Ultrafast x-ray sources*

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(Received 18 November 1992; accepted 20 January 1993)

Time-resolved spectroscopy (with a 2 psec temporal resolution) of plasmas produced by the interaction between solid targets and a high contrast subpicosecond table top terawatt (T^3) laser at 10^{16} W/cm^2, is used to study the basic processes which control the x-ray pulse duration. Short x-ray pulses have been obtained by spectral selection or by plasma gradient scalelength control. Time-dependent calculations of the atomic physics [Phys. Fluids B **4**, 2007, 1992] coupled to a Fokker–Planck code [Phys. Rev. Lett. **53**, 1461, 1984] indicate that it is essential to take into account the non-Maxwellian character of the electron distribution for a quantitative analysis of the experimental results.

I. INTRODUCTION

The advent of high-intensity subpicosecond lasers has opened up new horizons for the study of laser–matter interaction. These compact laser systems employing the technique of chirped pulse amplification[1] (CPA) are capable of generating powers in excess of 1 terawatt.[2–5] With ultrashort pulses (with very low prepulses) the plasma expansion during the interaction can be small or negligible[6,7] and it is thus possible to study solid density matter[8,9] and plasmas with very steep density gradients.[10–12] Among other aspects, these regimes are of great interest for the production of ultrashort x-ray pulses.[8,13] Furthermore under some conditions, the plasma heating and cooling occur on a time scale which can be short compared to the characteristic atomic excitation and relaxation times,[14] thus producing highly transient nonequilibrium plasmas. Thus the generation of maximum brightness and short x-ray sources by the means of heating a solid target with a subpicosecond laser has attracted a lot of attention recently.[14–19] The availability of ultrashort x-ray pulses will have a significant impact in areas such as time resolved x-ray diffraction, extended x-ray absorption fine structure,[13] probing of molecular dynamics[20] and pumping of x-ray laser plasmas.[21]

The spectroscopic studies realized up to now have been limited by the lack of the time resolution required to understand the basic processes controlling the transient plasma emission. The 1989 results on the overall keV emission obtained with picosecond time resolution[8] were a significant first step in the generation of ultrafast x-ray sources.

In this paper we present and discuss time resolved keV spectroscopy (with a temporal resolution of 2 psec) of plasmas produced by the interaction between solid targets and a high contrast subpicosecond terawatt laser at 10^{16} W/cm^2. Ultrashort emission (instrument limited) is observed in a short lived Be-like ionization state. We emphasize the strong effect on the x-ray pulse width of the plasma density at which the x-ray emission takes place. Time-dependent calculations of the atomic physics coupled to Fokker–Planck code appear to be essential tools in the quantitative analysis of the experimental results.

II. PLASMAS PRODUCED BY SUBPICOSECOND LASERS

Laser produced x-ray sources may have very interesting characteristics. The x-ray spectrum comes from highly stripped ions and is composed of x-UV or soft x-ray line radiation (from K, L, M, and even N shells), as well as continuum emission in the keV range. An important property of laser x-ray sources is the wavelength tunability.[22] While very large conversion efficiencies have been obtained with nanosecond drivers,[23] the efficient production of an ultrafast x-ray source is a more difficult task.

With picosecond lasers, it is now possible to obtain near solid density and ultrashort plasmas with $L/\lambda \ll 1$, where L and λ are, respectively, the characteristic gradient scale length and the wavelength. In these plasmas the density n_m for maximum absorption increases as the gradient scale length is decreased and is given[24] approximately (for an exponential density profile) by $n_m/n_c \approx 0.1(L/\lambda)^{-4/3}$ (provided $L/\lambda < 0.1$). As an example, for $L/\lambda = 10^{-2}$, $n_m/n_c \simeq 46$, which gives a density n_m near 10^{23} cm^{-3} with 0.5 μm irradiation. To attain such large values for n_m the incident laser pulse must be short enough (and also have a negligible prepulse) to ensure that there is a small plasma expansion and thus keeping $L/\lambda < 0.1$ during the laser pulse. At very high laser intensities, the ponderomotive pressure[25] will strongly affect the plasma dynamics,[26] tend-

*Paper 5I2, Bull. Am. Phys. Soc. **37**, 1468 (1992).
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 0899-8221/93/5(7)/2676/6/$6.00

ing to maintain and augment the initial steepness of the plasma density profile, even producing laser hole boring.[27] These elements indicate that subpicosecond high intensity lasers are excellent candidates to produce high-density plasmas (provided there is no prepulse).

