

BEAM-FOIL LIFETIMES OF CrII

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by

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Introduction

In 1962, the first formal proposal for a beam-foil experiment was made by Kay¹. Beam-foil Spectroscopy (BFS) has since come into wide use. Along with other applications BFS provides a new method for measuring the lifetimes of atomic states from which radiative transition rates may be derived. Previously arc, spark, and absorption spectra were the primary sources for this information. BFS presents problems of its own, of course, but it does not have the problem associated with it of the assumption of thermal equilibrium in the light source. In addition, one need not worry about plasma stratification, which is a difficulty encountered in shock tube experiments.²

A BFS lifetime measurement is performed by passing a beam of accelerated ions through a thin foil. As atoms pass through the foil the valence electrons are excited. Downstream of the foil one may observe the number of photons emitted as a function of distance from the foil. The velocity of the beam after the foil is then used to convert the distance scale to a time scale. Thus the final result is a plot of the number of photons emitted versus time. The slope of this plot is the lifetime of the emitting level.

Lifetimes of atomic energy levels may be used with branching ratios to find atomic transition probabilities, which are important to the study of elemental abundances in the solar and stellar atmosphere. Recent calculations of the chromium abundance, based on BFS lifetimes of CrI,³ make it desirable to calculate a similar abundance based on the lifetimes

of CrII. The reason for this is that at solar photospheric temperatures the principle ionic species is CrII. Therefore, a small error in the temperature-pressure stratification might lead to a large error in the fractional abundance of CrI and would produce a significant error in determining the total chromium abundance from CrI. The ratio of CrII to total chromium abundance should be much less sensitive to small errors in the temperature-pressure stratification.

The work presented in this thesis may be conveniently divided into four phases:

1) Some question as to the accuracy of BFS measurements has been raised in connection with the energy loss incurred by the beam.⁴ The problem has been examined to some extent in this work. The energy loss for 89 keV Ar⁺ has been measured and agrees surprisingly well with stopping power calculations.

2) In preparation for the lifetime experiments, spectral scans of the BFS spectrum have been taken between 1900 and 4100 Å. Lines originating in CrI and CrII exist in this region and have been identified.

3) Lifetimes of five lines representing four levels in CrII have been measured. These measurements were made at two energies to ensure adequate knowledge of the energy loss.

4) A preliminary calculation of the solar abundance of chromium has been made based on our CrII results.

Spectroscopic Background

The following discussion will define common spectroscopic terminology and relate the Einstein coefficient (A) to the oscillator strength (f), and to the atomic radiative lifetime (τ). The following definitions are based on the shell model of the atom and the LS coupling

scheme, and will consider only radiative transitions of the valence electrons.⁵ In this coupling scheme, a spectroscopic "term" is characterized by the total orbital angular momentum (L) and the total spin (S) of the valence electrons. The conventional notation for a term is ^{2S+1}L , where $2S+1$ is called the "multiplicity" of the term. The multiplicity is the number of possible orientations of S relative to L. One may think of the multiplicity as the number of ways L and S can couple to form the total angular momentum, J, but this is only true for $L \geq S$. For example, if $S = \frac{1}{2}$ and $L = 0$ as in the hydrogen atom, the term is 2S and there are two possible orientations of S, but only one possible coupling to form J ($J = \frac{1}{2}$). A "level" designates one possible value for J, and is written $^{2S+1}L_J$. One specifies the "sublevel" that an atom is in by specifying the orientation of the atom, M. Sublevels are degenerate in energy except in the presence of a magnetic or electric field. A transition between two sublevels produces a "line component". The blend of line components produced by all transitions between two particular levels form a "line". The set of all lines produced by transitions between two terms is called a "multiplet".

The lifetimes of atomic levels are the principal quantities with which this work is concerned. However, the literature often refers to measurements of "gA-values or gf-values" as a measure of electromagnetic transition probabilities. Therefore, it is appropriate to discuss these two quantities as an alternative means of expressing transition probabilities.⁶

Let us express the initial sublevel as $i(\alpha'J'M')$, where α' represents all internal quantum numbers, and the final sublevel as $f(\alpha JM)$. Also let us define $A(\alpha'J'M' \alpha JM)$ to be the transition rate for the transition

$i \rightarrow f$, that is the transition probability per unit time for $i \rightarrow f$. Since, in the absence of an electric or magnetic field the components blend to produce a single line, it is important to know the transition rate for the line. For each level there will be $2J+1$ sublevels, corresponding to the $2J+1$ possible values of M . To calculate the transition rate for the level ($A(\alpha'J' - \alpha J)$) it is necessary to sum the sublevel transition rate over all M and M' . Thus, $A(\alpha'J' - \alpha J)$ is defined by the equation

$$N(J') A(\alpha'J' - \alpha J) = \sum_{M'M} N(J'M') A(\alpha'J'M' - \alpha JM),$$

where $N(J')$ is the population of the level and $N(J'M')$ is the population of each of the sublevels $i(\alpha'J'M')$. Since the transition rate of the sublevel is not dependent on the orientation of the atom (M'), it may be factored from the sum over M' . Thus, suppressing M' in the transition rate,

$$N(J') A(\alpha'J' - \alpha J) = \sum_M A(\alpha'J' - \alpha JM) \sum_{M'} N(J'M').$$

Since it is clear that

$$\sum_{M'} N(J'M')/N(J') = 1,$$

we may write

$$A(\alpha'J') = \sum_M A(\alpha'J' - \alpha JM).$$

In a thermally excited source the sublevels are assumed to be equally populated. Thus, we may write

$$N(J') = (2J'+1) N(J'M') = gN(J'M'),$$

which is the defining equation for the statistical weight g . The observed intensity I is equal to the number of atoms multiplied by the transition rate. The intensities and populations are the quantities normally measured in thermal sources. One may express the intensity as

$$I = N(J') A(\alpha' J' - \alpha J) = gA(\alpha' J' - \alpha J) N(J'M').$$

The gA -values may be found experimentally by measuring the populations of the sublevel and the intensity of the line.

In BFS the lifetimes of atomic levels are measured directly. Thus a relation must be sought that will provide the total transition rate ($A(\alpha' J')$) from one particular level. This may be done by summing all the transition rates out of the level of interest. Therefore one has

$$A(\alpha' J') = \sum_{J, \alpha} A(\alpha' J' - \alpha J).$$

The lifetime (τ) of a level is defined to be the inverse of the transition rate, and it is the time needed for the population of the level to decrease by $1/e$ of its original value.

By observing the lifetime of a single line originating in the level of interest, when all transitions from that level are taking place, one is provided with the total lifetime of the level. This is true because

the number of transitions per unit time from the level of interest to a particular final level is proportional to the number of atoms ($N(\alpha'J')$) in the level at that time. But N obeys the time dependant equation

$$dN/dt = -N/\tau ,$$

and will be given by $N\lambda(\alpha'J' - \alpha J)$. Therefore, when all allowed transitions occur each transition out of level ($\alpha'J'$) will be characterized by a decay constant which is the total lifetime of the level. In order to transform the measured lifetime into the corresponding g_A -values for a specific transition. More experimental information is needed. This is the branching ratio which is defined as the ratio of the transition rate of $i(J') \geq f(J)$ to the total transition rate.

