THE LASER GENERATED PLASMA

AS A SPECTROSCOPIC LIGHT SOURCE

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ABSTRACT

The spectra emitted by laser-generated plasmas have been studied in the spectral region from about 40 Å up 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-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 which aid in the interpretation of spectra are discussed. The laser produced plasmas have several unique characteristics which prove to be desirable and convenient for spectroscopic work.

INTRODUCTION

The work described in this paper was undertaken to investigate 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. In the present work we have been interested in studying these laser-generated plasmas spectroscopically in order to help in understanding the various processes taking place within the plasma. We have also made use of the characteristics of the radiation emitted by the plasma 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 (10 to 500 Å) is shown schematically in Fig. 1. A 3 m grazing incidence spectrograph with a platinum coated concave grating possessing 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 L onto the target T, made of the element of interest. The distance between the target and the slit in the direction of the optical axis is 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 below, on the desired region of the plasma to be observed in each particular case.

The entrance slit, S_1, was about 0.5μ to 10μ wide for these experiments. A crossed slit, S_2, was employed to make the instrument less astigmatic, as described by Alexander, Feldman, Fraenkel and Hoory.[1] The crossed slit was varied in width from 0.1 mm to 1 mm depending on the spectral region being observed.

For the spectral range from 300 Å to the visible a 3 m focal length, normal incidence spectrograph 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 Fig. 2.

All of the experiments were performed using a TRG-104A, 100 MW ruby laser in both the normal and Q-switched modes. Energies of the order of 1 joule were delivered to the target in times of the order of 10-20 ns for the Q-switched mode. With the laser operating in the normal mode, energies of about 2.5 joules were delivered to the target in times ranging from 400 to 500 microseconds. Figures 3 and 4 show typical pulse shapes for the Q-switched and normal modes respectively. Typical photographic plate exposures required one 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 varied included the laser power, the distance of the target from 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 target thickness when deposited on glass as a thin film.

VARIATION OF LASER POWER

The power of the laser pulse was varied from about 10 MW to 100 MW in the Q-switched mode. The most important difference observed at different power levels consisted of 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 in such a way that it constitutes one of the jaws of the spectrograph slit. The laser beam was focused onto the target at a distance of some 0.5 mm from the slit opening in the direction of the optical axis. In this way it was possible to observe the plasma just on the target surface. For Be, a light element, relatively weak continuum radiation was observed in this region.

The power delivered to the target using 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 due to the lengthening of the pulse duration. 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 (a) and in the normal mode (b). In the first case, (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 second case (b in Fig. 6), with the laser operated in the normal mode, the emission spectrum appears and the continuum is weak.

DISTRIBUTION OF IONIC SPECIES IN THE PLASMA BALL

The distribution of ionic species in the plasma ball was studied in the region below 400 Å by introducing a crossed slit, S_2 in Fig. 1, between the principal slit, S_1, and the grating. In this way the astigmatism of the grazing incidence spectrograph is reduced and partial spatial resolution of the plasma ball is achieved. Figure 7 shows an Al spectrogram taken around 240 Å with the crossed slit. It can be seen that the spectral lines belonging to the lower stages of ionization appear longer than those radiated by the higher stages of ionization. It is also interesting to note that the width of the spectral lines is reduced towards 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

The presence of substantial continuum radiation is characteristic of the spectra emitted by laser generated plasmas. This continuum radiation increases in intensity as the atomic weight of the element of interest increases. This continuum gives rise to absorption lines that vary in intensity depending on the stage of ionization and on the particular atomic transition involved in each case. 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 can be observed as absorption lines in a strong continuous background. Figure 6a shows the continuum radiated by a Th target around 2500 Å. Again many absorption lines are observed.

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. The limitation of this method is 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 be desirable. In this way it would be possible to heat the plasma just enough to produce the stages of ionization of interest without "overheating" and consequently increasing the continuous background and other undesirable effects.

We have tried a different approach to this problem of creating emission spectra which consists of using a low atomic number target that contains the element of interest as a small impurity and the laser operating in the Q-switched mode. Figure 9 shows the spectra of Fe around 170 Å as obtained from an Al alloy containing 1% Fe. The Fe lines are in emission and are very sharp. However, this technique of producing emission spectra of good quality is limited by the problems of introducing impurities of certain test elements into a light element such as Al, for example. In many cases commercially available alloys may be employed. Our best spectra have been obtained using this "impurity" technique in so far as sharpness and lack of self reversal are concerned. The broad self-reversed lines observed in Fig. 9 are Al lines.

We have also tested, as originally proposed and demonstrated by Fawcett et al.,[2-6] the laser pulse (Q-switched) irradiation of targets formed by a thin film of the element of interest on a glass target. The most important consequence of using this technique was to reduce somewhat the intensity of the continuous background, thus increasing the contrast between the emission lines and the continuum. This fact made it possible to observe in emission lines of higher stages of ionization that had been previously masked by the continuum. However, the quality of the emission lines obtained in this way is still poor as compared with the emission lines obtained using the "impurity technique". The lines from thin film targets show strong broadening, self-reversal and asymmetries in many cases (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 Fig. 12 shows the continuum radiated by a Th target in the region from 300 to 4000 Å. The origin of these continua have been discussed by Fawcett et al.[2] Note that the height of the continua increases with wavelength which is in accord with the empirical model to be presented later. We have used these continua for absorption experiments in order to demonstrate the utility of the source for such applications. By filling the grazing incidence spectrograph with He and Ar at pressures ranging from 0.25 to 0.50 torr and using W as a target to provide a continuum in the soft X-ray region, we have been able to obtain the autoionization series of such elements. The spectra obtained in this experiment are shown in Fig. 13, which shows the autoionization series of He around 200 Å originally studied by Madden and Codling[7] using a synchrotron as a source of continuum radiation.

The laser plasma continuum source has the inconvenience of having present the absorption lines characteristic of the heavy element used as the target material. These absorption 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 angstroms have been reported.[8] 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

Esteva and Romand[9] and Fawcett, et al.,[2] have observed strong broadening and asymmetries in line shapes in laser-generated plasmas. 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 10.

The same general behavior has been observed in the 500-4000 Å region for Fe and Th. The magnitude of the shifts suggest caution when attempting to employ the laser-generated plasma light source for wavelengths determinations. For complex spectra this is particularly true if one tries to identify spectral lines from the isoelectronic sequences.

In such dense rapidly expanding plasmas these shifts and broadenings are, undoubtedly, the result of all of the several causes of such effects, including Stark, pressure, and Doppler phenomena, as well as self-absorption. However, in general, it is possible to obtain sharper lines for most stages of ionization by using occulting techniques. For example, the entrance slit of the spectrograph may be illuminated with only the light radiated from specific regions of the plasma drop. The viewed region may be varied, depending upon the stages of ionization to be investigated, extending from the very outer portions of the plasma for lower stages of ionization to the central, hotter portions of the plasma core for higher stages of ionization. The lines from the highest degrees of ionization produced, are obtained with minimum broadening. This technique has the serious disadvantage that since very small portions of the plasma ball are observed, very 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.

