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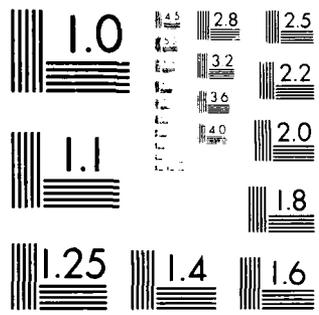
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19. ABSTRACT (Continue on reverse if necessary and identify by block number) The results of the Nd:YAG laser (1.064 m) experiments showed that the EUV output increased with laser pulse energy in a linear manner or slower. Therefore, there was no special incentive to go to higher pulse energies in the driver since increased problems of optical damage and of output saturation argued against moving in that direction. Because it was felt that higher repetition rates could translate directly into higher average outputs and published data indicated that the soft x-ray yield might be increased by use of shorter wavelengths in the laser driver, a 150 Hz excimer laser operating at 248 nm was purchased and installed as the driver for the laser plasmas. The peak pulse energy of this laser (Lambda Physik) with normal optics is 400 mj/pulse energy of 320 mj. Experiments with this system confirmed expectations and a reduction in exposure times for VUV and EUV emulsions of a factor of more than 20 when using the excimer rather than the Nd:YAG as a driver was observed. A major			
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disadvantage in the use of a laser plasma light source has been the generation of large amounts of debris from the target, which coats optics and lithograph masks. A series of studies intended to minimize the amounts and effect of target debris, using the excimer laser driver were performed. Experience gained from these studies provided the basis for the new generation laser plasma light source chamber just completed in the laboratory. A paper is being prepared describing the new generation light source in detail.

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Final Report: INTENSE XUV RADIATION SOURCES

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ABSTRACT

We have completed very high resolution studies of the XUV output of our laser produced plasma and have made photometric comparisons of different target materials in the 80 - 1000 Å region. In addition, we have installed a high average power excimer laser drive and have made preliminary comparisons of XUV outputs with excimer laser and Nd:YAG laser drivers. Finally, we have constructed and tested a new advanced plasma light source which should operate at a 150 Hz repetition rate for more than 1000 hours without maintenance, except for any required by the drive laser or the laser input window.

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TECHNICAL SUMMARY

This summary will be restricted to a brief outline of the major technical advances made under this contract. Detailed discussions of several portions of these efforts can be found in Appendices A through H. Five published papers form Appendices A through E while the title page and abstracts of papers presented at three meetings form Appendices F and H.

Earlier work demonstrated the usefulness of laser generated plasmas as intense light sources in the extreme ultraviolet (EUV) and soft X-ray spectral regions (10 - 300 eV). We have completed photograph and photometric comparisons of outputs from different target materials (Yb, Sm, Hf, Pb, W, Sn, Cu, steel, etc.) in the 80 Å to 1000 Å region using Nd:YAG or Ruby laser drivers. This work appears in Appendices A through D. We feel that the relative properties of Nd:YAG driven plasmas are now very well characterized in this spectral region. More recent work involved (a) installation of a high repetition rate and high average power excimer laser driver, (b) studies of various problems associated with laser generated debris from targets, and (c) construction of a new light source chamber designed for long term, maintenance free operation.

A few words of background relating to the more recent work may be helpful. The results of the Nd:YAG laser (1.064 μ) experiments showed that the EUV output increased with laser pulse energy in a linear manner or slower. Therefore, there was no special incentive to go to higher pulse energies in the driver since increased problems of optical damage and of output saturation argued against moving in this direction. Because it was felt that higher repetition rates could translate directly into higher average outputs and published data indicated that the soft x-ray yield might be increased by use of shorter wavelengths in the laser driver, a 150 Hz excimer laser operating at 248 nm was purchased and installed as the driver for the laser plasmas. The peak pulse energy of this laser (Lambda Physik) with normal optics is 400 mJ/pulse for 60 watts average power. The use of unstable resonator optics reduces the beam divergence to less than 600 microradians while reducing the peak pulse energy of 320 mJ. Experiments with this system confirm our expectations and we observe a reduction in exposure times for VUV and EUV emulsions of a factor of more than 10 when using the excimer rather than the Nd:YAG as a driver (see Appendix E).

LASER PRODUCED PLASMA LIGHT SOURCES FOR HIGH RESOLUTION XUV AND VUV SPECTROSCOPY

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Light from a laser produced plasma light source has been observed with the highest possible spectral resolution currently available in the 4-120 nm region. Plasma radiations observed from W, Yb, and Hf targets were found to be continuous with very few line emissions. Examples of highly resolved absorption spectra of He I and Ar I obtained using the plasma light source are presented.

Laser produced plasmas have been demonstrated to be convenient laboratory light sources for absorption spectroscopy below 50 nm [1]. While plasma emissions produced using rare earth (or heavier) elemental targets appeared to be predominantly continuous [1,2] (if viewing was restricted to the high density, high temperature focus region), it remained to be determined whether lines or other substructures would appear if the continua were observed at the highest possible spectral resolutions. To answer these questions we have observed (photographic detection) the output from a laser plasma light source [3] with (a) the 10.6 m grazing incidence spectrograph at the National Bureau of Standards in the 4.5-60 nm region and (b) the 6.65 m normal incidence instrument at the University of Maryland in the 30-120 nm region.

The plasmas were produced by focusing the output from a Nd:YAG laser (1.064 μm output) to a spot about 170 μm in diameter on a metal target using an $f = 30$ cm lens (see refs. [2,3] for details). For a typical laser pulse of 550 mJ in 25 ns, this spot size implies a maximum power density of about 10^{11} W cm^{-2} . The cylindrical metal targets could be rotated to produce fresh target material for every pulse, but for long exposures the targets generally were run in such a manner

that most target areas were used for several laser pulses. The plasmas were viewed at right angles to the incident laser beam with the normal to the target surface at the focal spot being approximately 45° to both the incident laser beam and the viewing direction. Line emission from surface impurities was minimized by cleaning the surface with low energy, superradiant pulses before taking the spectra.

A 10.7 m grazing incidence instrument with a 1200 grooves/mm grating was used in conjunction with a toroidal focusing mirror to study the short wavelength emissions from the plasmas (see fig. 1). Exposure times with a 50 μm slit on Kodak SWR plates typically were in the 10-180 s range (100-1800 laser pulses) when the laser was operated at 10 Hz. For comparison, about half the number of shots were needed to obtain similar exposures with a BRV spark source but, at one pulse every 20 s, the time required for an exposure was almost a factor of 100 longer. Emissions observed using W or Yb targets proved to be completely continuous in the 4.5-60 nm range studied, with very few emission lines (typically one every 2-3 nm). As is well known [1] these continua are most intense in the 10-20 nm region, and good exposures were obtained in the 6-25 nm region with W targets in 12 s (at 10 Hz and 640 mJ per pulse) with the 4.5-6 nm region weak but still usable. Exposures with Yb targets were similar, although exposures with Yb may require up to 50% more time to be comparable to W at the shorter wavelengths. To confirm that the observations were not predominantly of scattered light, the He I absorption spectrum was photographed in the 19-20.6 nm and 51-58.4 nm regions. During these helium absorption experiments, it was discovered that the source's continuum falls sufficiently rapidly in intensity above 50 nm that one can observe both the 51-58 nm spectrum and the third order of the 19-21 nm spectrum (first order near 60 nm) on the same exposures without order separation. The resolution in these third order absorption spectra is better

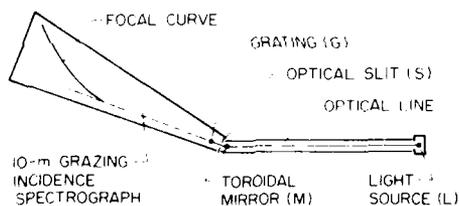


Fig. 1. Schematic diagram of the experimental arrangements used in the grazing incidence experiments. The distances between L and M and between M and S are 79 cm and 3 cm, respectively.

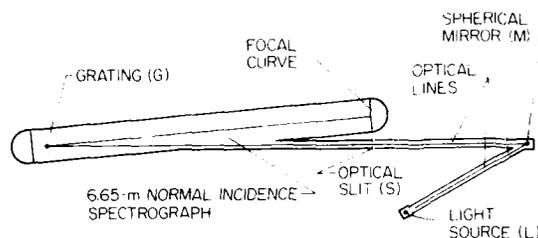


Fig. 2. Schematic diagram of the experimental arrangements used in the normal incidence experiments. The distances between L and M and between M and S are 275 cm and 300 cm, respectively.

than 0.001 nm, and the purely continuous nature of the Yb and W emissions in the approximately 20 nm region is confirmed to that level of observation. The reader is referred to ref. [4] for details and additional experimental results obtained with this system.

A 6.65 m normal incidence instrument with a 4800 grooves/mm grating (gold coated) blazed near 90 nm

was used in conjunction with a spherical focusing mirror (osmium coated) to study the longer wavelength emissions from Yb, W, and Hf plasmas (see fig. 2). First order exposures with a 50 μm slit on Kodak SWR plates ranged typically from 3 min at 50 nm to 20 min at 120 μm when the laser was operated at 10 Hz, with appropriate increases in exposure times when working with narrower slits or higher spectral orders. Emissions observed using W or Yb targets proved to be completely continuous in the approximately 30-120 nm region studied with very few line emissions - results entirely analogous to the shorter wavelength studies just discussed. The intensities of the continua decrease rapidly with increasing wavelength, so that we were able to observe the helium absorption transitions near 51 nm in both first and second orders (plate factors of 0.03 nm/mm and 0.015 nm/mm, respectively). Fig. 3 contains reproductions of typical first and second order absorption spectra of He I near the lowest ionization limit while fig. 4 provides a comparison of the absorption spectrum of Ar I observed near its lowest ionization limits taken with the plasma light source and with a

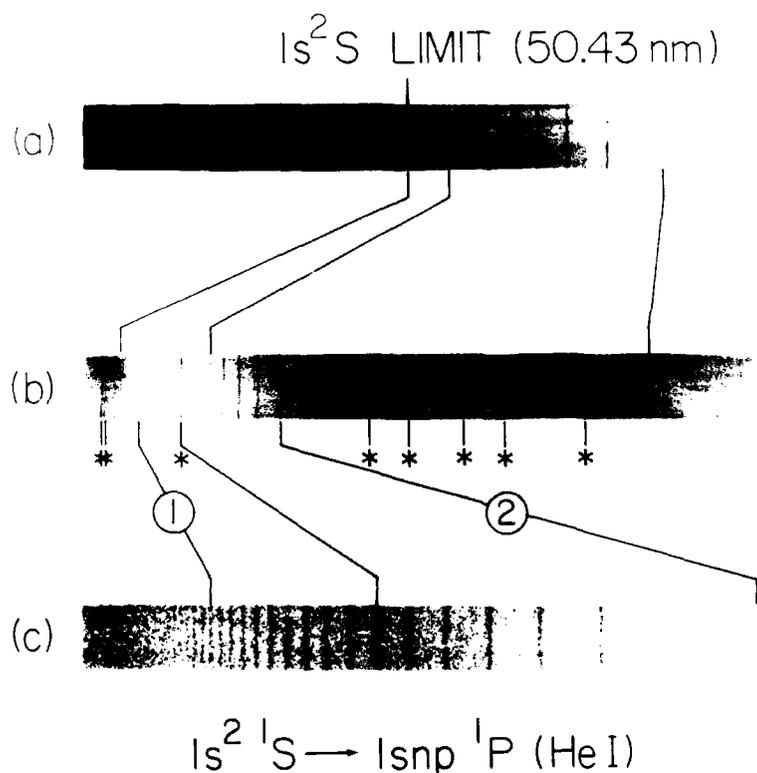


Fig. 3. Absorption spectra of the He I near the 50.43 nm ionization limit in the first (a) and second (b, c) orders of a 6.65 m spectrograph using a 4800 grooves/mm grating and a laser driven plasma light source with a Yb target. Lines 1 and 2 are the $1s^2 \ ^1S$ - $1s3p \ ^1P$ and $1s^2 \ ^1S$ - $1s14p \ ^1P$ transitions, respectively. Lines marked with an asterisk are impurities.

