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AUTOIONIZATION EFFECTS IN ULTRAVIOLET ABSORPTION SPECTRA OF HOT GASES

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Scientific Report #6

By

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A word of explanation on a deliberate obscurity in the title of the present review is appropriate, in the sense that "hot gases" implies nothing subtle, and has been inserted merely to denote that reference to autoionization effects observed in the absorption spectra of the inert gases at room temperature will not be mentioned. The beautiful examples found in these spectra, -- including prominent Beutler-Fano resonances observed by Madden and Codling by means of the synchrotron background continuum, -- are the subject of Dr. Madden's paper. The cases discussed here are drawn primarily from the absorption spectra of atomic gases which necessitate use of a furnace, shock-tube or something similar as an absorption vessel, placed in front of the slit of the spectrograph or monochromator. Though seemingly somewhat illogical, under the restriction of such heading, some initial remarks of effects observed in emission spectra seem worth brief mention.

Descriptions of autoionization effects, -- chiefly large natural breadths, line asymmetries and intensity peculiarities, -- have been accumulating since the early 1930's when Shenstone and Majorana independently suggested the explanations, in terms of Auger processes, of some singularities in optical line spectra. It will be recalled that Shenstone, who introduced the term "autoionization", was concerned with explaining Allen's measurements of large natural breadths of Cu I emission lines excited in a copper arc. Majorana gave an explanation in similar terms for what was thought, at that date, to be the absence of an expected line pair in each of the spectra: Zn I, Cd I, and Hg I. In each of these a group of four emission lines, superfluous to the normal series spectrum, had been recognized at the short wavelength end of the quartz ultraviolet, and ascribed to the combinations sp $^3P^\circ_{0,1,2}$ - p^2 $^3P_{0,1}$; transitions involving p^2 $^3P_2$ were thought to be missing. Majorana's explanation was that this level, -- as a result of mixing with sp^2 $^1D_2$, -- autoionized readily into the sE(d) $^1D_2$ continuum, with consequent extreme weakening of the combinations with sp $^3P_{1,2}$. Though Majorana's supposition that p^2 $^3P_2$ was subject to exceptional autoionization proved true, he was incorrect in thinking that the line pairs concerned were made unobservable, -- at least in the cases of Zn I and Pb I. In these spectra the reputedly missing lines must have been seen repeatedly for years by spectroscopists, until Dr. Rajaratnam in 1955 in my laboratory, obtained pictures of Zn and Cd arc spectra which cleared up the situation. In the case of Zn I, the line pair from p^2 $^3P_2$ had, -- understandably enough since autoionization had not been recognized, -- been mistakenly identified by Sawyer as due to p^2 $^1D_2$. In the case of Cd I, one line of the pair had been confused with a molecular band of Cd_2, which lies in the same neighborhood near 2200 Å, and the other member can easily be lost in the broadened wings of the nearby resonance line of Cd I at 2288 Å, unless special precautions are taken with regard to the light source used. Plate I shows the pp' groups of Zn I and Cd I mentioned. Photometric study of the Cd 2212 Å line, 5 $^3p^\circ_1$ - $5p^2$ $^3P_2$, (Garton and Rajaratnam, 1955), showed a good dispersion shape, which in terms of Fano's theory indicates a large value of the line profile index q. The case of Hg I still remains uncertain, as photographs of the spectral region between 1700 and 2100 Å, made by Rajaratnam at the time, showed nothing more than a mere suspicion of local blackening in the expected regions.* It

*Note added November 1965. The 6 $^3P_{1,2}$ - 6 p^2 $^3P_2$ transitions have now been confirmed by spectra obtained by R. C. M. Leaner and J. Morris at Imperial College, which contain roughly 50 new Hg I lines possessed of large autoionization widths.

seems in this case that autoionization does, indeed, make observation of the emission lines very difficult. The examples just quoted illustrate, however, that autoionization broadened lines are quite common in emission spectra. It is nevertheless true that the greater part of our systematic observational material on autoionization effects, at this date, associate with absorption spectra, -- particularly in the region of the vacuum ultraviolet, for the obvious reasons that strong lines obtained in absorption commonly start on the ground state of the atom concerned, and the ionization potentials of most elements are in excess of the six electron volt equivalent of 2000 Å. The emphasis on absorption spectroscopy stems partly from the fine pioneering experimental work of H. Beutler* in the 1930's. In this work, which produced many examples of autoionization broadening and asymmetries, Beutler emphasized the virtues of the absorption method to an extent which may have discouraged experiments in search of such effects in ultraviolet emission spectra. As shown recently by Tilford and Wilkinson (1964), in their work on O III, emission lines from strongly autoionizing upper states can be quite readily observed, if a high enough electron density is maintained in a suitable light source.

Returning now to the main theme of absorption spectra, the cases studied by Beutler were those of elements producible as monatomic gases with little difficulty, viz., the spectra of the inert gases Ar through Xe, Zn, Cd, Hg, Tl, Rb, Cs, and two lines of K. Most of Beutler's spectra were obtained on the background of the Hopfield He_2 continuum, stretching from 600 to 900 Å, and he invariably worked with the modest dispersion provided by a one-metre grating.