As has been shown, atomic lifetimes and transition rates are different quantitative measures of the same phenomena. The oscillator strength or f -value is yet another way of describing the rate of an atomic transition.⁷ The oscillator strength may be defined by comparing the strength of absorption by a quantum mechanical atom with that which would be incurred by an electron moving in a potential characterized by the resonant absorption frequency (ν). If, for a particular frequency, there are N such classical absorbers per unit volume, and N' atoms per unit volume, then f is defined by $N = N'f$, and is the number of classical absorbers per atom. In terms of the transition rate the f -value is given by

$$gf = g_A mc^3/8 \pi e^2 \nu^2 = 1.4493 \times 10^{-5} m^{-2} g_A/\nu^2 ,$$

where m is the mass of the electron and e is the charge on the electron in esu. Since the natural lifetime (T) of a classically absorbing electron is

$$T = 3mc^3/8e^2 \pi^2 \nu^2,$$

the preceding equation may be written as

$$gf = g_A T/3.$$

This relationship indicates the intrinsically classical nature of f -values. It is important to realize that the two values of g are not the same and may not be factored from the equation.

The Energy Loss: Procedure

The energy loss suffered by the chromium ions traversing the foil is of great importance in the calculation of the level lifetimes, as it is necessary to express the distance from the foil as a function of time. This distance is related to the time of flight of the ion by

$$t = d (m/2(E-\Delta E))^{1/2},$$

where E and m are the energy and mass respectively of the incident ion, d is the perpendicular distance between the foil and the axis of the lens, t is the time it takes for the ion to traverse d , and ΔE is the energy loss.

The problem of calculating the stopping power of a low energy ion traversing matter has been explored by Lindhardt.^{8,9} Calculations

of energy losses were made using the method of Lindhardt and a program written by Brand, Fox, and Keller.¹⁰ The results for several relevant choices of ion, energy, and thickness of carbon foil are listed in Table I.

Table I
Theoretical energy loss calculations for 89 keV Ar,
85 keV Cr, and 100 keV Cr

Ion	x (gm/cm ²)	E(keV)	E(keV)
Ar	2.4	89	11
Cr	2.0	85	13
Cr	2.0	100	13

In order to investigate the validity of these calculations, the energy loss for an 89 keV Ar ion was measured. Argon was used rather than chromium because the argon beam is easily produced. Since it has an atomic number ($Z=17$) close to that of chromium ($Z=24$), the results of the experiment provide a useful comparison to the calculation.

The beam for the energy loss experiment was supplied by a 150 kV linear accelerator, equipped with a Physicon universal ion source. The Physicon source is capable of producing ionized gas from either gaseous or solid materials. The argon beam was produced by allowing argon gas to enter the source at a rate which produced a partial pressure in the accelerator of approximately 6×10^{-6} mm of Hg. The gas was ionized in the source and extracted from it by a -10 kV potential on an extractor electrode. The beam was then accelerated to an energy of 89 keV, mass analyzed at the magnet, and directed into the target chamber, as shown

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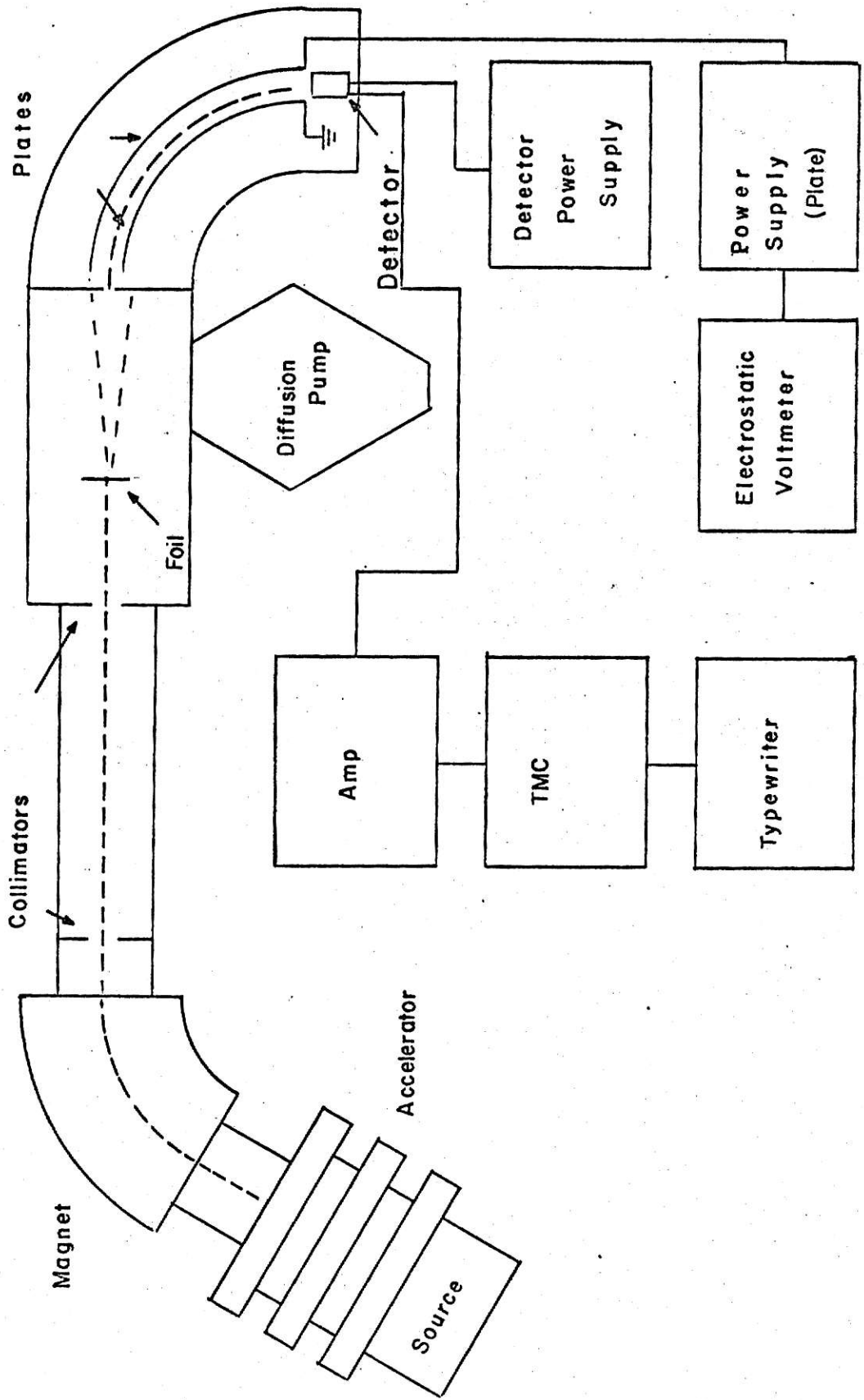


Fig. 1.--The apparatus for the energy loss experiment

in Figure I. The argon beam was identified by maximizing the beam current of the most intense beam at a Faraday cup just downstream of the magnet. The argon leak was then closed; if the beam current dropped dramatically, it was assumed that the argon beam had been identified.

The size of the beam spot was restricted by the use of two $\frac{1}{4}$ in. collimators, and vertical control of the beam was accomplished with the use of electrostatic deflection plates powered by a 400 V variable voltage supply. (The plates and power supply are not shown in Figure I.) A vacuum was maintained in the target chamber by a 4 in. diffusion pump located several inches downstream of the foil.

An electrostatic analyzer, built by Curnutte, was used to measure the energy of the beam before and after passage through the foil, the energy loss being the difference between the two values. The analyzer consisted of two curved parallel plates preceded by a 0.5 mm aperture. Particles were detected by a Bendix Spiraltron electronmultiplier located at the exit aperture of the analyzer.