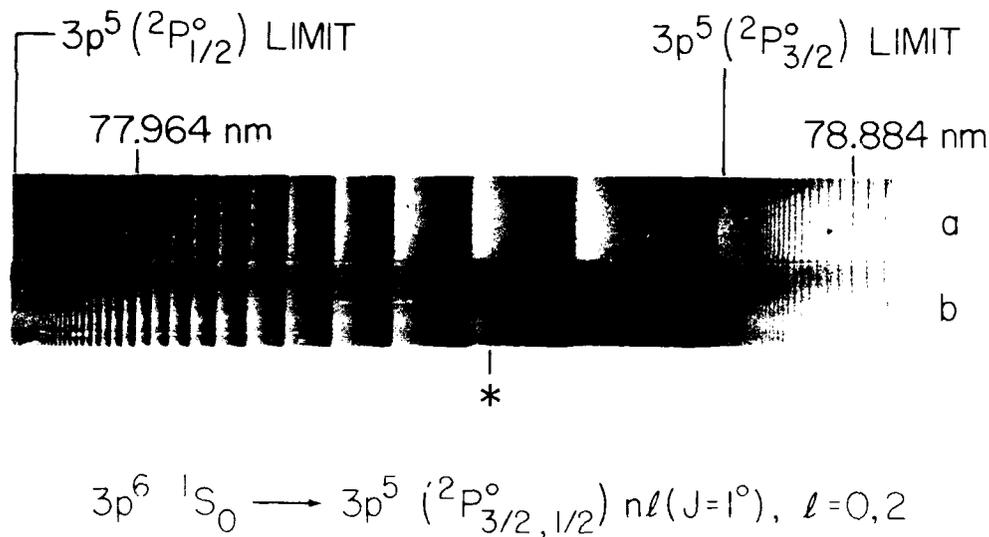


Fig. 4. Absorption spectra of Ar I near 78 nm using a helium continuum (a) and a laser plasma with Yb target (b) as background light sources. The only difference is the impurity line indicated by an asterisk.

helium (i.e., Hopfield) continuum light source. The resolution in the second order absorption spectra is better than 0.001 nm, so the purely continuous nature of the Yb, W, and Hf emissions in the approximately 50 nm region is confirmed to that level of observation. The reader is referred to ref. [5] for details and additional experimental results obtained with this system.

The work on the 10.7 and 6.65 μ m spectrographs was done in cooperation with P. Gohil and V. Kaufman, and with F. Orth and K. Ueda, respectively. This work was supported by the Air Force Office of Scientific Research under contract 4900-83-C-0130.

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High-resolution spectra of laser plasma light sources in the normal incidence XUV region

Frederick B. Orth, Kiyoshi Ueda, Thomas J. McIlrath, and Marshall L. Ginter

In proper conditions, laser-produced plasmas from Hf, Yb, W, or Pb targets are found to produce true continuum emissions which are essentially line free when studied with the highest spectral resolution currently available in the ~ 300 – 1200 -Å region. Examples of highly resolved absorption spectra taken in several orders of a 6.65-m spectrograph equipped with a 4800-groove/mm grating are included.

In recent years laser-produced plasmas using heavy metal targets have been utilized as intense sources of continuum radiation in the $\lambda < \sim 1000$ -Å spectral region.¹ The present work tests the continuous nature of these emissions using the highest spectral resolution currently available in the ~ 300 – 1200 -Å region, while a companion paper² describes similar tests in the ~ 45 – 300 -Å region. Specifically, we emphasize here observations in the 400 – 1200 -Å region of emission spectra from laser-produced plasmas using Cu, Hf, Yb, W, and Pb targets made using a 6.65-m normal incidence vacuum spectrograph equipped with a 4800-groove/mm grating. Our results establish that in proper conditions Hf, Yb, W, and Pb plasmas produce truly continuous emissions with negligible line contamination which are suitable for use in high-resolution absorption spectral studies. Using Cu targets we were unable to produce radiation suitable as a background for absorption spectroscopy.

The experimental arrangement employed is shown schematically in Fig. 1. Briefly, we used a 7.6-cm diam osmium coated concave mirror ($R = 3$ m) in near normal incidence to focus the image of the plasma onto the slit of the spectrograph and an $f = 30$ -cm lens mounted on a precision mechanical translation stage to position accurately the focus of the laser onto the surface of a cylindrical target. This arrangement allowed precise translation of the vertical image of the light from the plasma plume across the vertical entrance slit of the spectrograph. The focus of the laser beam was posi-

tioned on a cylindrical target so that the normal to the surface at the focal point was $\sim 45^\circ$ to both the incident laser direction and viewing direction of the plasma. The spectrograph and optical beam lines between the target and spectrograph slit were evacuated to pressures between 10^{-4} and 10^{-5} Torr for most experiments with low pressures of sample gases added to either the beam line or spectrograph when taking absorption spectra. The spectrograph is a 6.65-m Eagle mounting (MacPherson) equipped with a mechanically ruled³ gold-coated 4800-groove/mm grating with a gold coating which achieves⁴ resolving powers (RP) in excess of 180,000 in first order, and all spectra were recorded on Kodak SWR plates. The light source and its driver, a Nd:YAG laser with a 10-Hz repetition rate and ~ 600 mJ/pulse, are described in detail elsewhere.⁵ Estimates of power densities on the target for tightest focusing⁶ are in the 10^{11} – 10^{12} W cm⁻²/pulse range.

Studies were made in the first order (reciprocal dispersion = 0.3 Å/mm) using five targets: Cu ($Z = 29$); Yb ($Z = 70$); Hf ($Z = 72$); W ($Z = 74$); and Pb ($Z = 82$). The continuum intensities from Yb, Hf, W, and Pb were similar yet of somewhat different spectral distributions so that, for any specific wavelength interval within the range studied, emission from one or the other of the targets might appear stronger by as much as a factor of 2. We will not elaborate further on these relatively small emulsion exposure differences because photometric measurements made with comparable spectral RP and plasma imaging conditions will be necessary before such comparisons could be even qualitatively accurate. The four higher Z element plasmas can be made to produce clean continua with very few emission lines. Under continuum optimized conditions the few line emissions are very weak and diffuse, are common to all four plasmas, and can be assigned to transitions in ionized oxygen (i.e., $2s^2 2p^2 - 2s 2p^3$ in O III, $2s 2p^2 - 2p^3$ in O IV, $2s 2p - 2p^2$ in O V). It should be emphasized that to obtain intense line-free continua requires careful imaging of the hottest portion of the

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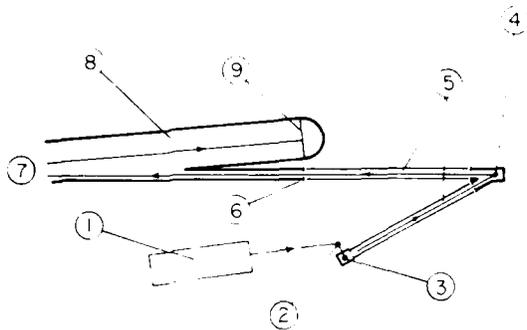


Fig. 1. Schematic diagram of the experimental arrangements: (1) Nd:YAG laser; (2) plane deflecting mirrors and focusing lens; (3) target and source point of plasma emissions; (4) concave mirror, radius = 3 m, Os coated; (5) optical beam line; (6) entrance slit of the spectrograph; (7) beam toward concave diffraction grating, 4800 groove/mm, Au coated; (8) main tank of 6.65-m spectrograph; (9) camera containing photographic plates on focal curve of grating. The distances between (3) and (4) and between (4) and (6) are 275 and 300 cm, respectively.

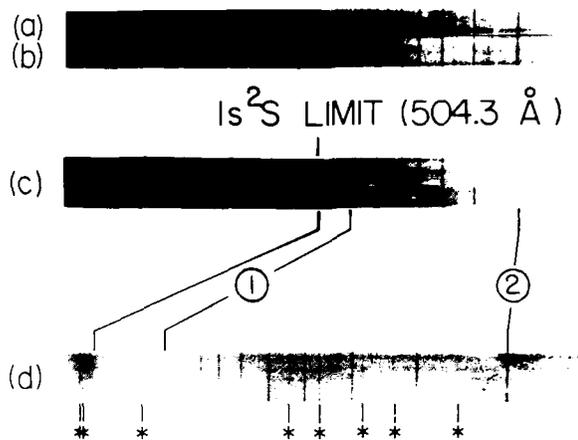


Fig. 2. Absorption spectra (black against white of continuum) for He I near the 504.3-Å ionization limit taken using a 6.65-m spectrograph with a 4800-groove/mm grating and a laser plasma light source with various targets. Spectra (a), (b) and (c) were taken in first order with 50- μ m slit widths and 6-min exposures (3600 plasma pulses) using W, Hf, and Yb targets, respectively. Spectrum (d) was taken in second order (36 min with a 20- μ m slit width) using a Yb target. Lines 1 and 2 are the $1s^2\ ^1S-1s18p\ ^1P^o$ and $1s^2\ ^1S-1s8p\ ^1P^o$ transitions, respectively, while lines marked with an asterisk are first-order impurity absorptions on the predominately second-order exposure.

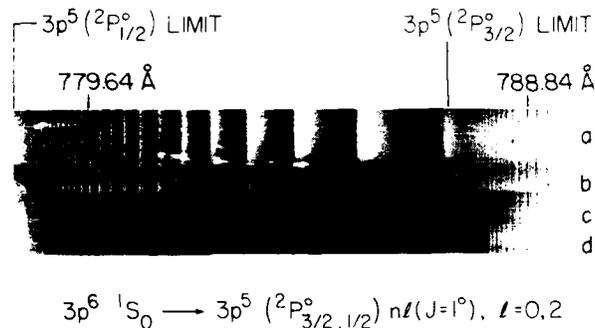


Fig. 3. Absorption spectra (dark lines) for Ar I in the 778-787-Å region using for background a helium continuum (a) and laser plasma radiation from Yb (b), Hf (c) and Pb (d) targets. Exposure times with 20- μ m slits were 6 min for the helium discharge and 30 min (or 18,000 plasma pulses) for the laser plasma spectra.



Fig. 4. Typical laser plasma spectra from a Hf target obtained as the laser plasma image is moved across a 50- μ m entrance slit of the spectrograph. The laser plasma image is \sim 1 mm in diameter when photographed using only VUV radiation [see Fig. 4(a) in Ref. 5]. Note that emission (line and/or continuum) appears white against black (see text). Spectra a, b, c, and d each were 4-min exposures taken at 60- μ m intervals from the optimal intensity positioning a. Spectrum e is identical to d, except the exposure time was increased to 12 min.

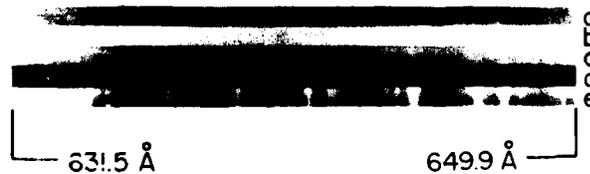


Fig. 5. Typical laser plasma spectra from a Pb target obtained as the laser plasma image is moved across the 50- μ m entrance slit of the spectrograph. Spectra a, b, c, and d each were 4-min exposures taken at 100- μ m intervals about an optimal intensity positioning near b. Spectrum e is identical to d except for increased exposure time (12 min). See caption to Fig. 4 and text for additional details.

plasma onto the entrance slit of the spectrograph (see below). Figure 2 contains examples of absorption spectra of the principal series of He I taken in the first order using optimal focusing of the laser on W, Hf, and Yb targets as well as a second-order spectrum taken without order sorting and using a Yb target to provide the background continuum. Figure 3 contains examples of absorption spectra of Ar I observed near the lowest two series limits taken in first order using as background continua (1) the plasma source with Hf, Yb, or Pb targets and (2) the Hopfield continuum from He₂ produced by a condensed discharge through a tube containing flowing He at $P \approx 25$ Torr placed directly in front of the slit of the spectrograph. In Figs. 2 and 3 absorption lines appear as dark lines against a light (emission) background. Finally, the continuum intensity from the Cu target was found to be weaker than those observed from the high Z element targets by approximately an order of magnitude with the Cu plasma emission dominated by strong line spectra.

It was noted in previous studies⁷ using Yb targets that line emission contamination near 1216 Å could be greatly reduced by focusing the image of the source on a slit. We find that both continuum output and emission line suppression can be optimized simultaneously by careful placement ($\sim \pm 100 \mu\text{m}$ for the source and optical system described above) of the plasma image onto the spectrograph slit with a width of 50 μm or less. Figure 4 shows the effect of translating the image of the plasma produced using a Hf target across the slit of the 6.65-m spectrograph for a typical spectral region. It should be emphasized that Fig. 4 is printed from the original plate (a negative) so that emission exposure (either continuum or line) appears as whitening rather than blackening (recall Figs. 2 and 3). In Fig. 4 spectra $a-d$ have identical exposure times and slit widths but different image placements on the slit, while exposure e has the same image placement and slit width as d but an increased exposure time. As can be seen from Fig. 5 (which is analogous to Fig. 4 in construction) the results using Pb targets are similar to those observed for Hf, although emission line suppression is slightly more critical to focal spot positioning in Pb than in Yb. The approximately $\pm 100\text{-}\mu\text{m}$ tolerance noted above corre-

sponds approximately to the measured⁶ spot size of the focused laser beam on the target ($\sim 170 \mu\text{m}$ in diameter) and, therefore, probably corresponds to sampling the image of the hot core of the plasma.