The use of the small-percentage-impurity low-atomic-number alloy target also 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 calculational models for the laser-produced plasmas have been proposed.[11] However, based on Dawson's model, and from 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 initial 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, which, for the case of ruby laser light ($\nu = 4.35 \times 10^{14}$ sec^{-1}) occurs at an electron density of 2.5×10^{21} cm^{-3} ($\nu_p = 8.9 \times 10^3\ n_e^{1/2}$).[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 is expanding, increasing its volume and consequently reducing the electron density and the plasma frequency. Once the plasma frequency drops to a value lower than the light frequency then the light starts penetrating the plasma again and is absorbed. The heating mechanism, once the plasma drop is formed, is presumably primarily inverse bremsstrahlung followed by electron-heavy particle collisions.

The energy losses 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 which 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 recombinations of the highly ionized species take place 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 in the outside of 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[10] 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 is also associated with the particular transitions involved. This fact makes it possible not only 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., the use of self reversed lines to help in locating the lower energy levels). This feature of the laser generated plasma proves to be of great help for spectroscopic work on highly ionized species. Figure 14 shows a microdensitometer tracing of a portion of the Al spectra around 250 Å. As seen in the figure, 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 note that 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 which 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 has been shown to be 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. Besides the advantages of this light source even the disadvantages of this inhomogeneous pulsed light source resulting in shifts, reversals, asymmetries, broadenings etc. of the spectral lines have proved useful in the solution of the classic problem in spectroscopy: that of identifying the states involved in the transitions giving rise to the observed spectral lines.

FOOTNOTES

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FIGURE CAPTIONS

1. Schematic 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.
2. Schematic of optical arrangement, normal incidence spectrograph. LL, laser; L, focusing lens; S, entrance slit; G, grating; P, plate.
3. Typical laser pulse shape, Q-switched mode, 10 nsec/div.
4. Typical laser pulse shape, normal mode, 50 μsec/div.
5. Principal series of Be III in the 100 Å region. Negative.
6. Normal incidence spectra of Th, 2500 Å region. (a) Q-switched mode. (b) Normal mode. Negative.
7. Grazing incidence spectrum of Al, Q-switched mode, 240 Å region, crossed slit inserted between entrance slit and grating. Positive.
8. Grazing incidence spectrum of pure Fe, Q-switched mode, 170 Å region. Positive.
9. Grazing incidence spectrum of Al alloy containing 1% Fe, Q-switched mode, 170 Å region. Positive.
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. Negative.
11. Grazing incidence spectra of (a) U^{238}, and (b) W, Q-switched mode, 330 Å region. Negative.
12. Normal incidence spectra of Th, Q-switched mode, zero order to 4000 Å region. Negative.

13. Grazing incidence absorption spectra of He in 200 Å region showing auto-ionization series in He. Continuum source is radiation emitted by W target irradiated by focused Q-switched laser pulse. Positive.

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

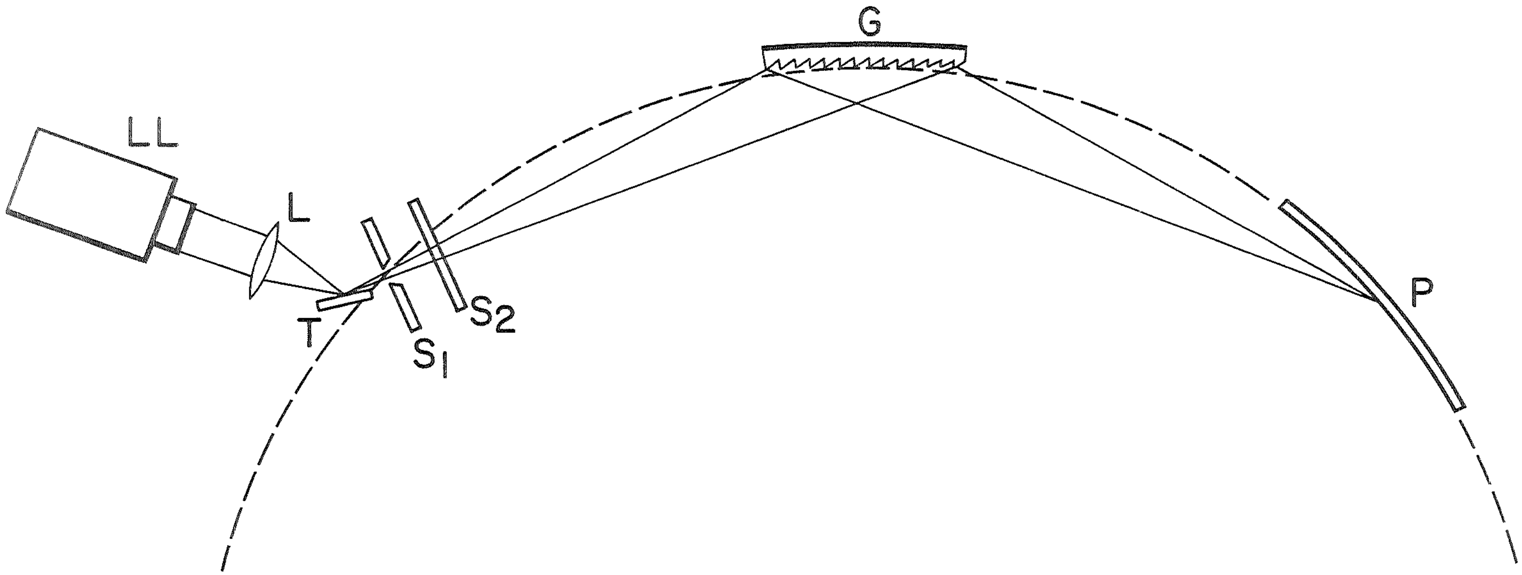


Figure 1.

