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Theoretical investigation of an ultrashort-pulse coherent x-ray source at 45 Å

S. J. Moon and D. C. Eder
Lawrence Livermore National Laboratory, Livermore, California 94550

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A scheme to obtain ultrashort-pulse coherent x rays, via population inversion following inner-shell photoionization, is analyzed for C at 45 Å. We calculate that a driving laser with a pulse duration of 40 fs, a 10 μm×1 cm line focus, and an energy of 1 J gives an effective gain length product (gl) of 10. At saturation ($gl \sim 18$) we expect an output of ~ 0.1 μJ per pulse. The short duration of x-ray lasing (<100 fs) combined with a 10-Hz repetition rate ($P_{\rm avg} = 1$ μW) makes this source of coherent x rays ideal for pump-probe experiments to study fast dynamical processes in chemistry and material science. [S1050-2947(98)05902-2]

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X-ray lasers driven by large optical lasers ($E \sim 1$kJ) with a pulse duration of order 100 ps have been generated over a wide range of wavelengths. Saturation has been obtained for wavelengths as short as 73 Å in Ni-like Sm [1]. Saturated x-ray lasers, at this wavelength and longer, can be used for single shot applications, e.g., interferometry in hohlraums [2]. While lasing has been demonstrated at 45 Å using this approach [3], saturation at this wavelength has not been achieved and only μJ pulses have been generated. However, such energy per pulse is sufficient for many applications provided one has a reasonable repetition rate. The large optical lasers used in conventional x-ray lasers do not have a sufficient repetition rate.

Within the last 5 years there have been major advances in obtaining high optical intensity through chirped pulse amplification. Pulse duration as short as 20 fs [4,5] and powers >1 PW, with longer pulse duration (~ 500 fs), have been demonstrated [6]. The repetition rate for these ultrashort-pulse (USP) lasers depends on the total energy per pulse. Lasers with energies of order 1 J have been operated at repetition rates of 10 Hz. Using an USP driver, x-ray lasing at long wavelengths ($\lambda = 326$ and 418 Å) [7,8] has been demonstrated and there has been some evidence for lasing at somewhat short wavelength ($\lambda \sim 135$ Å) [9–11]. However, the pulse duration at $\lambda \sim 135$ Å is greater than 10 ps, making it too long for many applications. For wavelengths down to ~ 67 Å [12], there is a mechanism to produce short duration ($\leqslant 100$ fs) coherent x rays by using high harmonics. Experiments in material science have shown the usefulness of such a short pulse of coherent x rays [13]. However, high harmonics have a very low conversion efficiency at short wavelengths [14]. In this paper we discuss the inner-shell photoionized x-ray lasing scheme. This scheme promises both a short wavelength and a short pulse source of coherent x rays with high average power.

Inner-shell photoionization (ISPI) x-ray lasing is a very attractive approach to short wavelength lasing ($\lambda < 50$ Å). In this approach, an incoherent x-ray source with a fast rise time is used to selectively ionize inner-shell electrons of the lasant material. The scheme was originally proposed by Duguay and Rentzepis [15] but problems of collisional ionization associated with the relatively long pulse optical lasers available at that time caused x-ray lasing to never be realized. Alternatives to this approach have been proposed [16–18] that require additional pumping or Auger transitions to populate the upper laser state. More recent work, using an assumed blackbody source with a specified rise time, done by Kapteyn [19] and Strobel *et al.* [20] concentrated on very short wavelengths, $\lambda \leqslant 15$ Å, where x-ray lasing has not been demonstrated. We present results, using the conventional ISPI scheme, for C at 45 Å as a representative low-Z element where lasing can be tested using current high energy USP lasers. Lasing at 45 Å requires less energy, at least an order of magnitude less than for 15 Å.