In the work described above for the four higher Z elements the apparent spectral intensity variations with wavelengths (i.e., the observed intensity distributions uncorrected for grating efficiency, grating and mirror reflectivities, or plate sensitivities) show maxima around 500–600 Å with rapid falloff toward both shorter and longer wavelengths. The shorter wavelength falloff in apparent intensity almost certainly is due to decreases in reflectance and grating efficiency,^{3,4} since the peak in the output from the plasma source is known to occur at shorter wavelengths.¹ We estimate that the relative apparent intensities of the continua at 600, 800, and 1000 Å are 6, 3, and 1, respectively. Thus the apparent intensity of the second order of the 500–600-Å region is slightly more intense than the first order of the 1000–1200-Å region, which permits the combined light source and optical system to be used for higher-order absorption studies (see, for example, Fig. 2) without order sorting.

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High-resolution spectra of laser plasma light sources in the grazing incidence region

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Studies of emission from laser-produced plasmas have demonstrated their utility as sources of intense XUV and soft x-ray radiation.¹ To be useful as a laboratory spectroscopic light source, it is desirable that the source have high intensity, good reproducibility, and emit a clean continuum with a minimum of contaminating lines. Because the most promising targets are the high-Z rare earth materials radiating from ionized systems with partially filled shells,² any line emission is likely to be extremely dense and easily confused, at low resolution, with continuum emission. In this Letter we report on studies of the emission from Cu, Yb, and W targets in the region from 45 to 250 Å with the NBS 10.7-m grazing incidence spectrograph. The Yb and W produce very intense and very clean continua in the 45-250 Å region. The Cu spectrum is heavily contaminated by emission lines at the longer wavelengths but produces a relatively clean continuum below 150 Å.

The laser used in this experiment was a Nd:YAG producing a nearly diffraction-limited beam at 1.064 μm. The measured spot size using a single 300-mm focal length plano-convex lens was 170-μm FWHM or approximately three times the diffraction limit. The laser pulse energy was ~630 mJ in 20 ns at a 10-Hz repetition rate giving a power density on target of 10¹¹-10¹² W cm⁻², although one series of exposures was made with a single pulse energy of 100 mJ. The target was a cylindrical rod which was rotated between shots to provide a fresh surface on each shot. The incident laser beam and the viewing direction were orthogonal and the target normal was ~30° from the viewing direction. Both the laser and the target chamber are described in more detail elsewhere.³

The target was placed 82 cm from the entrance slit to the spectrograph and focused onto the slit with an aluminized toroidal mirror placed ~3 cm from the slit. The target surfaces were cleaned with a weak laser pulse before spectra were taken in order to reduce emission from surface oxides. The target materials used were solid copper rod, sintered tungsten rod, and ytterbium sheet molded around a cylindrical

center rod. The spectrograph was the National Bureau of Standards 10.7-m grazing incidence photographic instrument with a 1200-grooves/mm grating. This instrument has resolved spectral lines that were separated by $\Delta\lambda = 0.005$ Å. Although the resolution was reduced by the use of a 50-μm wide slit ($\Delta\lambda = 0.04$ Å), the resolving power was greater than any previously reported for laser plasma continuum light sources. The spectra were recorded on 101-05 plates.

Sample spectra for copper, tungsten, and ytterbium targets are shown in Fig. 1. The copper target gave an intense continuum, but it was heavily contaminated with emission lines above 150 Å. At shorter wavelengths, and especially below 100 Å, the emission is predominantly continuum. Because of their freedom from line emission, most of our effort was spent on tungsten and ytterbium targets.

The W and Yb emissions were excellent sources of continua below 250 Å. The density of emission lines was typically one every 20 or 30 Å on the plate. In addition, the continuum was extremely intense. Good exposures were obtained in the 60-250-Å region with W targets in 12 s at 10 Hz and 640 mJ/pulse delivering a total of 77 J to the target; exposures from 45 to 60 Å were weak but still usable at these energies. The continua from Yb targets were comparable in intensity.

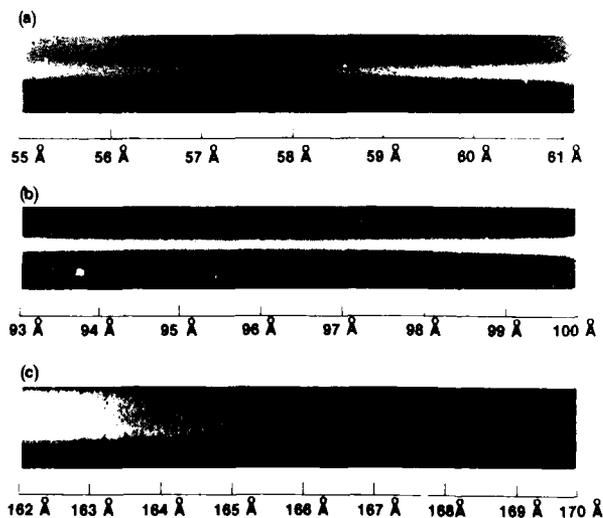


Fig. 1. Spectra of (a) copper 55 Å to 61 Å; (b) tungsten 93 Å to 100 Å; (c) copper 162 Å to 170 Å.

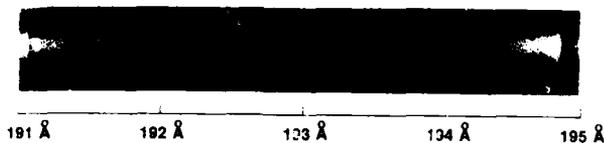


Fig. 2. Absorption spectrum of He, 191 Å to 195 Å, taken in third order with a ytterbium continuum and no order separator

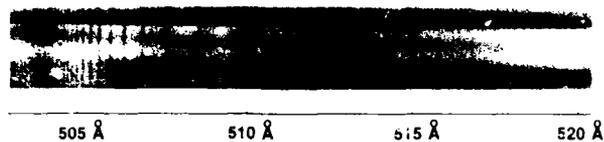


Fig. 3. Absorption spectrum of He, 501 Å to 518 Å, taken in first order with a ytterbium continuum.

possibly requiring up to 50% more exposure time at short wavelengths. Both sources were comparable in the uniformity of the continuum.

One of the questions associated with the use of laser plasma light sources is how the character of the spectra changes with incident power density. To study this, we recorded Yb spectra with the laser intensity reduced by a factor of 6 to 100 ml/pulse. Good exposures were obtained by increasing the exposure time by a factor of 3. The spectrum was not visibly changed and there was no increase in line contamination. The one visible change was the apparent decrease in source size. This manifested itself most obviously in the 45–100-Å region where the spectrograph toroidal mirror combination has a low astigmatism. This reduced source size probably accounted for the good exposures at half of the delivered energy used at higher laser powers.

One feature of these sources is the localization of the output in broad spectral regions. In the case of Yb and W the exposures were especially weak above 500 Å. Part of this undoubtedly reflects the properties of the spectrograph; however, absorption spectra of He were obtained and it was seen that most of the continuum observed in the region of the first order was in fact third order of the 200-Å emission. This is seen in Fig. 2 where the 195 Å and the next four ionizing resonances are observed in third order. The contrast across the line indicates the paucity of first-order radiation. Second-order radiation was presumably ab-

sorbed by the He continuum. The resolution in third order is ≈ 0.01 Å, and the purity of the continuum shown in this figure is typical of the Yb and W outputs. Figure 3 shows the absorption spectrum of neutral He in the region above 500 Å. These indicate that continuum emission is still usable at 500 Å but becomes quite weak at ~ 600 Å.

To allow a comparison of intensities the same experimental setup was used with a BRV spark source to obtain exposures in the same spectral region. Exposures obtained with 150 shots, requiring 40 min, were significantly weaker than the laser plasma spectra and were not usable below ~ 150 Å. The BRV source also had considerable line emission overlying the continuum. The comparison is made more difficult by the wandering of the location of the BRV spark which spreads the short-wavelength stigmatic spectra over a considerably larger region of the plate than the laser source. Work with an imaging system on a normal incidence spectrograph (Orth *et al.*⁴) indicates that the ability to image a small, fixed region of the source onto the spectrograph entrance slit is an important factor in eliminating line emission.⁴

This work was partially supported by Air Force grant F496201-83-C-0130. P. Gohil acknowledges support from the Science & Engineering Research Council (U.K.) in the form of a postdoctoral research fellowship.

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Investigation of a laser-produced plasma VUV light source

J. M. Bridges, C. L. Cromer and Thomas J. McIlrath

An investigation was conducted on the VUV radiation from laser-produced plasmas using a channel electron multiplier detector and a 1.5-m grazing incidence spectrometer. High-resolution quantitative spectra from 8 to 40 nm were obtained from the plasmas generated by a 0.5-J Nd:YAG laser focused on nine different target materials. The effects on the plasma emission of laser energy and focus were measured.

I. Introduction

When a high-energy pulsed laser is focused onto a solid target a dense high-temperature plasma is generated. Spectral line radiation as well as continuum radiation is generally emitted from such plasmas, and their value as vacuum ultraviolet (VUV) and soft x-ray sources was recognized soon after the development of high-power lasers. Early studies of laser-produced plasma radiation concentrated on line emission originating from multiply ionized species. More recently, several studies have been directed toward the continuum radiation originating from bremsstrahlung and recombination radiation and from a large number of blended spectral lines from targets having complex ionic configurations. Breton and Papoular¹ used both Nd:glass and ruby lasers with tantalum targets to produce intense radiation in the region of 121.5 nm; their studies include measurements of the absolute spectral intensity of the generated radiation, its angular distribution, and its dependence on laser power. Several workers have extended the studies to the soft x-ray region. Nagel *et al.*² investigated emission between 1.4 and 30 nm produced by a Nd:YAG laser on several target materials.

Much of the work to date has been directed toward maximizing intensities over a broad spectral region, and studies have been carried out with low spectral resolution using target materials which yield a rich mixture of line and continuum radiation. Recent work by Carroll *et al.*,³ however, has included high-resolution studies between 4 and 200 nm, which have shown that with the proper choice of target material a virtually line-free continuum can be produced over extended spectral regions.

The absolute spectral irradiances of laser-produced plasma continua have been measured from 115 to 200 nm with low resolution by O'Sullivan *et al.*⁴ and from 7 to 100 nm by Fischer *et al.*⁵ These measurements also showed the irradiance to be quite reproducible, generally as good as or better than the reproducibility of the driving laser power. These factors have led to consideration of laser-produced plasmas as background light sources for VUV spectroscopy and materials studies and as radiometric standards in the spectral region from 1 to 100 nm where no portable standards currently exist.

Further work is required, however, to establish the most effective sources for different spectral regions both in terms of spectral purity and maximum intensity and to establish this type of source as an irradiance standard. In the present work, we report spectra between 7 and 40 nm obtained from nine target materials. In contrast to previous qualitative photographic observations or monochromator scans, we have obtained photoelectric intensity measurements with high resolution. This was accomplished by use of a recently constructed instrument using a photoelectric array detector.⁶ The measurements were made with a 0.5-J low-divergence Nd:YAG laser and include investigations of the effect on the radiation of laser energy and focus. In addition some measurements were made with a 4-J ruby laser for comparison.

II. Experimental Arrangement

The details of most components of the apparatus have been described elsewhere⁶ so that only a summary of the system is given here. The laser was a Q-switched Nd:YAG laser generating ≈ 0.5 J in ≈ 25 ns. The output had a focused spot size of approximately four times the diffraction limit in diameter. The radiation was focused onto the target with a 100-mm focal length plano-convex lens giving a measured focal spot diameter, full width at half maximum, of 60 μm . However, because of the possibility of aberrations intro-

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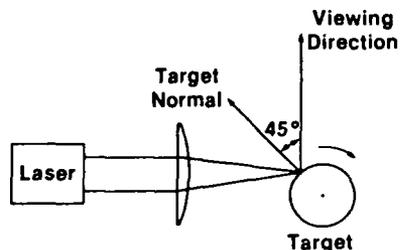


Fig. 1. Diagram of target alignment.

duced in the measurement process by the reflectors and neutral density filters used for this determination, this figure should be considered an upper limit. We assume the diffraction diameter of $15\ \mu\text{m}$ as a lower limit. Thus the peak power on target was 10^{11} – $10^{12}\ \text{W cm}^{-2}$.