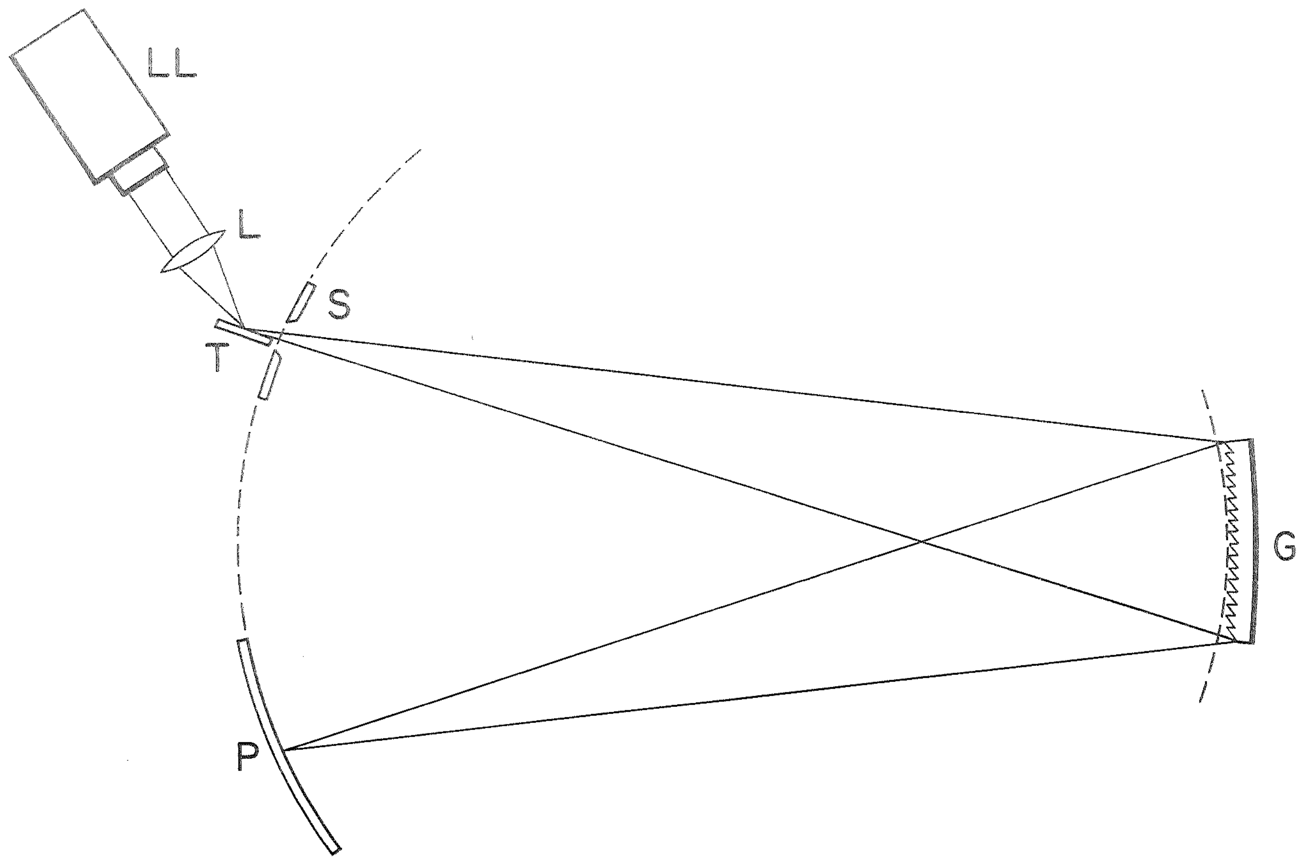


Figure 2.

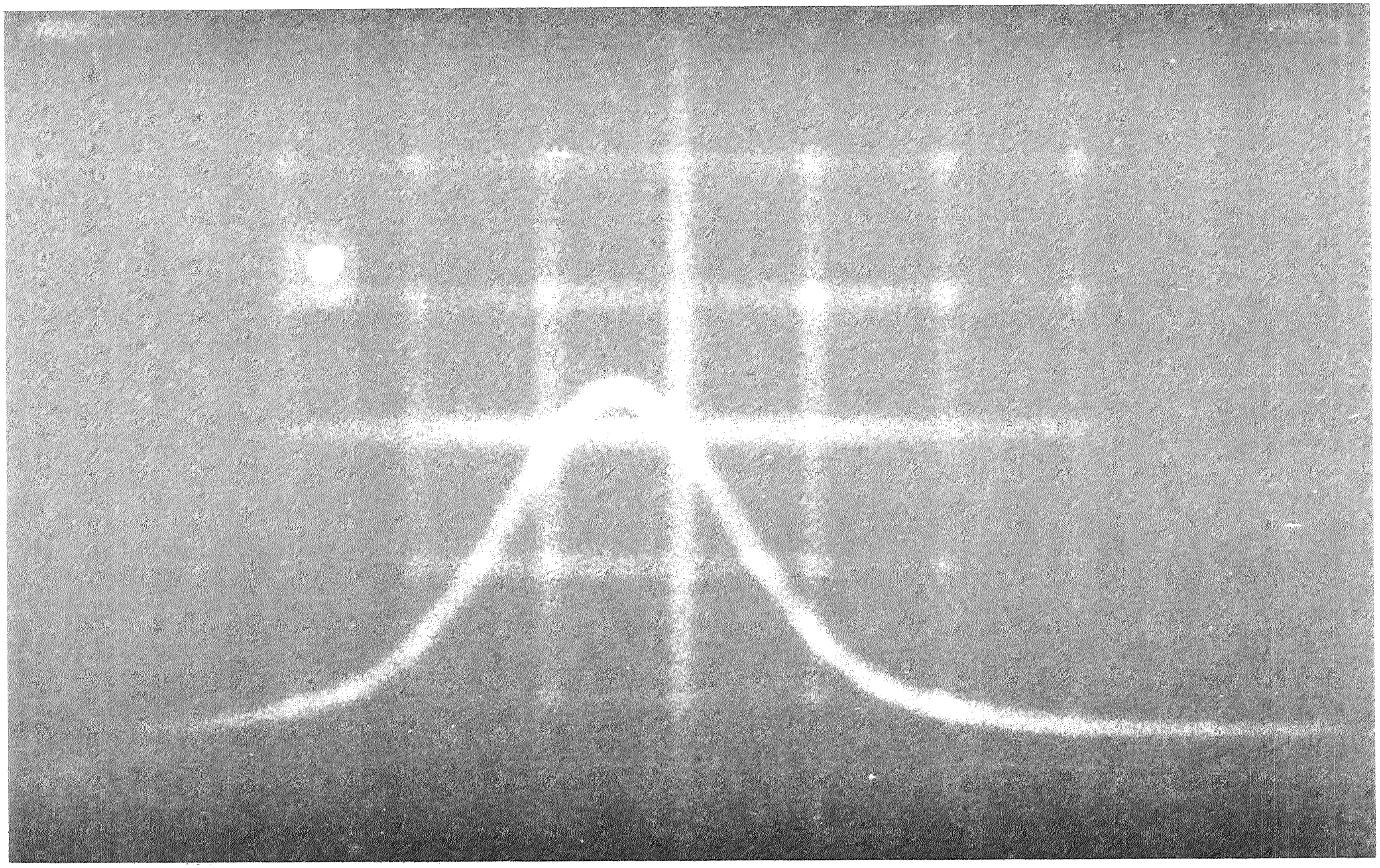


Figure 3.

