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LASER-GENERATED PLASMA AS A SPECTROSCOPIC LIGHT SOURCE

by Francisco P. J. Valero, David Goorvitch, Boris Ragent, and Benjamin S. Fraenkel

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16. Abstract

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LASER-GENERATED PLASMA AS A SPECTROSCOPIC LIGHT SOURCE

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SUMMARY

The spectra emitted by laser-generated plasmas have been studied in the spectral region from about 40 to 4000 Å. The radiating regions of the plasma have been spatially resolved using either a stigmatic normal incidence spectrograph, or an astigmatic grazing incidence spectrograph and crossed slit apparatus, depending upon the wavelength region. Both emission and absorption spectra were observed for highly ionized atoms. Both line shifts and strong asymmetries were observed. The continuum spectra radiated by the core of the plasma have been used to perform absorption experiments in the soft X-ray region. Features of the laser generated plasma light source that aid in the interpretation of spectra are discussed. The laser produced plasmas have several unique characteristics that prove to be desirable and convenient for spectroscopic work.

INTRODUCTION

This paper describes an investigation into the possibilities of using a laser-generated plasma as a spectroscopic light source.

Several investigators have proposed the use of the focused radiation from a giant pulse Q-switched laser to produce very hot plasmas. Various laboratories have instituted programs to investigate and to exploit the unique characteristics of this source of high temperature gases and its possible applications to plasma physics, thermonuclear reaction studies, etc. (refs. 1-8). In the present work laser-generated plasmas were studied spectroscopically in order to help in understanding the various processes taking place within the plasma. Also the characteristics of the radiation emitted by the plasma were used in studying the spectra of ionized species in the vacuum ultraviolet and soft X-ray regions.

OPTICAL ARRANGEMENT

The optical arrangement used for the experiments in the grazing incidence region (40 to 500 Å) is shown schematically in figure 1. A 3-m grazing incidence spectrograph with a platinum coated concave grating possessing

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1200 lines per mm and blazed at 5°10' was used at an angle of incidence of 82°. The laser beam was focused by a lens onto the target made of the element of interest. The distance between the target and the entrance slit in the direction of the optical axis was about 1 mm. The distance of the target from the optical axis in the direction normal to both the optical axis and to the slit was varied from 0 to 5 mm, depending, as discussed in the section on broadening and shifts of spectral lines, on the region of the plasma to be observed in each particular case.

For these experiments, the width of the entrance slit was varied from 0.5 μ at 40 Å to 10 μ at 500 Å. The crossed slit was used to make the instrument less astigmatic, as described in reference 9. The width of the crossed slit was varied from 0.1 to 1 mm, depending on the spectral region being observed.

For the spectral range from 300 Å to the visible, a normal incidence spectrograph with a 3-m focal length was employed. This instrument provided nearly stigmatic spectrograms so there was no need for any additional slits. The optical arrangement and physical orientation of the grating are presented schematically in figure 2.

In all the experiments a TRG-104A, 100 MW ruby laser was used in both the normal and Q-switched modes. Energy of the order of 1 J was delivered to the target in times of the order of 10 to 20 ns for the Q-switched mode, and about 2.5 J in times from 400 to 500 μs for the normal mode. Figures 3 and 4 show typical pulse shapes for the Q-switched and normal modes. Typical photographic plate exposures required 1 to 20 laser pulses.

EXPERIMENTS

The spectra radiated by laser-generated plasmas of Li, Be, Al, Fe, W, Th and U have been observed in the grazing incidence region. The spectra of Fe and Th were also photographed in the normal incidence range (300 to 4000 Å). The experimental parameters that were varied included the laser power, the distance between the target and the slit in the direction normal to both the slit and the optical axis, the density of the test element appearing as an impurity in a light element alloy target, and the thickness of the target deposited on glass as a thin film.

VARIATION OF LASER POWER

The power of the laser pulse was varied from about 10 to 100 MW in the Q-switched mode. The most important difference observed at various power levels was a general decrease in intensity of the line emission and a proportionately greater decrease of the continuum emission as the power was decreased. Figure 5 shows the principal series of Be III in the 100 Å region. The lines of this series show strong self-reversal when the laser is used in the Q-switched mode and the target is located so that it constitutes one of the jaws of the spectrograph slit. The laser beam was focused onto the target at about 0.5 mm from the slit opening in the direction of the optical axis. In this way it was possible to observe only the plasma on the target surface. For Be, a light element, relatively weak continuum radiation was observed in this region.

The power of the laser in the normal mode was also varied. In this mode the total energy delivered to the target is increased but the power is greatly reduced because of the lengthening of the pulse. Figure 6 shows the radiation from a Th plasma in the normal incidence region as obtained with the laser operating in the Q-switched mode and in the normal mode. In the Q-switched mode (fig. 6(a)) a strong continuum is radiated by the plasma with resonance lines appearing in absorption. The absorption is stronger for lines belonging to the lower stages of ionization. In the normal mode (fig. 6(b)), the emission spectrum appears and the continuum is weak.

DISTRIBUTION OF IONIC SPECIES IN THE PLASMA BALL

To study the distribution of ionic species in the plasma ball in the region below 400 Å a crossed slit (S_2 in fig. 1) was introduced between the principal slit S_1 and the grating. In this way the astigmatism of the grazing incidence spectrograph was reduced and partial spatial resolution of the plasma ball was achieved (ref. 2). Figure 7 is an Al spectrogram taken at about 240 Å with the crossed slit. The spectral lines belonging to the lower stages of ionization appear longer than those radiated by the higher stages of ionization. Also the width of the spectral lines tapers toward the ends. From the relative length of the lines it is apparent that, as expected, the highest stages of ionization in the plasma ball are concentrated in the hottest central core of the plasma.