The targets were cylindrical in shape, 15 mm in diameter \times 6 mm long, mounted on a vertical threaded rod. After each laser pulse the target was moved to provide a clean surface for the following shot. To provide a comparison of the various target materials, several targets were mounted on the same shaft to allow the targets to be changed without breaking vacuum.

As shown in Fig. 1, the incident laser beam was normal to the viewing axis with each axis being 45° from the normal to the target material. The radiation from the target was viewed by the spectrometer either directly through an entrance slit 50 cm from the target or by using a toroidal focusing mirror placed 10 cm from the spectrometer slit and used at 5° incidence angle. The spectrometer was a 1.5-m grazing incidence instrument with a 1200-groove mm^{-1} grating. The detector assembly was mounted in a chamber attached to the spectrometer scanning carriage in place of the normal exit slit and could be moved over the entire usable spectral region of the spectrograph without breaking vacuum. A zero-order trap and baffles were used to reduce zero-order and other scattered radiation.

The detector was a channel electron multiplier array (CEMA) which has been described in detail elsewhere.⁶ The CEMA provides amplification into a phosphor which is connected to a self-scanned diode array by a fiber-optic bundle. The CEMA channels are separated by $15\ \mu\text{m}$, and the diode array elements are $25\ \mu\text{m}$ in width. Electron repulsion and imperfections in the fiber coupler limit the CEMA spatial resolution to 1.2 pixels for a combined instrumental resolution of 0.01 nm (or 3 pixels with 1200-line/mm grating and $10\text{-}\mu\text{m}$ entrance slit). The output of the diode array was digitized and stored by a computer for processing. A spectrum was obtained with a single laser shot, but at least 20 shots were averaged to improve the SNR.

Structure in the output due to spatial gain variation across the CEMA was found to be $\sim 20\%$ of full scale. It was determined that the gain profile was sensitive to the angle of incidence of the radiation presumably because of changes in the penetration of the incident radiation into the microchannels. Thus it was impossible to measure the gain variation by uniformly illuminating the CEMA at normal incidence. The gain profile was determined by continuously scanning the CEMA carriage over a large spectral range while firing the laser and accumulating data. In this way each array element was exposed to approximately the same integrated spectral irradiance at approximately the same angle of incidence. The accumulated gain profile was stored and used to divide into each measured segment of spectra, thereby eliminating apparent structure due to the gain profile itself.

To obtain the emission spectra for a given target material, the CEMA was successively placed in twelve overlapping positions to cover the entire 7–40-nm spectral range. For each position, the data from twenty laser shots were accumulated, detector background signal subtracted, and gain fluctuations divided out. The spectra for adjacent CEMA positions did not usually match perfectly in the overlapping regions because of laser power drifts and overall changes in CEMA efficiency with angle of incidence. Thus the relative normalization for each position was changed to match the adjacent position. This normalization process, combined with the spectral variation of the system efficiency, precludes drawing conclusions concerning the relative intensities from any target material at widely separated wavelengths, although relative intensities at a particular wavelength were obtained for all the target materials and are accurate to better than 20%.

III. Results

A. Spectra

The spectra from the various targets are reproduced in Figs. 2 and 3. The intensity scales at a given wavelength are the same for all materials. Significant differences between the various target materials are immediately apparent. Qualitatively they range from aluminum, which is predominantly a line spectrum, through samarium and ytterbium which are almost pure continua. Common lines in the samarium and ytterbium spectra at 22.5 and 17.0 nm are oxygen lines from surface oxide layers. In general, the rare earths and tungsten give the most line-free continua in agreement with the reports of Carroll *et al.*³ The rare earth continua show similar broad maxima with the peaks occurring at successively longer wavelengths with decreasing Z as discussed by O'Sullivan.⁷ The line spectra of aluminum and copper give good illustrations of the absence of second-order spectra. The known strong features at 16.0 nm in aluminum, for example, are not seen at 32.0 nm; the signal at 32.0 nm cannot, therefore, be due to second-order radiation.

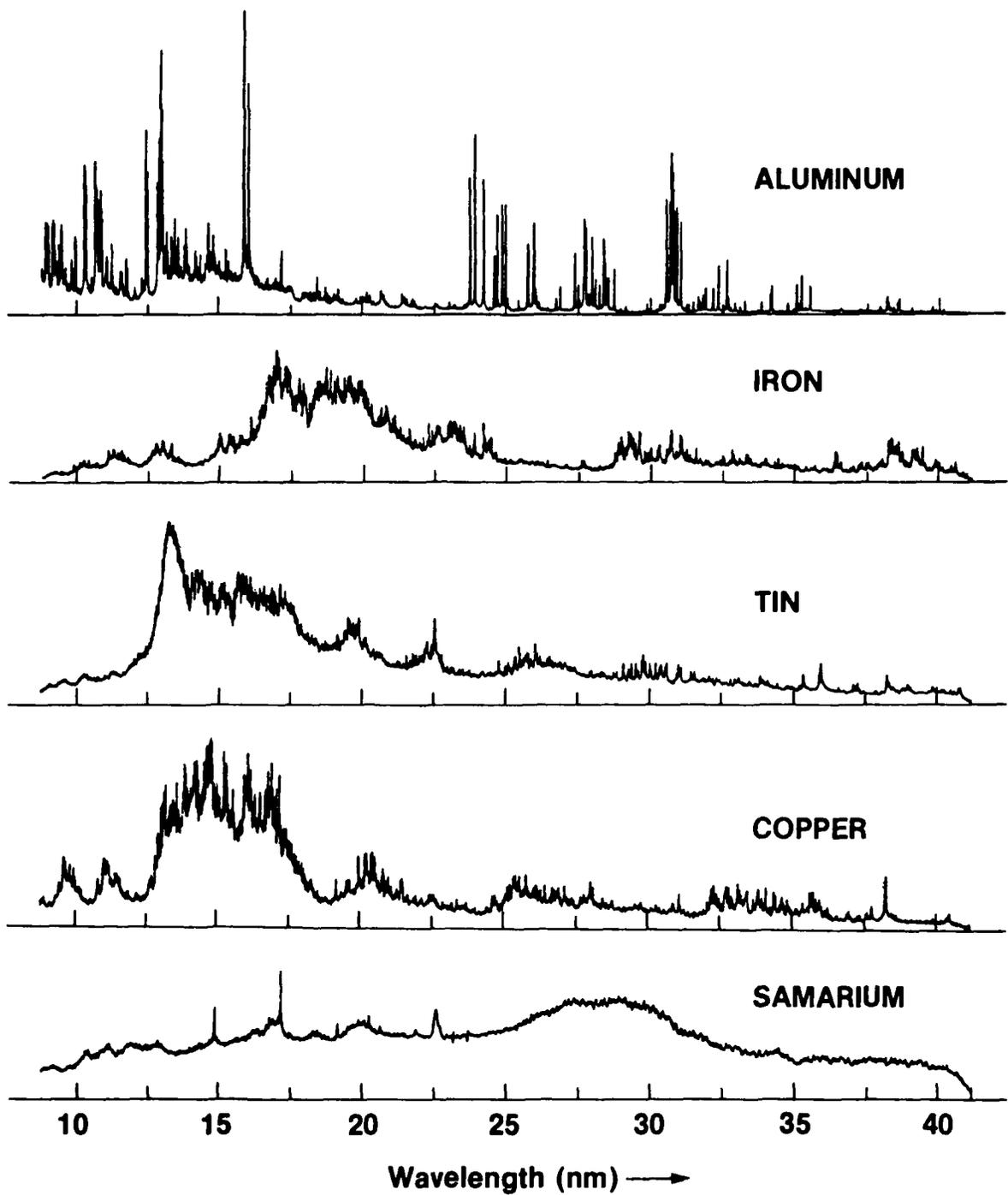


Fig. 2. Spectra from various target materials. The intensity scales are the same for all spectra.

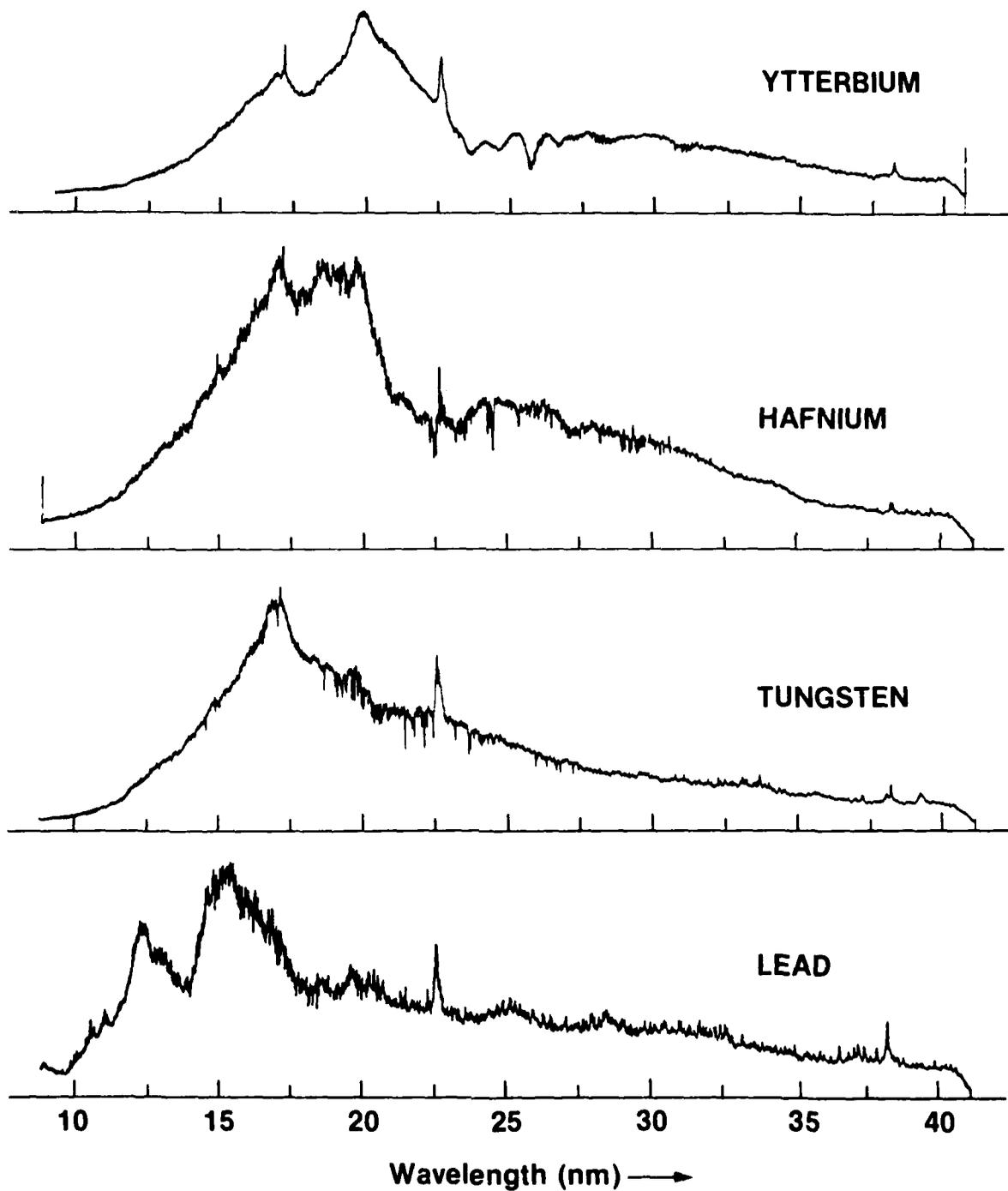


Fig. 3. Spectra from various target materials. The intensity scales are the same for all spectra.

B. Intensities

The use of photoelectric detection with the CEMA, as opposed to photographic detection, provides accurate intensity information. Since our spectrometer system has not been calibrated, the data are valid only for relative intensities, and no absolute irradiance values can be given. Also, the variation with wavelength for each target reflects the wavelength response function of our system, but the sensitivity should vary slowly with wavelength so that local variations in intensity are accurately portrayed. Moreover, Fischer *et al.*⁵ have calibrated the spectrum for a tungsten target irradiated by a Nd:YAG glass laser. Comparing our spectrum from a tungsten target with their calibrated spectrum should give a good estimate for the response of our system as a function of wavelength. Such a comparison indicates that our system has its peak efficiency at 19.0 nm and drops off smoothly to a value of 0.2 times the peak efficiency at 11 nm on one side and at 40 nm on the opposite side.

As mentioned previously, a toroidal mirror focused the radiation from the target onto the spectrometer slit. The mirror was used to increase the intensity at the slit to allow a smaller slit width and higher spectral resolution. For the tungsten target, a scan was also made with the mirror removed, and direct irradiation from the target to the slit was used. For this condition the observed spectral distribution of the radiation was unchanged from the distribution observed using the mirror, showing that with tungsten targets the spectrum is not critically dependent on the observed area of the source. In fact, the difference in the observed areas of the source with or without the mirror is limited by the large astigmatism and aberrations of the toroidal mirror.