As seen in Fig. 1 an x-ray source creates a K-shell hole in C creating C$^+$ where an allowed K to L ($2p-1s$) radiative transition can take place yielding a 45-Å photon. For x rays above the K-shell energy only a small fraction $\leqslant 5\%$ of the ionizations are out of the L shell. Therefore the possibility arises for inversion between C$^+$ states with K- and L-shell holes. The dominant decay channel out of the upper-laser state is a nonradiative Auger transition to C^{++}, with a lifetime of 10.3 fs compared to the radiative lifetime of 293 fs [21]. Both the photoionization and the Auger transition produce a population of energetic free electrons. These electrons can ionize an L-shell electron in the neutral atom and populate the lower-lasing state. Consequently, the pump must have a fast rise time to achieve significant population inversion before electron ionizations destroy the inversion. We can estimate the requirement of the incoherent x-ray source

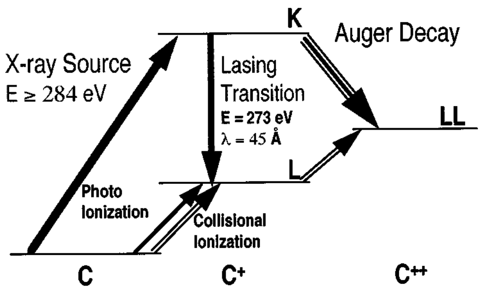

FIG. 1. Energy level diagram for inner-shell photoionization x-ray lasing in C at 45 Å. A high-energy x-ray photon can preferentially photoionize an inner-shell electron creating a K-shell hole where a K-L shell transition can take place.

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rise time and intensity by simple arguments. Current USP laser systems can operate at high power with sub-100-fs pulses. The rise time of the x-ray source is determined primarily by the rise time of the USP pump. Assuming a rise time, collisional ionization considerations restrict the neutral density, which combined with a desired gain coefficient determines the needed intensity. The time scale of collisional events is given by $\tau \sim 1/N_0 \sigma_{Le} v$, where N_0 is the neutral density, σ_{Le} is the L-shell electron ionization cross section [22,23], and v is the average electron velocity. To have a time scale greater than 50 fs requires a neutral density of less than $10^{20}\ \mathrm{cm}^{-3}$, using a representative electron thermal velocity corresponding to 100 eV. This density combined with the pump rate (cross section [24] times x-ray flux), $\sigma_K \Phi$, and the Auger lifetime, τ_{Auger}, enable us to estimate the required source intensity. To do so, we assume that lasing takes place before any collisional effects occur and that photoionization of the L shell by the filtered x-ray source is negligible. We can approximate the lower-lasing state as empty (numerical calculations below do not make this assumption) and the laser gain as

$$g = \sigma_s N_U \approx \sigma_s (\sigma_K \Phi N_0 \tau_{\mathrm{Auger}})$$

where σ_s is the stimulated emission cross section. Absorption of the lasing transition scales linearly with lasant density and will reduce the gain by $4.5\ \mathrm{cm}^{-1}$ for a neutral density of $10^{20}\ \mathrm{cm}^{-3}$. This absorption leads to ionization of the L shell, yet the photoionizations of the $2s$ electrons are dominant, leading to a small depletion of the neutral density and very little additional population of the lower-lasing level. An effective gain coefficient, accounting for absorption, of $10\ \mathrm{cm}^{-1}$ requires an x-ray flux of $\sim 10^{29}$ photons/(sec cm^2) assuming a neutral density of $10^{20}\ \mathrm{cm}^{-3}$ and using one-half of the peak K shell cross section, σ_K, to account for a broad band incoherent x-ray source (assuming an average photon energy ≈ 400 eV). Thus, we have shown in order to limit collisional ionization of the lower-laser state, with an assumed source rise time of 50 fs, a neutral density of $\sim 10^{20}\ \mathrm{cm}^{-3}$ is required. When coupled to the desire to have gains $> 10\ \mathrm{cm}^{-1}$ this leads to the requirement that the x-ray source be $> 10^{29}$ photons/(sec cm^2) or $> 10^{13}$ W/cm^2 for representative photons above the K shell. For a faster rising source there will be a reduction in the required intensity associated with a higher allowed neutral density.

