Relative ion expansion velocity in laser-produced plasmas

S. Goldsmith,[a] J. C. Moreno, and H. R. Griem
Laboratory for Plasma and Fusion Energy Studies, University of Maryland, College Park, Maryland 20742
Leonard Cohen
Laboratory for Solar Physics, Goddard Space Flight Center, Greenbelt, Maryland 20771
M. C. Richardson
Laboratory for Laser Energetics, University of Rochester, Rochester, New York 14623-1299

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The spectra of highly ionized titanium, Ti XIII through Ti XXI, and C VI Lyman lines were excited in laser-produced plasmas. The plasma was produced by uniformly irradiating spherical glass microballoons coated with thin layers of titanium and parylene. The 24-beam Omega laser system produced short, 0.6 ns, and high intensity, 4×10^{14} W/cm,^{2} laser pulses at a wavelength of 351 nm. The measured wavelength for the $2p$-$3s$ Ti XIII resonance lines had an average shift of $+0.023$ Å relative to the C VI and Ti XX spectral lines. No shift was found between the C VI, Ti XIX, and Ti XX lines. The shift is attributed to a Doppler effect, resulting from a difference of $(2.6\pm0.2)\times10^7$ cm/s in the expansion velocities of Ti XIX and Ti XX ions compared to Ti XIII ions.

I. INTRODUCTION

Ion expansion velocity in laser-produced plasmas has been the subject of many investigations since use was made of intense laser beams to generate such plasmas. Generally, the plasma is produced either by focusing the laser radiation on a point inside a fluid medium or on the surface of solid targets. When a solid target is irradiated by a single beam laser, a conical plasma jet is ejected from its surface, expanding outwardly. The axis of the jet is generally perpendicular to the surface. In the case of a spherical target illuminated uniformly by several laser beams, a spherical plasma shell is formed. Various observations have shown that the laser-produced plasma expands from the solid surface at velocities up to about 10^8 cm/s, corresponding to ion kinetic energies ranging from a few keV to hundreds of keV.[1] The expansion velocity of the plasma depends on the irradiation intensity and plays an important role in the energy balance of the target laser system. It has been shown in the case of flat targets that the total kinetic energy of the ions can be about 70% of the original laser energy.

The methods employed in determining the ion expansion velocity include the use of velocity analyzers, time of flight measurements, and the determination of the Doppler shift in the spectrum emitted by the moving ions.[2-6] In the study of plasma flow from planar targets, particular attention has been given to the dependence of the expansion velocity on the direction relative to the laser beam path or the normal to the target surface.[4] Spectroscopic and time of flight methods were also used to determine the ion velocity field in the expanding plasma plume, i.e., obtaining a spatial mapping of the velocity vector.[4,6] In some cases the dependence on time of the velocity field was also obtained. Another factor which determines the expansion velocity is the degree of ionization. This effect was established by spectroscopic methods in a plasma produced by focusing a ruby laser of 3×10^{11} W/cm^2 flux onto a polyethylene foil. Different flow velocities were measured for C VI, C V, and C IV ions, 2.4×10^7, 2.2×10^7, and 2.1×10^7 cm/s, respectively. It was observed that velocity increases with the degree of ionization.[1,3,4]

From a spectroscopic point of view the determination of relative expansion velocities is of great significance for a proper wavelength measurement. In recent years laser-produced plasmas (LPP) have become one of the major radiation sources for spectra of highly ionized atoms.[7] In a way, LPP sources replaced the former high-voltage sparks used for this purpose. The LPP spectroscopic source emits not only the new spectral lines but also some already well-known lines which are used as references to calibrate the new ones. In many cases the reference lines do not correspond to the same element whose ions are under study, but to other atoms. The reference lines may be emitted by impurities in the target material, e.g., oxygen or carbon, and in such cases they will typically correspond to hydrogenlike or heliumlike ions. Hydrogenlike transitions are known theoretically and allow accurate wavelength determination. However, a relative velocity between the ions emitting the new lines and those emitting the reference lines, results in a Doppler shift that reduces the accuracy of wavelength measurement. It is important, therefore, from a spectroscopy perspective, to study gradients in expansion velocities and to be able to estimate their effect on wavelength determination.