An energy deposition at high density is important to obtain x-ray pulses which both turn on and shut off very rapidly,[28,29] because it favors ionization and produces very fast cooling. For instance, at an electron temperature of 400 eV the characteristic time for collisional ionization of Li-like ions is, for Al, less than 2 psec only if the electron density is higher than 10^{22} cm^{-3}. The collisional excitation time for the He-like (AlXII) $1s2p(^1P)$ level, from the $1s^2$ ground state, is still long (a few tens of picoseconds) and at this temperature the ionization of heliumlike ions is negligible. Naturally, when the plasma density is high, the plasma cooling is very rapid. Cooling by expansion for instance scales as $\tau=\delta/c_s\sim 1/(n_eT_e)^{1/2}$, where δ is the skin depth (the width of the energy deposition zone). We have typically $\tau\simeq 2$ psec at 100 eV and 10^{22} cm^{-3}. Then the emission coming from the highest ionization state produced in the plasma, and having the shortest wavelength, will decrease very rapidly following the temperature. However, the duration of longer wavelength emission should be eventually fixed by recombination processes. The three-body recombination rate scales[30] as $n_e^2/T_e^{9/2}$ and typical recombination time is roughly 10 psec for 100 eV and $n_e=10^{23}$ cm^{-3}.

III. ULTRAFAST LASER AND DIAGNOSTICS

We use a table top terawatt (T^3) laser system which employs the chirped pulse amplification (CPA) technique.[1,31] This technique which requires a lot of pulse manipulation (spectral expansion followed by temporal stretching, low peak intensity amplification and final pulse recompression) gives the possibility to generate laser pulses which could be ultraclean (peak to background contrast ratio of 10^7:1), very short (down to 300 fsec) and very intense (power greater than 1 terawatt). Another advantage of this technique is that it makes compact size (high peak power) laser available with high repetition rates (greater than 1 Hz). The experiments presented here have been carried out using the laser[2] at the University of Michigan. The 400 fsec laser pulses of wavelength 1.05 μm were generated in a Nd:glass laser system which incorporates[31] a passive pulse cleaner and a wideband Ti:sapphire regenerative amplifier. The intensity peak to background contrast ratio was measured (at 1.05 μm), using a third-order autocorrelator, to be 5×10^5:1 in these experiments, as shown in Fig. 1. The contrast ratio has been further approximately squared by frequency doubling ($\lambda=0.53$ μm) the laser pulse using a potassium dihydrogen phosphate (KDP) crystal. Maximum energies of 400 and 100 mJ have been used on targets at, respectively, 1.05 and 0.53 μm.

The laser beam is focused at normal incidence with an $f/6$ spherical fused silica lens on Al thick solid plane targets. The focused spot size of the laser beam has been measured at *full power* in vacuum with a 30$\times$ magnifying imaging system and a digital charge-coupled-device (CCD) camera. For 1 and 0.5 μm irradiations focal spot diameters of, respectively, 100 and 50 μm have been measured. This gives a maximum intensity of about 2×10^{16} W/cm^2 (at 1 μm) with a prepulse (longer than 300 psec) at an intensity of about 4×10^{10} W/cm^2. At 0.5 μm, the maximum intensity is about 10^{16} W/cm^2 for the main pulse, *without any significant prepulse.*

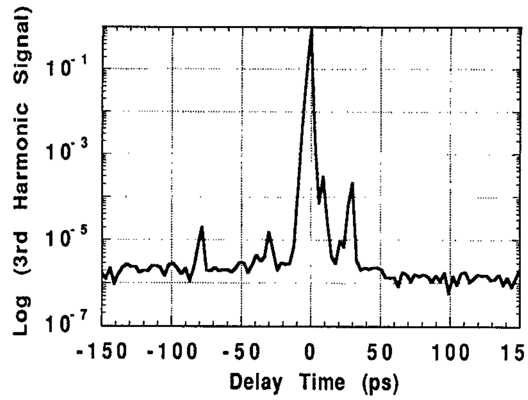

FIG. 1. Laser beam characteristics. Third-order autocorrelator trace showing a peak to background contrast ratio of 5×10^5:1.

