PHYSICAL REVIEW A VOLUME 39, NUMBER 1 JANUARY 1, 1989

Kilovolt x-ray spectroscopy of a subpicosecond-laser-excited source

J. A. Cobble, G. A. Kyrala, A. A. Hauer, A. J. Taylor, C. C. Gomez, N. D. Delamater, and G. T. Schappert
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
(Received 18 April 1988)

A subpicosecond laser is focused to 10^{17} W/cm^2 to create an extremely transient Al plasma with a kV radiation time short compared to most of the excitation times. High-resolution spectroscopy is used to analyze the kV line emission from the He- and H-like ions and to determine that the plasma reaches a temperature near 1 keV and cools in less than 10 ps.

INTRODUCTION

The advent of high-intensity, picosecond lasers has introduced a new dimension for the study of laser-matter interactions.[1] Irradiance levels above 10^{17} W/cm^2, for which the electric field strength of the laser approaches 100 V/Å, are now available. This offers the possibility of studying a new regime of laser-plasma interactions in which plasma heating and cooling occur on a time scale that is short compared to the characteristic excitation and ionization times of the observed states. This is very different from the case of (0.1–1)-ns laser-driven plasmas in which equilibrium models may be applied.[2]

In the present work, we report on the observation and analysis of x-ray emission produced by high-intensity laser irradiation of a solid Al target. Copious x-ray emission is observed from 100 eV to 2 keV. We emphasize high-resolution spectroscopic analysis of the kV line radiation. This makes possible studies of the detailed atomic structure of highly stripped ions and the development of related time-dependent plasma diagnostics. Previous spectroscopic work with solid targets was at lower irradiance and dealt with much softer radiation.[3] In addition, in the present case, target ions are more highly stripped, e.g., He- and H-like Al.

EXPERIMENTAL CONDITIONS

The laser is a high-brightness, small-aperture, KrF-based system which routinely produces irradiances greater than 10^{17} W/cm^2 when focused on target.[4] Subpicosecond 248-nm seed pulses, generated by up-converting the output of a visible dye laser, are amplified at 3 Hz in two commercial KrF discharge lasers separated by a vacuum spatial filter. The amplified pulses have an energy up to 25 mJ and are 700 fs in duration. The output beam, expanded to 2.5 cm in diameter, is focused on target with an $f/3$ off-axis parabolic mirror. The spot diameter on target has been determined by several independent means[5] to be about 4 μm—twice the diffraction limit.

The main pulse is superimposed on a 10-ns pedestal of amplified spontaneous emission (ASE) which contains 5–10% of the total energy. The ASE produces a plasma with which the main pulse interacts. By adjusting the timing of the 700-fs pulse within the ASE window, the prepulse energy fraction can be reduced to as little as 2%—about 400 μJ.

The x-ray emission is studied with a variety of diagnostics. Spectroscopy is typically done at an angle of between 60° and 70° with respect to the target normal. A free-standing, gold-foil, transmission-grating spectrograph was employed to survey time-integrated plasma radiation between 100 and 4000 eV with a resolution of 1–2 Å. Typically, spectra are accumulated in several hundred shots. High-resolution, time-integrated spectra in the (1.5–2)-keV range were obtained with a flat pentaerythritol (PET) crystal spectrograph. The resolving power of this instrument is about 2000. The very small emission region in this work causes the source-size contribution to instrument broadening to be only about 0.3 eV. In addition, due to the very short duration of the keV radiation, there is no hydrodynamic expansion of the effective source size. The high resolution of the kV line spectra is a notable feature of this experiment.

A pinhole camera was used to characterize the emission region. It consists of a Be-filtered, 3-μm pinhole, and direct exposure film (DEF). The camera has a magnification of about 15 and a spatial resolution of 6 μm. The Al radiation above 1 keV is seen to come from a region limited by the spatial resolution of the camera, which is consistent with the focal spot size. Thus, the keV x-rays are emitted only from the region of highest laser intensity.