The data in Figs. 2 and 3 show the large enhancement in continuum intensity as the target moves to heavier Z. The most intense output was obtained using Hf with Yb providing an excellent choice for high intensity and line-free continuum. Samarium also produced good intensities above 25 nm with a greatly reduced output near 15–17.5 nm compared with Hf or Yb. The primary value of Al is seen to be as a line source. Copper is strongly contaminated with lines but has a substantial underlying continuum, especially at short wavelengths. These results are consistent with the reports on photographic spectra by Carroll *et al.*³ but give a quantitative measure of the relative intensities of different source materials.

C. Dependence on Focus and Laser Energy

By moving the laser focusing lens along the optical path, the dependence of the signal on focus was measured. The result is shown in Fig. 4 for an ytterbium target with a wavelength bandpass of 3 nm centered at 20 nm. To avoid the effects of small displacements of the imaged source relative to the spectrograph slit, the intensities for varying focus conditions were measured

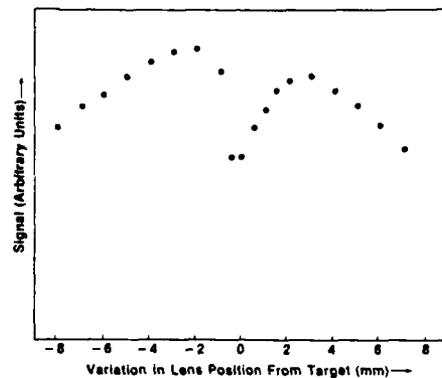


Fig. 4. Dependence of signal on the lens-target separation (Nd:YAG laser). Positive abscissa values correspond to lens closer to target.

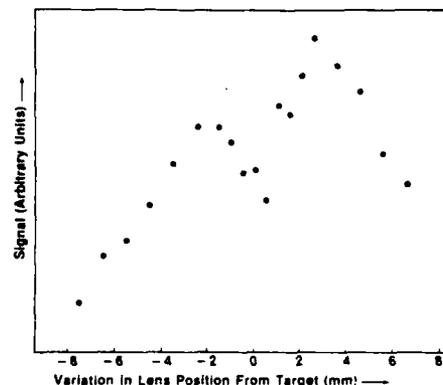


Fig. 5. Dependence of signal on lens-target separation (ruby laser). Positive abscissa values correspond to the lens closer to target.

without the toroidal mirror. Similar results were obtained with the mirror in place, however.

The lens was a simple plano-convex 100-mm focal length glass singlet, and the measured focal spot diameter, FWHM intensity, was 60 μm . The theoretical spot diameter for a diffraction limited beam is 15 μm , and the confocal beam parameter is 4 mm. The data in Fig. 4 were taken with a Yb target and the Nd:YAG laser at 500 mJ. Lower numbers of the lens position correspond to focusing outside the targets. Figure 5 shows a similar scan using a ruby laser at 3 J and a tungsten target. The results are qualitatively the same, although the beam quality of the ruby laser beam is markedly inferior to that of the Nd:YAG laser. It is not clear what is the cause of the narrow intensity dip. At the best focus the power density on target is a maximum, but the irradiated area is a minimum. It would appear that the maximum VUV intensity occurs with a larger than minimum emitting area and a reduced incident power density. This implies a kind of saturation effect. However, Fig. 6 discussed below shows a linear dependence of the VUV output vs laser energy for the Nd:YAG laser. Analysis of the dependence of output signal on the position of the focusing

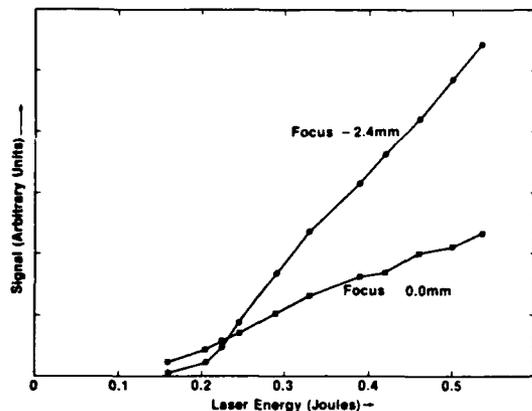


Fig. 6. Signal vs laser energy. Nd:YAG laser on ytterbium target; $\lambda = 20$ nm. The two curves are for the lens positioned for maximum signal at 500-mJ energy (focus -2.4 mm) and for the lens positioned to give the dip in the signal (focus 0.0 mm).

lens is complicated by the fact that the laser image is not a diffraction-limited Gaussian spot. We have no detailed knowledge of the intensity distribution or of the variation of the distribution near the focus. Some inherent astigmatism could even give separated intensity maxima. For a more definitive study of this effect, one would need to have a well-characterized laser beam. For the purpose of providing a reproducible radiation source, however, the best lens-target separation is that which gives a maximum signal. This distance is easily found and gives the least sensitivity of signal to motion of the lens or target.

The dependence of the VUV output and spectra on laser energy were investigated by varying the incident laser energy for fixed focus conditions. Figure 6 shows a plot of the integrated signal between 18.5 and 21.5 nm using a Yb target and Nd:YAG laser energies from 160 to 540 mJ. Two curves are shown, one for which the lens was positioned for maximum signal at 500-mJ incident energy (focus -2.4 mm) and one with the lens positioned at the intensity dip shown in Fig. 4 (focus 0.0 mm). Both curves show a general linear dependence with an energy threshold, but the two curves have different slopes. They cross at ~ 230 mJ so that the lens position giving a reduced output at higher energies gives a greater output at lower energies. This is consistent with the dip occurring at the focus position. The laser energy was varied in these experiments by varying the amplifier voltage while keeping the oscillator conditions fixed to minimize variations in beam character. Figure 7 gives the results of some measurements of signal dependence on laser energy for the ruby as well as Nd:YAG laser. These data are for the VUV output near 18 nm from a tungsten target. The focus was adjusted for maximum signal for each laser. These data show the threshold VUV output using the ruby laser to be considerably higher than that using the Nd:YAG laser and also show evidence of output signal saturation at higher incident powers using the ruby laser.

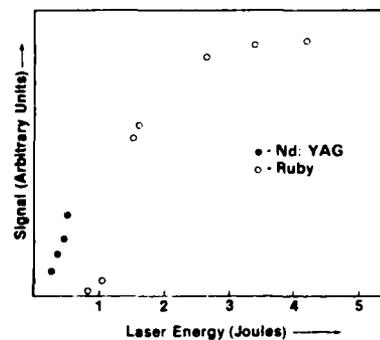


Fig. 7. Signal vs laser energy. Nd:YAG and ruby lasers; tungsten target, $\lambda = 18$ nm. The lens was positioned for maximum signal for both lasers.

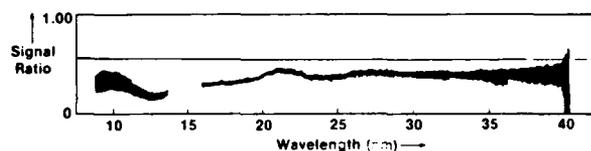


Fig. 8. Ratio of signals from Hf target for laser energies of 290 and 520 mJ, respectively. The horizontal line is drawn to give the ratio of these two laser energies.

To study the effect of incident power on the spectral distribution of the output, Hf was irradiated at both 290- and 520-mJ pulse energy. Figure 8 shows the ratio of the resulting spectra. The spectrum at 520 mJ is shown in Fig. 2. It is clear that above 20 nm the spectra are essentially identical with an output proportional to the input energy. Below this wavelength there is a decrease in the relative intensity of the output in the 290-mJ incident energy spectrum indicating a faster than linear drop in output at short wavelengths. There is no obvious increase in the line content of the spectrum at lower incident laser powers.

IV. Target Debris

One major disadvantage of laser plasma sources is the production of large amounts of splattered material from the target. The Nd:YAG laser was operated at a 1-Hz repetition rate for the Q -switched pulse in order not to overload the capabilities of the data acquisition system but with a 10-Hz repetition rate of the flashlamps to maintain a constant heat load on the laser system which was aligned at 10 Hz. On each of the nine pulses which were not Q -switched a superradiant pulse of $\approx 25 \mu\text{J}$ and long duration was observed. Although the energy of the superradiant pulse was orders of magnitude less than the 500-mJ Q -switched pulse, it was found to produce comparable cratering on the target and to contribute comparable amounts of debris as the Q -switched pulse. Figure 9 shows target cratering for different target materials in various conditions. It was found that use of a mechanical shutter to block the superradiant pulses significantly reduced the debris problem in the system. In addition, it was found

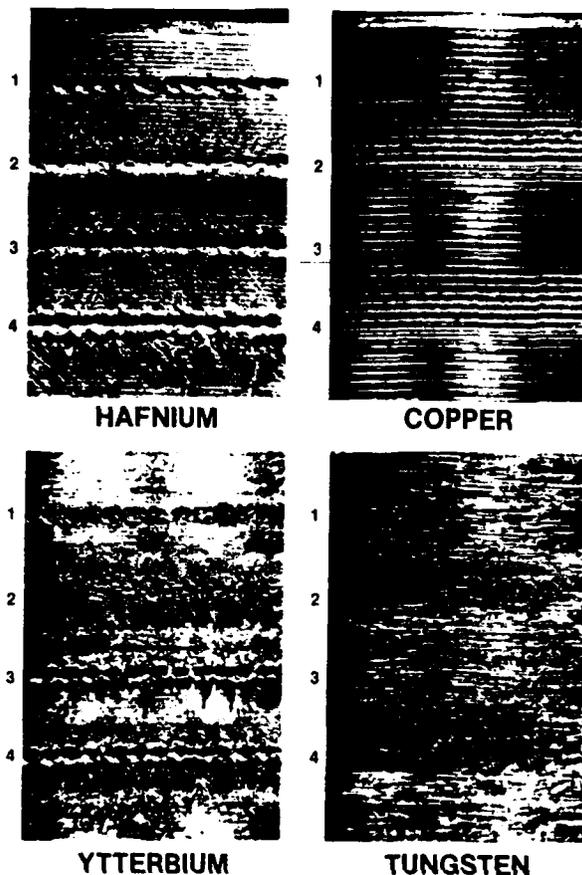


Fig. 9. Target damage for hafnium, ytterbium, copper, and tungsten. Each target was irradiated with four rows of laser shots as follows. From top to bottom, at each target position there was (1) one superradiant pulse; (2) one Q-switched pulse; (3) ten superradiant pulses; (4) nine superradiant pulses followed by one Q-switched pulse. The horizontal grooves in the nonirradiated areas of the targets are from machining.

that if only the Q-switched pulses are allowed into the system, the incident laser pulse kept the central portion of the input window clean by ablating any accumulated debris with each shot.

The rare-earth targets showed deep cratering from the superradiant pulses indicating significant melting of the material. The copper and tungsten targets showed no significant cratering and some surface degradation from the Q-switched pulses.

It is interesting to note that the tungsten target exhibited a shower of sparks on each Q-switched pulse. This may have been because the tungsten used was sintered during fabrication, and large particles may be dislodged during the rapid heating of the target during the laser pulse.

V. Summary

We have studied the VUV output of laser-produced plasma light sources with high resolution in the 8–40-nm spectral region, using a grazing incidence spectrometer with a channel electron multiplier array detector. Spectra have been obtained for Al, Cu, Fe, Sn, Sm, Hf, Yb, W, and Pb, which can be directly compared for the relative intensity at each wavelength. Several rare earths and tungsten show very clean continuum outputs with little line contamination. The outputs were linear with incident laser energies up to >1 J/pulse, and the dependence of the output on focus conditions was shown to produce maxima away from the apparent point of minimum spot size. There was no significant degradation of the continuum nature of the spectra with decreasing laser energies down to 200 mJ/pulse. The target debris problem was seen to be manageable, but the debris produced by low-energy superradiant pulses was surprisingly high. In general, these sources seem to be fulfilling their promise as VUV laboratory light sources and seem to have all the required properties for secondary intensity transfer sources in the difficult VUV spectral region.

We gratefully acknowledge the assistance of D. Morgan. The assistance of P. Gohil and the guidance and assistance of T. B. Lucatorto were essential in carrying out this work. This experiment was partially supported by Air Force Office of Scientific Research (AFOSR) contracts F49620-83-C-0130 and ISSA85-0033.

Thomas McIlrath is also on the staff of the University of Maryland at College Park.