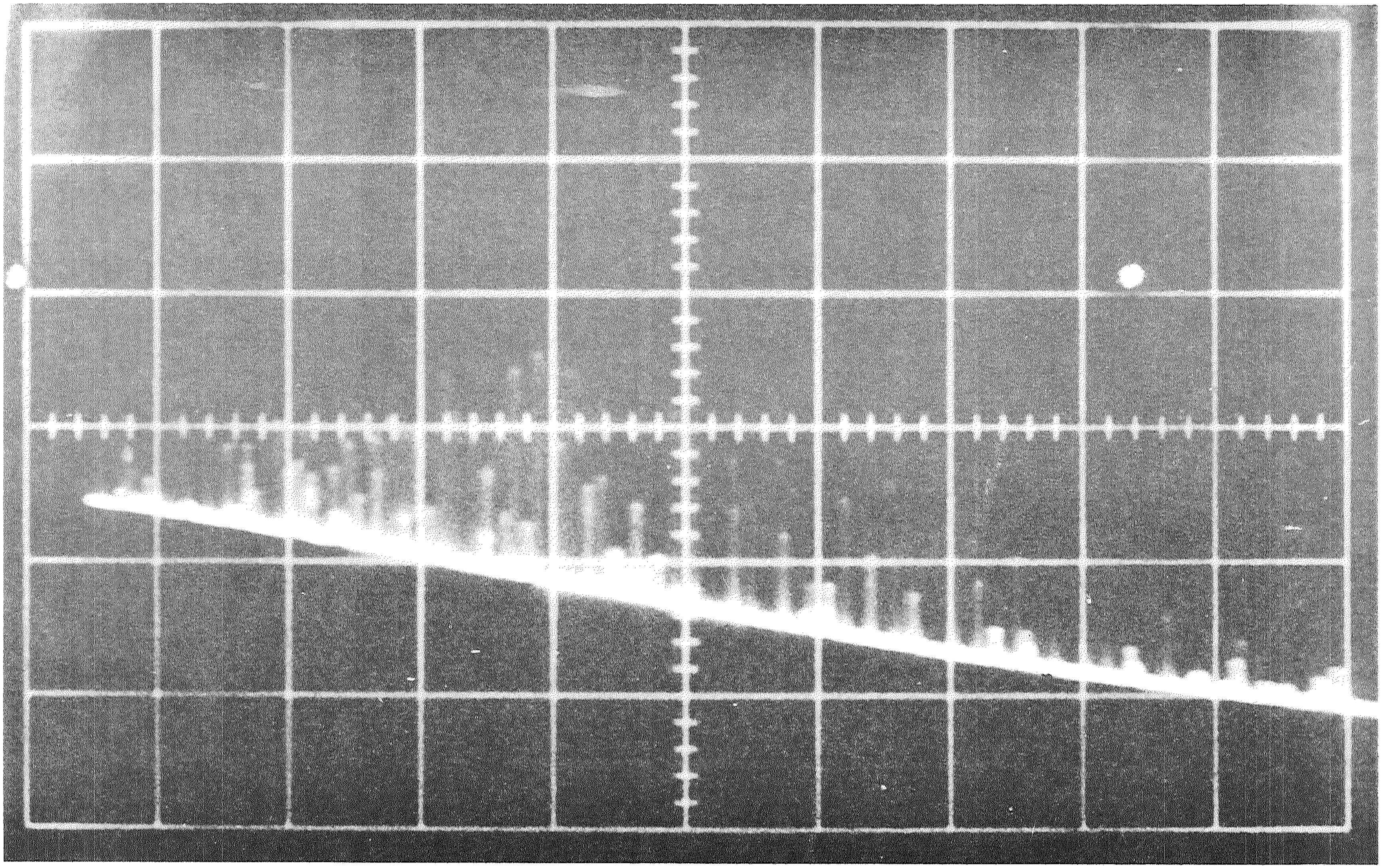


Figure 4.

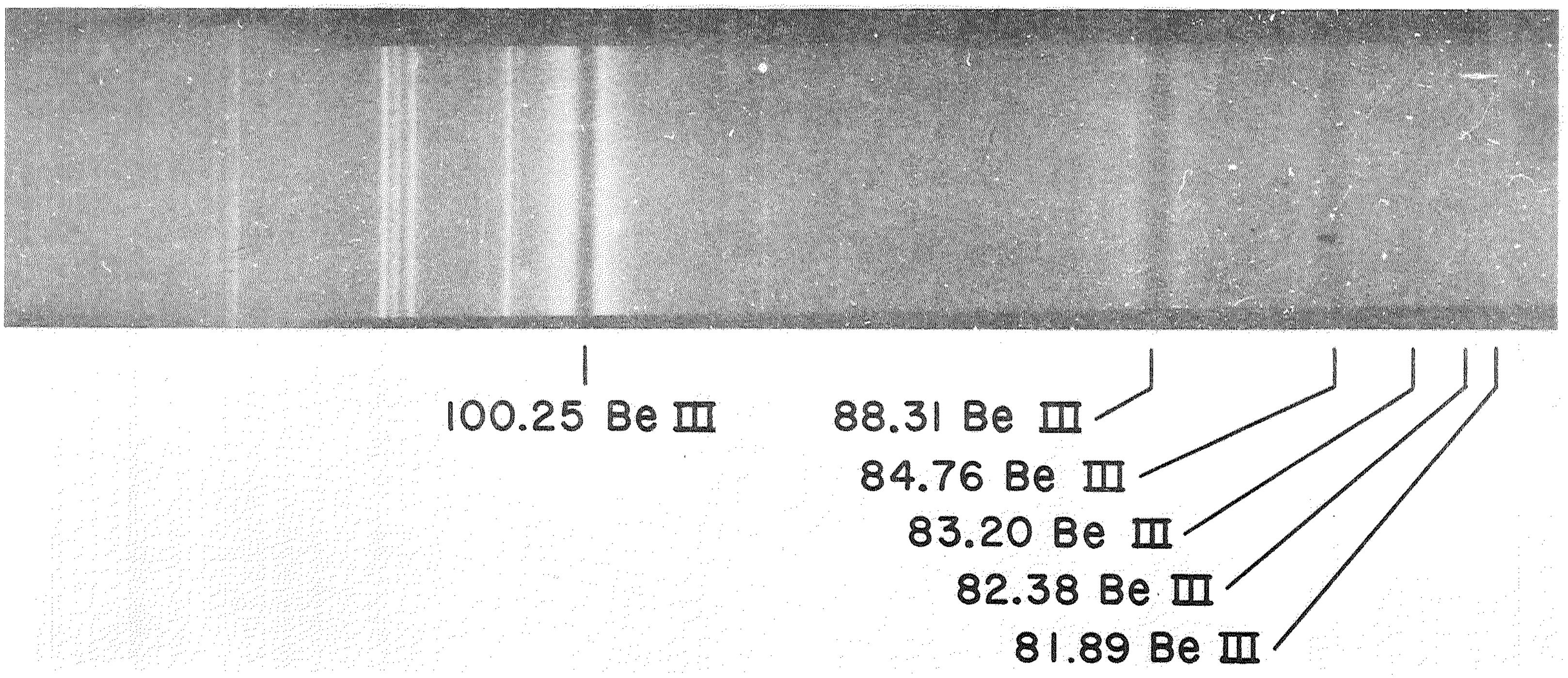

100.25 Be III
88.31 Be III
84.76 Be III
83.20 Be III
82.38 Be III
81.89 Be III

Figure 5.