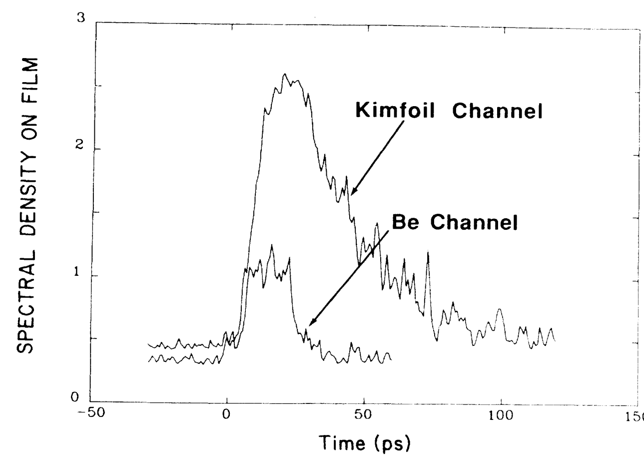

FIG. 1. Densitometered streak camera record of the Be and Kimfoil channels. The instrument broadening corresponds to about 20 ps.

An x-ray streak camera with a CsI photocathode was used to set a limit on the radiation lifetime. The camera slit was divided into three zones: (1) unfiltered, (2) filtered with 25-μm of Be, and (3) filtered with 270 g/cm^2 of aluminized Kimfoil ($C_{16}H_{14}O_3$). The three channels turn on simultaneously with a few picosecond rise time. The streak camera response to the kV radiation (Fig. 1) is essentially the 20-ps instrument function. Therefore, the lifetime of the actual kV emission is much shorter. Deconvolution suggests an upper bound on the x-ray pulse length of less than 10 ps. The Kimfoil channel, which is predominantly 200–284 eV radiation, lasts 20–50 ps.

RESULTS AND ANALYSIS

The ASE prepulse sets the initial conditions for the interaction of the main pulse with the target plasma. The most important component of ASE is that which originates in the first amplifier and is coupled through the spatial filter to the second amplifier. Thus, the ASE will have approximately the same focal properties as the picosecond pulse. An ion time-of-flight detector has been employed to measure the velocity of the ASE-produced blowoff plasma in the absence of the main pulse. It indicates that the target plasma expands no more than the measured focal depth (about 50 μm) before illumination by the main pulse. Thus, the irradiance on target is not reduced by target expansion. Taking transmission losses into account, the ASE irradiance on target is less than 1×10^{12} W/cm^2. With ASE only, no keV x rays are detected. Both hydrodynamic modeling and previous experiments[6] indicate that the prepulse plasma has a temperature T_e of about 5 eV. When the main pulse arrives, the irradiance climbs to $1-2\times10^{17}$ W/cm^2, and the plasma in the focal spot is further ionized, thus, increasing the electron density about fivefold.

In Fig. 2, we show the grating spectrum of an Al target irradiated at 10^{17} W/cm^2. One of the brightest features is the unresolved emission lines of He- and H-like ions near 7 Å or 1600–2000 eV . Many lines of Be- and Li-like aluminum were also observed in the range of 35–55 Å (225–350 eV). Taking grating efficiency and detector sensitivity into account, the Li-like lines at 48 and at 52 Å (250 eV) both have approximately the same photon flux as the blended 7-Å line.

This band of radiation from 1600–2000 eV is resolved with the crystal spectrograph. Figure 3 shows the $1s$-$2p$ transition of H-like Al as well as several lines of He-like Al. In another spectrum (Fig. 4), the He-like $1s^2$-$1s2p$ singlet transition and various Li-like satellites are resolved.

To analyze the transient plasma, the rates important for establishing the radiation need to be identified. As a starting point, we assume that the electron density n_e is near the critical density for 248 nm: about 2×10^{22} cm^{-3}. Since transitions requiring 2 keV for excitation are observed, a T_e of at least several hundred volts is anticipated. For the density assumed, Table I shows relevant time scales for plasma and atomic processes at 300 and 1000 eV. We see that electron equilibration can occur within the 700-fs main pulse. Thus, the electron distribution is thought to be Maxwellian. Ionization of Li-like Al to the He-like state is relatively fast for the given range of T_e. However, ionization to H-like Al at 300 eV is slow compared to the duration of the keV radiation as determined by the streak camera. The excitation of He-like states except for the $n=2$ levels is slow for either temperature. There is scarcely time to populate the $n=3$ levels of He-like Al. Collisional mixing of excited states is fast, comparable to the radiative lifetime of the $1s4p$ state, for example. These considerations indicate that a time-dependent, nonlocal thermodynamic equilibrium model must be used to predict plasma parameters from the line ratios. Recombination times, long compared to the streak record of the keV radiation, are not important for this ionizing plasma. Multiphoton processes and high-field effects may play a role during the intense laser pulse[7] in setting the initial ionization conditions and T_e. For the ionization and excitation of the observed highly charged ions, these effects are judged to be unimportant, and they are not included in our present analysis.