The signal from the Spiraltron was amplified by a spectroscopic amplifier and recorded on a TMC (Technical Measurements Corporation) multi-channel analyzer, model 404-6. The voltage for the analyzer plates was supplied by a Spellman high voltage power supply, and was calibrated with an electrostatic voltmeter to ± 0.1 kV (corresponding to approximately ± 1.06 keV in particle energy). In order to record the energy spectrum on the TMC, the plate voltage was varied manually at a rate of .03 kV/sec, while the TMC address was advanced internally by 1 channel/sec.

The beam was electrostatically analyzed without a foil from which the ratio of the beam energy to the plate voltage of the electrostatic analyzer was determined to be 0.0945 kV/keV of particle energy. Several energy loss measurements were then made on a single foil. It was noted that as time elapsed the energy loss of the beam increased. Therefore energy loss measurements were made using a new foil noting carefully the time elapsed during each run. The foil used for these runs had been damaged in mounting and thus small holes were present in the foil. Under these circumstances, the energy spectrum contained a peak for the beam of particles that passed through the foil and a second peak for the unperturbed particles, which passed through the holes. Three additional runs were made with unused foils to make possible a calculation of the average energy loss.

The foils used throughout the experiments were provided by the Arizona Foil Company. The thicknesses quoted here, ranging from $10 \mu\text{gm}/\text{cm}^2$ to $2.0 \mu\text{gm}/\text{cm}^2$, are those measured by the supplier. Thin carbon foils are not durable if self-supporting, and therefore were mounted on 80% transmission electroplated nickel mesh. Several foils could be placed in the chamber simultaneously, and could be moved in and out of the beam while the system was under vacuum.

The Energy Loss: Results

The results of the energy loss experiment may be divided into two parts: first, the thickening of the foils; second, the actual energy loss incurred by the beam.

Figure II shows the gradual thickening with time of a carbon foil, which was initially $2.4 \mu\text{gm}/\text{cm}^2$ in thickness. This time dependence is probably better described as a dependence on the amount of charge

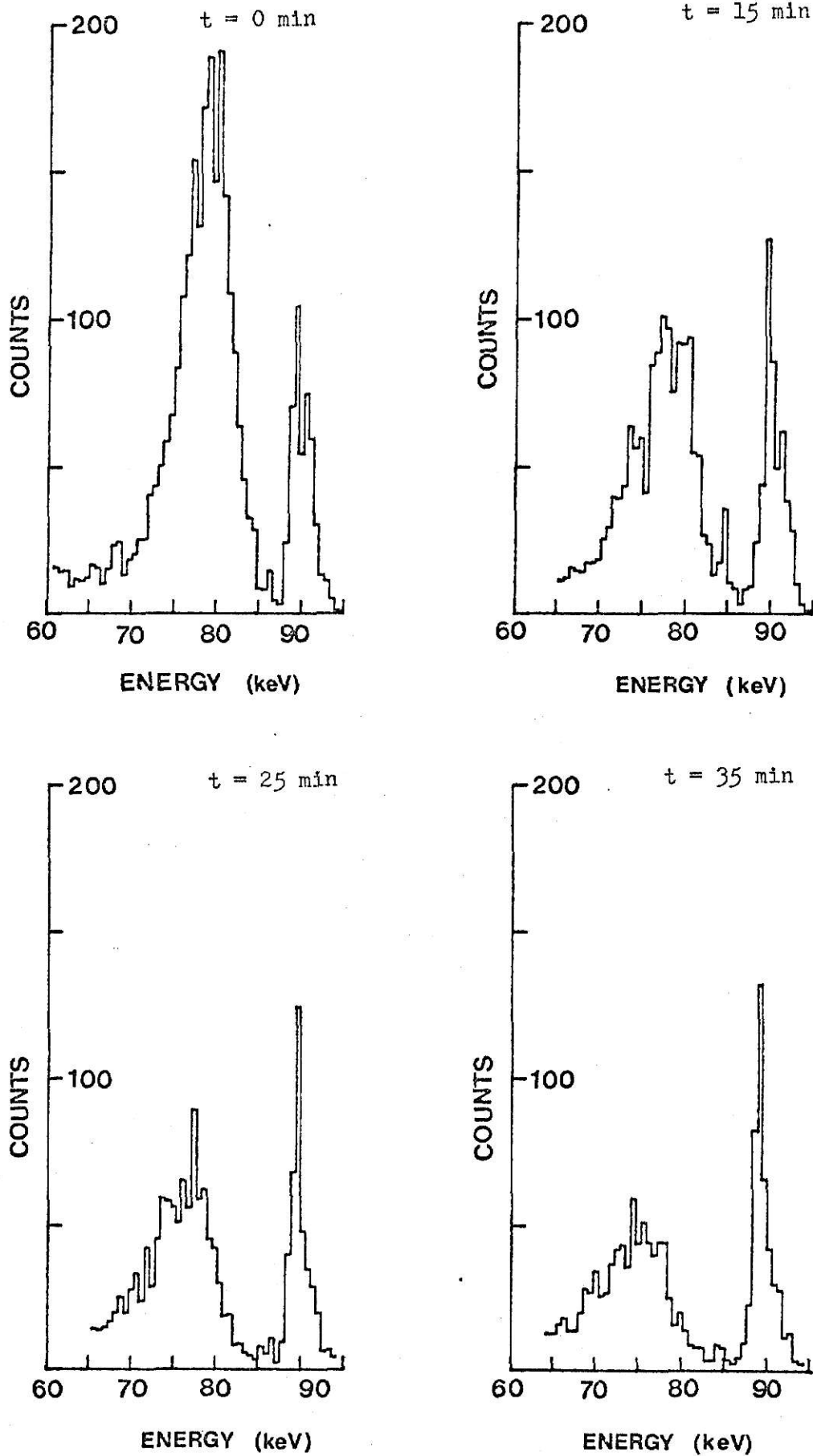


Fig. II.--The energy spectrum of Ar after passage through a carbon foil

delivered to the foil. The incident beam was 89 keV Ar⁺, and corresponds to the peak of higher energy in each graph. The pressure in the system was 10⁻⁵ mm of Hg. The time indicated is the time at which each run began, a typical run duration being 3 minutes. The beam current could not be measured directly while the electrostatic analyzer was in use, but it was measured at a forward Faraday cup before the experiment. The percentage of current transmitted to the foil from this cup is normally about 30%, but transmission has been as high as 50%. Therefore from the measurement of 13 μ A at the Faraday cup, one may conclude that the foil actually received a current on the order of several microamperes. It was found that the energy loss increased from 10 keV to 14 keV in a period of 35 minutes. This implies that the foil thickened at a rate of 0.015 μ gm/cm²-min. This effect is attributed to the presence of hydrocarbons (pump, oil, etc.) in the system. The build up probably occurs when a molecule very close to the foil is hit by an incoming ion. Dissociated, the molecule may form new bonds with the foil or other molecules. It is hypothesized that better vacuum would reduce the size of the effect. Precautions were taken both in subsequent energy loss experiments and in the lifetime experiments to avoid this change in foil thickness. For the energy loss measurements each foil was used only once, while in the lifetime experiments the time each foil was exposed to the beam was kept below 15 minutes.

An average energy loss of 10 keV was measured using 2.4 μ gm/cm² foils. The foils were bombarded for approximately 2 min. The agreement between this measurement and the previous calculation of 11 keV (see Table I) strongly supports the validity of our use of the Lindhardt calculations for the chromium lifetime analyses.