Johann and Von Hamos spectrometers have been used at 45° from the laser axis to record the time integrated spectra in the 8 Å range. The time resolved keV spectroscopy is obtained with a modified Kentech x-ray streak camera coupled to a Von Hamos high brightness spectrometer.[32] Extremely good temporal resolution (2 psec) has been obtained, following the route of the Berkeley group,[8] by the use of (i) a high extraction voltage (up to 50 kV/cm), (ii) a photocathode having a very narrow photoelectron energy distribution,[33] and (iii) a calibrated sweep speed (8 psec/mm). The contribution of the unswept image of the slit to the instrument time resolution is 1.5 psec. The camera has been calibrated directly from the keV x-rays emitted by two adjacent plasmas produced by two low intensity beams (at 1.053 μm) having a known relative time delay between them.[32,34] The spectral dispersion is about 70 mÅ/mm along the camera entrance slit in the 8 Å range.

IV. TIME RESOLVED X-RAY EMISSION RESULTS

The presence of a prepulse has been considered as a crucial issue for obtaining ultrafast x-ray pulses. Several experiments[8,15,32,35,36] have shown that a prepulse leads (i) to much higher total x-ray emission, due to a larger laser absorption length and to an increase of the plasma size (ii) to changes in the spectral distribution due to changes in temperatures and ionization stages, and (iii) to an increase of the x-ray pulse duration, the emission coming from a lower-density plasma.

In the present experiments at 1 μm the prepulse intensity is above the ionization threshold of the target. The main ultrashort pulse is interacting with a low temperature, long gradient scale-length preplasma. The mean den-

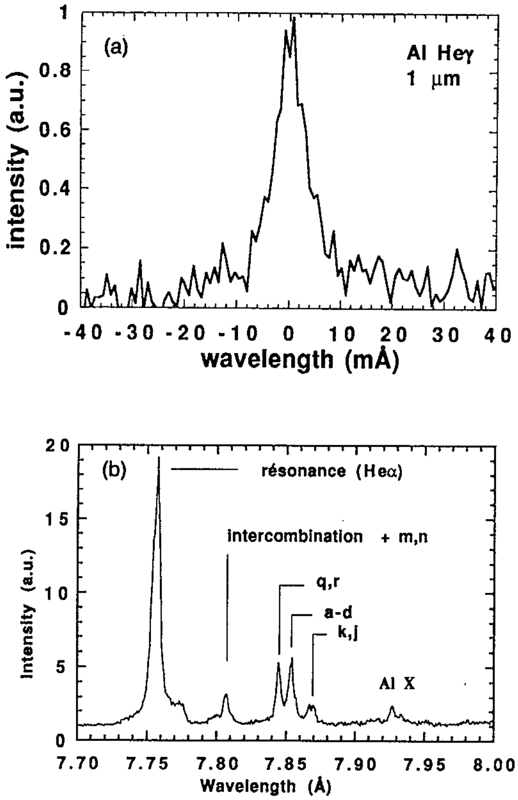

FIG. 2. Time integrated 1 μm irradiation results. (a) experimental He_γ line profile; (b) Al spectrum near the resonance line.

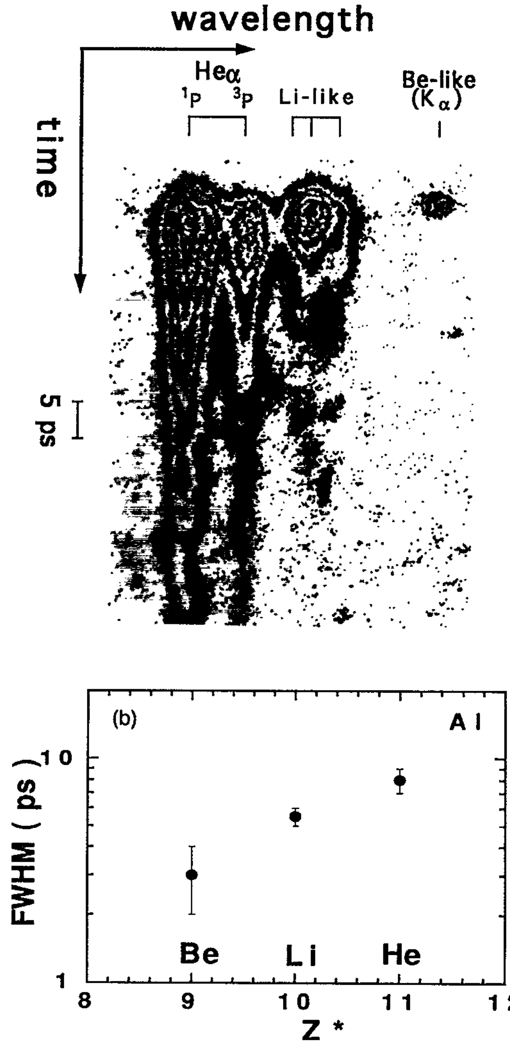

FIG. 3. Time resolved 1 μm irradiation results; (a) time resolved spectrum obtained with a 2 psec time resolution; (b) x-ray pulse FWHM as a function of the emitting ionization state.