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APPENDIX E

DEBRIS AND VUV EMISSION FROM A LASER-PRODUCED PLASMA
OPERATED AT 150 Hz USING A KRYPTON FLUORIDE LASER

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The output of a KrF laser (248 nm) operating at 150 Hz was focused onto metal targets to produce plasmas which emitted strongly in the VUV and XUV regions. Quantitative measurements of target debris produced in a laser-plasma light source show (1) that low pressures (~ 100 mtorr) of He buffer gas reduce the debris collected 150 mm from the target by more than an order of magnitude and (2) that the amount of debris collected rises faster than linearly with laser pulse energy in the 100-300 mj range. This observed suppression of debris is explained in terms of a counterstreaming plasma mechanism. High resolution absorption spectra observed in the 50-100 nm range using the laser-plasma light source showed (1) that VUV continua produced using the KrF laser or a Nd:YAG laser (1064 nm) as a driver are equally line free and (2) that exposure times using SWR plates were an order of magnitude shorter for the KrF driver operating at 150 Hz and 300 mj/pulse than for the Nd:YAG driver operating at 10 Hz and 600 mj/pulse.

I. INTRODUCTION

Plasmas using high repetition rate lasers focused onto heavy metal targets have been demonstrated to be high average power sources of VUV ($\lambda < 200 \text{ nm}$) and soft X-ray radiation. The use of laser produced plasmas as sources of line spectra goes back to the 1960's¹ and the use of rare earth targets to obtain nearly line free continua was introduced by Carroll and coworkers in the 1970's.² Recent studies of spectral outputs from 3 nm to 200 nm using a variety of high resolution instruments further demonstrated the source's utility for absorption spectroscopy³ and provided measurements of the relative VUV emission intensities obtained using different metal targets.⁴ The same laser plasma light sources have proved useful for exposing microlithographs.⁵ Hence, it now is firmly established that laser plasma light sources are useful laboratory tools for a variety of applications requiring short wavelength radiation.

Most of the recent experiments³⁻⁵ mentioned above were performed using a Nd:YAG laser operating at 10 Hz and 1064 nm as the source's driver. We were interested in trying to increase the source's VUV output by increasing driver laser repetition rate or peak pulse power or both. In addition, we were interested in remedying or at least reducing a persistent and major problem plaguing the use of laser plasma sources: contamination of windows and other surfaces by debris thrown out from the target during operation. Advances in both of these areas are the subject of this article.

The use of a nearly transparent buffer gas to suppress the debris reaching critical surfaces has been suggested by several authors. Momentum transfer cross-sections indicate that several torr-cm of pressure-path length product (PL) would be necessary for a neutral-neutral collision mechanism to be effective using a heavy buffer gas, with significantly higher pressures

required if a light gas such as He or H₂ were used. On the other hand, a lighter gas is more desirable from the point of view of VUV and soft X-ray transparency. In this paper we present quantitative measurements of the suppression of debris from heavy metal targets using low pressures of He gas in the experimental chamber. The debris was collected on a sample plate in the target chamber and measured after each run using quantitative wet-chemistry and photometric techniques. We demonstrate a major reduction of measured debris with PL < 1.5 torr-cm of He. This result will be discussed in terms of collisionless interactions between the moving debris plasma and a stationary buffer gas plasma. Finally we show that operation at higher repetition rates and lower pulse energies reduces the debris while increasing the average output power of the source in the VUV region.

II. EXPERIMENTAL ARRANGEMENTS

The conditions and apparatus used for the debris and the high resolution spectroscopic experiments are summarized in the block diagrams in Fig. 1. The driving laser was a KrF excimer laser (Lambda Physik Model 202 MSG) operating at 248 nm with an unstable resonator configuration in the laser cavity. The output for this system had a rectangular image ~ 20 mm × 6 mm with 50% of the output energy falling within an 0.5 × 0.5 mrad square solid angle. The 20 ns pulse length output could be varied from 100 mj to 350 mj per pulse with a repetition rate of 150 Hz. The laser output was focused by a 300 mm focal length lens onto the surface of a rotating metal target through a quartz window placed 280 mm from the target. The evacuable target chamber, which has been discussed elsewhere,⁶ used cylindrical targets 16 mm in diameter with 2500 mm² of target surface available to the laser. The target was attached to a 3.2 thread per mm screw driven by a stepping motor which moved the target

under the laser focal spot to provide a fresh surface for each shot. On the first pass over a target the resulting target damage appears as a helical array of about 30,000 spots extending from top to bottom of the cylinder's length. After this first pass the direction of the rotation was reversed automatically and the helix retraced with a slight offset in initial position. This process usually was repeated until the surface of the target showed obvious wear, at which time the target was removed and the surface rejuvenated by removing between 0.08 mm and 0.25 mm from the target's diameter on a lathe, the amount depending on the target's material and length of service.

In the debris experiments a plastic sample plate with an exposed region 41 mm in diameter was placed 150 mm from the plasma source in an evacuable chamber. The line from the source to the collecting plate was at right angles to the incident laser beam, and the normal to the target surface at the plasma production point bisected this right angle. Thus, both the angle of incidence and the angle to the center of observation were 45° with the plane of both angles perpendicular to the target's cylinder axis. For most experiments steel targets were used, with the amount of Fe deposited on the sample plate determined after each run. The target chamber was evacuated to < 5 mtorr with a well trapped mechanical pump before the He buffer gas was admitted into the chamber. A typical run consisted of 90,000 plasma pulses, which took 10 minutes operating at 150 Hz. When chamber pressures were low ($P < \sim 50$ mtorr), the entrance window to the target chamber became visibly clouded from debris deposited during the tests. The window was changed after each 10 minute run to minimize power density reduction on the target caused by window contamination. Window transmissions were measured before and after a run and the results of these measurements are discussed in detail below. The laser power

also was measured at the beginning and end of each run, as was the buffer gas pressure.

The amount of Fe deposited on a sample plate was measured using standard analytical procedures⁷ for spectrophotometric color comparisons. The Fe deposit was dissolved in concentrated HCl and the resulting solution diluted by a factor of ~ 10 with distilled water. Excess potassium ferrocyanide was added in solution to complex all of the Fe ions and this final solution diluted with distilled water to a standard volume selected for use with all samples. The absorbances of the resulting blue solutions were then spectrophotometrically compared with absorbances obtained from a set of similarly prepared standard samples of known Fe concentration to determine quantitatively the amounts of iron deposited during each exposure. When using targets other than iron only relative amounts of debris were determined, and we will make only qualitative statements in such cases. For our samples, the total amounts of debris were small (micrograms) and attempts to determine them by direct weighing were unsuccessful.

The experimental arrangements for the high resolution spectroscopic experiments were similar to those employed previously.^{3a} Briefly, the light source was placed 2800 mm from a concave mirror of 3000 mm radius (75 mm diameter, osmium coated) which imaged the laser produced plasma on the slit of a 6.65-m vacuum spectrograph equipped with a 4800 grove/mm grating (blaze 90 nm, gold coated). The center lines between the entrance slit and the focusing mirror and between the plasma light source and the focusing mirror were 30° apart and in the same plane as the Rowland circle of the spectrograph. Only the heavy metal targets Pb, Yb, and W were used with the 150 Hz driver to check the ability of the excimer laser to produce background continua^{3a,6} suitable for high resolution photoabsorption studies. As in the debris studies, a 90°

angle was maintained between the direction of plasma viewing and laser input and between the target's cylinder axis and the viewing input plane. However, in the spectroscopic experiments two dielectric coated mirrors were used to steer the laser beam into the optical plane of the spectrograph and focusing mirror while in the debris experiments the central laser ray and debris viewing centerlines were aligned without mirrors by directly positioning the laser relative to the light source and collector plate.

III. RESULTS

Typical results, obtained from the debris measurements of Fe from steel targets, have been collected in Figs. 2 and 3. Fig. 2 illustrates the central and most significant portion of our observations on debris production as a function of He buffer gas pressure. In Fig. 2 the open circles and crosses are from measurements made with static He gas and with the He flowing counter to the direction of the debris travel, respectively. As can be seen from Fig. 2 there is no significant difference between the static and flowing gas conditions. Each data point in Fig. 2 represents an Fe debris measurement made after a 90,000 pulse run with the laser energy at 200 mj per pulse and with the laser operating at a 150 Hz repetition rate (30 W average power). The pressures for successive runs were varied randomly to ensure that no systematic errors related to laser or window deterioration were introduced. The laser energy was stable typically to $\pm 10\%$ or better during any given run.

At low gas pressures the quartz window through which the laser beam entered the light source clouded visibly during a run except for what appeared to be a clear rectangular region where the laser beam passed through the window. The clean quartz windows used in our experiments had a measured transmission of $94 \pm 5\%$. The transmission of the laser through the apparently

clear rectangular region of our windows was measured after each run. For conditions with low buffer gas pressures ($P \sim 5$ mtorr) the transmission dropped to $62 \pm 5\%$ after a run of 90,000 shots. At 100 mtorr pressure the window transmission after the run was $90 \pm 5\%$, while at 30 mtorr pressure the transmission dropped to $75 \pm 5\%$. Thus, the fall in measured window transmission with decreasing buffer gas pressures is consistent with the more quantitative measurements shown in Fig. 2. It should be emphasized that the data in Fig. 2 have not been corrected for this gradual decrease in window transmission during each experiment. While such corrections would be negligible for data taken with $P > \sim 80$ mtorr, they could be quite significant at lower pressures. For example, for $P \sim 5$ mtorr a 200 mj laser pulse initially delivers 190 mj through the clean window to the steel target, while 90,000 pulses later the same 200 mj pulse is delivering only about 130 mj through the now dirty window. Since the amount of debris depends strongly on the per pulse energy of the driver (see below and Fig. 3), near the end of this 90,000 pulse example the rate at which Fe would be deposited on our collector plate would be down by at least 50% from the rate of deposition at the start. Thus, the $P < \sim 60$ mtorr debris measurements in Fig. 2 are lower than they would have been if the effects of debris on window transmission could have been removed, and a truly window independent curve would rise more steeply at low pressures than the curve in Fig. 2.

It is important to note that the transmission measurements were made on that portion of the window which appeared to be kept clear by energy absorbed from the incoming laser pulse, the surrounding area being more strongly clouded by debris. In the Fe experiments, windows could be cleaned to their original UV transmission if cleaned before they had accumulated debris from more than about 100,000 shots. Windows used for several runs with $P < \sim 100$ mtorr

before cleaning did not clean to their original transmission in the portions of the window exposed to the laser beam.

The effect of laser pulse energy on Fe debris from steel targets is shown in Fig. 3. The open circles and crosses represent measurements with 60 mtorr and 15 mtorr of He buffer gas, respectively. As with Fig. 2, the data in Fig. 3 have not been corrected for losses in window transmission produced by debris deposition. These window transmission effects of debris deposition discussed in the preceding paragraph lead us to believe (1) that the 15 mtorr data would be shifted to higher debris values and probably would rise more rapidly with increasing pulse energies if there were no progressive window transmission losses and (2) that the 300 mj points would be the only points significantly affected for the 60 mtorr data (possibly displaced upward) by removal of window losses. Using a Nd:YAG laser (1064 nm) we previously have shown that down to at least 100 mj per pulse, the XUV output scales approximately linearly with laser energy. Since we find that the spectral outputs from the plasmas produced by either the KrF or the Nd:YAG drivers are the same (see below), the amount of debris could be reduced and a fixed XUV output maintained by operating at higher repetition rates and lower pulse energies. As the spectral region of interest extends further into the soft X-ray region, it is expected that high laser powers will be necessary, but it is probable that the debris problem can be minimized by producing high powers with shorter pulse durations rather than higher pulse energies.

Having presented quantitative information for iron debris produced by the 150 Hz KrF excimer driver laser from steel targets, we now compare qualitatively these results with observations made using other target materials. We found that Cu and Yb targets produced less debris than Fe while Pb produced more. These statements are based on relative rates of debris accumulation on

the source's laser entry window and on total target damage produced by similar accumulations of laser pulses. The sample of W tested apparently was originally sintered but not fully remelted or annealed, because its debris was mainly small pieces of sputtered metal rather than the thin film coatings produced by the other target materials.

It should be noted that at a 150 Hz repetition rate and with 15-50 W average power, there was significant heating of the light source's targets. For example, this heating was sufficient during a 10 minute run to discolor (strongly blue) steel targets. Accumulating debris for 90,000 pulses broken into 30,000 pulse sets separated by intervals of 15 minutes produced 10-20% less debris than was obtained from nonstop accumulation. Thus, target heating also can be a factor and we provided a fixed 15 minute interval between each 10 minute (90,000 pulse) run to allow for target cooling and to reduce the risk of a systematic bias as a series of runs progressed. Since target heating appears to be a significant problem when high repetition rate and average power drivers are employed, we have incorporated internal cooling in new targets now under construction.