The Spectral Scans: Apparatus

The same acceleration, collimation, and pumping systems were utilized for the spectral scans as for the energy loss experiment (see Figure III). Solid chromous chloride was used in the source to produce the chromium beam. The Cr^+ beam was identified by first producing an Ar^+ beam as in the energy loss experiment. A Hall probe was then used to measure the magnetic field of the deflection magnet. The voltage measured across the Hall probe is proportional to the magnetic field. The magnetic field needed to turn an ion of given energy through a specified radius of curvature is proportional to the square root of the mass of the ion. Thus, for Ar^+ and Cr^+ beams the following formula gives the projected Hall probe voltage (V_2) for the chromium beam,

$$V_2 = 1.14 V_1,$$

where V_1 is the Hall probe voltage measured when argon was used.

An adjustable LiF lens, with a focal length of 4.5 in. at 4000 Å, was placed perpendicular to the beam line and downstream of the foil. The purpose of this lens was to focus the light emitted by the decaying ions on the slits of the monochromator. Survey scans were made using $6 \mu\text{gm}/\text{cm}^2$ foils. The position of the foil was such that a maximum amount of light was focused on the slits of the monochromator. The monochromator used was a McPherson model 218 vacuum ultraviolet monochromator. Two gratings were available for use with the monochromator; a 1200 line per mm grating blazed at 3000 Å which produced a dispersion in the system of 26.5 Å/mm of slit width, and a 2400 line per mm grating blazed at 1500 Å which produced a dispersion of 13.3 Å/mm.

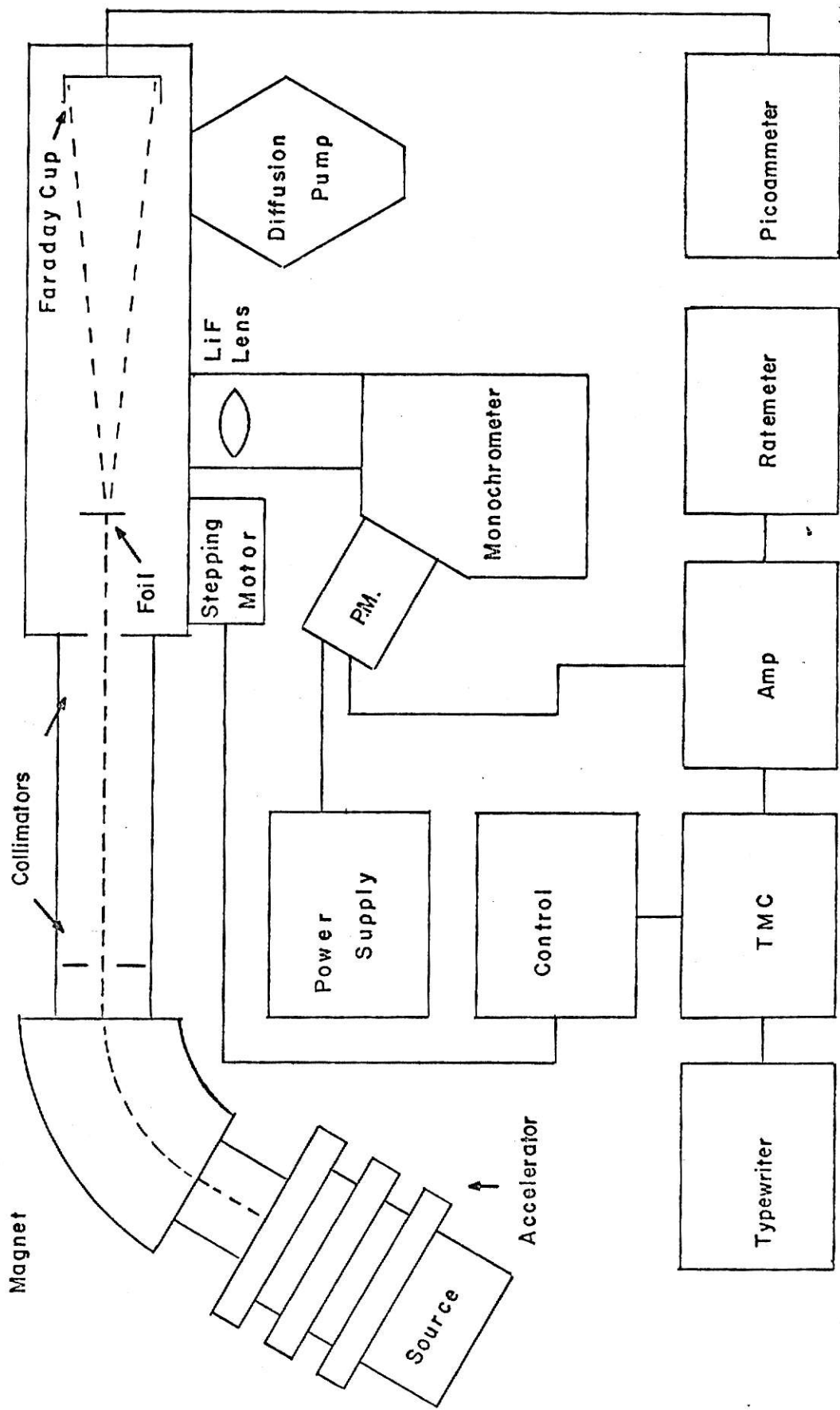


Fig. III.--The apparatus for the spectral scans and for the lifetime experiment

Photons of the proper wavelength were detected with an EMI 6256S photomultiplier tube. The tube was enclosed in a copper case and was cooled by means of a thermoelectric cooling device. A voltage of 1.5 kV was supplied to the tube by a Fluke model 404M power supply. A Nuclear Data Corporation dual channel amplifier and single channel analyzer model ND500, was used to amplify the photomultiplier signal and discriminate against low level noise. This amplifier supplies two output signals, positive pulses which were counted by the TMC, and negative pulses which were monitored on a count rate meter. The electrostatic analyzer had to be removed, and was replaced by a Faraday cup which was used to monitor the beam current.

Two sets of scans were made:

1) Low resolution scans were made using the 2400 line/mm grating. The monochromator slits were set at 250 microns to produce an instrumental resolution of 3 Å. The scanning rate was 50 Å/min. and the TMC was operated in the internal channel advance mode at one channel per second. Thus the TMC recorded all counts produced within a wavelength range of $5/6$ Å in a single channel. Seven overlapping scans of 400 channels (330 Å) each were taken to cover the wavelengths between 1900 and 4100 Å. The results of these scans are presented in Figures IV - IX.

2) High resolution scans of selected regions were taken in a similar manner using the 1200 line/mm grating. Slit widths for these scans were reduced to 150 or 200 microns depending on the resolution needed. The TMC was paced at 1 channel/sec., and the monochromator scan rate was 5 Å/min. The length of these scans was approximately 50 Å. The resulting spectra are shown in Figures IX - XIII.