CHARACTERISTICS OF SPECTRA

Substantial continuum radiation is characteristic of spectra emitted by laser-generated plasmas. The intensity of this continuum radiation increases as the atomic weight of the element of interest increases. Absorption of this continuum causes the intensity of the absorption lines to vary, depending on the stage of ionization and on the probability of the particular atomic transition involved. Figure 8 shows the spectra of Fe near 171 Å, as obtained with the laser operating in the Q-switched mode. Lines belonging to Fe VI, VII, VIII, and IX appear as absorption lines in a strong continuous background. The continuum radiated by a Th target at about 2500 Å is shown in figure 6(a). Again many absorption lines appear.

In many cases, however, emission spectra are of interest. One way to obtain the emission spectrum is to use the laser in the normal mode, as mentioned above. This method is limited, however, in that only the lower stages of ionization are produced, presumably because of the lack of flexibility in adjusting the rate of rise of the laser pulse. A laser capable of producing pulses of variable rise time would make it possible to heat the plasma just enough to produce the stages of ionization of interest without "overheating" and consequently increasing the continuous background and causing other undesirable effects.

In a different approach to this problem of creating emission spectra, the target had a low atomic number and contained the element of interest as a small impurity. The laser was operated in the Q-switched mode. Spectra of Fe at about 170 Å obtained from Al alloy containing 1 percent Fe are shown in figure 9. The Fe lines are in emission and are very sharp. The broad self-reversed lines are Al. While this technique produced the best spectra, insofar as sharpness and lack of self-reversal are concerned, it is limited by the difficulty of introducing impurities of certain test elements into a light element, such as Al, for example. In many cases commercially available alloys may be used.

Tests were also made of the laser pulse (Q-switched) irradiation of targets formed from a thin film of the element of interest on a glass substrate. This technique, originally proposed and demonstrated in references 1 to 5, reduced somewhat the intensity of the continuous background, increasing the contrast between the emission lines and the continuum. It was thus possible to observe emission lines of higher stages of ionization that previously had been masked by the continuum, but the quality of the emission lines obtained in this way is still poor compared with the quality of those obtained by the "impurity technique." In many cases, the lines from thin film targets show strong broadening, self-reversal, and asymmetries (see fig. 10). Also some lines of the lower stages of ionization that appear in absorption when observed in the solid targets and in the thicker films become emission lines for the thinner films. Some lines of the higher stages of ionization that are not present in the thicker films appear in the thinner films.

CONTINUOUS RADIATION FROM HEAVY ELEMENTS

As mentioned above, Q-switched laser-produced plasmas of the heavy elements emit strong continuum radiation. Figure 11 shows typical continua emitted by U^{238} and W plasmas in the grazing incidence region, and figure 12 shows the continuum radiated by a Th target in the region from 300 to 4000 Å. The origin of these continua has been discussed in reference 1. Note that the height of the continua increases with wavelength, which is in accord with the empirical model presented later. These continua have been used in absorption experiments to demonstrate the utility of the source for such applications. Filling the grazing incidence spectrograph with He and Ar at pressures from 0.25 to 0.50 torr and using W as a target to provide a continuum in the soft X-ray region, made it possible to obtain the autoionization series of such elements. The autoionizing series of He at about 200 Å, originally studied by Madden and Codling (ref. 6) using a synchrotron as a source of continuum radiation, is shown in figure 13.

An inconvenience of the laser plasma continuum source is the presence of the absorption lines characteristic of the heavy element used as the target material. These lines may be taken into account by comparison with the spectrogram of the heavy element taken with no absorber present. In addition, because of the pulsed nature of the source, detailed quantitative measurements require either very careful sensitometric methods with photographic recording, or repetitive pulses with point by point scanning techniques using high frequency detector and electronic recording apparatus.

The principal advantages of this continuum source are its relative ease of production, moderate cost, and simplicity of application. Spectral outputs of about 10^8 photons per angstrom at 700 Å have been reported.[2] The continuum source has a useful spectral output extending from the visible to at least as low as 40 Å.

BROADENING AND SHIFTS OF SPECTRAL LINES

Strong broadening and asymmetries in line shapes in laser-generated plasmas were reported in references 1 and 7. We have observed the same phenomena and also noticeable relative shifts between the emission and the corresponding absorption lines both in the grazing incidence and in the normal incidence region for several different elements and stages of ionization. Our observations of these shifts amounting to approximately 500 cm^{-1} for Fe, Al, and Be plasmas in the soft X-ray region were reported in reference 8. The same general behavior has been observed in the 500-4000 Å region for Fe and Th. The magnitude of the shifts suggests the need for caution when the laser-generated plasma light source is used for determining wavelength, particularly from isoelectronic sequences.

In such dense rapidly expanding plasmas, these shifts and broadenings undoubtedly have several causes, including Stark, pressure, and Doppler phenomena, as well as self-absorption. However, in general, occulting techniques will give sharper lines for most stages of ionization. For example, the entrance slit of the spectrograph may be illuminated solely by the light radiated from specific regions of the plasma drop. The viewed region may be varied, depending on the stages of ionization to be investigated, from the outermost portions of the plasma for lower stages of ionization to the central, hotter portions of the plasma core for higher stages. The lines from the highest degrees of ionization are obtained with minimum broadening. A serious disadvantage of this technique is that since very small portions of the plasma ball are observed, localized conditions in the plasma will seriously affect the spectral lines. For example, spectral lines may not be broadened but they may be seriously shifted, depending on the local conditions in the particular portion of the plasma being observed.

[2]Private communication from G. L. Weissler

The use of the small-percentage-impurity low-atomic-number alloy target reduces the line widths and self-absorption effects and apparently also reduces the line shifts as well as introducing little continuum radiation.