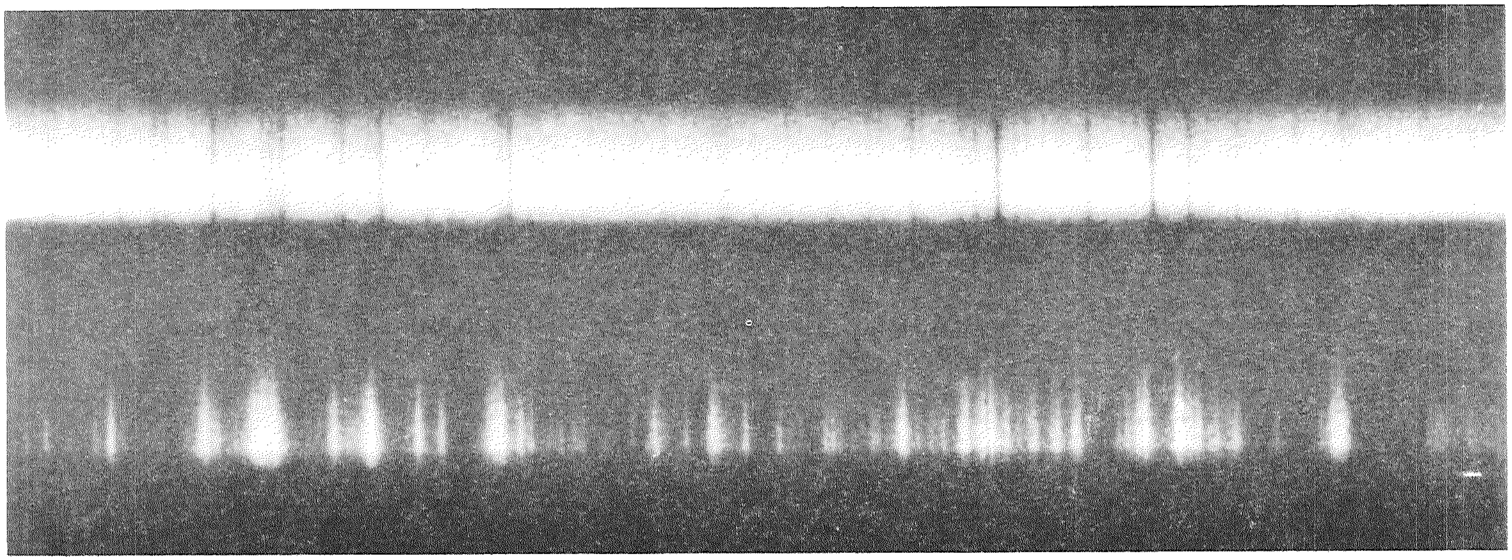
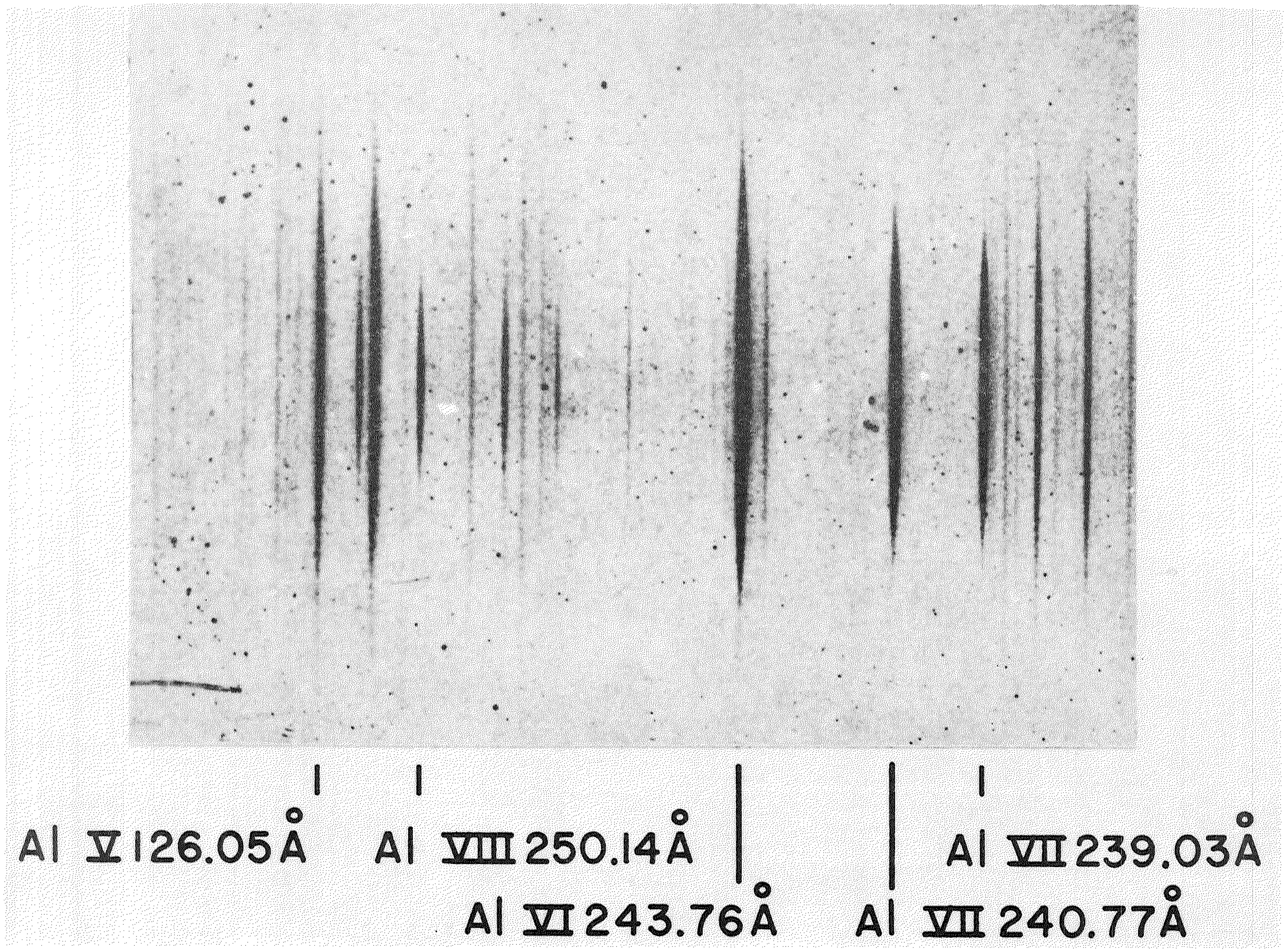


Figure 6.



Al V 126.05Å
Al VIII 250.14Å
Al VII 239.03Å
Al VI 243.76Å
Al VII 240.77Å

Figure 7.