A laser heated high-density plasma can be used as an incoherent x-ray source to provide the needed flux and rise time of x rays determined above. To estimate the input laser energy needed to heat the plasma and the output energy on saturation, we assume a line focus of $10\ \mu\mathrm{m} \times 2$ cm. The x-ray flux calculated above of 10^{13} W/cm^2 with a duration of 50 fs translates into a requirement of 10^{-3} J of incoherent x rays near the K-shell energy. Detailed calculations show that a laser produced plasma produces a broad band x-ray source with an efficiency of 5% while only 2% of these x rays are at energies with appreciable K-shell cross section. Our simple estimate assumes no population in the lower-laser state but detailed calculations show that including the lower state results in a factor of approximately 2 higher flux requirement. This results in conversion efficiencies of 5×10^{-4} or ~ 2 J of laser energy. We calculate a saturation intensity, $I_{\mathrm{sat}} = 4 \times 10^{12}$ W/cm^2, with a gain length product (gl) of 18. Using a gain duration of ~ 50 fs and an area of $10\ \mu\mathrm{m} \times 10\ \mu\mathrm{m}$ yields an x-ray laser energy output of $\sim 0.2\ \mu$J.

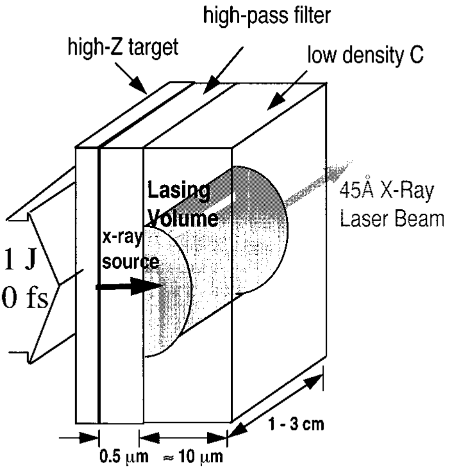

FIG. 2. The required large flux of x rays is obtained from a high-Z target heated by a high-intensity ultrashort-pulse laser. The incoherent filtered x-ray source creates a population inversion in the low-density C resulting in x-ray lasing at 45 Å.

The required low neutral carbon density of order $10^{20}\ \mathrm{cm}^{-3}$ can be achieved by a number of different approaches. In low density carbon foam the average cluster size is on the order of 100 Å. While the clusters have much higher local densities, where energetic electrons have a mean free path on the order of angstroms, a prepulse can be used to heat the clusters, causing them to expand, filling the voids and creating a uniform low density target. Alternatively, the use of a methane gas jet can also provide the needed densities of carbon. Dissociation of the methane molecule does not take place on the time scale of x-ray lasing. Therefore, the lasing photons are from the molecular transition $(1a_1)^{-1}$ to $(1t_2)^{-1}$, which has similar energy and linewidth as the $2p$-$1s$ atomic transition [25].

Figure 2 shows our proposed x-ray laser target with a line focused laser beam incident on a high-Z material. This laser-produced plasma emits a broad spectrum of x rays. A high-pass x-ray filter is required to remove low energy x rays, which would lead to significant L-shell ionization, prior to the x-ray source being incident on the low density C. As shown in Fig. 2 a lasing volume in C is produced and x-ray lasing can take place. Due to the USP nature of the ISPI x-ray laser a traveling wave pump is needed to achieve a large gain length. In this manner output in a single direction is achieved and the length of the laser is only dependent on the driving laser and tolerances in the optical system. Since the lasing is on a time scale of 50 fs this requires tolerances in the pump wave front on the order of 15 μm. Reliable predictions of the gain expected from such an x-ray laser requires an accurate model of both spectral emission and risetime of the pump source.