In addition to its contribution to spectroscopy, the study of gradients in the ion expansion velocity in LPP is also significant for improving the models of the interaction between the laser radiation and the target. Basic semiempirical modeling of the laser-target interaction, e.g., energy absorption, thermal conduction, and plasma parameters, entails a dependence of T_e (electron temperature) and V_i (ion escape velocity) on I (laser intensity). Both T_e and V_i should in-

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[a] On sabbatical leave from School of Physics and Astronomy, Tel Aviv University, Tel Aviv 69978, Israel.

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crease nonlinearly with I.[8] As the average degree of ionization in the plasma generally depends strongly on T_e, this also implies some correlation between the average degree of ionization and the ion expansion velocity.

The present study of ion expansion velocity in laser-produced plasmas was made in conjunction with a wider research project devoted to the study of thermal transport of 351-nm laser energy flux through spherical parylene layers.[9] The transport of the laser radiation energy into the target from the external critical density region in the laser-produced plasma is accomplished by electron conduction.[8] Studies of electron thermal conductivity have often been performed in the past using composite targets, consisting of two different material layers.[10,11] The process of heat transport is studied in the external layer, while the function of the inner layer is to register the penetration of the laser energy by emitting its characteristic spectral radiation. Aluminum and titanium internal layers were used by us in separate experiments to signal energy penetration. Ions of Al and Ti expand from these laser-produced plasmas and the observation of a change in the titanium ion expansion velocity with the degree of ionization and its implications are the main subject of this report.

II. EXPERIMENTAL ARRANGEMENT

The titanium plasma was produced with the Omega laser system at the Laboratory for Laser Energetics at the University of Rochester (LLE). The laser system consists of 24 Nd:glass laser beams frequency tripled to a wavelength of 351 nm. Typical incident intensities used in this investigation were 4×10^{14} W/cm^2, with a pulse length of 0.6 ns. To improve the uniformity of target irradiation, the laser light was focused at 8 times the spherical target radius behind the target. The spherical targets consisted of glass microballoons, 400 μm in diameter and approximately 10 μm in wall thickness (Fig. 1). These microballoons were coated with a thin (2 μm) layer of Ti to provide signature line emission for the thermal transport study. A final overcoat (0–11 μm) of parylene was deposited on the targets.

The target element spectra were recorded by a 3 m grazing incidence spectrograph[12] using Kodak 101–05 photographic plates. The grating had 1200 lines/mm and a blaze angle of 2°35′. At an 88°-angle of incidence, the blaze wavelength is close to 60 Å. The radiation from the plasma was focused onto the entrance slit of the spectrograph with a cylindrical concave mirror, positioned at a grazing angle of incidence to the incoming radiation and adjustable to focus the light. Each spectral plate was exposed with a single laser shot. The positions of the lines on the plates were measured by use of a Grant comparator to within 1 μm.

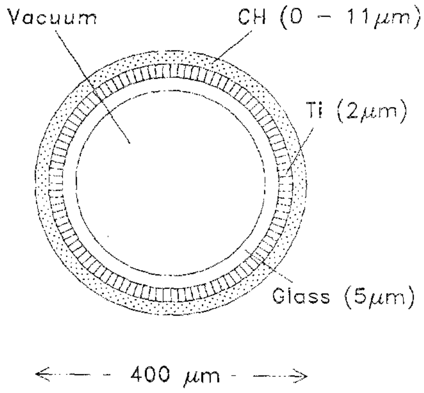

FIG. 1. The structure of a typical target used in this experiment.