sity (time integrated) at which the emission takes place has been deduced from the shape[37] of the $AlHe_\gamma$ line ($1s^2$-$1s4p$) (using modeling code SPECTRA.[38] At 1 μm, the experimental He_γ profile [Fig. 2(a)] indicates that the He-like emission is radiated at a density of 3×10^{21} cm^{-3}. The Fig. 2(b) shows the time integrated spectrum recorded near the resonance $AlHe_\alpha$ line. We see that the intercombination line is relatively small and that the time integrated *a–d,q,r* (Gabriel notations[39]) inner shell Li-like satellites [$1s^22p^2P$-$1s2p^2\ ^2P$ and $1s^22s^2S$-$1s2s2p(^1P)^2P$ transitions] are greater than the *k,j* dielectronic Li-like satellites ($1s^22p^2P$-$1s2p^2\ ^2D$ transitions). Figure 3 shows an example of an Al time resolved spectrum (in the same spectral range, around the $1s^2$-$1s2p^1P$ He_α resonance line) obtained in one shot [Fig. 3(a)] (laser wavelength of 1.05 μm at 2×10^{16} W/cm^2). This observed spectrum is very different from previous time resolved spectra obtained with long pulses.[40] The duration of the x-ray pulse varies with the emitting ionization state as shown in Fig. 3(b) and very short x-ray pulses are obtained in spite of the presence of a prepulse. The He_α resonance line has a 3 psec rise time and a full width at half maximum (FWHM) of 8 psec while the Li-like satellite emission lasts only a few picoseconds (the FWHM is around 5 psec), and the line at 7.94 Å which is radiated by short-lived Be-like ionization state has a FWHM of only 2–3 psec. These results highlight an interesting new aspect of ultrafast x-ray sources. Ionization states produced at low or intermediate temperature will *only* exist during the laser pulse rise time, when the plasma density is high, and before any expansion. Then x-ray pulses shorter than the laser pulse can be produced during the ionization rise time.

Frequency doubling the laser pulse allows us to work without any prepulse. The experimental He_γ profile [Fig. 4(a)] for 0.5 μm laser wavelength indicates that the mean density at which the emission takes place is around 7×10^{22} cm^{-3}. When the characteristic gradient scale length is shortened, by using 2 ω irradiation, the He_α (^1P line) pulse width decreases. Figure 4(b) presents the resonance line width, at 10^{16} W/cm^2, as a function of the mean electron density of the main emitting zone. We verify this behavior by using two collinear 0.53 μm beams incident on the target with an adjustable time delay between them, and with the same energy in each beam (25 mJ). The delay between the two beams allows us to adjust the gradient scale length seen by the second pulse and thus to control the plasma density at which the second pulse produces x-ray emission. We observe[32] an increase of the x-ray pulse width from 3 to 10 psec as the delay between the two beams changes from

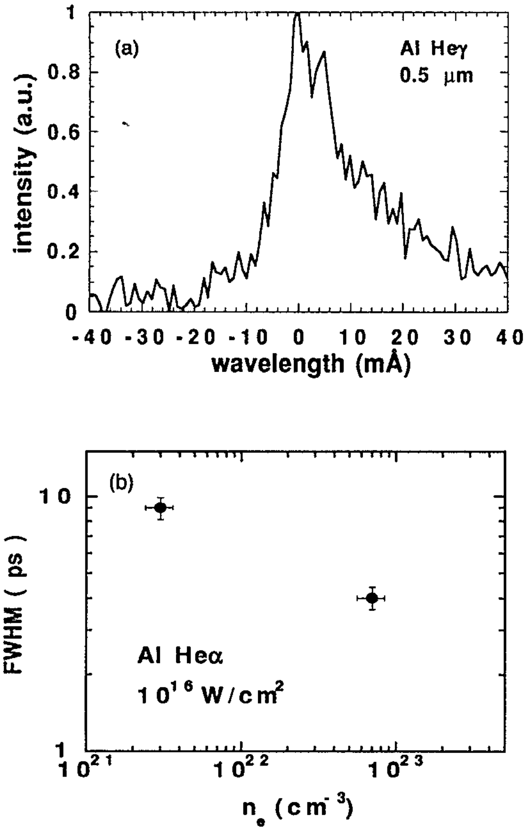

FIG. 4. Effect of the density of the emitting zone. (a) time integrated experimental He_γ line profile for 0.5 μm irradiation; (b) FWHM of the $He_\alpha(^1P)$ resonance line as a function of the mean electron density of the emitting zone.