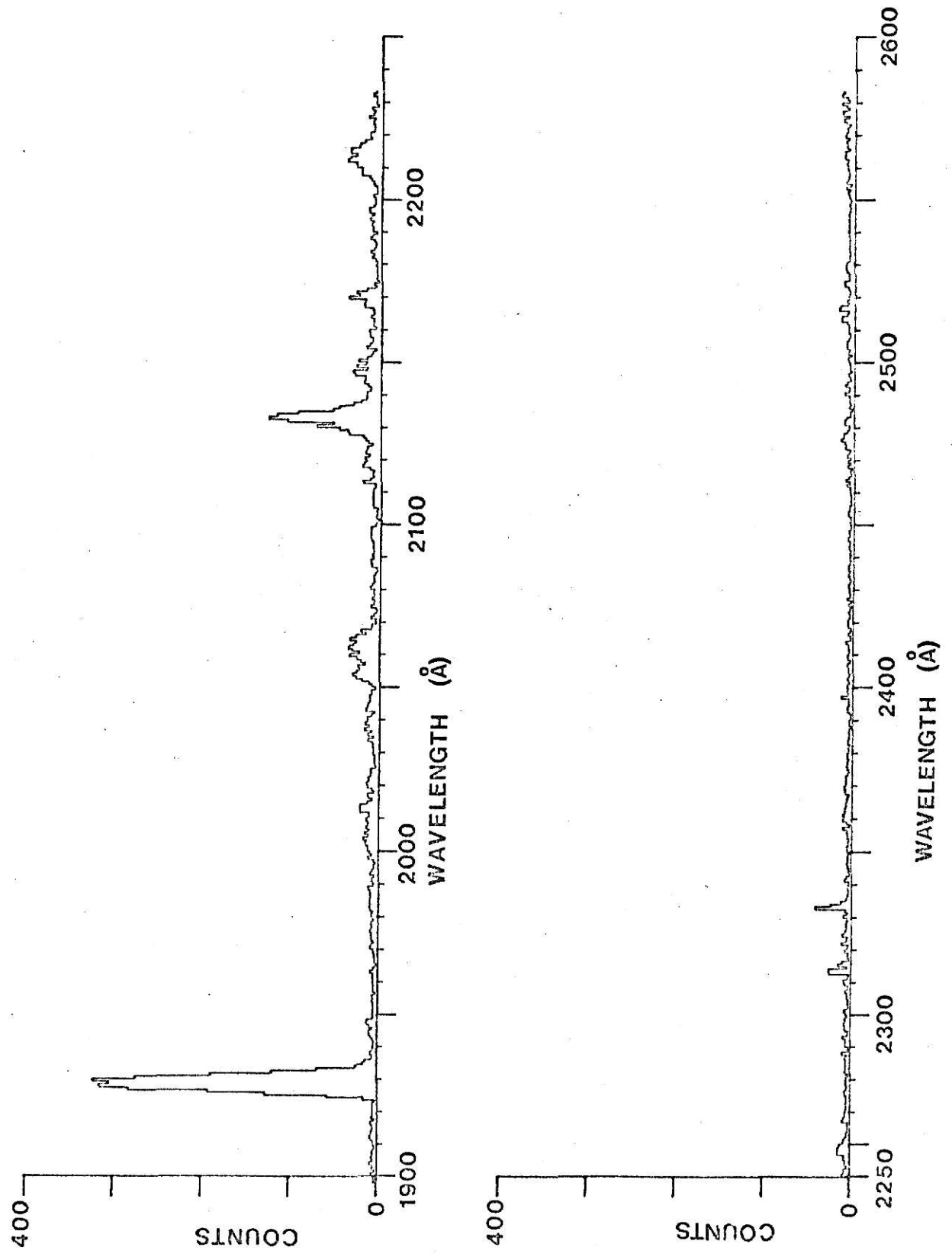


Fig. IV.--The spectrum: 1900 to 2580 A

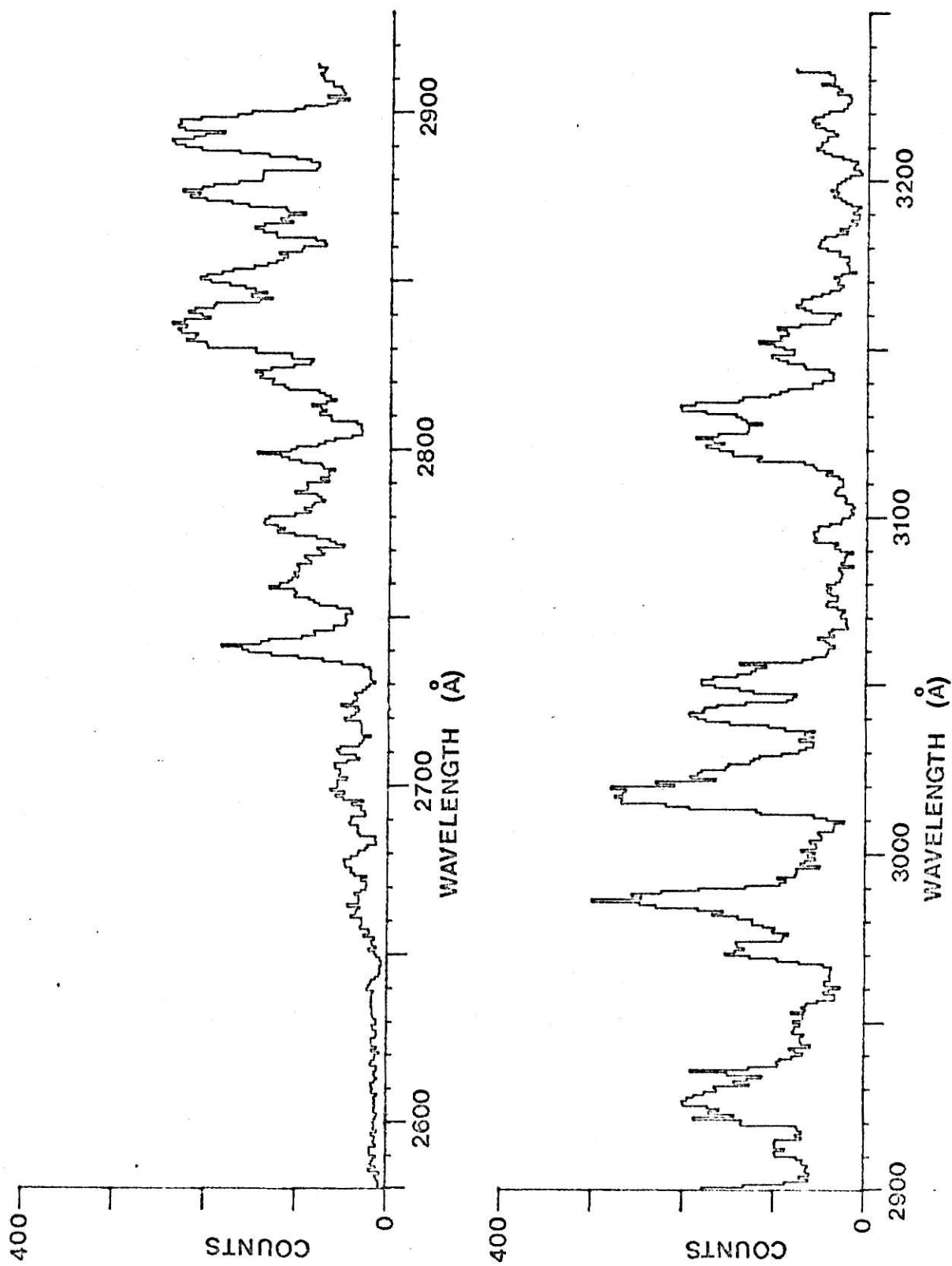


Fig. V.--The spectrum: 2580 to 3230 Å

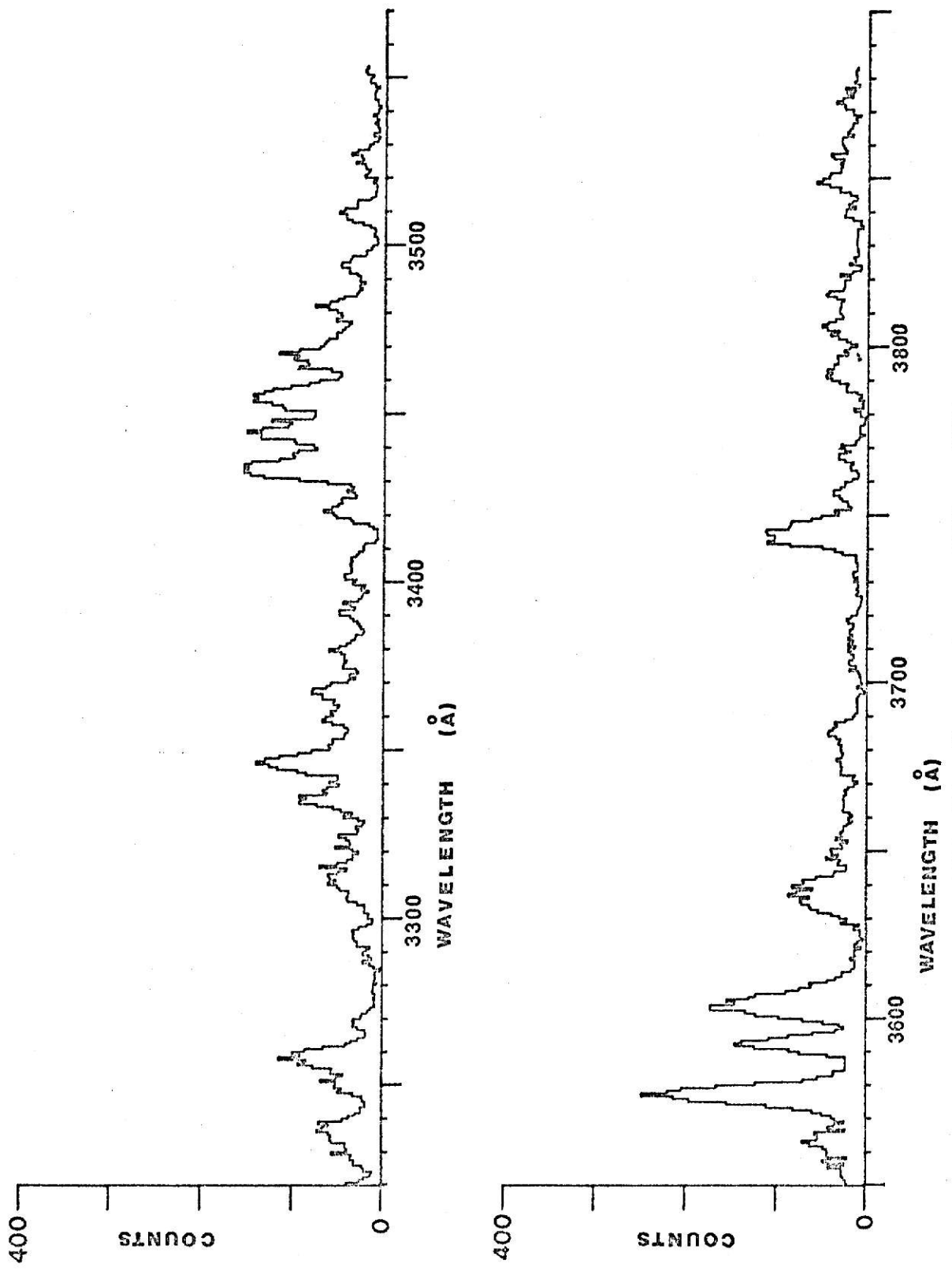