The line profiles were used to determine n_e. The theoretical calculation of the profiles is only weakly

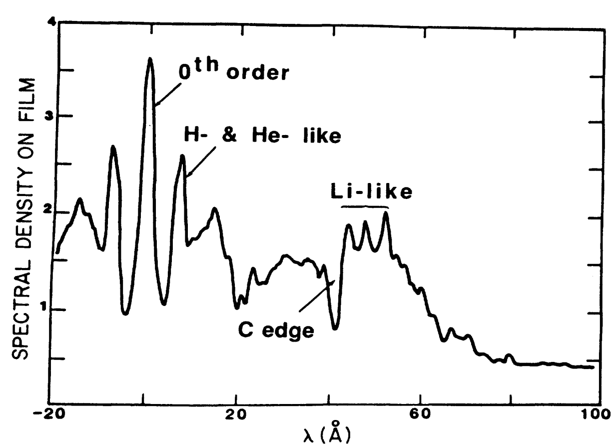

FIG. 2. Transmission-grating spectrum of Al at 10^{17} W/cm^2.

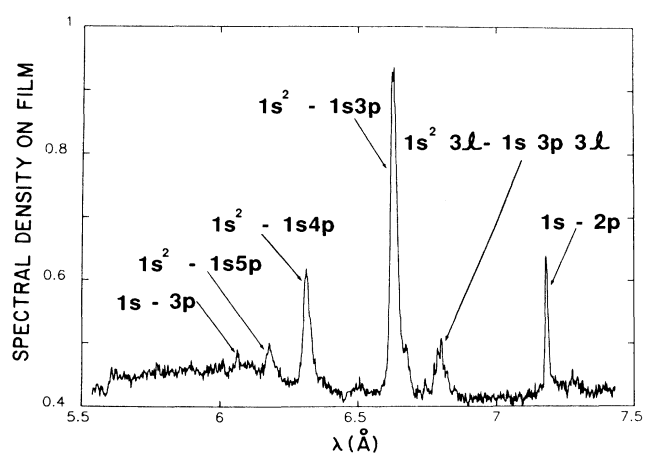

FIG. 3. PET crystal spectrum of kV Al radiation.

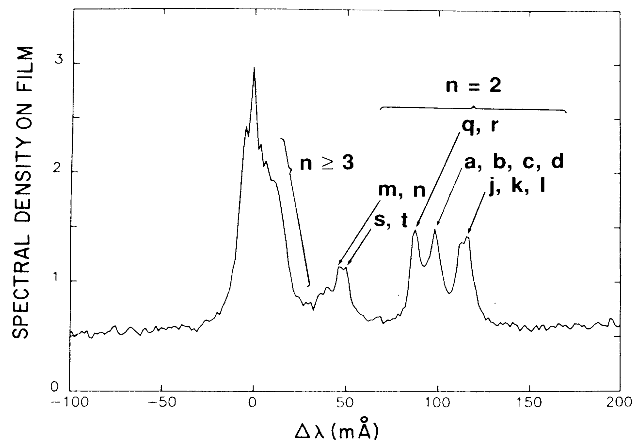

FIG. 4. The $1s^2\,{}^1S-1s2p\,{}^1P$ line of He-like Al with its doubly excited Li-like satellites. The quantum number of the spectator electron for the satellites is indicated by n.

dependent on T_e; hence, the time history of T_e is not very important. The theoretical calculation[8] takes into account all known broadening mechanisms such as Stark, Doppler, instrumental, etc. The density certainly does not change while the He-like ions are radiating. We concentrate on the $1s^2$-$1s4p$ transition because having a lower oscillator strength and being broader, it is more likely to be optically thin than the transitions from lower states. The small emission volume will mitigate opacity effects too. The result of the fitting analysis gives $n_e \sim (2-3) \times 10^{22}$ cm^{-3}, which is equal to or slightly greater than critical density for KrF.

The determination of the temperature is more difficult because of the transient nature of the plasma. Early in an ionizing plasma, the relative abundance of H- to He-like ions is roughly equal to the ratio of the radiation time to the ionization time. From line intensities in Fig. 3, the H-like population is about 10% of the He-like population. Assuming the H-like radiation comes from the same (or smaller) volume, this requires a temperature of 1 keV for 1 ps or 500 eV for 10 ps to provide the necessary collisional ionization. The streak record suggests that the plasma cools sufficiently in 10 ps to turn off collisional excitation of the He- and H-like species.