*A bibliography of Beutler's papers has been given by Boyce (1941).

Starting in the early 1950's with rather primitive apparatus, which has been improved and supplemented progressively during the last ten years, a number of absorption spectra of atomic gases, rather more difficult to handle, have been studied at Imperial College. This work has been done chiefly with King-type evacuable furnaces serving as absorption vessels. During the last few years a fruitful collaboration has developed with the group of Professor L. Goldberg at Harvard College Observatory where we have been able to supplement the furnace techniques very effectively by using shock-tubes to produce the absorbing column of atomic gas of interest. While furnace methods have proved very successful for development of long series of absorption lines, and can be used to temperatures of about 2800°C, the spectra are usually restricted to absorption lines which start on the ground term, -- or at most deep-lying excited states, -- of the neutral species. Drs. Parkinson and Reeves at Harvard have developed powerful combinations of shock-tubes and vacuum spectrographs, which enable us to record absorption lines with lower levels lying several thousands of wave-numbers above the ground state of an atom, as well as absorption lines of ionic species in some cases. Fig. 1 provides a block diagram of the arrangement used for photographic recording of an absorption spectrum, and simultaneous determination of the reversal temperature of the shock-heated plasma. The remainder of this review will mainly concern effects found in absorption spectra obtained with the furnace, or shock-tube, technique, and occasionally from combinations of the two. The spectra to be mentioned contain examples of intensity or line profile peculiarity which, previously extremely puzzling, now seem containable within the framework of recent theory (Fano, 1961, Fano and Cooper, 1965).

The absorption spectra of the alkaline earths, at the short-wavelength end of the quartz-ultraviolet and in the Schumann region, obtained by Dr. K. Codling at Imperial College around 1960, contain many fascinating details. The spectrum of Ca I reproduced in Plate 2 shows the region of the converging principal series, and six series of two-electron transitions, classified as $4\ ^1S_0$ - 3dnp $^{1,3}P_1^o$, $^3D_1^o$ and 3dnf $^{1,3}P_1^o$, $^3D_1^o$ converging on the 3d ^2D metastable levels of the ion. In this case, we have the complications of mutual mixing of series levels converging on the two levels of the ion, and with the 4sE(l) continua; an extra complication arises from the presence of the 5s4p $^1P_1^o$. The extreme effects of autoionization, on breadths and intensities of the lower members of the doubly excited series are obvious, and closer study of the spectra shows that additional perturbations are prominent amongst the higher lines (Garton and Codling, 1965). Working at rather lower dispersion than was used for the spectrum of Plate 2, Ditchburn and Hudson (1960) have measured the integrated oscillator strengths of the stronger lines of the doubly-excited series of Ca I, as well as the course of the absorption cross-section in the underlying continua. The latter proves to be very small, which probably explains the general absence of pronounced Beutler-Fano profiles from the Ca I spectrum, on the interpretation that the lines concerned have large values of q.

The corresponding spectrum of Ba I (Garton and Codling, 1960) contains an example of the Beutler-Fano effect which, until recently was unique. Here, the transition $6\ ^1S_0$ - 5d8p $^1P_1^o$ occurs as a very broad feature practically coincident with the first ionization potential of Ba I, the maximum of the resonance profile falling just within the continuum, and the "transmission window" amongst the converging lines of the

6snp $^1P_1^\circ$ series. These effects can be clearly seen in the densitometer trace of this region of the Ba I furnace absorption spectrum, made at Harvard College Observatory, and reproduced in Fig. 2; some additional perturbations in the intensities of the converging series lines due to the presence of 5d8p $^1P_1^\circ$ and $^3P_1^\circ$ are indicated by the extension of the broken line to the right.

The first example of what may be suitably named "forced autoionization" was observed in the absorption spectrum of Ba obtained with the Harvard shock-tube apparatus (Garton et. al., 1962). Plate 3 contains a comparison of the absorption spectra obtained (a) by means of a furnace and (b) with a shock-tube. Whereas in (a) the lines 2433 and 2445 Å are quite sharp, the upper levels lying below the ionization limit, in (b) these lines have become distinctly broadened, -- particularly the 2433 Å line, -- and accompanying this the higher members of the converging principal series, -- which are prominent in (a), -- have disappeared. A reasonable interpretation seems that the microfields of the charged particles in the shock-heated plasma depress the ionization potential sufficiently to drown out the higher series members, and simultaneously permit the doubly-excited 5d8p $^3P_1^\circ$ and $^3D_1^\circ$ levels to autoionize.