Fig. VI.--The spectrum: 3220 to 3880 A

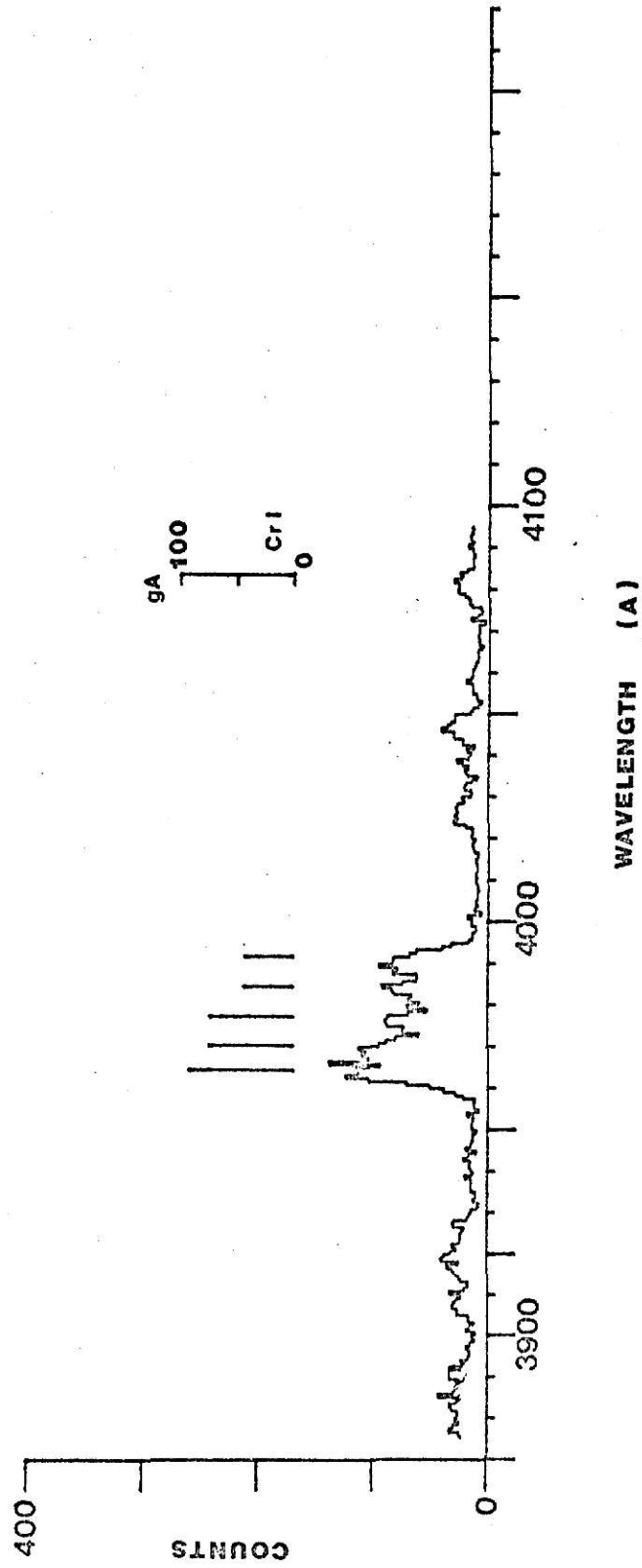


Fig. VII--The spectrum: 3870 to 4100 A

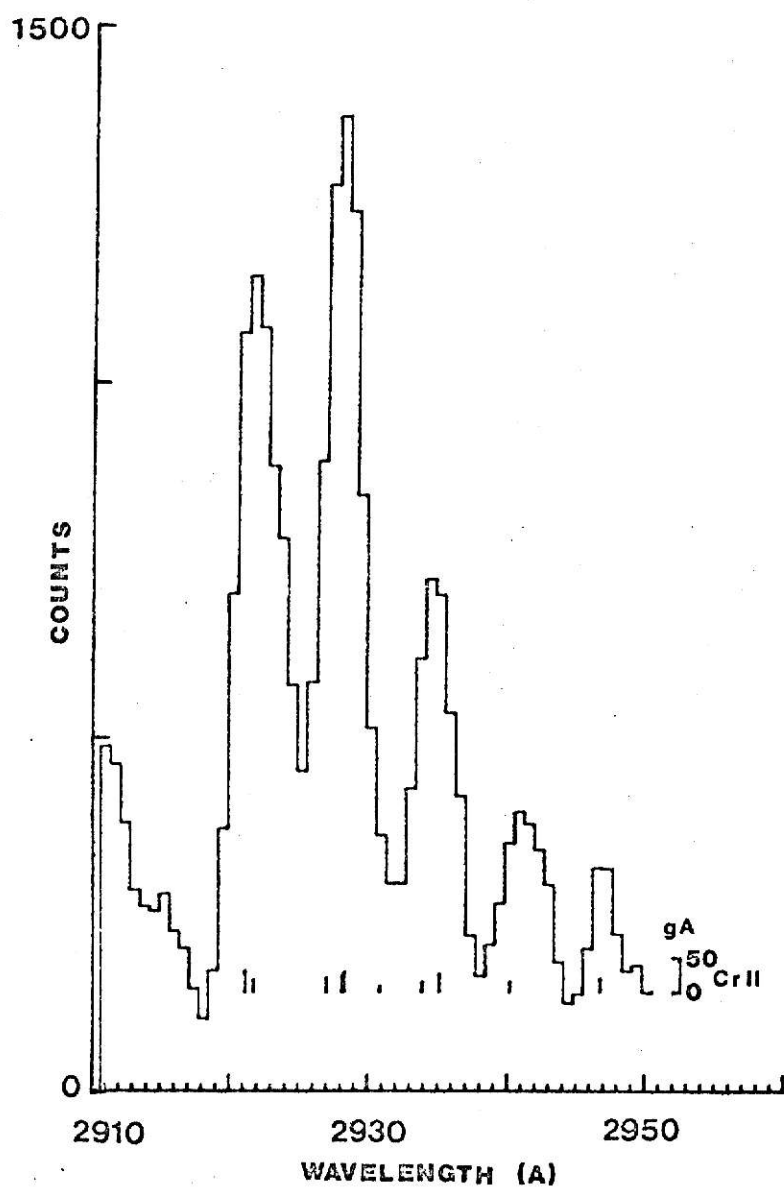


Fig. VIII.--The high resolution spectra of CrI and CrII:
2910 to 2950 Å