0 to 60 psec. Then the x-ray pulse width (in the keV range) can also be controlled by adjusting the plasma characteristic gradient scale lengths.

V. TIME-DEPENDENT CALCULATIONS

The present ability to diagnose the temporal evolution of x-ray emissions coming from various ionization states will enable testing of some crucial issues in time-dependent modeling. The ultrashort laser matter interaction has been modeled with a Fokker–Planck kinetic code (code FPI)[41] which has been successful in describing the thermal transport in the context of laser-driven inertial confinement fusion. We recently used the new diagnostic of x-ray line polarization spectroscopy[42] to test how well we model the interaction of a short pulse with a preplasma. In this regard, our results display our success. Although these kinetic calculations have not the versatility of hydrodynamic calculations, Fokker–Planck (FPI) calculations do give us the exact shape of the electron velocity distribution at each position and time step that we will use in a time dependent calculation of the atomic physics. The calculations (one dimension, planar geometry) include the self-consistent (E) field for quasineutrality, electron–electron, and electron–ion collisions, ionization from ground state, collisional excitation, and ionization from excited levels. For $L/\lambda < 1$ an electromagnetic solver[24] is used to calculate the absorption above n_c. Geometrical optics is used at normal

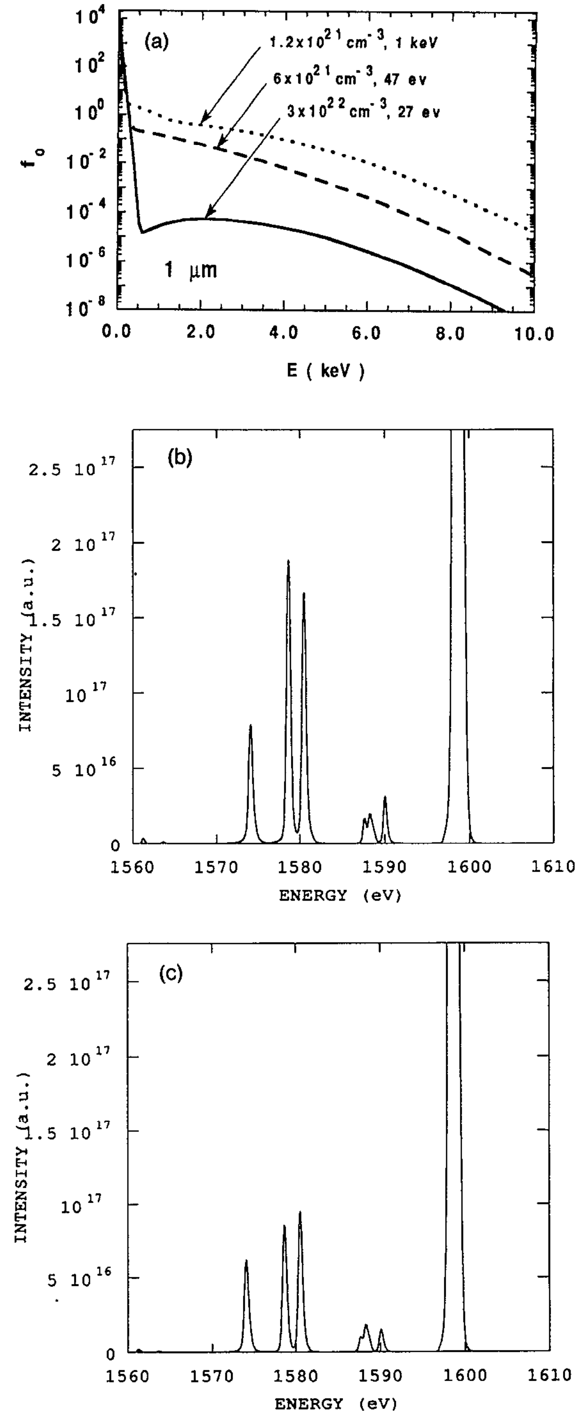

FIG. 5. Results of time dependent calculations: 400 fsec pulse, 1 μm wavelength, 10^{16} W/cm² incident on a preformed plasma ($L/\lambda = 1$); (a) electron distribution f_0 calculated at various densities, with FPI code at the maximum of the pulse; (b) Li-like satellite spectrum calculated with the non-Maxwellian electron distributions; (c) Li-like satellite spectrum calculated with a Maxwellian distribution (local plasma parameters).

incidence for $L/\lambda \geqslant 1$. The calculations include the Langdon kinetic heating operator.[43] A time-dependent atomic physics (coded TRANSPEC)[44] has been coupled to the FPI code.