Each spectrum of the group Al I, Ga I, In I, Tl I contains interesting effects of configuration mixing, including autoionization, in the levels of the sp^2-configuration. This configuration gives ^4P, ^2D, ^2P and ^2S. Starting with Al, the ^4P is well recognized and lies deep, and the ^2D appears from the quantum defect of the $3s^23d$ ^2D-series to lie deep also.* In the Schumann region easily recognized

*This is confirmed by unpublished theoretical results of Dr. R. J. S. Crossley, Harvard College Observatory, who finds the level hitherto listed as $3s^2$ 3d $^1D_2$ to be of predominently $3s3p^2$ $^1D_2$ character.

combinations of ^2P and ^2S with the ground term 3 ^2P occur, the 3 $^2P^o_{1/2,3/2}$ – $3s3p^2$ $^2S_{1/2}$ pair occuring also as prominent Fraunhofer lines in the solar spectrum near 1932 Å. As will be mentioned later, the f-values of the components of this important doublet have been measured by means of the Harvard shock-tube apparatus.

In Ga I (Garton, 1952) we again find a group of six lines in the 1500 to 1650 Å region, which seem to be similarly ascribable to sp^2 ^2P and ^2S. Also, in this spectrum the ^4P term lies deep, but the location of ^2D is still uncertain. The spectrum of In I, in the Schumann ultraviolet, contains decided peculiarities. The course of the quantum defect of the s^2nd ^2D-series (Garton and Codling, 1961), together with the fact that the photoionization cross-section does not set in with a large value at the series limit but takes a blunt maximum about 2700 cm^{-1} from it, suggests that the sp^2 ^2D term is buried in the continuum by very extreme autoionization. If we follow-up this interpretation with the inquiry as to the whereabouts of the Beutler-Fano minimum, we notice a curious intensity perturbation which, until recently, appeared isolated and inexplicable. In the long absorption series $5s^2$ 5p $^2P^o_{1/2,3/2}$ – $5s^2nd$ $^2D_{3/2,5/2}$ a marked local weakening between n = 8 and 10 is found, the n = 9 lines being completely absent, even if the density of the absorbing vapour is made quite high. This effect seems tentatively explicable as a Fano "window", which lies displaced by about 5100 cm^{-1} from the corresponding maximum of the profile. A similar situation probably exists in Ga I, though careful long series measurements of furnace spectra, at Imperial College by Dr. Reeves (unpublished), do not show so pronounced a fall in the quantum defect near the limit, as that found in In I; nevertheless there occurs an intensity anomaly in the $4s^2nd\ ^2D$ series similar to that in In I.*

The In I spectrum contains other curiosities, the most striking being that only five lines are observed, instead of six, ascribable to $5s^25p\ ^2P^\circ - 5s5p^2\ ^2S,\ ^2P$. In classifying these lines some years ago (Garton, 1954) the author gave the explanation that, because of approach to J-J coupling conditions, the member $5\ ^2P^\circ_{1/2} - 5s5p^2\ ^2S_{1/2}$ could be missing from this spectrum. In a preliminary announcement of the results of recent f-value measurements on the In I $5s5p^2$ line group, Dr. G. V. Marr of Reading showed that, -- if this interpretation were correct, -- the line attributed by the present author to $5\ ^2P^\circ_{3/2}$ - $5s5p^2\ ^2S_{1/2}$ would have the improbably high f-value of 1.3; a much more reasonable f-value (0.08) is obtained if $5\ ^2P^\circ_{1/2}$ is exchanged for $5\ ^2P^\circ_{3/2}$ as the lower level. Also some recent unpublished calculations of Dr. J. M. Wilson of Imperial College suggest that $sp^2\ ^2S$ ought to lie much higher in In I, but in this event we are faced with the position that there is no sign of an absorption line pair suitably placed at shorter wavelengths.**

Results of some recent experiments on the last member of this group of elements, Tl, illustrate the benefits of

*Note added November 1965. The proposal that in both Ga I and In I, $sp^2\ ^2D$ lies buried in the $s^2E(d)\ ^2D$ continuum is now supported by calculations of J. M. Wilson at Imperial College, based on diagonalization of the complete energy matrix and least-squares adjustment of the Slater parameters.

**Note added November 1965. Further theoretical calculations by Wilson and experimental work at Harvard have cleared-up the $sp^2\ ^2S_{1/2}$ problem in In I. The "missing" 6th line of the group $5\ ^2P^\circ_{1/2,3/2} - 5s5p^2\ ^2S_{1/2},\ ^2P_{1/2,3/2}$ has now been found at 1854.7 Å in shock-tube spectra and percentage compositions of the three J = 1/2 levels now known have been calculated in terms of ^2S, ^2P, ^4P contributions.