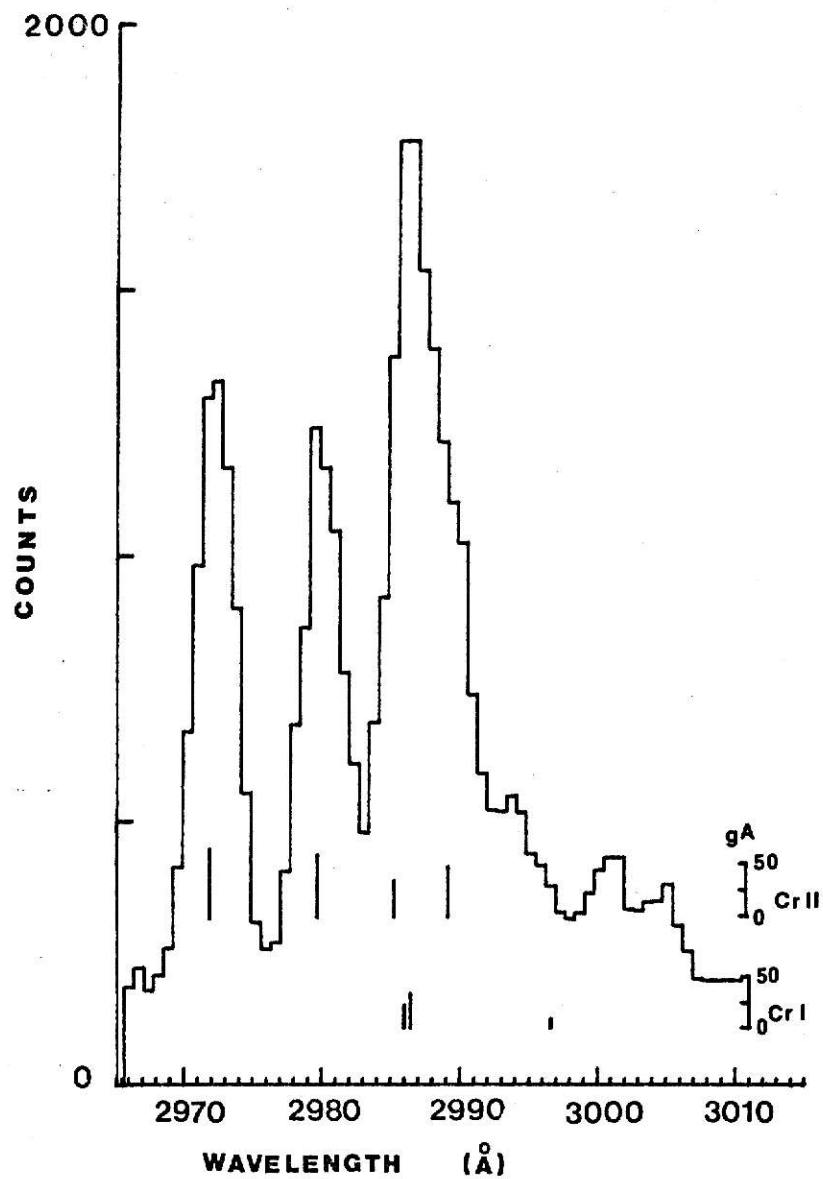


Fig. IX.--The high resolution spectra of CrI and CrII:
2970 to 3010 Å

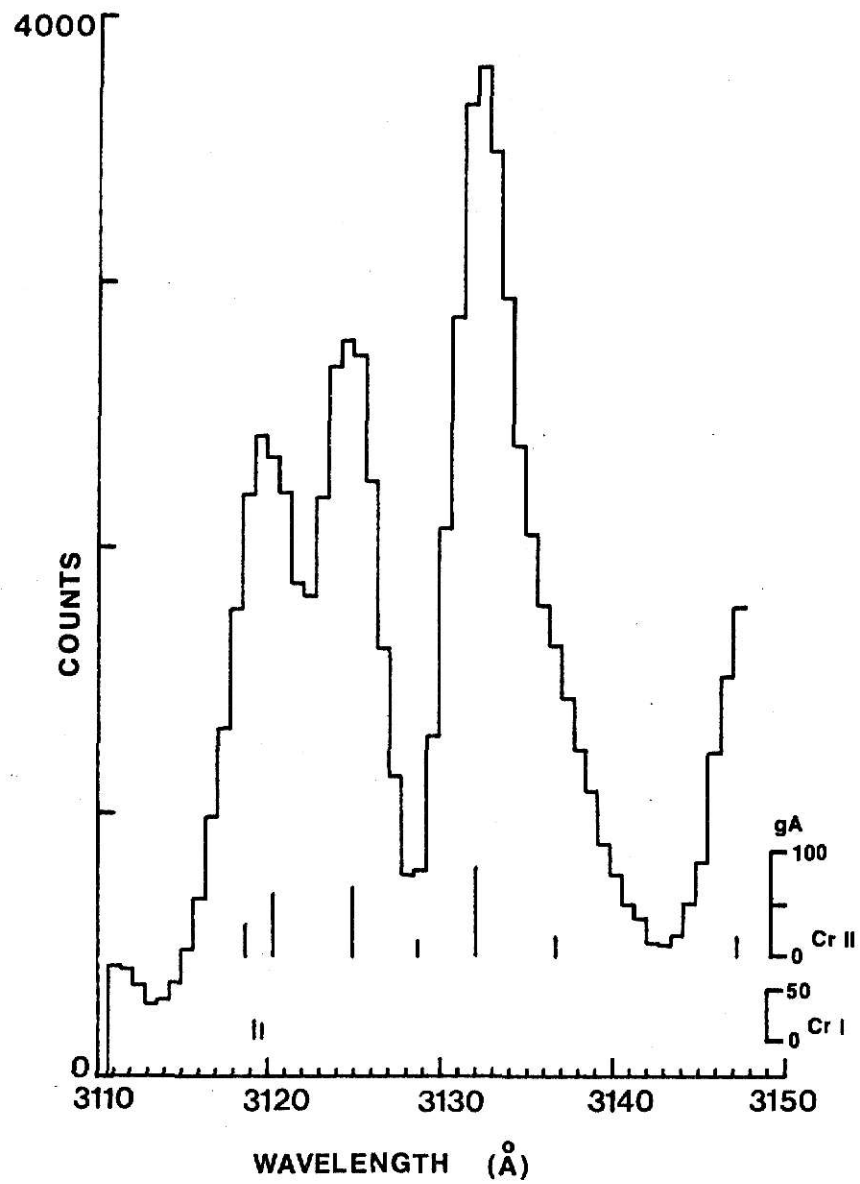


Fig. X.--The high resolution spectra of CrI and CrII;
3110 to 3150 Å