We performed high resolution studies of plasma emissions produced from Pb, Yb, or W targets by the KrF excimer driver operating at 150 Hz in the 250-300 mJ per pulse range. Emission from these plasmas were photographed in the ~ 50-100 nm region using Kodak SWR plates and the first order of the 6.65m spectrograph (plate factor 0.03 nm/mm). These emissions were found to be continuous, line free, and in every way identical to comparable spectra^{3a} obtained from similar targets using a Nd:YAG driver operating at 10 Hz with per pulse energies in the 500-600 mJ range. Under the conditions stated above, we found that exposure times for a given plate blackening were 10 times shorter when using the excimer laser as a driver than they were when using the

Nd:YAG laser.

IV. DISCUSSION

The effectiveness of low pressure He gas in reducing debris effusion from the target region is clear from Fig. 2. Using a geometric collision cross-section of 10^{-16} cm⁻² and a pressure of 50 mtorr, one obtains a mean free path of over 5 cm. Combining this result with the Fe to He mass ratio of 56:4, it is surprising that He could buffer Fe vapors anywhere close to as effectively as shown. The explanation for this unexpected effectiveness probably lies in the fact that the intense VUV and XUV emission from the laser plasma completely photoionizes the He buffer gas. Thus, the ionized debris plasma from the target must flow through a stationary plasma of He⁺ and He⁺⁺ ions rather than a low pressure gas of neutral atoms. The problem of counterstreaming laser-produced plasmas has been studied earlier by Koopman and coworkers under conditions very similar to those present in our experiment.⁹ In Koopman's experiment a Cu (A=29) target was used to produce a plasma in a H₂ (A=2) or He buffer gas. Koopman et al.⁹ found that although Cu ions have a large mean free path in a neutral He or H₂ background gas, background gas exposed to the laser plasma is ionized and swept along by the expanding Cu⁺ plasma. The coupling of the Cu⁺ plasma to the ionized background gas results in the background gas piling up in front of the expanding Cu⁺ plasma to produce what is termed a "snow plow" effect. The result of this snow plowing is a collisionless transfer of momentum out of the target atoms into the buffer gas atoms. The use of a light buffer gas enhances the snow plow effect since the light nuclei are easily swept up in the Coulomb field of the expanding debris plasma, with the result that there is little penetration of the debris by the ionized background gas. At large distances from the plasma source the

background gas ion density falls off as r^{-2} due to the decreased ionization efficiency, but the debris density falls off as r^{-3} due to expansion. Thus, debris continues to snow plow the ionic background and lose momentum to the buffer gas. At lower relative velocities the Coulomb coupling increases, enhancing the effect, so that once the buffer gas atoms are swept up they remain strongly coupled to the debris plasma. At large distances the coupling is increasingly between the near stationary background gas plasma and background gas ions which have been accelerated in the "snow plow" front. Measurements⁹ of the target ions with a spectrometer and a time of flight mass spectrometer 15 cm from the target showed a strong reduction of the target ion velocity and a disappearance of highly charged ions as a result of charge exchange with the remaining neutral background gas. Conservation of mass coupled with the isotropy of the experiment would indicate that the same number of target nuclei ultimately would reach the sample plate, but as neutral atoms or clusters with a greatly reduced translational velocity which could significantly change the sticking coefficient of the target material.

Further experiments by Koopman and other investigators focused on the interaction of counterstreaming plasmas in the presence of a strong (kilogauss) magnetic field.¹⁰ In the presence of the magnetic field the electron motion is strongly inhibited across the field lines. The resultant restriction on the electron motion, combined with the strong coupling of the ions with the electrons, leads to a strong collisionless coupling of the two plasmas. This mechanism of coupling to ion motion in plasmas is much more effective than would be expected from the cyclotron motion of a single free ion in a magnetic field, and it is possible that the addition of magnets in appropriate configurations would further inhibit effusion of target atoms from the immediate vicinity of the target along directions critical to particular experiments

(i.e., toward windows, masked photoresists, etc.).

V. SUMMARY

We have made quantitative measurements of the debris from a laser-plasma light source and have shown that this debris can be strongly quenched by the addition of He at sufficiently low pressures and short pathlengths to be transparent throughout most of the VUV and XUV. The buffer gas pressure necessary to reduce the amount of debris by over an order of magnitude is less than 100 mtorr for a distance of 15 cm. The amount of debris produced for a fixed average XUV output also can be reduced by operating at a higher repetition rate and a lower pulse energy.

The cause of this effective quenching is most likely the coupling of the expanding debris with a stationary plasma of gas atoms photoionized by the VUV and XUV radiation from the laser plasma. The primary coupling is Coulombic and the light background atoms are readily swept up and absorb momentum from the expanding debris. We expect that application of a strong magnetic field will significantly enhance the interaction of the debris with the ionized background. Thus, a significantly cleaner source of XUV and soft X-ray radiation can be made if the laser plasma light source is operated at low pressures of He buffer gas.

High resolution absorption spectra observed in the 50-100 nm range showed that the VUV continua from plasmas produced by the KrF laser operating at 300 mj/pulse and 150 Hz were the same as those produced by a Nd:YAG laser operating at 600 mj/pulse and 10 Hz. The only differences were that the SWR plate exposures were an order of magnitude faster with the excimer driver system than with the Nd:YAG system.

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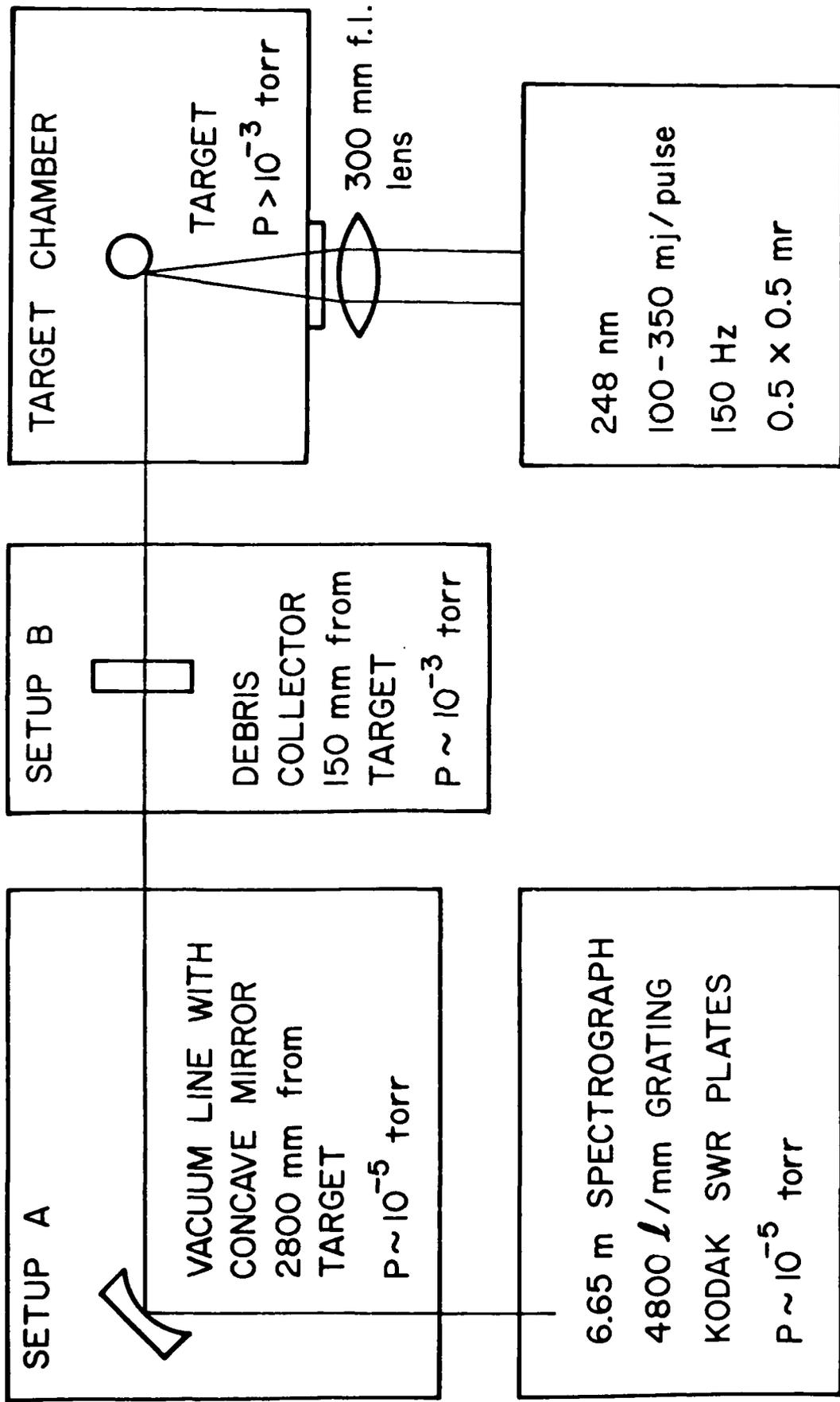
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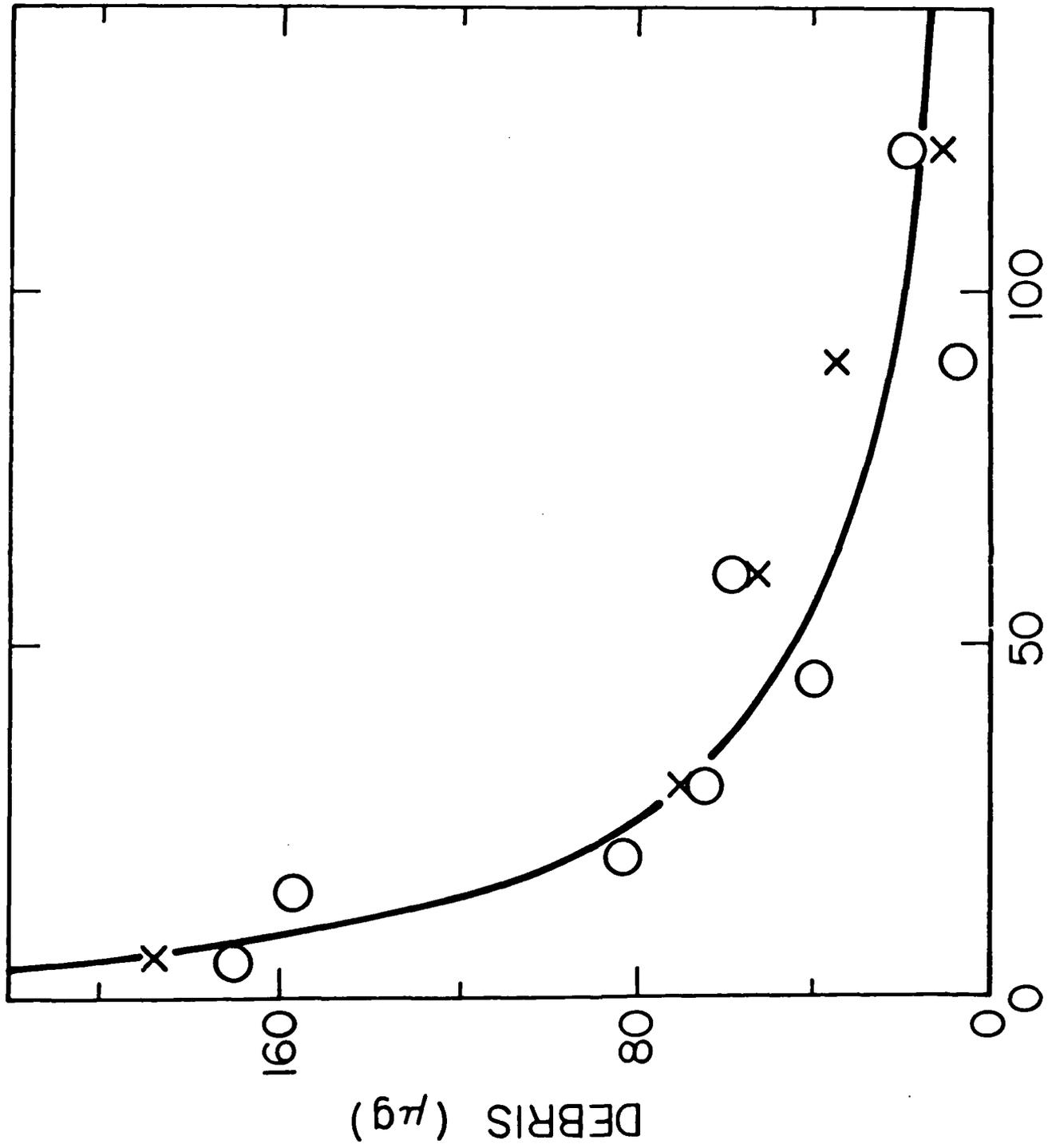
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FIGURE CAPTIONS

- Fig. 1: Schematic diagram of experimental apparatus used in both the debris (setup B) and spectroscopic (setup A) experiments.
- Fig. 2: Debris (micrograms) collected on sample plates with 1340 mm² of exposed area as a function of He pressure. Open circles represent static gas, crosses are data with gas flow counter to the direction of debris travel. Each data point required 90,000 pulses of the KrF laser operating at 150 Hz and 200 mj/pulse.
- Fig. 3: Debris (micrograms) collected on sample plates with 1340 mm² exposed area as a function of laser pulse energy. Open circles were taken with 60 mtorr of He, crosses with 15 mtorr of He. Each data point required 90,000 pulses of the KrF laser operating at 150 Hz.