combining furnace and shock-tube methods. In Tl I the $6s6p^2\ {}^4P$ term is unusual in that its component levels bridge the $6\ {}^1S_0$ limit of the series spectrum, -- in fact, ${}^4P_{5/2}$ is still of very uncertain location; ${}^4P_{1/2}$ lies below the limit, ${}^4P_{3/2}$ combines with the ground level $6\ {}^2P_{1/2}$ to give a classic Beutler-Fano profile (Garton et. al., 1965), which satisfactorily conforms with the expected autoionization into a single available continuum $[s^2E(d)\ {}^2D_{3/2}]$. The $6\ {}^2P^o_{1/2}$ - $6s6p^2\ {}^4P_{3/2}$ transition was found by Beutler himself, together with the lines involving $6s6p^2\ {}^2D_{3/2}$ and ${}^2P_{1/2}$. The oscillator strengths of these three lines have been measured by Marr (1954). In furnace experiments as usually performed, it is not possible to get the temperature high enough, -- before boiling the material, -- to populate appreciably the upper level of the Tl I ground term, viz. $6\ {}^2P^o_{3/2}$ which lies nearly 7800 cm^{-1} above ${}^2P_{1/2}$. Consequently, furnace spectra reveal fewer of the sp^2 combinations. In a shock-tube plasma the $6\ {}^2P^o_{3/2}$ metastable level can be well populated, and in some very new absorption spectra, taken at Harvard with the shock-tube apparatus and a 1-metre vacuum spectrograph, we have found three new lines, one of which confirms Beutler's $6s6p^2\ {}^2P_{1/2}$ level, the other line pair being combinations with $6s6p^2\ {}^2S$ which, in Tl I has reversed its position with respect to $sp^2\ {}^2P$, as compared with the situation in Al I. We are still left with some problems, for example the location of $6s6p^2\ {}^2P_{3/2}$ and ${}^2D_{5/2}$. Possibly, details of coupling conditions, when examined sufficiently carefully, may explain these remaining curiosities in In I and Tl I absorption spectra.

Absorption spectra of the group IV B elements Si, Ge, Sn, and Pb, have been photographed during the last few years by Dr. J. M. Wilson at Imperial College, with the King furnace, and almost all lines have been assigned. In Si I, though many new series lines have been found, we do not regard the results as satisfactory because the small intervals of the 3 ^3P ground term and between the ^2P levels, which form the series limits, necessitate that this spectrum be re-examined at higher dispersion than at present available at Imperial College. The other three spectra have been almost completely analysed.

In this group of elements, the ground state is $s^2p^2\ ^3P_{0,1,2}$. We obtain an absorption spectrum in the Schumann region, which in each of the cases, Ge and Sn, is of rather complex appearance due to the overlapping of series from the three levels of the ground state and convergence to the double $s^2p\ ^2P$ limit, with some additional confusion due to lines involving other odd configurations, especially sp^3. Moreover, the use of LS-symbolism becomes of doubtful validity, particularly in Pb I where the level spacings approximate more closely to those of J-J coupling. The spectrum of Sn I contains the second recorded example of a Beutler-Fano resonance overlying a series limit, as illustrated above in the spectrum of Ba I.

In Pb I, the ground term intervals are large enough for the furnace absorption spectrum to become much simpler, there being little sign of absorption lines starting of 6 $^3P_1$ and 6 $^3P_2$; the $^2P^o_{1/2,3/2}$ splitting in the ion is large also, so that the region between the two limits of the absorption spectrum extends from 1353 to 1672 Å. Plate 4 reproduces the absorption spectrum photographed with a 3-metre vacuum spectrograph (Garton and Wilson, 1965); the picture contains immediately striking examples of Beutler-Fano transmission windows, all of which can be readily classified. The expanded view of the region of the $^2P^o_{3/2}$ convergence limit, shown in the lower part of the Plate, reveals that the resonances present correspond very nearly to q = 0. This work on Pb I has, just recently, been supplemented by absorption spectra obtained with the Harvard shock-tube apparatus, which differ from those obtained with the furnace in containing many lines which start on the $^3P_1$ and $^3P_2$ ground levels. This work is so recent that at the date of the Goddard Conference, the spectra have not been analysed. The analyses of the Ge I and Sn I spectra will soon be published.

Mention has been made above of the occurence of the Al I 3 $^2P_{1/2,3/2}$ - $3s3p^2$ ^2S line pair in the solar spectrum, the profiles being fairly well resolved even at the low dispersions so far used with rocket-borne equipment in the 1900 Å region. These lines, until quite recently, were the only ones, showing strong autoionization broadening, recognized in the solar spectrum, and it was plainly of likely usefulness to solar physics that we obtain a measurement of f-values, -- or rather of $\frac{df}{d\nu}$ over their profiles. This was done last year (Garton et. al., 1964), by photometric reduction of the profiles of the absorption lines, photographed by means of the shock-tube and the "powdered-solid technique" described by Nicholls et. al. (1963). In order to determine the particle density of neutral Al in the shock plasma, we used a mixture, of known ratio, of Ca and Al salts, and utilized Ditchburn and Hudson's (1960) value for the integrated $\left(\frac{df}{d\nu}\right)$ of the Ca 1885 Å line, which has a very large autoionization breadth (see Plate 5). The temperature of the plasma, which was needed for application of the Boltzmann and Saha equations, was obtained as usual, by the line reversal method (Parkinson and Reeves, 1964). One of the major uncertainties in this determination of Al I f-values concerns the correction which has to be applied to the ideal ionization potential, on insertion in the Saha equation. Though this problem is usually distinquished from the question of how to terminate the series of terms representing a partition function for an atomic species present in a high-temperature plasma, the two are at least closely related and the present situation in theory and experiment needs improvement.