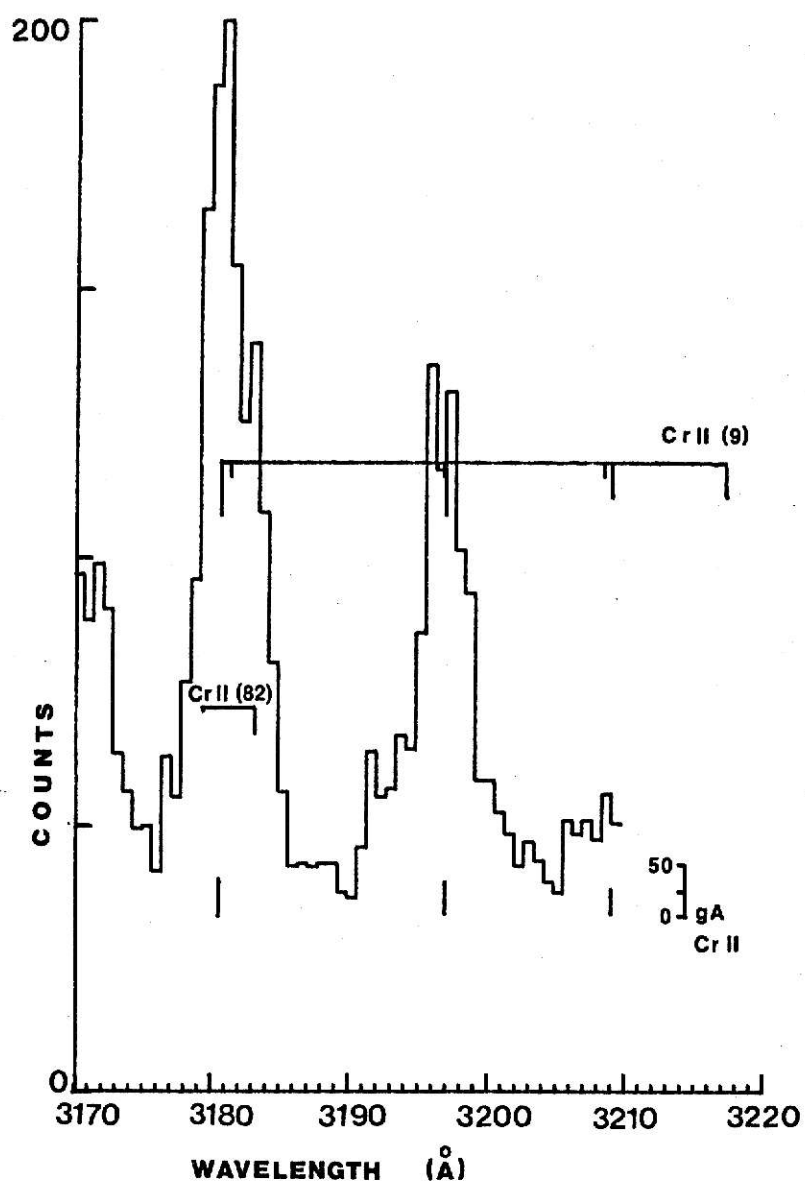


Fig. XI.--The high resolution spectra of CrI and CrII:
3170 to 3220 Å

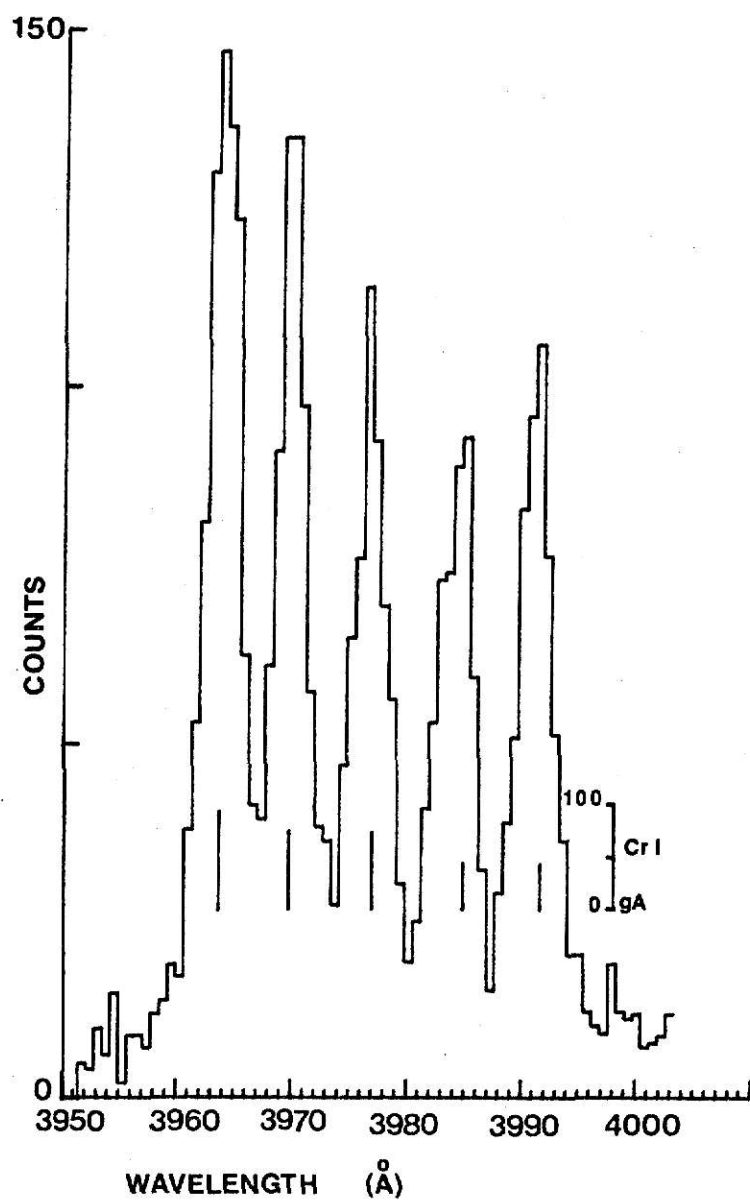


Fig. XII.--The high resolution spectra of CrI and CrII:
3950 to 4000 Å