CVI ions.

Electrons in the high-energy tail of the distribution function, due to their high velocities and low cross section in interaction with ions at such velocities, can quickly escape the narrow plasma channel without participating in the recombination processes. After a short time, in the order of a few picoseconds, these electrons



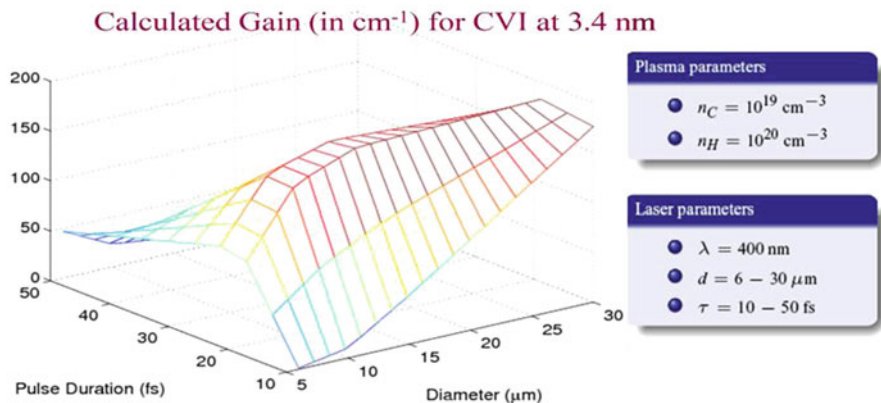
**Fig. 2** Calculated highly non-Maxwellian electron distribution (in *blue*) after fully stripped carbon ions off electrons by means of OFI; corresponding Maxwellian distribution is shown (in *red*)

proceed to thermal equilibrium, which reach Maxwellian electron distribution in about 15–20 ps with it maximum energy at about 250–300 eV, much too high for effective three-body recombination process.

Therefore, the very short and very intensive pulses can create fully ionized, high density carbon plasma with very non-Maxwellian electron energy distribution, leading to a 3.4 nm gain generation in CVI followed by 4.03 nm gain in CV. The gain generation in CVI takes place in a very short time, hence potentially leading to a very short lasing time, during which the ground state of CVI would rapidly populate, followed by its fast recombination to highly excited states of CV. Due to this process gain in the singlet states of the CV ions is expected to be obtained, while the electrons are still highly non-Maxwellian.

### 3 Predicted Gain for $2 \rightarrow 1$ Transition in CVI Ions and Experimental Setup

**Gain** generation in H-like ions, as we have already indicated, requires the atoms to be totally stripped of electrons, as the first step. Next, these atomic nucleus in the plasma have to recombine very fast by three-body, non-radiative process (nucleus + 2 electrons), providing H-like ions in highly excited states (three-body recombination is approximately proportional to main quantum number  $n$  to



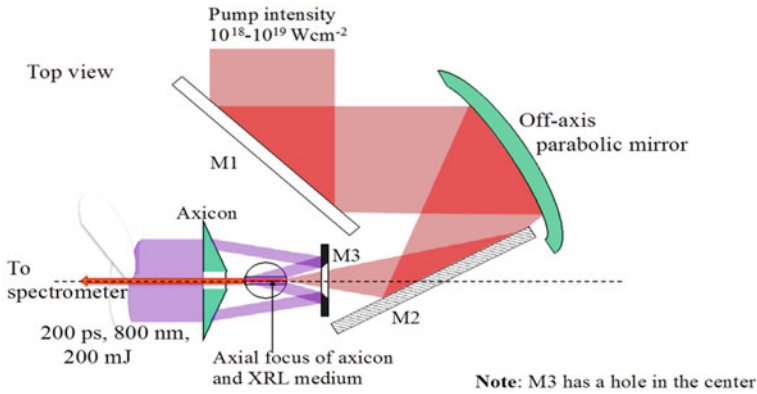
**Fig. 3** Predicted gain (from [18]) in H-like CVI at 3.4 nm as a function of pumping beam diameter (in  $\mu\text{m}$ ) and its pulse duration (in fs) for initial carbon and hydrogen densities  $n_C = 10^{19} \text{ cm}^{-3}$  and  $n_H = 10^{20} \text{ cm}^{-3}$