This survey has included reference to all the published work, known to the writer, concerning autoionization effects in atomic absorption spectra, which have been studied in any detail, except those found in the spectra of the inert gases dealt with by Dr. Madden. So far as concerns line profiles, half-widths and f-values the existing data are very scanty as yet. In fact, if we summarize these we have f-values and breadths of only three Tl I lines (Marr, 1954), of the stronger $4\ ^1S_0 - 3dnp\ ^{1,3}P_1^o$ transitions of Ca I (Ditchburn and Hudson, 1960), of the Al I $3\ ^2P^o - 3s3p^2\ ^2S$ pair (Garton et. al., 1964), the breadth of the Cd I $5p^2\ ^3P_2$ level (Garton and Rajaratnam, 1955), and the early work of Allen (1931) on Cu I lines. In supplement to these published quantitative results, Dr. Hudson communicates that he has reliable unpublished measurements on the profiles of the double-electron transitions in Sr I and Ba I, a preliminary report has been made by Dr. Marr of Reading University concerning the sp^2 combinations in Ga I and In I, and an unpublished study, at Harvard, of the profile of the Tl I $6\ ^2P_{1/2}^o - 6s6p^2\ ^4P_{3/2}$ line may be included. It seems certain that the papers of Fano and Fano and Cooper cited, along with other theoretical contributions mentioned at the Goddard Meeting, will greatly stimulate the efforts of experimental spectroscopists towards observation, classification and accurate photometric measurement of autoionization broadened lines in many more atomic spectra and over a range extending farther into the vacuum ultraviolet.

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It is both fitting and pleasant to record here a personal acknowledgement of the beneficial outcome of repeated visits to Harvard College Observatory since 1961, under arrangements made by Professor L. Goldberg, leading to establishment of a close collaborative effort in laboratory astrophysics between the two institutions, and in particular to successful joint experimental work with the shock-tube apparatus assembled at Harvard College Observatory by Drs. W. H. Parkinson and E. M. Reeves.

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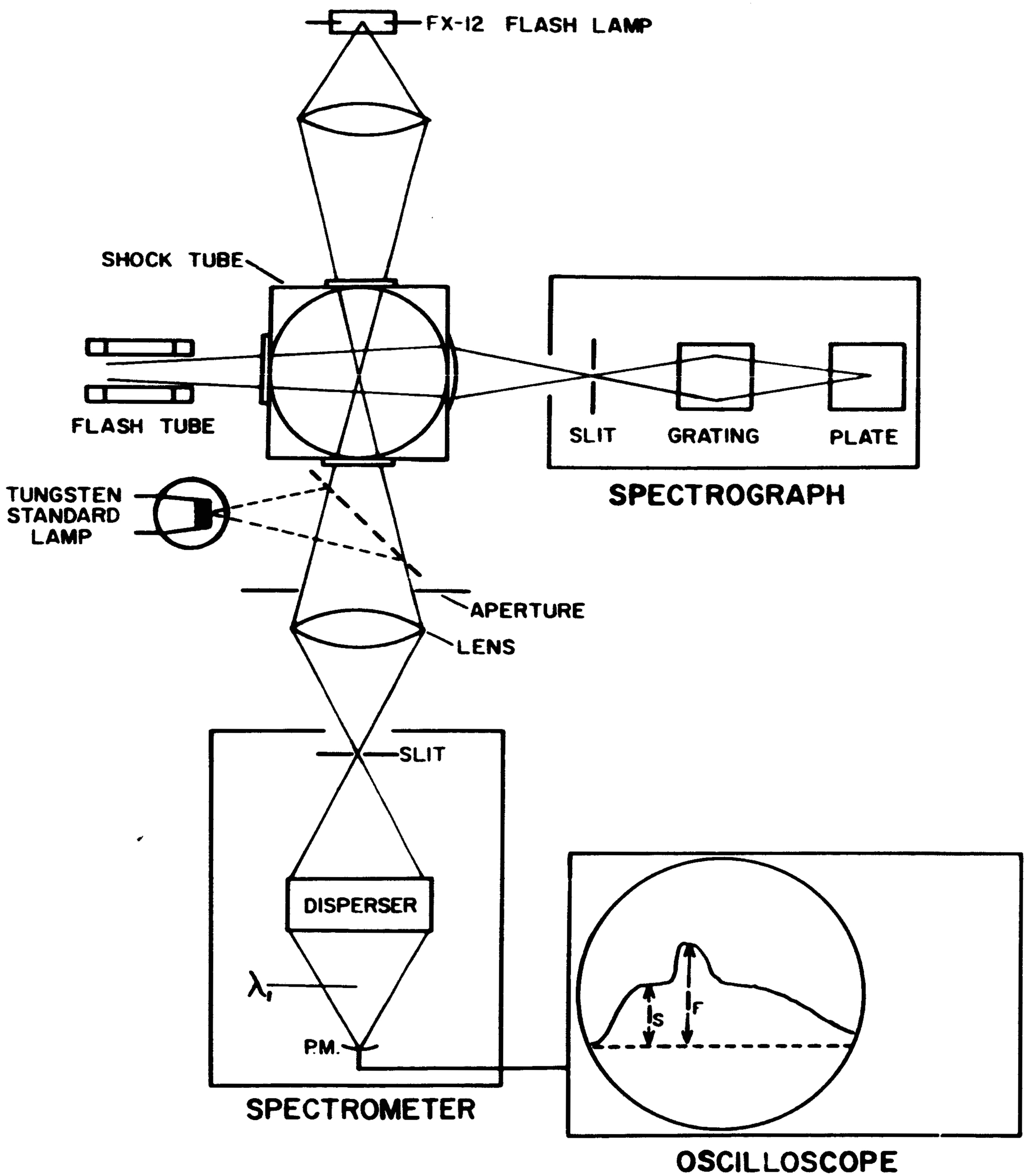