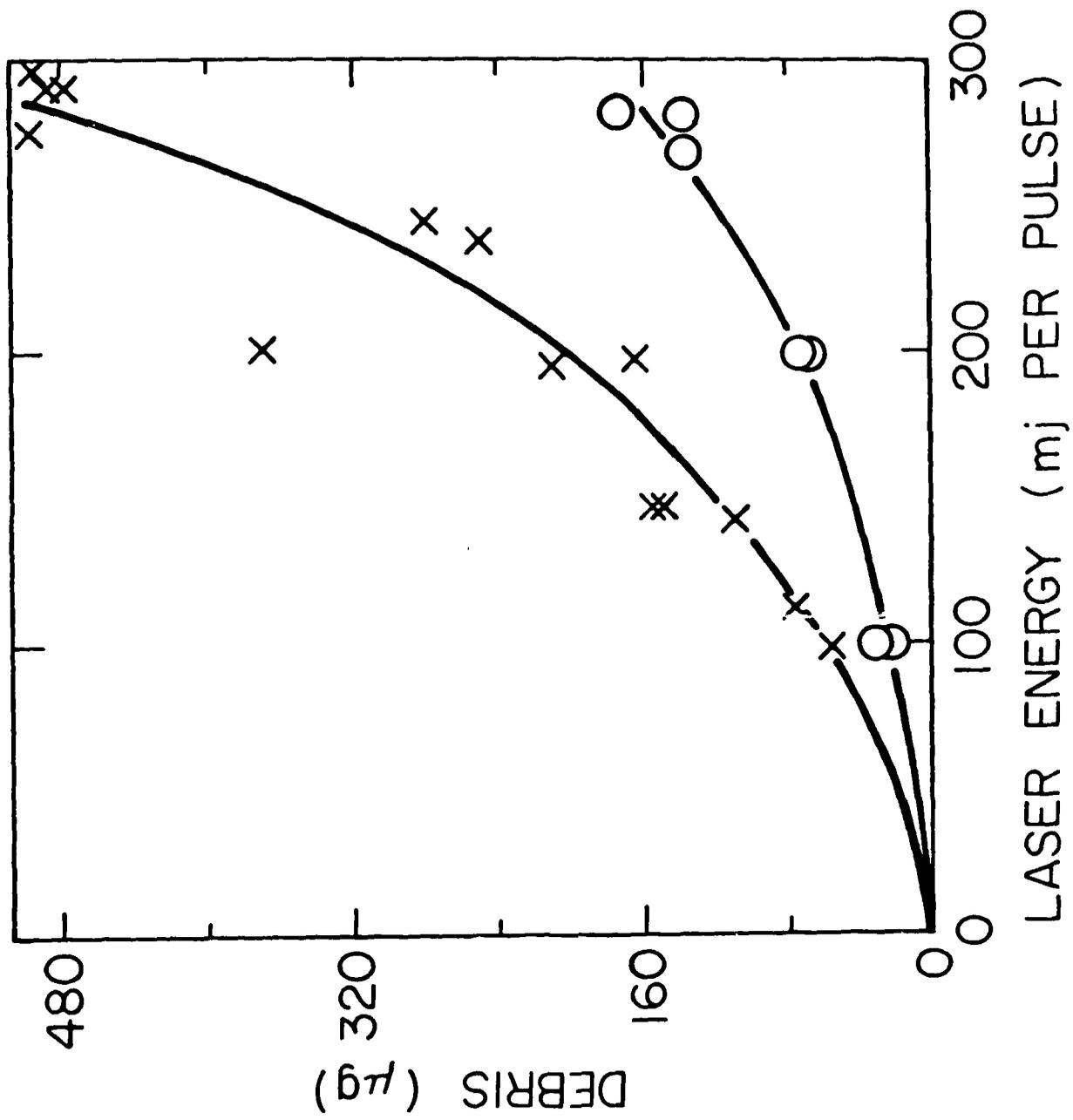


- McIlrath & Ginter, Fig. 1



- - - GAS PRESSURE (mtorr)

- McIlrath & Ginter, Fig. 2



- McIlrath & Ginter, Fig. 3

APPENDIX F

Abstract of invited paper presented to the Annual Meeting of the Optical Society of America, October 14-18, 1985, Washington, D.C.

EUV SPECTROSCOPY USING LASER-GENERATED PLASMA LIGHT SOURCES

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Laser-produced plasmas have been shown to be excellent laboratory sources of continuum radiation in the XUV and soft x-ray spectral region (10-1000 Å). We present high-resolution photographic and photoelectric spectra of the continua between 40 and 500 Å, comparing different target materials for cleanliness and relative intensity of the continua. Photographic spectra have been obtained on a 10.7-m grazing incidence spectrograph and a 6.67-m normal incidence spectrograph and a photoelectric spectra obtained with a 1.5-m grazing incidence spectrograph using a multichannel detector array. Spectra have been obtained using both a ND:YAG (10-Hz) laser and a ruby (0.1-Hz) laser. The XUV output is characterized by excellent intensity stability, high intensity, and an almost line-free continuum. A comparison with other short wavelength light sources is given. The source has been used for atomic absorption spectroscopy of ground state and excited state neutral atoms and for ground state atomic ions by placing samples in a metal vapor heat pipe and using a resonant dye laser for excitation and ionization of the atomic vapor. Inner shell absorption spectra of Ca, Ca⁺, and other atomic systems are presented.

APPENDIX G

Abstract of paper presented to the Eighth International Conference on Vacuum Ultraviolet Radiation Physics, August 4-8, 1986, Lund, Sweden.

A LASER PRODUCED PLASMA LIGHT SOURCE FOR HIGH RESOLUTION SPECTROSCOPY AND SOFT X-RAY LITHOGRAPHY

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Laser produced plasmas have been demonstrated /1/ to be convenient laboratory light sources for absorption spectroscopy below 500 Å. Recently, we have shown /2/ that under proper conditions no line structure appears in the intense continua from our laser plasma sources when observed at the highest possible spectral resolutions and have used this source for high resolution spectroscopy. The source also is used with modified conditions /3/ for soft x-ray lithography.

Two laser systems were used in this work: a Nd:YAG laser (1064 nm) with ~600 mj, 25 ns pulses and a 10 Hz repetition rate and a Krypton fluoride laser (249 nm) with ~300 mj, 20 ns pulses and a 150 Hz repetition rate. Power densities in the incident pulses at the focal spot on the plasma source targets generally were around 10^{11} W cm⁻². The source's cylindrical metal targets can be rotated to provide fresh material for every shot /3/ and the plasma is viewed at right angles to the incident laser beam with the normal to the surface at the focal spot being ~45° to both the laser beam and the viewing direction. The source provides a simple, inexpensive laboratory competitor to synchrotron radiation for both spectroscopy and lithography, as demonstrated by the examples presented.

The work was supported by Air Force Grant AFOSR-85-0174.

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APPENDIX H

Abstract of paper presented at the Second International Conference on Laser Science, October, 1986, Seattle, Washington

A LASER PRODUCED PLASMA LIGHT SOURCE FOR HIGH RESOLUTION SPECTROSCOPY AND SOFT X-RAY LITHOGRAPHY

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ABSTRACT

Laser produced plasmas have been demonstrated¹ to be convenient laboratory light sources for absorption spectroscopy below 50nm. Under proper conditions no line structure appears in the intense continua from laser sources when observed at the highest possible spectral resolutions² and we have used this source for high spectroscopy. The source also was used with modified conditions for soft x-ray lithography³.

INTRODUCTION

Laser produced plasmas using heavy metal targets and high repetition rate lasers are high average power source of XUV (<100nm) and soft x-ray radiation. We have made intensity measurements and studied outputs under very high resolution in order to characterize these sources for several applications. The plasmas were produced by focusing the output of a Nd:YAG laser (1064nm, 600mj/pulse, 10Hz) or an excimer laser (248nm, 300mj/pulse, 150Hz) onto a metal target using an f=30cm or an f=10cm lens. The maximum power density on the target with both sources was 10^{11} - 10^{12} W cm⁻². The cylindrical metal targets⁴ were rotated to produce fresh target material for every pulse. The plasmas were viewed at right angle to the incident laser beam with the normal to the target surface at the focal spot being approximately 45° to both the incident laser beam and the viewing direction.

SOURCES FOR HIGH RESOLUTION XUV SPECTROSCOPY

The outputs of the laser plasmas sources have been studied in the normal incidence spectral region (~30-100nm) using a 6.65m spectrograph equipped with a 4800g/mm grating⁵. Targets included Cu(Z=29), Yb(Z=70), Hf(Z=72), W(Z=74) and Pb(Z=82). The continuum intensities from Yb, Hf, W and Pb were similar yet of somewhat different spectral distributions. The four higher Z element plasmas can be made to produce clean continua with very few emission lines. However, in order to obtain intense line-free continua it is necessary to carefully image the hottest portion of the plasma onto the entrance slit of the spectrograph. Optimization of the continuum output and emission line suppression required careful placement (~ 100 microns for the source described above) onto the 50 micron entrance slit of the spectrograph. The

continuum intensity from the Cu target was found to be weaker than those obtained from the high Z element targets by approximately an order of magnitude with the Cu plasma emission dominated by strong line spectra.

The emission spectra from W, Yb and Cu targets were also observed on a 10m grazing incidence spectrograph⁶ equipped with a 1200g/mm grating. The spectral region from 4.5nm to 60nm was studied. Both the W and Yb sources produced almost line free continua with comparable intensities. The intensity appeared to be strongest in the 10-20nm region. The falloff of intensity at longer wavelengths was such that absorption spectra of He in the 17-21nm region could be recorded in third order with no order separator and no significant contamination by first or second order light.

Intensity studies have been made in the 8 to 40nm region with a 1.5m grazing incidence spectrometer and a channel electron multiplier detector⁷. Nine different elements were compared in intensity and spectral purity. These studies confirmed the desirability of using high Z targets and provide a basis for choosing the target most suited to the particular spectral region being studied.

SOFT X-RAY LITHOGRAPHY

In soft X-ray lithography the continuum purity of the source is no longer of concern. We have studied the exposure of photoresists using Cu, W or steel targets driven by the Nd:YAG laser⁸. The photoresist used was a copolymer of polyglycidyl methacrylate and ethyl acrylate (COP), which was spun to a thickness of 4700Å on the surface of 75-mm diameter silicon wafers. COP is a negative photoresist with a sensitivity of 15mj/cm². Masks consisted of polyimide membranes overlaid with circuit patterns in gold. The shortest exposure times were obtained with steel (Fe) with Cu targets being only slightly less effective. Exposure times with W targets were approximately twice as long. The lithographs showed greater uniformity of exposure than that obtained using synchrotron radiation exposure. Exposure times were ~10 times longer with the laser plasmas than with the synchrotron. Preliminary studies of the output using the 150Hz excimer laser suggest that exposure times would be reduced by a factor of ~10 over the time using the 10Hz Nd:YAG laser. There were no obvious differences in the quality of the lithographs made using either light source.

SOURCE DEBRIS

A major problem in using laser plasma sources is the production of large amounts of debris. We have begun studies of the effect of buffer gases in the source chamber on the resultant debris. A glass sample plate was placed 10cm from the source

which was irradiated by the 150Hz excimer laser driver. A steel target was used and the Fe deposited on the sample plate was quantitatively analyzed by chemical means. It was found that the introduction of 20 microns of He gas reduced debris by a factor of 2 and above 100 microns pressure the debris was reduced by more than a factor of 10 to near our detection limits. It was also found that raising the laser pulse energy from 100mj to 300mj increased the debris level by more than a factor of 6. Since the XUV output is nearly linear with the driving energy, this indicates that the debris problem can be reduced by using a higher repetition rate with a lower pulse energy. Work is continuing on optimizing the output with minimum debris.

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