III. OBSERVED SPECTRA

The observed spectra consisted of lines corresponding to CV, CVI, and the titanium ions TiXIII through TiXXI in the region 8 to 50 Å.[13] In CVI we observed the first five members of the Lyman sequence, while in CV only the first resonance transition, $1s^2\ {}^1S_0$-$1s2p\ {}^1P_1$ and some lines corresponding to transitions from doubly excited states are observed. The latter transitions are located near the CVI Lyα line. Some new transitions in titanium ions were identified and classified,[14] among them $2s$-$3p$ and $2p$-$3d$ transitions identified for the first time in TiXXI, heliumlike Ti. The differentiation between lines of different titanium ions utilized the effect that the external parylene layers had on the intensity of the various lines. Increasing the thickness of the parylene reduced the penetrating flux of the laser energy, and led to the lowering of the highest degree of ionization observed in the titanium spectrum.

The spectra of highly ionized Ti were studied in the past using LPP or vacuum sparks as spectroscopic sources.[15] A reasonable agreement in wavelength is obtained between the lines measured with both sources (see, e.g., Ref. 16). In both cases impurity lines were used as reference for calibration. The LPP spectroscopic sources generally utilized a single beam laser directed normally to a flat target, and the spectrograph axis was normal to the laser beam. The fact that wavelength determination in various experimental systems agreed, indicates that even if plasma expansion took place, the impurity ions and the Ti ions either had the same velocity, or the relative velocity between the different ions was too small to affect wavelength determination.

IV. LINE SHIFT OBSERVATION

In the present experiment we have first determined line wavelengths for the transition arrays $1s^22s$-$1s^23p$ and $1s^22p$-$1s^23d$ in TiXX. The wavelengths of the lines are well known experimentally and theoretically,[15,16] and as they were rather intense in second order they were well situated for calibration relative to the CVI lines. The wavelengths obtained on several plates are presented in Table I, showing a good agreement (well within the estimated experimental error of 0.005 Å) with the previous wavelength determination.[15,16] Thus, even if we assume a possible difference between the expansion velocities of the CVI and TiXX ions, the agreement with the previous wavelength measurements and the agreement with the theoretical values indicate that it should be negligible. Using the lines of Table I as reference lines, wavelengths of other TiXIX and TiXX lines in the region 15–17 Å were determined, and again the agreement with the previously

TABLE I. Wavelengths (Å) of four lines of TiXX, lithiumlike ion, corresponding to the transition arrays $1s^23p$-$1s^22s$ and $1s^23d$–$1s^22p$, respectively. The first and second columns define the transition. The columns marked I, II, and III present the wavelengths determined in the present experiment, using different plates, in second order. The spectrum of GFOC (Ref. 15) was obtained with a vacuum spark, while ABPF (Ref. 16) used LPP as a source.

Trans.	$j-j'$	I	II	III	GFOC	ABPF
$2s$-$3p$	0.5–1.5	15.214	15.216	15.210	15.217	15.211
$2s$-$3p$	0.5–0.5	15.255	15.258	15.255	15.252	15.253
$2p$-$3d$	0.5–1.5	15.910	15.912	15.910	15.914	15.907
$2p$-$3d$	1.5–2.5	16.055	16.055	16.054	16.059	16.049

TABLE II. Measured wavelengths of two resonance transitions in TiXIII. (a) $2s^22p^6\ {}^1S_0$-$2s^22p^53s\ {}^1P_1$ and (b) $2s^22p^6\ {}^1S_0$-$2s^22p^53s\ {}^3P_1$. Columns I–IV present measurements on four plates (ET = Ref. 18).

Trans.	I	II	III	IV	ET
a	26.662	26.663	26.665	26.666	26.641
b	···	···	···	26.981	26.960

published values was within the experimental error. Finally, a least-squares fit of all observed and measured lines between 10 and 20 Å to the grating dispersion relation resulted in a standard deviation smaller than 5 mÅ, indicating once more that the present wavelength determination for TiXIX and TiXX agrees very well with previous data.[13] In addition to this, the identification of the new lines in TiXXI,[14] whose measured wavelengths agreed well with their accurately calculated values,[17] provide a fiducial point for absolute wavelength calibration, showing again that wavelength determination for the TiXIX through TiXXI and for the CVI Lyman series lines is proper and not affected by the plasma expansion.