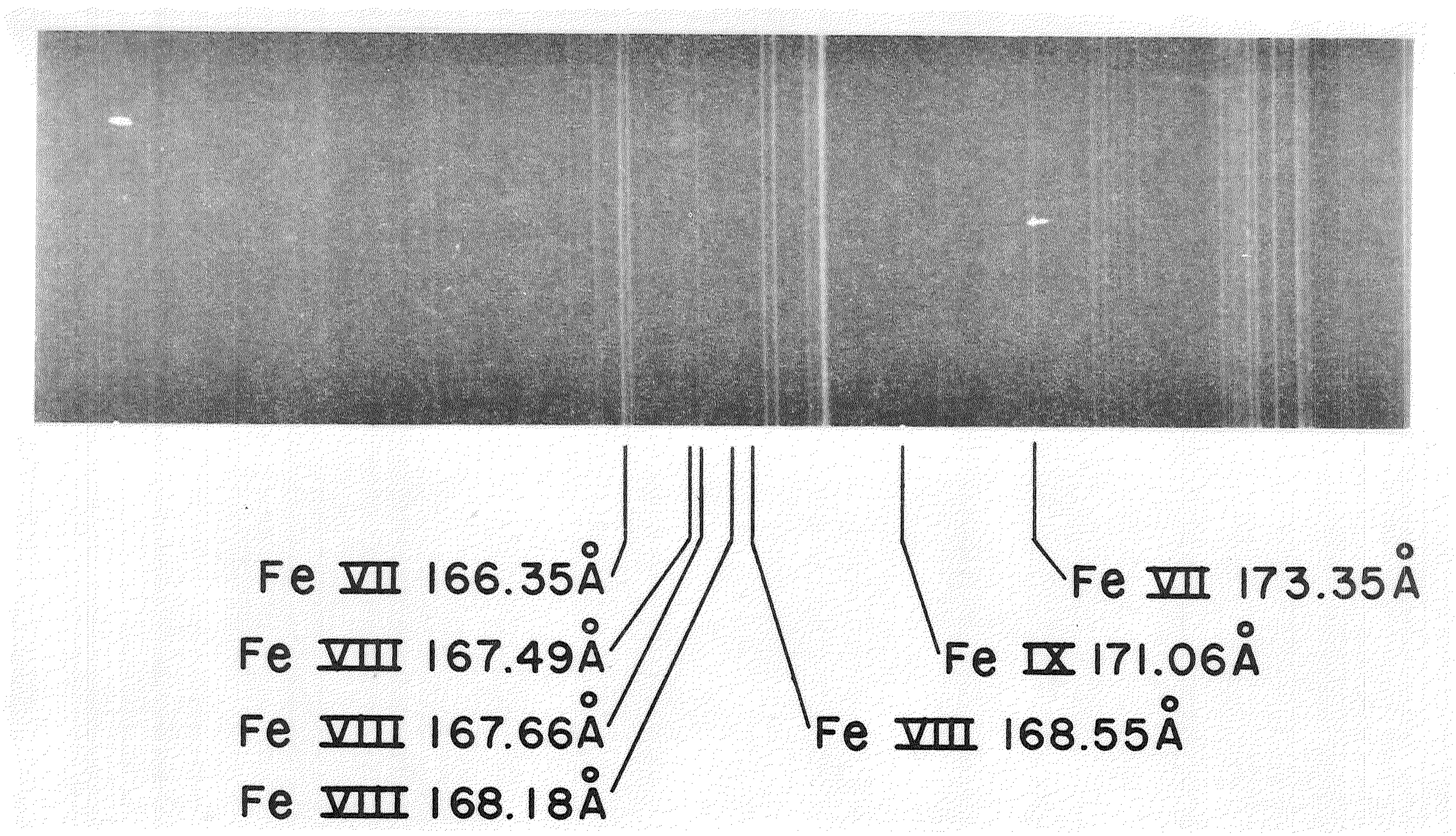

Fe VII 166.35Å
Fe VIII 167.49Å
Fe VIII 167.66Å
Fe VIII 168.18Å
Fe VIII 168.55Å
Fe IX 171.06Å
Fe VII 173.35Å

Figure 8.

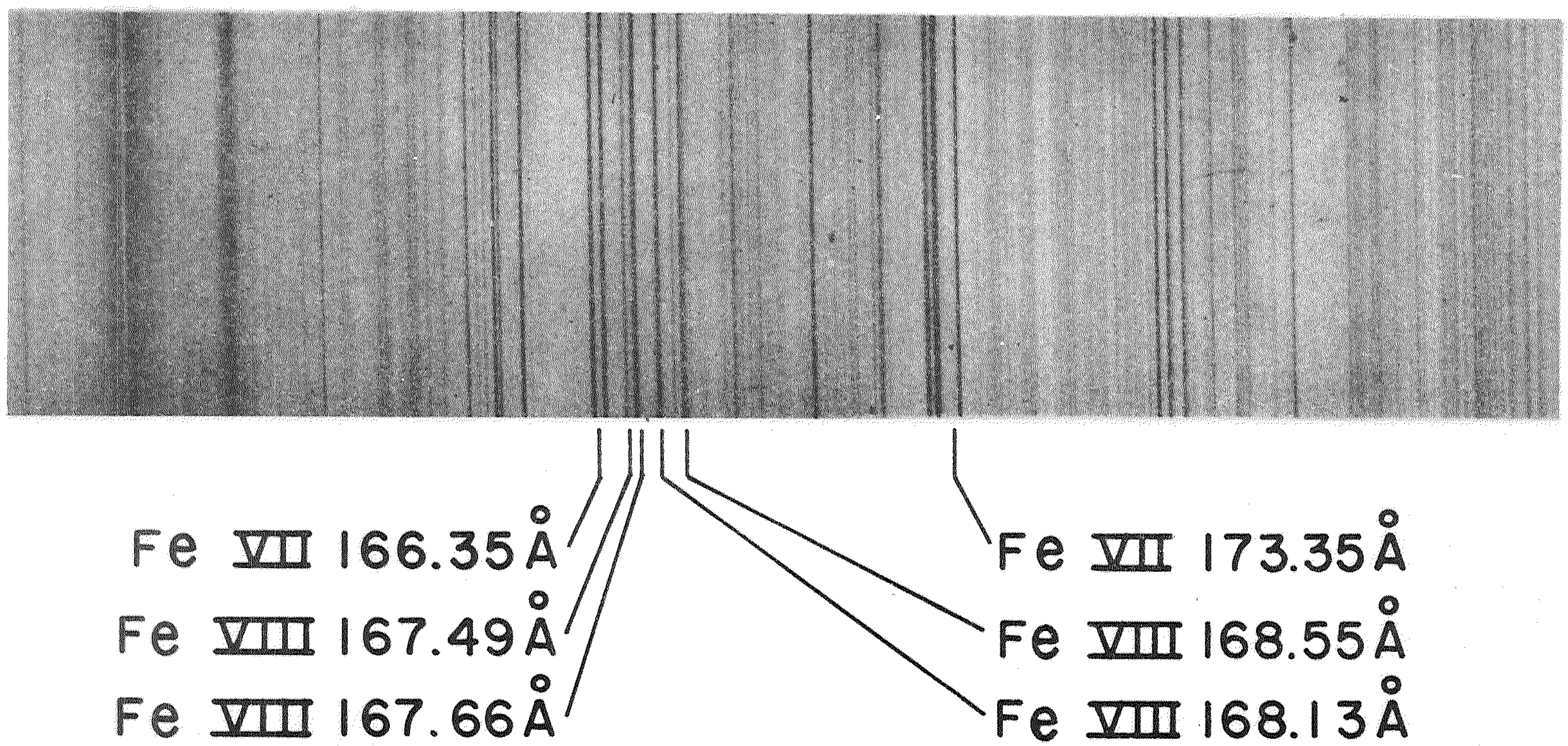

Fe VII 166.35Å
Fe VIII 167.49Å
Fe VIII 167.66Å
Fe VII 173.35Å
Fe VIII 168.55Å
Fe VIII 168.13Å

Figure 9.