Figure 1. Block diagram of apparatus for photographic recording of absorption spectrum and measurement of reversal temperature of shock-heated plasma.

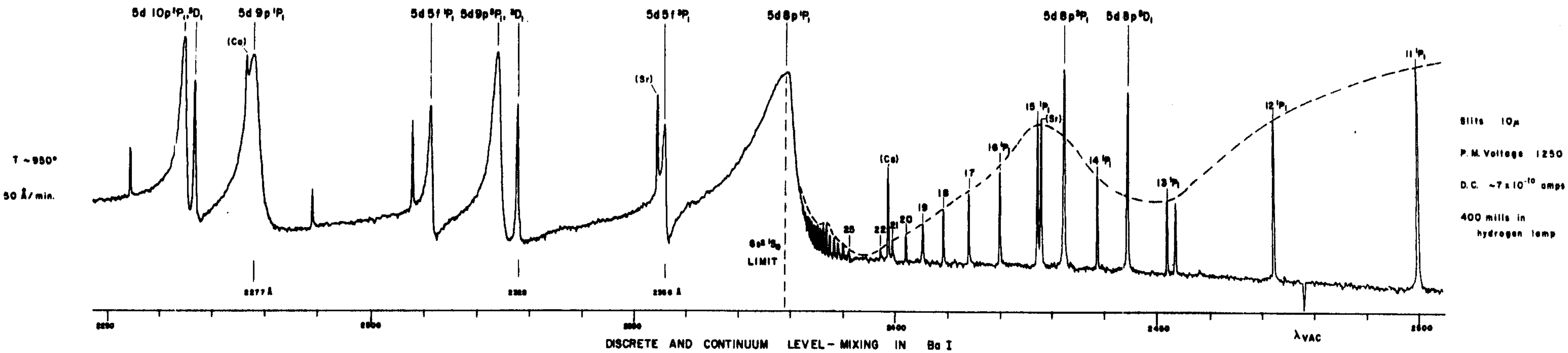


Figure 2. Densitometer tracing of Ba I absorption spectrum, showing autoionization broadening of the two-electron transitions, and Beutler-Fano resonance effects near the first ionization limit.