A different situation occurs in the case of TiXIII, neonlike ion, resonance transitions. Edlén and Tyrén[18] first identified five lines of the transitions $2s^22p^6$-$2s^22p^53l$, $l = 0,2$. In the spectrum obtained with an uncoated target, the two $2p$-$3s$ lines appear strong and unperturbed, while the $2p$-$3d$ lines are hardly observed. On that plate the wavelengths of the two $2p$-$3s$ transitions were determined by use of the second-order TiXX lines as references. When the spectrum is produced with targets coated by parylene, the TiXIII line at 26.960 Å is strongly perturbed by the Stark broadened CVI Lyman line at 26.990 Å and its wavelength cannot be determined. With such targets the wavelength of only one $2s$-$3p$ TiXIII transition can be measured relative to the CVI reference lines. The obtained wavelengths are presented in Table II. There is a significant shift on all plates between the present wavelengths and those reported previously.[18] As we shall discuss in the next section we attribute this shift to a difference between the expansion velocities of TiXIII and TiXX ions. As the average value of the shift is $+0.023$ Å, the TiXIX, XX, and CVI ions move towards the spectrograph slit with a velocity larger by $(2.6 \pm 0.2) \times 10^7$ cm/s than that of the TiXIII ions.

V. DISCUSSION

In general, line shifts in dense plasma sources may be caused by several effects, including the Stark effect, plasma polarization shift, and Doppler shift. A calculation of the TiXIII line shift due to the two plasma effects results in negligibly small values, much smaller than the observed shift, and we therefore assume that the Doppler effect is responsible for the observed line shift. The difference in the ion expansion velocity mentioned above is actually between the averages of the components of the expansion velocity in the direction of the spectrograph axis. Spherical shells of LPP generally have a radial expansion velocity with spherical symmetry. Such spectral sources when they are transparent to the studied lines, do not produce a Doppler shift but only line broadening due to the spread in the velocity along the spectrograph axis. The center of mass velocity along this axis is zero. If, on the other hand, the radiation from half of the plasma shell is blocked from the spectrograph, the center of the line will be Doppler shifted. A similar argument is also valid in the case of cylindrical plasma expanding from a flat target.[5] In the present experiment an asymmetry in the expansion velocity distribution in the spectrograph direction is produced because only the radiation emanating from that half-sphere of the laser-produced plasma, which is on the same side of the spectrograph slit, is recorded. The blocking of the radiation emitted by the other half-sphere is caused by that part of the titanium layer which is not affected by the laser.

The existence of a blocking titanium layer is suggested experimentally by the absence in our spectra of any line of ionized silicon, although we do see two weak lines of highly ionized oxygen. Previously, spectra taken with the same experimental arrangement but using either simple glass microballoons or glass microballoons coated with a thin layer of parylene, had always contained the lines of highly ionized silicon, even of SiXIV, and rich spectra of highly ionized oxygen.[19] The absence of any ionized silicon lines indicates that the laser flux does not penetrate the titanium layer to reach the glass substrate and produce a silicon and oxygen plasma; a conclusion supported also by the general study of heat transfer.[9] In contrast, the observed but very weak OVIII Lyα line and the faint first resonance line of OVII are attributed to the presence of titanium oxide on the surface of the titanium layer. Thus, the opacity of the titanium layer which is only partially penetrated by the laser enables the observation of the different expansion velocities of titanium ions.

The observation on the same plate of the spectra of titanium ions with very different ionization energies raises some questions. First, it is significant to note that the present spectra were obtained by time integration and with no spatial resolution. As a result we can not experimentally determine where and at what point in time the lines of TiXIII and those of higher ionization, e.g., lines of TiXX, were generated. If it is assumed, however, that the plasma is approximately at ionization equilibrium, it is difficult to maintain that lines of both ions are emitted simultaneously from the same location. In Fig. 2 we present a plot giving our calculation of the dependence of the relative abundances of titanium ions in a plasma at coronal equilibrium on the electron temperature, T_e. The observation of strong lines of TiXX suggests that T_e should be in the vicinity of 1000 eV, while the presence of TiXIII indicates that T_e should be close to 100 eV. It is clear from the plot that the relative abundances of TiXIII and TiXX will be significant only in regions of the plasma with markedly different T_e.