EMPIRICAL MODEL

A number of detailed models have been proposed for calculating laser-produced plasmas (refs. 10 and 11). However, on the basis of Dawson's model, and the above observations, the following simple phenomenological model of the plasma ball can be constructed. This model is in accord with most of the models discussed and is useful for planning experiments. The initial portions of the laser pulse cause the surface of the target to emit atoms and electrons in the high energy density region of the focal spot. This cloud of partially ionized atoms and electrons is further heated by the subsequent portions of the laser pulse for as long as the light can penetrate the plasma. The laser light will penetrate the plasma ball as long as the plasma frequency is less than the light frequency (ref. 10); for ruby laser light ($\nu = 4.35\times10^{14}$ sec^{-1}) penetration will continue until the electrons reach a density of 2.5×10^{21} cm^{-3} ($\nu_n = 8.9\times10^3\ n_e{}^{1/2}$) (ref. 12). At this electron density, with which a certain degree of ionization will be associated, the plasma ball will start reflecting the radiation. Simultaneously, the plasma drop will expand, increasing its volume and consequently reducing the electron density and the plasma frequency. Once the plasma frequency drops below the light frequency, the light penetrates the plasma again and is absorbed. The heating mechanism, once the plasma drop is formed, is presumably primarily inverse bremsstrahlung followed by collisions between electrons and heavy particles.

The energy-density changes in the plasma are due to expansion, thermal conduction and radiation (free-free, free-bound, and bound-bound). Thus the plasma is spatially inhomogeneous, containing in its initial phases a very dense, small, hot core that produces intense continuum radiation, from bremsstrahlung and recombination processes, and line radiation from transitions by bound electrons in ionized species. The line radiation from the stripped ions in the hot core of the plasma may show strong broadening effects and shifts caused by the high particle density and mass motion. As the plasma expands, it cools, and the highly ionized species recombine in the outer regions of the plasma drop. Simultaneously, matter may be added to the expanding plasma from its interaction with the adjacent target surface. The interaction of the cooler matter outside the plasma drop with the continuum and line radiation from the inner regions of the plasma results in absorption lines of the lower stages of ionization and strong self-reversal effects for intermediate stages of ionization. This observation refers to viewing the plasma in a radial direction. If the plasma drop is seen in the direction of a chord, the effects of absorption and self-reversal tend to disappear and sharp emission lines for the lower stages of ionization appear in the outermost regions of the plasma ball. The characteristics of the shifts observed between emission and absorption lines (ref. 8) implies the presence, in the laser-generated plasma, of at least two very different regions. A shock wave developing inside the plasma is consistent with the observed phenomena, which is characterized by spectral line shifts greater than line broadenings.

APPLICATIONS

One of the major problems confronting the experimentalist in investigating spectra in the vacuum ultraviolet is the separation of lines according to the stage of ionization from which they were radiated. The characteristics of laser-generated plasmas may be exploited to help in solving this problem. As mentioned above, when the plasma drop is viewed radially, the lower stages of ionization appear in absorption while the intermediate stages show self-reversed lines and the higher stages of ionization show emission lines. The amount of absorption and the degree of self-reversal in lines belonging to the lower stages of ionization are also associated with the particular transitions involved. This fact not only makes it possible to separate the lines according to the stage of ionization by inspection of the characteristics of the lines but also helps in classifying the particular transitions involved simply by extending the techniques commonly used in the visible region of the spectra (i.e., using self-reversed lines to help in locating the lower energy levels). This feature of the laser-generated plasma is of great help in spectroscopic work on highly ionized species. As figure 14 shows, it is relatively simple to separate lines according to the state of ionization by directly relating the magnitude of the self-reversal to the degree of ionization. In conjunction with this technique the crossed slit can be used in the grazing incidence region. Figure 7 shows the spectra of Al in the region of 240 Å obtained using a crossed slit which produces partial spatial resolution of the radiating regions of the plasma drop. As mentioned earlier, lines belonging to the lower stages of ionization are longer than those radiated by higher stages of ionization, consistent with the phenomenological model given above.

Another interesting spectroscopic application of laser-generated plasmas involves the generation of highly ionized species that are not easily excited in spark discharges or magnetic compression devices. For example, since this source requires only that the species be available in a solid, powder, or even liquid state without reference to chemical state or physical shape, it is possible to use compounds, irregularly shaped pieces, microstructures or even materials embedded in some type of holding matrix.

Another application involves using the strong continuum emitted by heavy elements irradiated by the beam from a focused Q-switched laser to perform absorption experiments in the vacuum ultraviolet down to at least 40 Å.

CONCLUSION

The laser-produced plasma is a valuable light source for work in the vacuum ultraviolet, especially as a source of radiation from highly stripped ionic species and for the production of continuum radiation. Even the shifts, reversals, asymmetries, broadenings, etc., of the spectral lines have proved useful in solving the classic problem in spectroscopy; namely, that of identifying the states involved in the transitions that cause the observed spectral lines.

Ames Research Center
National Aeronautics and Space Administration
Moffett Field, Calif., 94035, June 25, 1970

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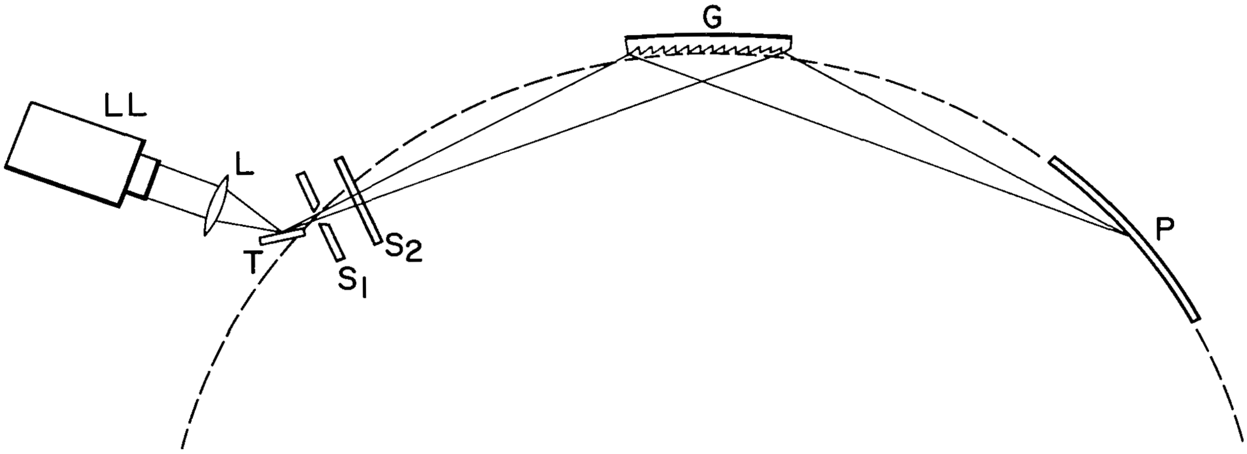