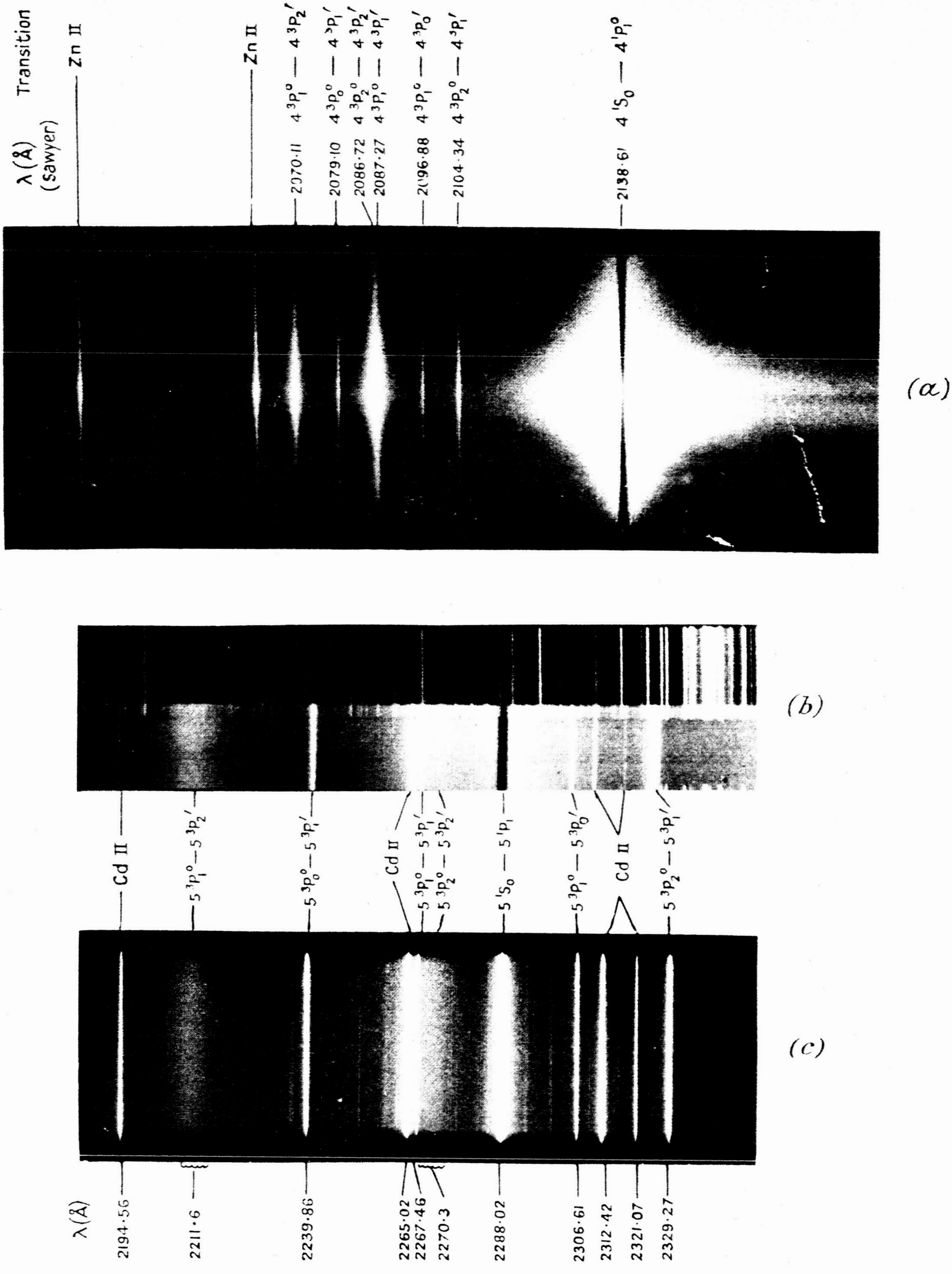


Plate 1. The PP' -groups of Zn I and Cd I, showing autoionization broadening of $p^2\ ^3P_2$.