An investigation of x-ray emission from targets heated by an USP high-intensity optical laser was conducted using the hydrodynamics-atomic kinetics code LASNEX [26]. In Fig. 3 we show typical spectral results from a 40-fs USP driving

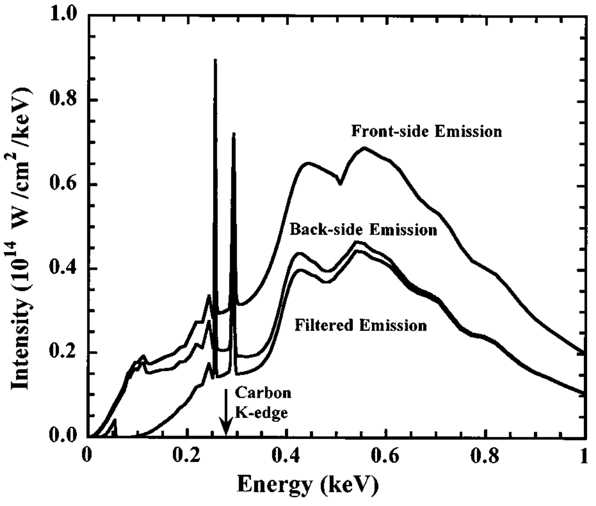

FIG. 3. Spectral results of front side, back side, and filtered emission from a 1 J, 40-fs USP driving laser, at time of maximum gain, incident on a flat target of thin Au layered on a 0.5 μm lithium hydride filter.

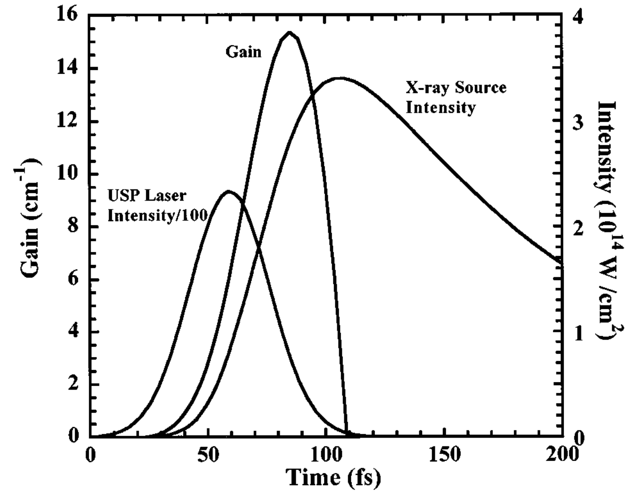

FIG. 4. The gain as a function of time along with the time-dependent optical USP laser intensity divided by 100 and the filtered intensity of the x-ray source are shown.

laser with an energy of 1 J incident (assuming 20% absorption based on experiments [27]) on a flat target of thin (200 Å) Au layered on a 0.5-μm lithium hydride filter (see Fig. 2). When we model this system, the energy is assumed to be deposited exponentially in an optical skin depth and the atomic kinetics and x-ray emission are calculated with an average-atom atomic model that includes spin-orbit coupling. The laser-produced plasma emits a broad spectrum, which is shown at time of maximum gain. We show the emission from the front side of the Au target emitted back in the direction of the USP laser, the back side of the Au target and finally through the high-pass filter. The thickness of the Au target has an effect on the emission around the C *K* edge. Gain cannot be achieved without filtering and the filtering of low-energy x rays also results in the reduction of x rays at and above the *K* edge by 25% where filtering is not desired. During the time of positive gain, the ionization front has penetrated the Au and 0.04 μm of the filter. This reduction in effective filter thickness of less than 10% does not lead to significant changes in gain but is included in our model. We have investigated the use of 0.2-μm Ti as a filter layered on Au and performed back-side emission calculations from micrometer thick targets of Cu and Zn, which self-filter low-energy x rays. We find that these targets also provide the needed x-ray emission to achieve high gain in C at 45 Å.