power 4). Moreover, the electron densities have to be high (in range  $10^{17}$ – $10^{20} \text{ cm}^{-3}$ , depending on the nucleus charge  $Z$ ), and in order to create a  $2 \rightarrow 1$  population inversion, the ionizing pulses have to be of very high intensity and very short duration. Short pumping pulse duration prevents significant plasma heating, which the main source for OFI process is the residual heating, or the above threshold ionization (ATI) heating. This heating arises from the variation in the oscillation phase  $f_d$  between the free electrons from the ionization of atoms and ions and the phase of the laser electric field. The average residual energy is proportional to the quiver energy of the electrons in the laser field,  $\varepsilon_q = e^2 E^2 / 4m_e v^2$ , where  $e$  is the electron charge,  $E$  is the laser peak electric field,  $m_e$  is the electron mass and  $v$  is pumping laser frequency.

Because three-body recombination rate is proportional to the square of the electron density,  $N_e^2$ , and quantum number  $n$  to power 4, the transition from fully ionized carbon,  $\text{C}^{6+}$ , to H-like CVI occurs primarily to the states with high  $n$ , while collisional and radiative transitions to level  $n = 2$  are faster than to ground level  $n = 1$ , creating a population inversion between  $n = 2$  and  $n = 1$ . In addition to minimizing plasma heating, the ultrashort pumping pulses are also necessary due to the very short radiative lifetime,  $\tau$ , of the first excited levels of ions and decreasing this time as  $Z^{-4}$  ( $Z$  is atomic number of ions). In fact, this time has to be much shorter than the radiative decay time in order to be shorter than the collisional decay time at high  $N_e$ . It was also shown that the gain can be enhanced and become less stringently—dependent on exactly matching the required experimental parameters, if hydrogen is added into the carbon plasma.

The gain (in  $\text{cm}^{-1}$ ), which was obtained by computer calculations [18], is shown in Fig. 3 for  $2 \rightarrow 1$  transition at 3.4 nm in CVI for the range of pumping beam intensity  $I \approx 7 \times 10^{18}$ – $1 \times 10^{19} \text{ W/cm}^2$ , where the lower intensity was for the 50 fs pumping pulses and the higher intensity was for the 10 fs pulses. The initial carbon

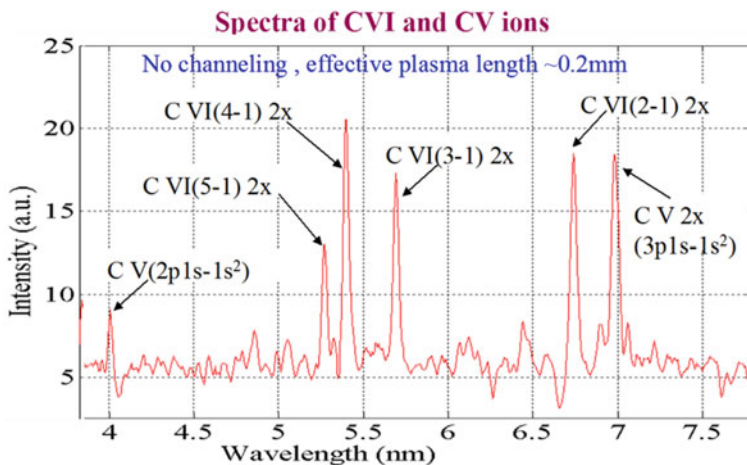
## Pumping and X-ray beams with optics in XRL chamber



**Fig. 4** Experimental setup

density was  $n_C = 10^{19}\text{ cm}^{-3}$  with added hydrogen density of  $n_H = 10^{20}\text{ cm}^{-3}$ . The gain is presented as a function of the pumping beam diameter and its pulse duration. It is apparent that the maximum gain,  $g \approx 150\text{--}180\text{ cm}^{-1}$ , is obtained for pulse duration of 20–30 fs, and this gain is also less dependent on the pumping beam diameter. For pumping pulses  $\sim 50$  fs and up to 100 fs the predicted gain drops to  $50\text{ cm}^{-1}$  and below, but still is quite high.