As an example, we present in Fig. 5 the results of simulations realized assuming that a 400 fsec pulse (1 μm wavelength) is incident at 10^{16} W/cm² on a low-temperature preformed plasma having an initial gradient scale length $L/\lambda = 1$. The isotropic part of the electron distribution f_0 calculated at various densities [Fig. 5(a)] significantly deviates from a Maxwellian distribution. The Li-like *a–d,q,r* satellites calculated with these distributions are enhanced [Fig. 5(b)] compared to what is expected with a Maxwellian (local temperature) distribution [Fig. 5(c)], because these lines are produced in a very narrow high-density region (the He_α line is unchanged, being produced at a lower mean density). The calculations with the non-Maxwellian distributions [Fig. 5(b)] are in very good agreement with the experiments [Fig. 2(b)]. Other important experimental features (Li-like line shorter than He_α line, small intercombination, satellites to resonance line intensity ratio) are also recovered when only non-Maxwellian effects are taken into account. Experimental and calculation results are in good qualitative agreement. However, some effects not included in the present calculations (hot electrons, such as those which could be produced by nonlinear processes at n_c or $n_c/4$, ponderomotive force) have to be considered for a more quantitative analysis of the data.

VI. TOWARD HIGHER INTENSITIES

In the present experiments at 10^{16} W/cm^2, the shortest x-ray pulse has the lowest brightness. The use of higher laser intensities (10^{18} W/cm^2 range) will not only allow a hotter plasma and a brighter source to be obtained, but also to take advantage of the ponderomotive force which will maintain an ultrashort gradient scale length required to produce emission at high density and still very short x-ray pulses. Preliminary results have been obtained by focusing the beam (at 0.53 μm) with a parabolic mirror. A focal spot diameter of 10 μm has been measured at full energy giving intensities of 5×10^{17} W/cm^2 on target (at 0.53 μm and without prepulse). The shape of the He_γ line [similar to the profile presented in Fig. 4(a)] indicates that the emission is mainly produced at 6×10^{22} cm^{-3} (as at 10^{16} W/cm^2) and the observation of stronger Ly_α line (1*s*-2*p*) clearly indicates a hotter plasma. X-ray diodes (filtered with Be and protected by strong magnets) indicate that the keV x-ray emission increases by around an order of magnitude when the intensity is increased from 10^{16} W/cm^2 to 5×10^{17} W/cm^2.

VII. CONCLUSIONS

In this work, we have isolated a small number of parameters which are critical for the development of an ultrafast x-ray source. As previously recognized in several works, the plasma density must be very high, compared to the critical density in order to have x-ray emission pulses that shut off very rapidly. Furthermore, the time resolved (2 psec temporal resolution) spectroscopy appears to be an essential diagnostic to understand quantitatively the plasma dynamics. In particular, this work indicates that ionization states produced at low or intermediate temperature can have a very brief lifetime during the laser pulse rise time. Then x-ray pulses shorter than the laser pulse could be achieved by judiciously selecting the emission wavelength. The analysis of the time resolved spectra shows that the non-Maxwellian character of the electron distribution has to be taken into account to properly model the x-ray emission and design an efficient x-ray source. The present results have been obtained at 10^{16} W/cm^2. Using higher intensities (10^{18} W/cm^2 range) will allow the production of hotter plasmas increasing the source brightness, while maintaining an ultrashort gradient scale length and very high density in the emitting regions due to the ponderomotive force. This conjunction of high density and high temperature is necessary to obtain x-ray sources adapted to the various planned applications.

ACKNOWLEDGMENTS

One of us (J. C. Kieffer) acknowledges many helpful discussions with L. Da Silva and A. K. L. Dimoke Bradshaw.

This work is supported in part by Natural Sciences and Engineering Research Council of Canada and Ministére de l'Education du Québec.

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