Figure 1.– Sketch of optical arrangement, grazing incidence spectrograph. LL, laser; L, focusing lens; T, target; S_1, entrance slit; S_2, crossed slit; G, concave grating; P, photographic plate.

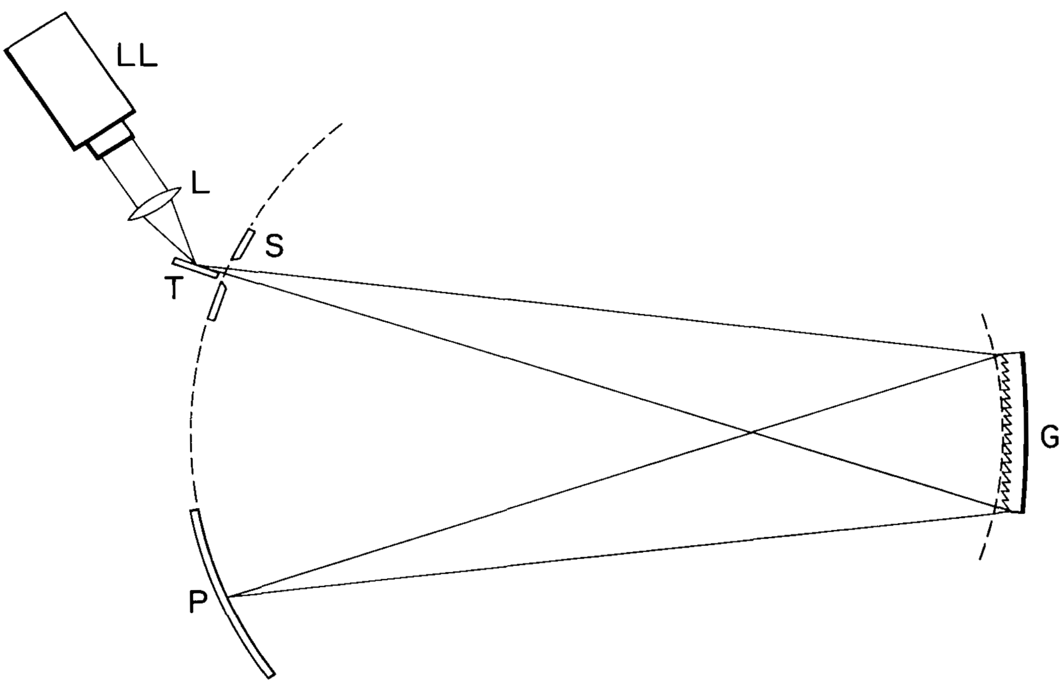

Figure 2.– Sketch of optical arrangement, normal incidence spectrograph. LL, laser; L, focusing lens; S, entrance slit; G, grating; P, plate; T, target.

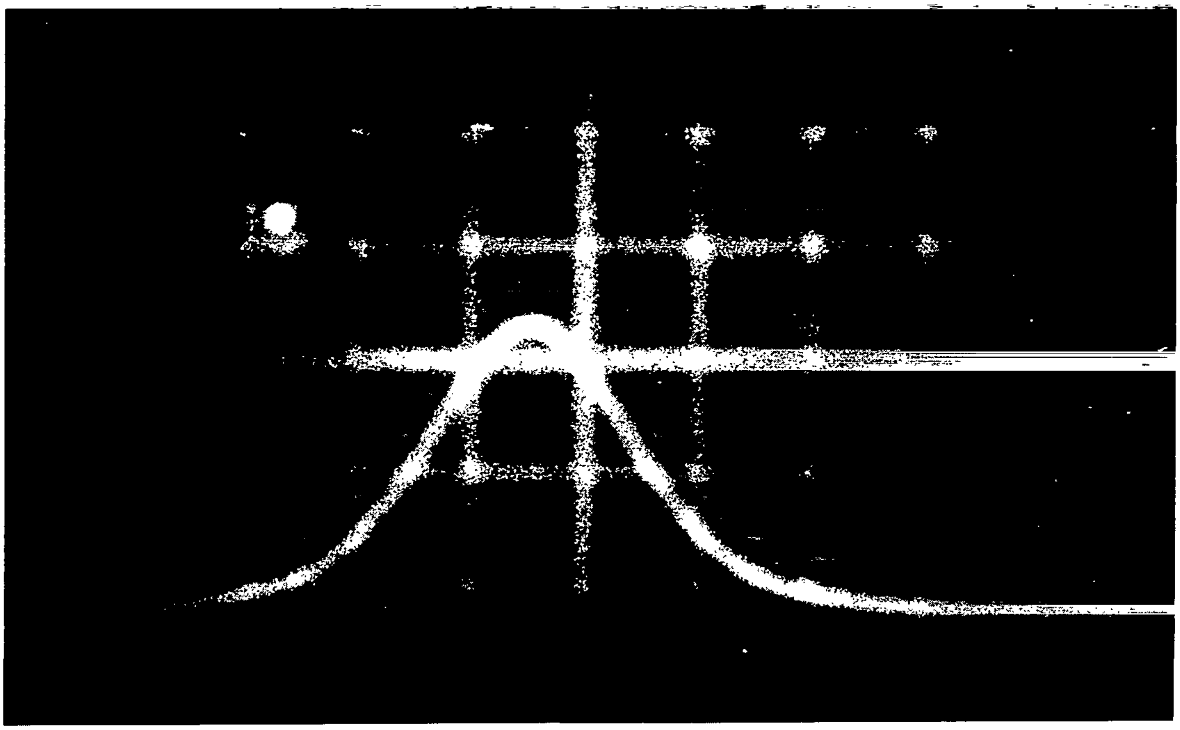

Figure 3.— Typical laser pulse shape, Q-switched mode, 10 ns/div.