**Experimental setup**, which is shown schematically in Fig. 4, was designed following computer gain predictions for optimal laser pulses and plasma medium parameters as well as gained experience with development high gain in  $2 \rightarrow 1$  transition in H-like LiIII ions [15]. The multipass Ti:Sa amplifier provided 2 J, 800 nm 200 ps pulses at 5 Hz repetition rate. The pulse was optically compressed down to 100 fs and was focused by an off-axis parabolic mirror ( $f = 150\text{ mm}$ ) into the pre-formed plasma in the gas jet. The plasma was initiated by an optical breakdown using part of a 200 ps pulse, splitted before the compressor. It was focused by an axicon lens in order to create a pre-formed plasma waveguide, whose purpose was to enable the propagation of the ultra-intense pump pulses over a much longer distance than its Rayleigh length, pioneered by Milchberg and his group [19, 20] and provided computer modeling by Sprangel et al. [21]. In these experiments, the near diffraction-limited focus of the pulse was achieved and enabled reaching  $2 \times 10^{19}\text{ W/cm}^2$  intensities with 300 mJ of pulse energy. The extensive diagnostics were implemented to monitor absolute intensity distribution of the 100 fs pulses in the plasma channel, the electron density distributions as well as the neutral gas density in the gas jet was monitored using a shear-type of interferometer with time and spatial resolutions of 200 fs and  $\sim 5\text{ }\mu\text{m}$ , respectively. The spectra in “water window” region in direction of fs beam propagation in plasma were recorded using a grazing incidence flat-field spectrometer having a diffraction grating of 1200 grooves/mm with a variable density of grooves. The spectra were registered using a  $1024 \times 1024$  back illuminated CCD camera. Two  $150\text{ }\mu\text{m}$  stainless steel pinholes were placed



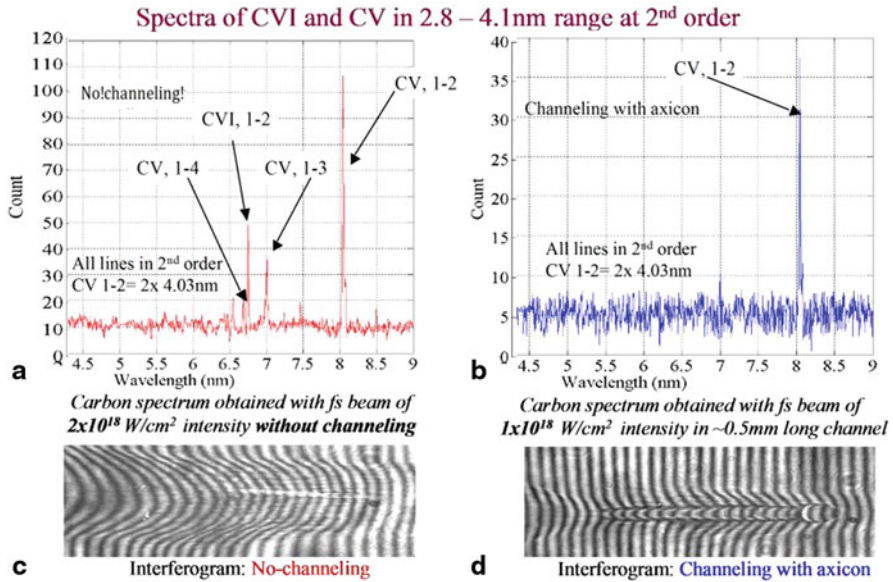
**Fig. 5** Spectra of CV and CVI in “water window” wavelength range created with 100 fs beam without channeling and with pulse energy/intensity: 200 mJ/ $\sim 4-5 \times 10^{18}$  W/cm<sup>2</sup>, effective plasma length was:  $l_{\text{eff}} \sim 0.2$  mm, gas jet mixture: 20% of C<sub>2</sub>H<sub>6</sub> and 80% of H<sub>2</sub> note: notation 2x, for example in CVI(2 – 1) 2x, indicates *second* order of CVI(2 – 1) line

between the spectrometer slit and the plasma to reduce the amount of visible light from the plasma and stray light from the laser from reaching the detector, very significantly improving signal-to-noise ratio in the recorded spectra.