We assume that ideally the plasma is formed in the following manner. The laser energy is absorbed by the target through inverse Bremsstrahlung producing a relatively steep heat front which penetrates into the target. Each successive layer upon heating ablates and flows out with little recombination, thus preserving its original ionization state. The velocity of expansion increases with the radius so that the charge states reflect the temperature distribution within the target before the expansion. If mixing of the layers occurs this would result in a broadening of the velocity distribution for the charge states and in turn produce larger Doppler broadening. However, in this case a Doppler shift was observed which, when related to a velocity of expansion proportional to $T_e^{1/2}$, results in a velocity difference of $(2.6 \pm 0.2) \times 10^7$ cm/s between TiXIII and TiXIX, XX.

This velocity difference indicates that there is little or no mixing between highly ionized titanium (TiXIX, XX) and neonlike titanium (TiXIII). One might expect that nonuniformities in laser irradiation could lead to mixing of all the charge states. However, it has been observed[20] that high-Z layers (barrier layers), such as titanium, act to mitigate the effect of laser nonuniformities. On the other hand low-Z layers, such as CH, are seriously affected by laser hot spots and can have significantly different electron thermal transport[9] and mixing of temperature layers.

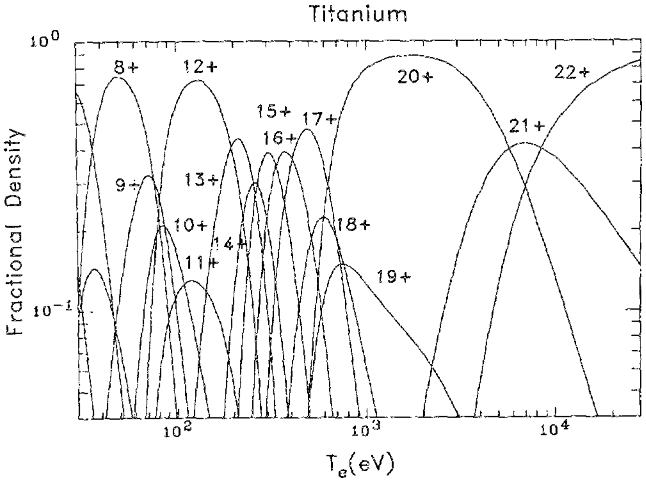

FIG. 2. The dependence of the relative population density of titanium ions on electron temperature in coronal ionization equilibrium.

Recombination (including dielectronic recombination) in the expanding plasma is believed to be the dominant process for the generation of CVI ions in the carbon plasma produced from the external parylene layer. The external layer is exposed to the maximum laser intensity and should have T_e exceeding 1000 eV,[8] resulting in a carbon plasma that consists initially of completely stripped carbon atoms. As the carbon plasma then expands and cools, CVI ions are formed and line radiation is emitted. This model correlates well with the observation that CVI and TiXIX, XX have very close expansion velocities, indicating that the source of these ions is in a plasma region with approximately the same high T_e. In addition, mixing of the low-Z carbon plasma can also contribute to producing similar expansion velocities.

VI. SUMMARY AND CONCLUSIONS

In the present experiment the spectrum of highly ionized titanium was studied in laser-produced plasmas. Significant relative shifts between the lines of TiXIII and TiXIX, XX were observed and attributed to Doppler shifts. The high ionization states TiXIX and TiXX were expanding outward more rapidly than TiXIII resulting in the observed Doppler shift. This observation is consistent with no charge state mixing of the titanium plasma. Other issues discussed here were the observation of a wide range of degrees of ionization in the laser-produced plasma and some of the relevant processes involved in producing these ions. Additional experiments are needed, with appropriate temporal and spatial resolution, to study the exact role played by the various processes involved in the generation of ions with different degrees of ionization. Such a study would be significant for efforts to develop a short wavelength recombining plasma laser and in understanding the effects of laser uniformity on laser-produced plasmas.

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