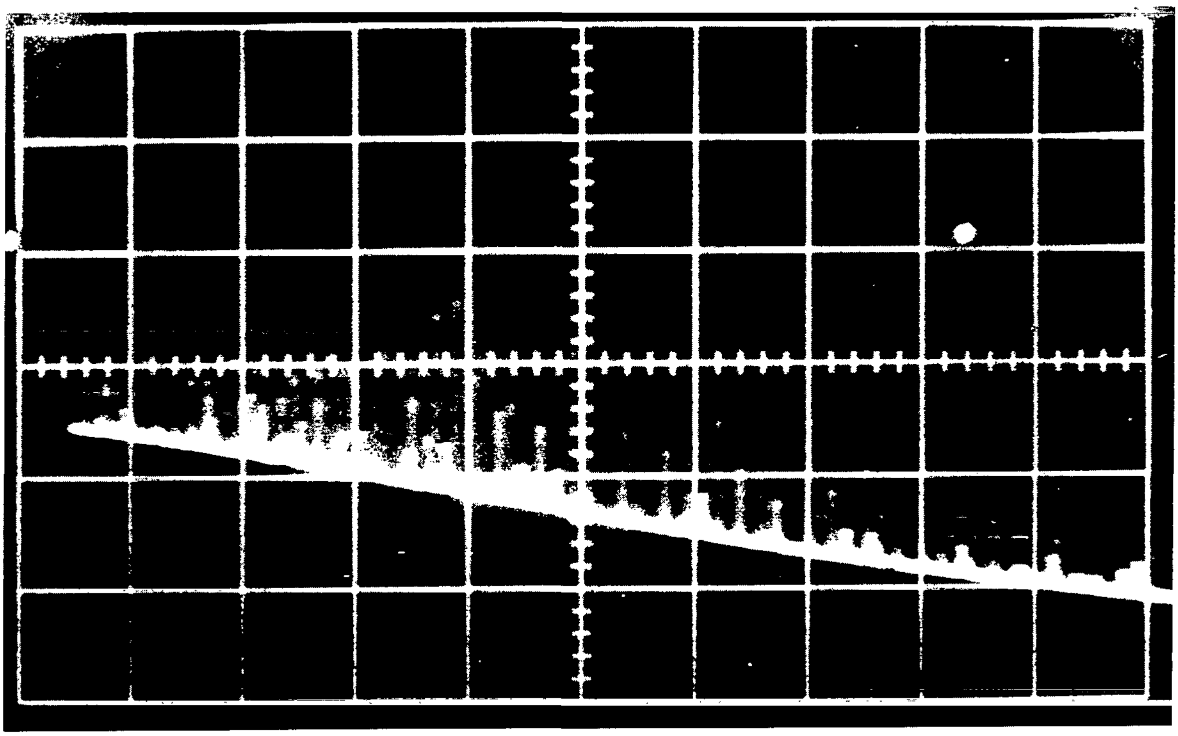

Figure 4.— Typical laser pulse shape, normal mode, 50 μs/div.

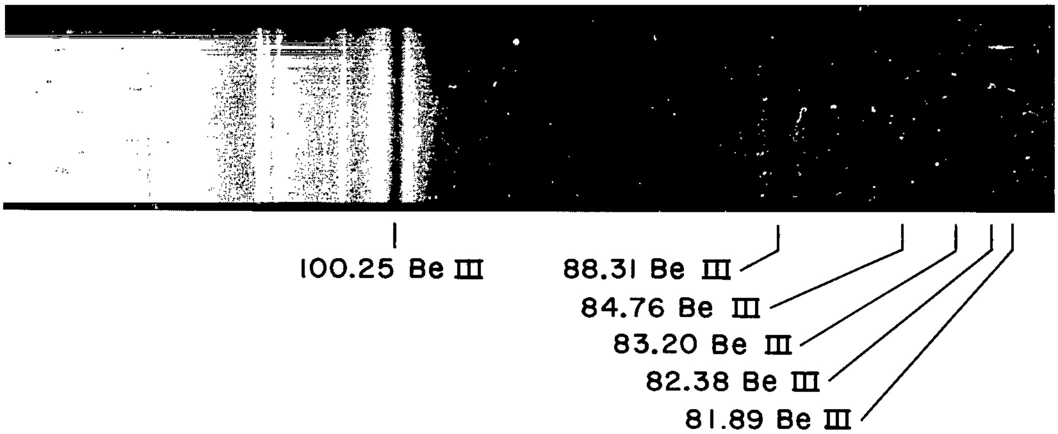

Figure 5.– Principal series of Be III in the 100 Å region. Negative.