#### 4 Recorded CV and CVI Spectra in “Water Window”

The spectra were recorded in the wavelength range of 2.3–4.4 nm, covering the “water window”, in search for conditions to generate gain in transitions  $2 \rightarrow 1$  at 3.4 and 4.0 nm in CVI and CV ions, respectively. For this purpose the spectra were taken along the propagation path of the laser pulses, perpendicularly to the gas jet’s nozzle slit and both the neutral gas density and the plasma density were monitored simultaneously in a single shot. The example of CV and CVI line emissions in “water window” wavelength range is shown in Fig. 5 for tightly focused 100 fs beam onto thin plasma layer of the effective depth  $l_{\text{eff}} \approx 0.2$  mm without channeling. The plasma was created in a gas jet mixture of 20% C<sub>2</sub>H<sub>6</sub> and 80% H<sub>2</sub>. The CVI lines dominate the spectra for these pumping laser pulses of intensities in the range of  $\sim 4-5 \times 10^{18}$  W/cm<sup>2</sup> for 200 mJ pulse energy, although the CV ( $3p-1s^2$ ) in second order is intensive. Moreover, the intensity ratio of CVI (4 – 1)2x to CVI (3 – 1)2x indicates quite strong population inversion between levels four and three when taking into account  $A_{41}g_4/A_3g_3 \approx 0.2$ , where  $A_{nm}$  are spontaneous emission coefficients from upper level  $n$  to lower level  $m$ , and  $g_n$  are statistical weights of levels  $n$ . Similar population inversion was observed for levels three and two in CVI





**Fig. 6** **a** Carbon spectrum using only a fs beam (no channeling), the pulse energy: 150 mJ, gas jet mixture: 20% of  $C_2H_6$  and 80% of  $H_2$ ; **b** Carbon spectrum with channeling (waveguiding with axicon), energies of channeling and fs beams were 150 mJ each, gas jet mixture as in (a); **c** Interferogram of plasma created by 100 fs laser pulses with 150 mJ pulse energy; **d** Interferogram of plasma created by the 100 fs laser pulses propagated in pre-formed plasma waveguide, which was created by axicon *line*-focused of 200 ps pulses with 150 mJ pulse energy; the delay between fs and ps pulses was 1.2 ns, gas jet mixture was as in (a). The laser pulses propagate from *right to left*

and CV, when longer wavelengths were monitored to see simultaneously all these lines in second order.

For lower pumping laser intensities the He-like CV lines became dominant. For example, for pumping intensity of  $\sim 2 \times 10^{18}$  W/cm<sup>2</sup> with pulse energy  $\sim 150$  mJ and the same gas jet mixture as in Fig. 5, the spectrum in Fig. 6a is dominated by CV lines, which is reasonable considering that the peak intensity is sufficient to create predominately CV ions but much less CVI ions. Such dominance of CV ions was even more pronounced when the axicon created plasma channel has been used to enable propagation of the fs pulses in longer plasma, as shown in Fig. 6b, while the intensities of fs pulses were twice lower than in Fig. 6a. Under this condition, only the line from the CV  $2 \rightarrow 1$  transition could be observed. This indicates the encouraging possibility of achieving lasing in  $2 \rightarrow 1$  transition in CV ions at 4.03 nm, which is within the “water window”. It is very interesting and provides additional encouragement for reaching our goal that the spectrum in Fig. 6b is very similar to the spectrum of H-like LiIII in Fig. 1 in [16], which was recorded in a series of experiments leading to high gl for  $2 \rightarrow 1$  transition at 13.5 nm.



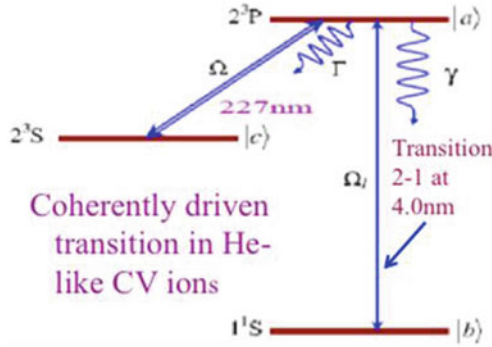
The recording spectra shown in Fig. 6a, b and the neutral particles and plasma densities were simultaneously monitored with an interferometry. The interferogram of the plasma created by a single ultra-intense 100 fs pulses with 150 mJ pulse energy taken immediately after it passed through the jet is shown in Fig. 6c. Although the focal spot of the fs pulse had a diameter of  $\sim 5 \mu\text{m}$ , the resulted plasma was over 200  $\mu\text{m}$  wide presumably due to the ionization-induced refraction and other nonlinear effects during the propagation of the fs pulse with effective plasma length  $l_{\text{eff}} \approx 0.2 \text{ mm}$ .