The use of flat targets offers the absorption of only a fraction of the incident energy, yet with the use of structured targets one can achieve nearly complete absorption. Structured targets can be composed of grooves, clusters, or cylinders. All these have approximately 100% absorption of the incident energy with high x-ray emission. Cluster targets, e.g., gold-black, are inexpensive but hard to model due to their fractal properties and there are issues of slower rise times associated with potential low-density emission [28]. Grooved targets, in general, are expensive but easy to model. A third choice is a two-dimensional lattice of cylindrical absorbers where work done by Marjoribanks *et al.* [29] shows high x-ray conversion efficiencies for such structured targets. We model the system by considering individual cylinders and calculate the emission normal to the surface. We estimate the radiation emitted from the front side or the back side, through a thin base, as the normal emission divided by 3 to account for geometrical effects. In general, we obtain approximately a factor of 3 higher gains using structured targets as compared to flat targets.

Modeling shows that a driving laser with energy of 1 J, 40 fs full width at half maximum (FWHM), incident on a structured target composed of vertical cylindrical absorbers ($d = 600$ Å) is sufficient to produce a large gain-length product. For a 10 μm×1 cm line focus, a density of 1.2×10^{20} cm^{-3}, and a 0.5-μm lithium hydride filter a gain of 15.6 cm^{-1} is found. In Fig. 4 we show the gain as a function of time along with the time-dependent filtered intensity of the x-ray source. The duration of the x-ray source is threetimes that of the positive gain. Collisional ionization to the

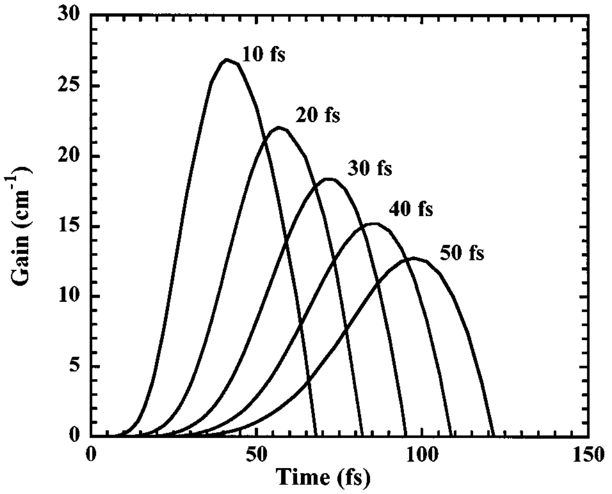

FIG. 5. The calculated gain coefficient depends on the input pulse duration and we show the effect of changing the pulse duration from 50 to 10 fs FWHM for a constant density of 1.2×10^{20} cm^{-3} and a constant energy source of 1 J.

lower-lasing levels limits the duration of lasing to of order 40 fs FWHM.

The calculated gain coefficient depends on the pulse duration as shown in Fig. 5 where the pulse duration was varied from 50 to 10 fs FWHM. We find an increase in the gain from 13 to 28 cm^{-1} as the pulse duration is reduced from 50 to 10 fs. The resulting gain widths are similar $\sim$40 fs FWHM. Allowing for absorption, the effective gain is $\sim$23 cm^{-1} for 10 fs, $\sim$10 cm^{-1} for 40 fs and zero for pulses longer than 80 fs. Thus, a 40-fs FWHM USP laser with twice the energy can obtain a similar gl to a 10-fs laser, yet a laser with a pulse length longer than 80 fs cannot achieve x-ray lasing at any power.

Our calculations show that a driving laser with a pulse duration of 40 fs, a 10 $\mu\mathrm{m}\times 1$ cm line focus, and energy of 1 J gives a gain length of 15.6 in C at 45 Å or an effective gain length of 10 accounting for absorption. We demonstrate that such a short duration for the optical driving pulse is required to control collisional filling of the lower laser state. Optical lasers with the required attributes and with repetition rates of order 10 Hz are becoming available, making this approach realizable in the near term.

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