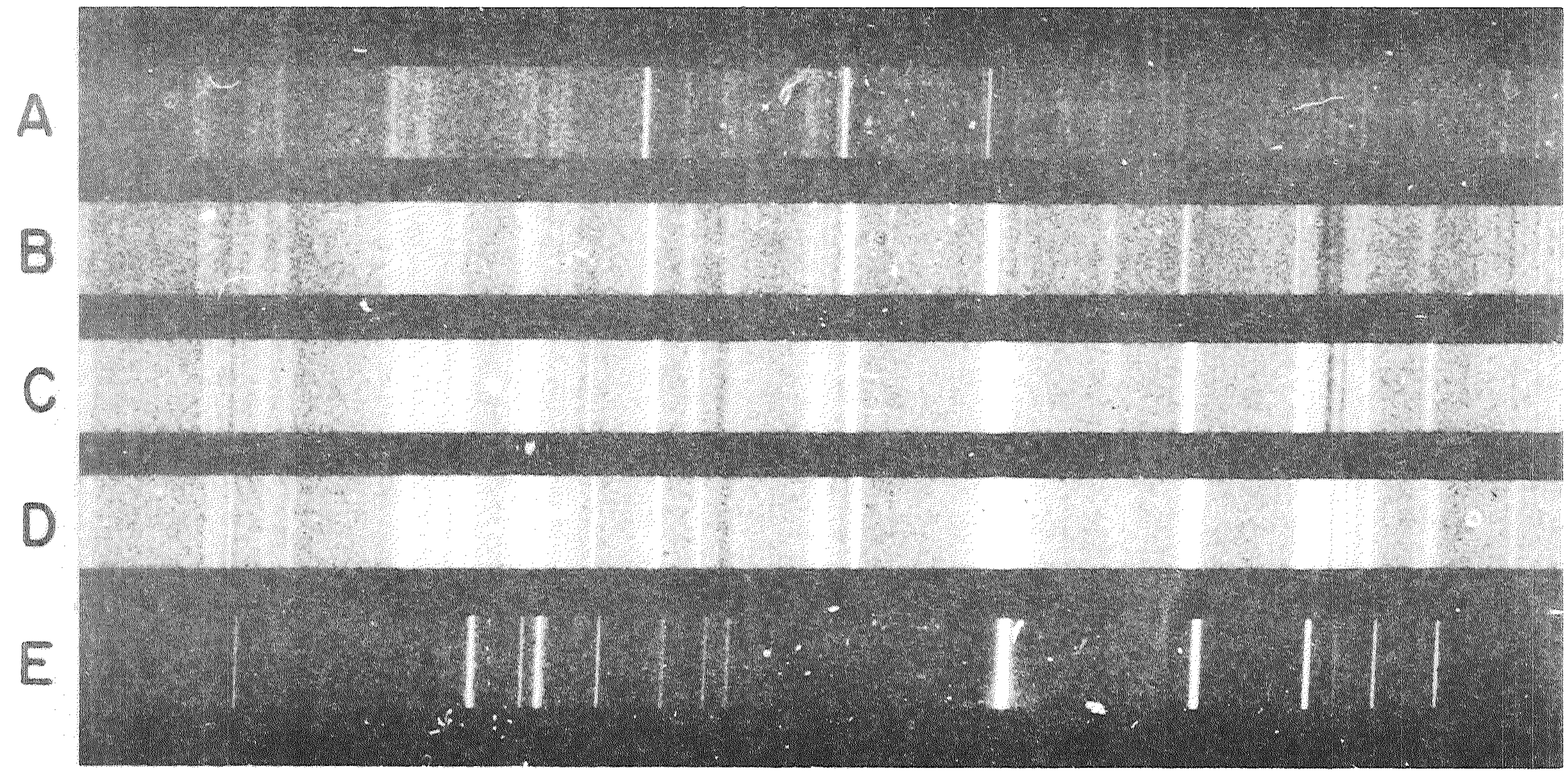


Figure 10.

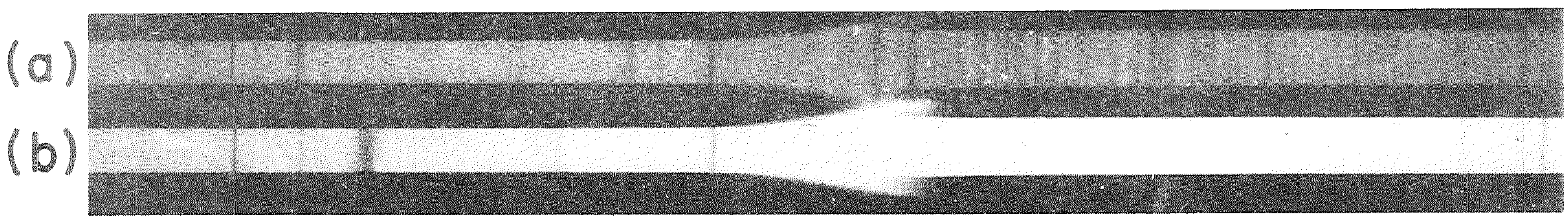


Figure 11.