The interferogram of the plasma created by propagating the same 100 fs pulse, but through the preformed plasma waveguide is shown in Fig. 6d. The 200 ps pulse had 150 mJ energy and arrived at the gas target 1.2 ns prior to the arrival of the fs pulse. The gas target used in this set of data was the same as in Fig. 6a. Diluting the  $\text{C}_2\text{H}_6$  gas with  $\text{H}_2$  lead to two advantages: (1) to suppress parametric instabilities during the ionization by the Bessel beam and (2) to provide larger density of low energy electrons due to much lower ionization potential of hydrogen atoms than that of CVI and CV ions, which is favorable for fast three-body recombination of  $\text{C}^{6+}$  to CVI ( $\text{C}^{5+}$ ). The pre-formed plasma waveguide effectively confined the propagating the fs pulse maintaining its high intensity over distance  $l_{\text{eff}} \approx 0.5 \text{ mm}$ , apparently already having a very important effect on the intensity of the CV(2 – 1) line in Fig. 6b spectrum.

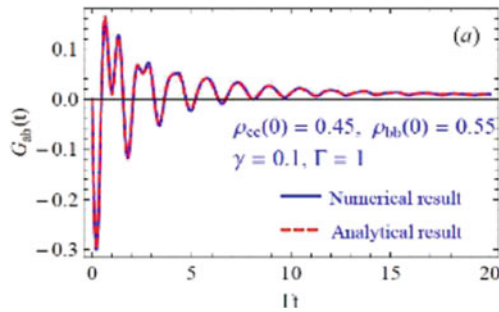
## 5 Possibility of Gain Increase in CV (2 $\rightarrow$ 1) Transition at 4.0 nm Via Quantum Coherence

Recent work on quantum coherence control of transitions in He atoms and He-like ions has predicted the possibility of generating lasing action in the XUV and X-ray regions via superradiance radiation enhancement and even generation gain without population inversion [22–24]. In general, the quantum coherence phenomenon in atomic and radiation physics has led to many interesting and unexpected results. For example, by preparing an atomic system in a coherent superposition of states, under certain conditions, it is possible for atomic coherence to cancel absorption but not emission, which is the basis for lasing without inversion, LWI. Frequently this is accomplished in three- or four-level atomic systems in which there are coherent routes for absorption that can destructively interfere, thus leading to the cancellation of absorption. Moreover, it was shown that when the excitation of an ensemble of atoms is swept in the direction of lasing (gain-swept superradiance, GSS), so that atoms are prepared in the excited state just as the radiation from previously excited atoms reaches them and yields superradiant emission [25, 26], which has common features with Dicke superradiance [27] for three-level system and can yield soft X-ray lasing in, for example, He-like ions of  $\text{C}^{4+}$  at 4.0 nm, schematically presented in Fig. 7.

It can be seen from Fig. 7 that strongly driving (with high Rabi frequency  $\Omega$ ) transition  $|a\rangle \rightarrow |c\rangle$  in CV ions make feasible to coherently stimulate transition to



**Fig. 7** Illustration of quantum coherence application to gain increase in recombination scheme for transition to ground state of He-like CV ions at wavelength of 4.0 nm