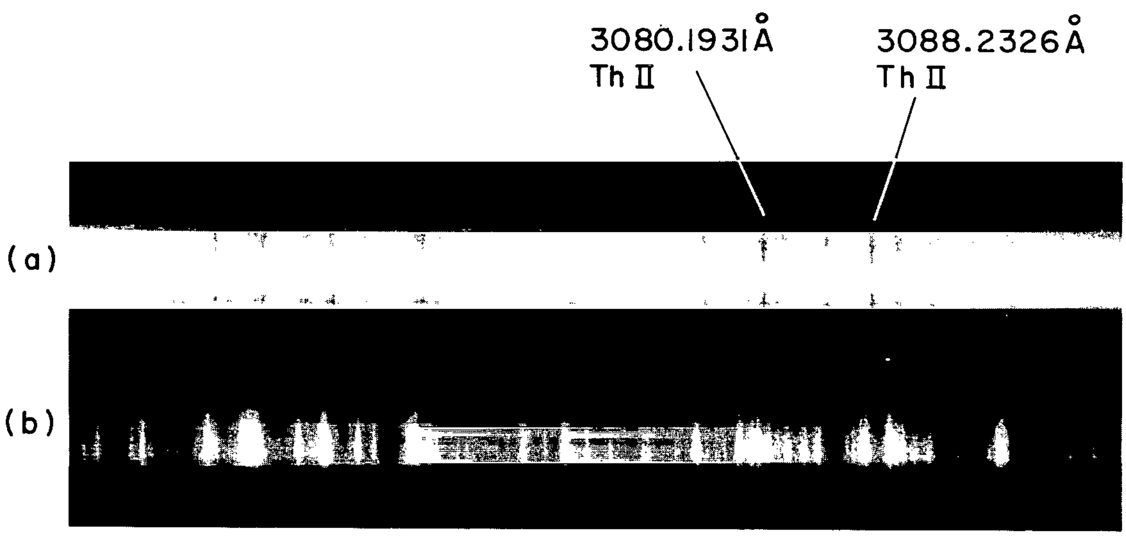

Figure 6.– Normal incidence spectra of Th, 3000 Å region. (a) Q-switched mode with an exposure of 6 pulses. (b) Normal mode with an exposure of 12 pulses. Negative.

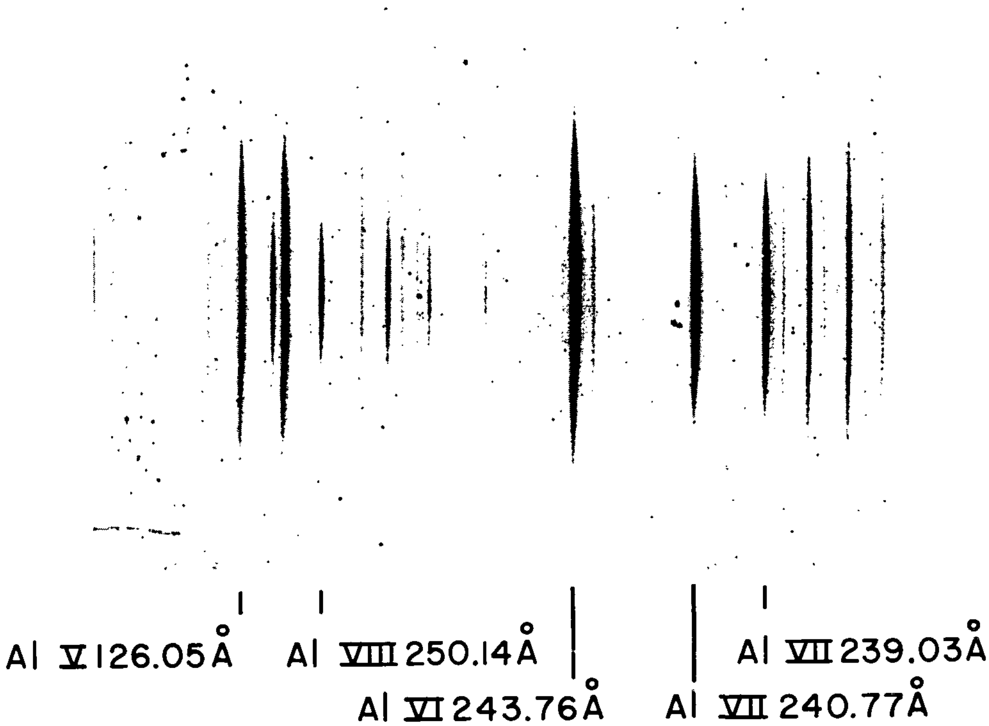

Figure 7.– Grazing incidence spectrum of Al, Q-switched mode, 240 Å region, crossed slit inserted between entrance slit and grating. Total exposure of 30 pulses. Positive.

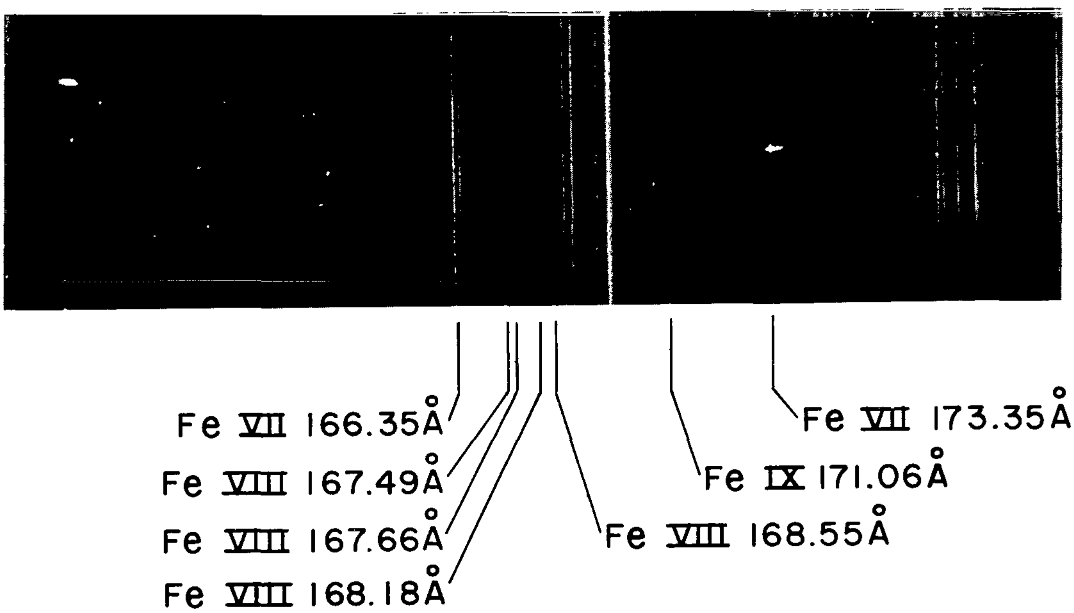

Figure 8.–Grazing incidence spectrum of pure Fe, Q-switched mode, 170 Å region. Total exposure of 6 pulses. Positive.