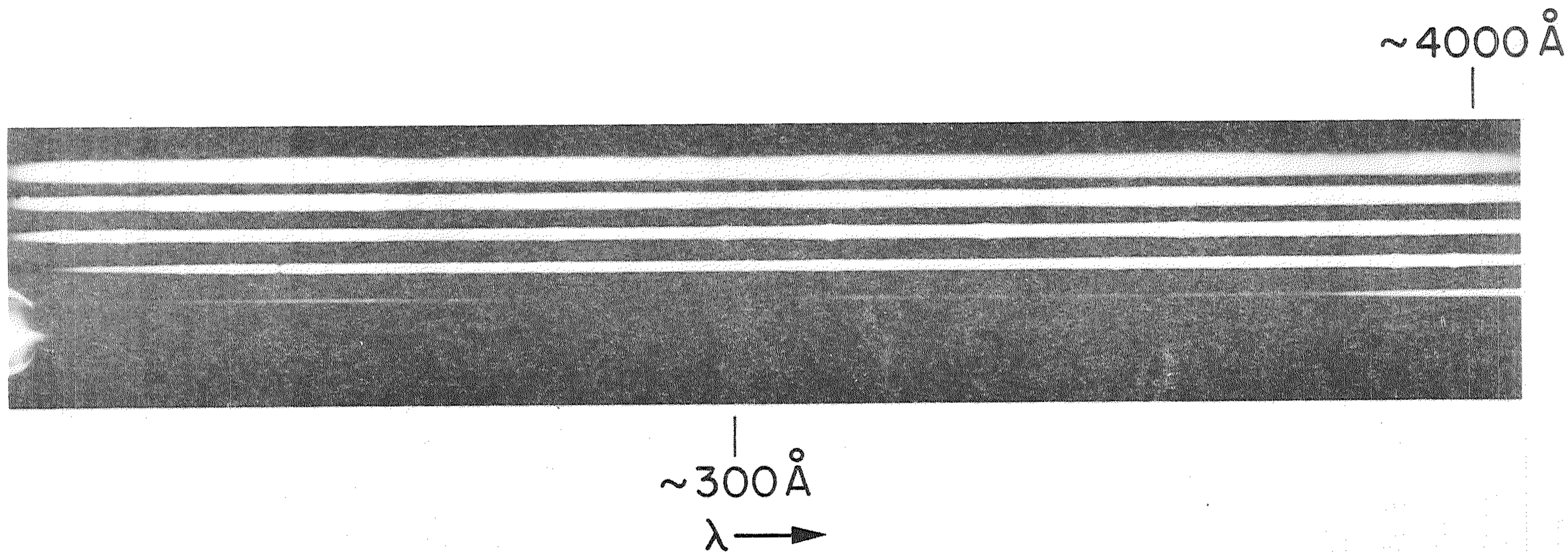


Figure 12.

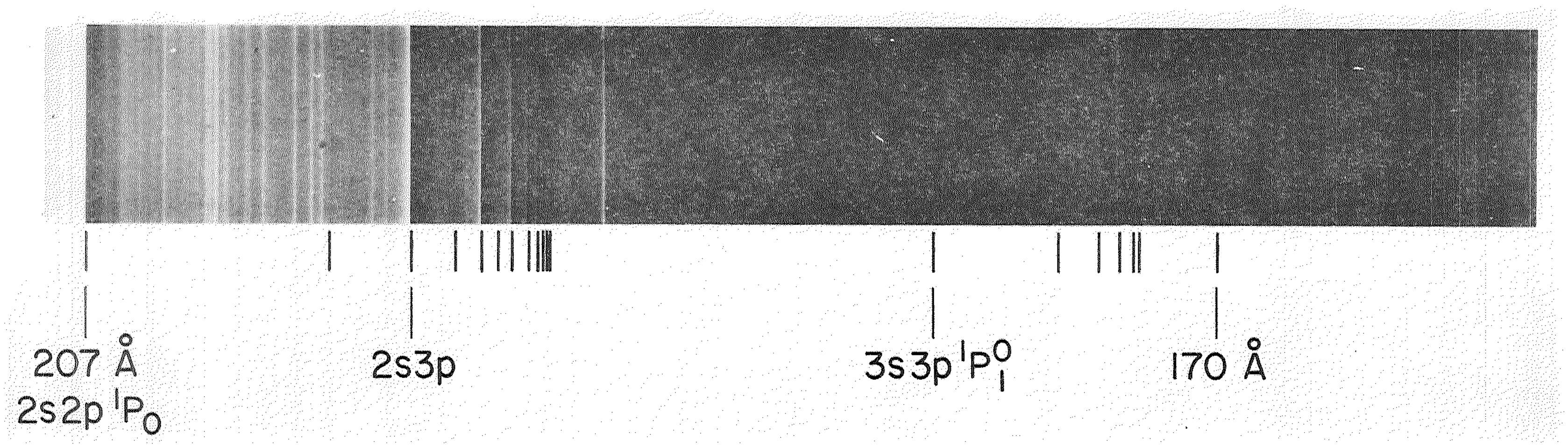


Figure 13.

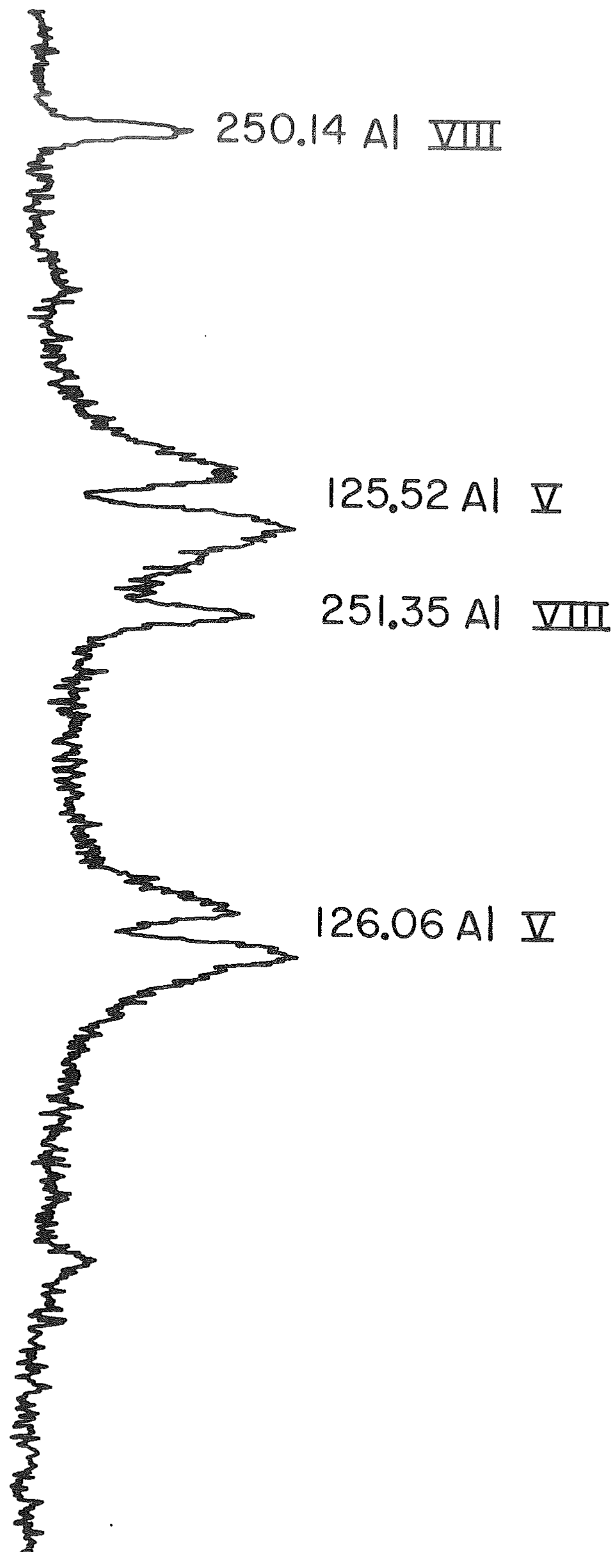


Figure 14.