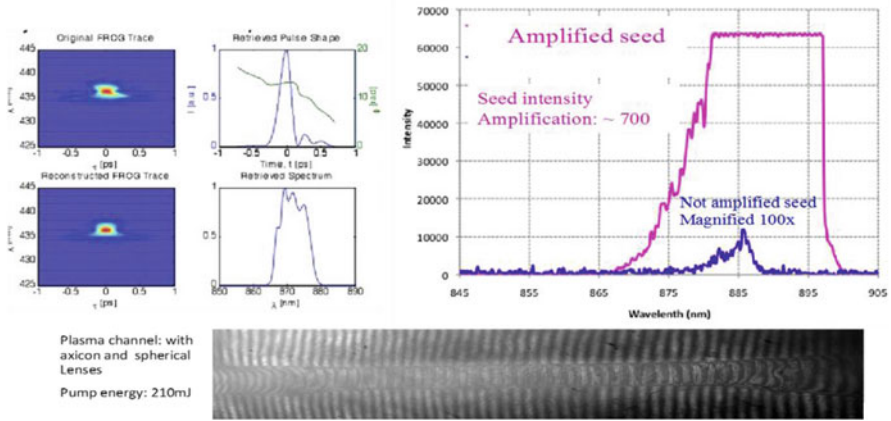
**Fig. 8** Plots from [25] of dimensionless gain/absorption in the lasing transition  $|a\rangle \rightarrow |b\rangle$  vs dimensionless time  $\Gamma t$ . *Blue* and *red* curves show, respectively, numerical and analytical results, where total decay and radiative decay frequencies of *upper state*  $|a\rangle$  are  $\Gamma = 1$  and  $\gamma = 0.1$

ground state  $|a\rangle \rightarrow |b\rangle$  with superradiance radiation and possible LWI at 4.0 nm, as it is illustrated in Fig. 8. The calculations presented in Fig. 8 were performed for total and radiative dimensionless decay frequencies  $\Gamma = 1$  and  $\gamma = 0.1$  (normalization factor is  $\Gamma = 4.5 \times 10^7 \text{ s}^{-1}$ ) of upper state  $|a\rangle$ . The plots for analytical and numerical calculation results indicate excellent agreement between both of them and are very encouraging for significant gain increase for 4.0 nm radiation.

## 6 Ultraintense Femtosecond Plasma Laser for Pumping XRLs

This last section of the paper is related to the application of the ultraintense fs plasma laser, which is currently in the development process at Princeton, as the pump for compact XRLs operating in the “water window” and also at shorter wavelengths. The development of the ultraintense fs plasma laser, in which the large and expensive optics for pulse amplification and compression will be replaced by plasma

SRBS Amplifications and Spectra with Large Diameter Plasma Channels



**Fig. 9** Seed intensity amplification of  $\sim 700$  and with a relatively low pump energy of 210 mJ in the plasma channel. Interferogram shows very good channel  $\sim 4$  mm long, although effective length of  $\sim 2.5$  mm was used here. The figure on the right shows the input and output seed spectra (input seed is magnified 100x to be visible, whereas the amplified seed saturated detector is due to insufficient signal attenuation. In the left low corner the interferogram shows a plasma channel of good quality, and at the left the time profile of output amplified seed from single pulse autocollimator FROG measurement is shown.

column, is based on stimulated Raman backscattering (SRBS) for creating a plasma amplifier and compressor. This experimental research was conducted for a number of years [28, 29] and was preceded by extensive theoretical research [30–32].

The very encouraging earlier results of seed intensity amplification of 10,000 and its compression from 550 fs to  $\sim 90$  fs in a single pass, and increases this intensity by 2x and further decreases the pulse duration down to  $\sim 50$  fs in the two passes experiment [33], stimulate present research towards practical fs plasma laser. With a much larger plasma column diameter of  $\sim 400\text{--}500 \mu\text{m}$  for waveguiding large diameter pump and seed pulses, which diameters of  $200\text{--}250 \mu\text{m}$  are expected to ensure a compact plasma laser with intensities at the focus of  $\sim 10^{20} \text{ W/cm}^2$ , the ideal pump for XRLs in the “water window” and down to a wavelength of  $\sim 1\text{--}2$  nm.

Figure 9 shows recently obtained, still preliminary, seed intensity amplification in the order of  $\sim 700$  using large diameter seed and pump pulses, propagating in a corresponding large diameter plasma channel, as discussed above. The pump energy of 210 mJ, hence its intensity, was still much below the required value to reach amplified output seed intensity in focus in the order of  $\sim 10^{20} \text{ W/cm}^2$ , which is one of the main goals of the present research.

**Acknowledgements** We are thankful to Q.Chen and N.Tkach for their help with the experiments. The research was supported by NSF/Phys Grant 1068554, NSF/DOE Grant 1064465 and DOE/NSA Grant DE-FOA-0000611

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