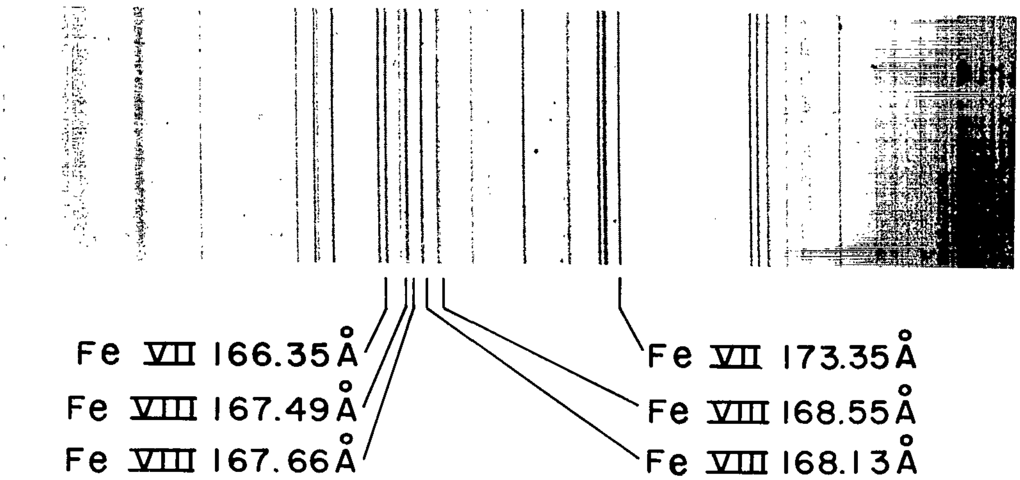

Figure 9.– Grazing incidence spectrum of Al alloy containing 1 percent Fe, Q-switched mode, 170 Å region. Total exposure of 6 pulses. Positive.

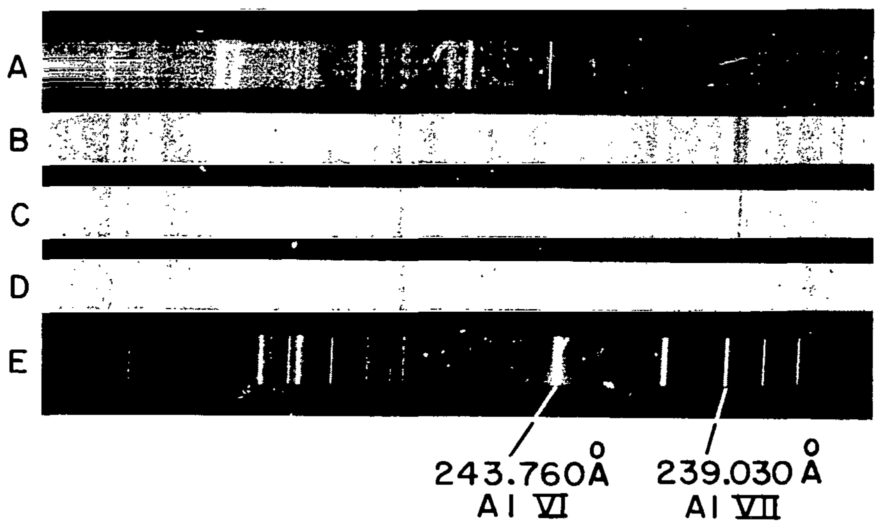

Figure 10.– Grazing incidence spectrum of Al film deposited on glass sheet, Q-switched mode, 240 Å region. Film thicknesses: (a) solid target; (b) 3000 Å; (c) 1000 Å; (d) 500 Å; (e) 200 Å. Total exposure of each track is 6 pulses. Negative.

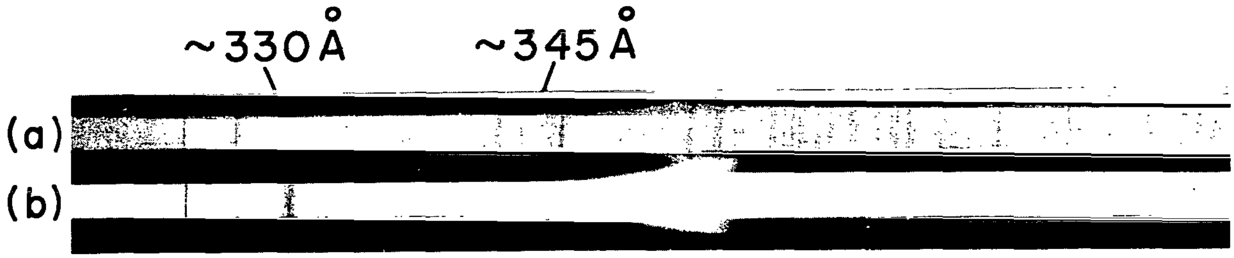

Figure 11.— Grazing incidence spectra of (a) U^{238}, and (b) W, Q-switched mode, 330 Å region, a total exposure of 15 pulses for each spectrum. Negative.