The electron temperature was also estimated from the slope of the continuum radiation at energies between 1500 and 2000 eV. Because of the high average ion charge, the continuum is dominated by free-bound transitions, and because of the long recombination time (Table I), the resulting 350-eV time-averaged temperature is weighted toward times late in the evolution of the plasma after it has cooled significantly. The peak temperature is certainly much higher.

An upper bound on T_e may be found too. Using the estimated plasma volume from the pinhole camera, we calculate the stored energy in the electrons and the energy investment in ionization to be about 2 mJ for a 1-keV T_e. The absorbed laser energy is not measured, but with allowance for losses, T_e cannot be expected to exceed a few keV. We add that were T_e as high as 2 keV, the He-like radiation must be reduced with respect to that of the H-like ions as the plasma is further stripped. Therefore, the conclusion that T_e peaks near 1 keV seems consistent and reasonable.

Figure 4 shows a rich spectrum of satellites from doubly-excited, Li-like Al in the vicinity of the $1s^2\,{}^1S$-$1s2p\,{}^1P$ line. The (j,k,l) satellites are usually associated with dielectronic capture whereas the (a,b,c,d) and (q,r) satellites are generally associated with inner-

TABLE I. Comparison of relevant plasma and atomic time scales in ps for $n_e = 2\times10^{22}$ cm^{-3}.

Process	$T_e = 300$ eV	$T_e = 1000$ eV	
Electron-electron equilibration time[a]	0.01	0.05	
Ionization time: Li to He[b]	0.9	0.3	
Ionization time: He to H[b]	> 1500	10	
Radiative recombination: He to Li[b]	100	190	
Dielectronic recombination time[c]	> 500	> 500	
Three-body recombination[d]	16	150	
Excitation time[e]: $1s^2$-$1s2p$	110	4	
Excitation time[e]: $1s^2$-$1s3p$	> 1500	28	
Excitation time[e]: $1s2s$-$1s3p$	0.6	0.3	
Radiative lifetime: $1s2p\,{}^1P$			0.035
Radiative lifetime: $1s3p\,{}^1P$			0.13
Radiative lifetime: $1s4p\,{}^1P$			0.31

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shell excitation. For the $\leq$ 10-ps interval for which T_e is high, inner-shell excitation dominates dielectronic recombination.[9] The collisional excitation rates for these three groups are roughly equal; however, the (j,k,l) group has a net radiative transition rate only a tenth of the others because of its higher autoionization rate. Because the transitions all have equal intensity, the (j,k,l) line clearly has a significant contribution to its intensity from recombination at later times. The resonance line is broader than expected from the plasma electric field. This is due to the presence of Li-like satellites for which the spectator electron is in the $n \gtrsim 3$ level. These transitions are rarely observed to be so strong in dense, laser-produced plasmas. Finally, note that in this plasma, the intercombination line should be completely quenched by electron collisions.

Considering the excitation times from the He-like ground state (Table I), we expect that little of the total radiation comes during the 700 fs when the high electric field of the laser is present. The laser field, at 90 V/Å, exceeds the plasma field by 30 times. If appreciable radiation were emitted during this time, one would expect effects like ac Stark shifts and Baranger-Mozer-like satellites for the resonance line.[10] These effects, if present, are difficult to identify because of overlap with the numerous satellites.

The high intensity of the laser system, especially the ability to focus to a spot less than 5 μm across, has enabled high resolution x-ray spectroscopy seldom realized in laser-produced plasmas. The spectra of highly charged ions may be investigated experimentally and conveniently. The capabilities of the laser system to create a point-like, multi-kV radiation source at 3 Hz is noteworthy too. This work demonstrates the potential for many unique discoveries in both atomic spectroscopy and in plasma diagnostics. Work is in progress to apply a time-dependent model to characterize the plasma in detail. The role of multiphoton processes needs further investigation to see if they should be incorporated into the model.

ACKNOWLEDGMENTS

In this work, we have been greatly assisted by T. R. Hurry, J. Grosso, J. Roberts, M. Maestas, S. Harper, J. Clifford, and H. Halbig and we extend our thanks. Thanks are also due to K. LaGattuta for calculating rate coefficients and to H. Griem for fruitful discussions. Finally, the enthusiasm and encouragement of P. Goldstone and J. Trainor is greatly appreciated. This work was performed under the auspices of the U.S. Department of Energy.

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