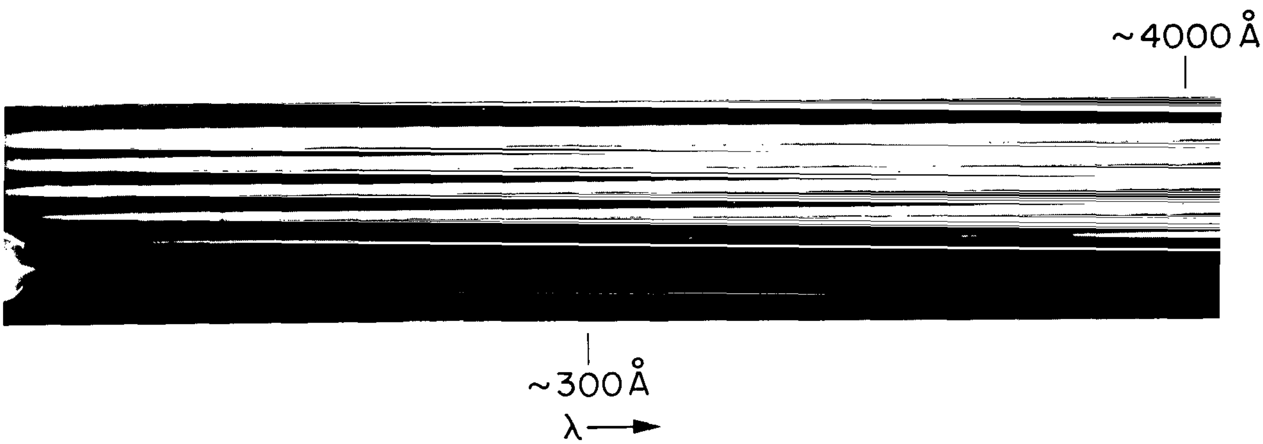

Figure 12.— Normal incidence spectra of Th, Q-switched mode, zero order to 4000 Å region. The total exposure for each track is 10 pulses. Negative.

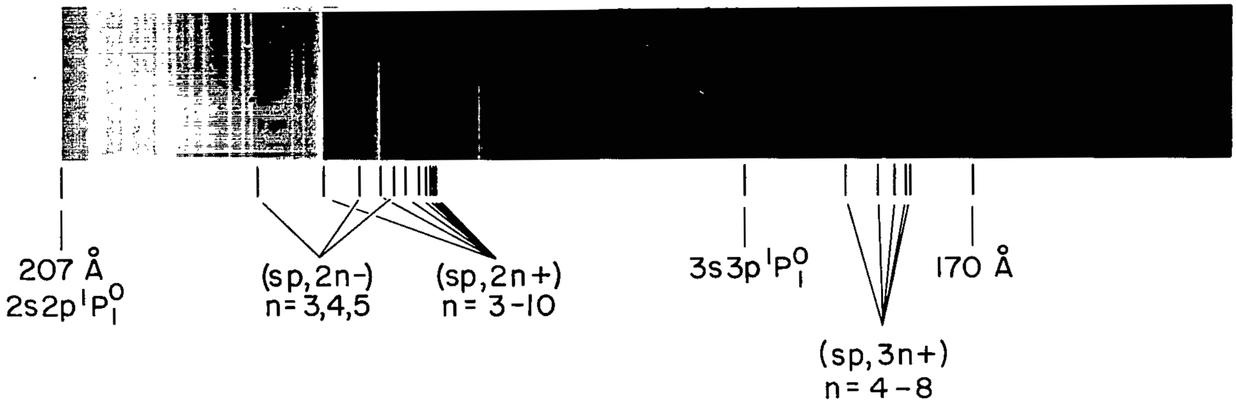

Figure 13.– Grazing incidence absorption spectra of He in 200 Å region showing autoionization series in He. Continuum source is radiation emitted by W target irradiated by focused Q-switched laser pulse. The total exposure is 20 pulses. Positive.

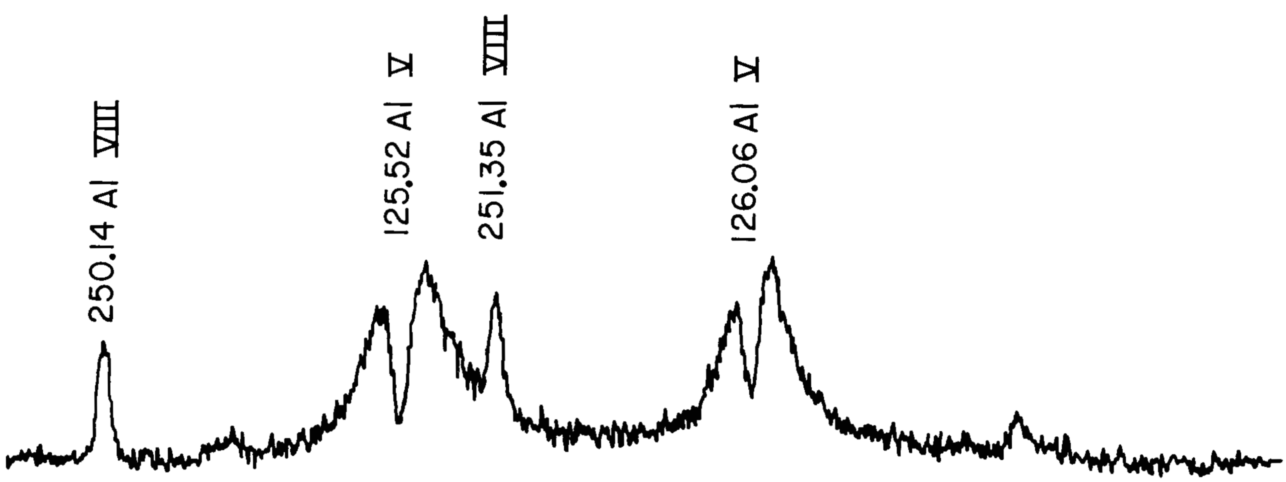

Figure 14.– Microdensitometer tracing of grazing incidence aluminum spectrum, 250 Å region.

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