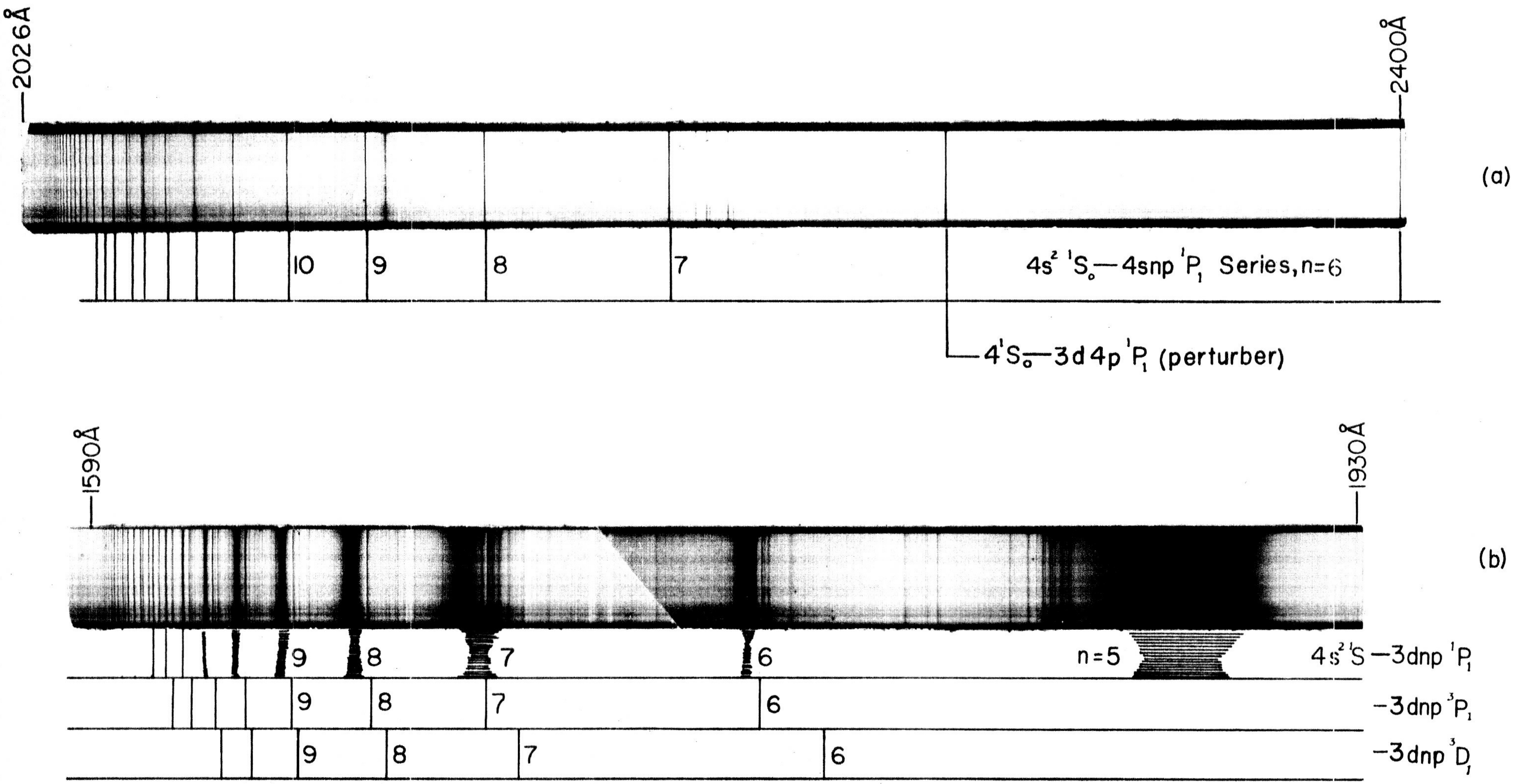


Plate 2. Ca absorption spectrum: (a) converging principal series and perturber, (b) series due to simultaneous excitation of two electrons.

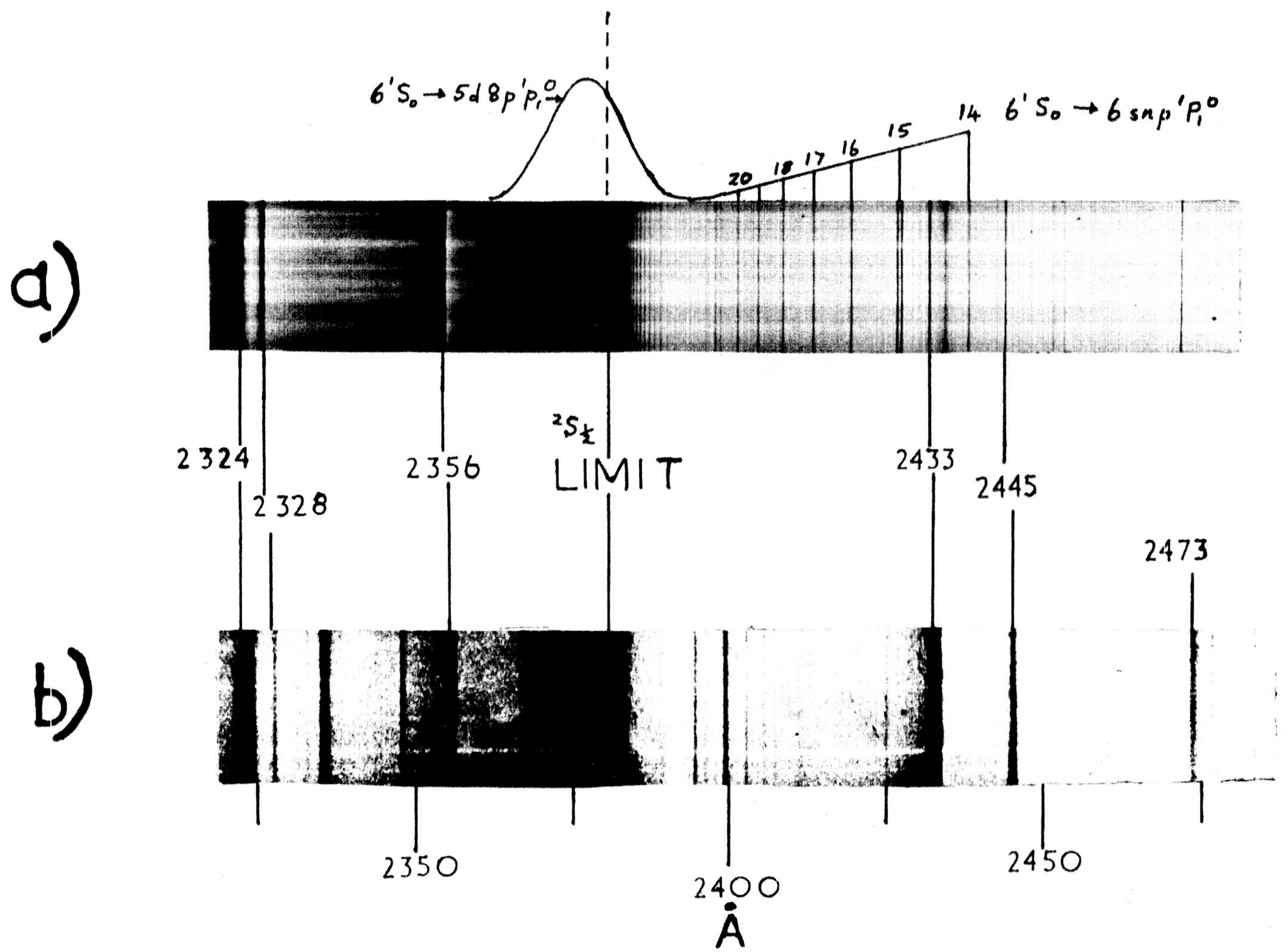


Plate 3. Forced autoionization in the Ba I spectrum: (a) furnace absorption spectrum, (b) shock-tube absorption spectrum.

PbI ABSORPTION SPECTRUM SHOWING TRANSMISSION WINDOWS

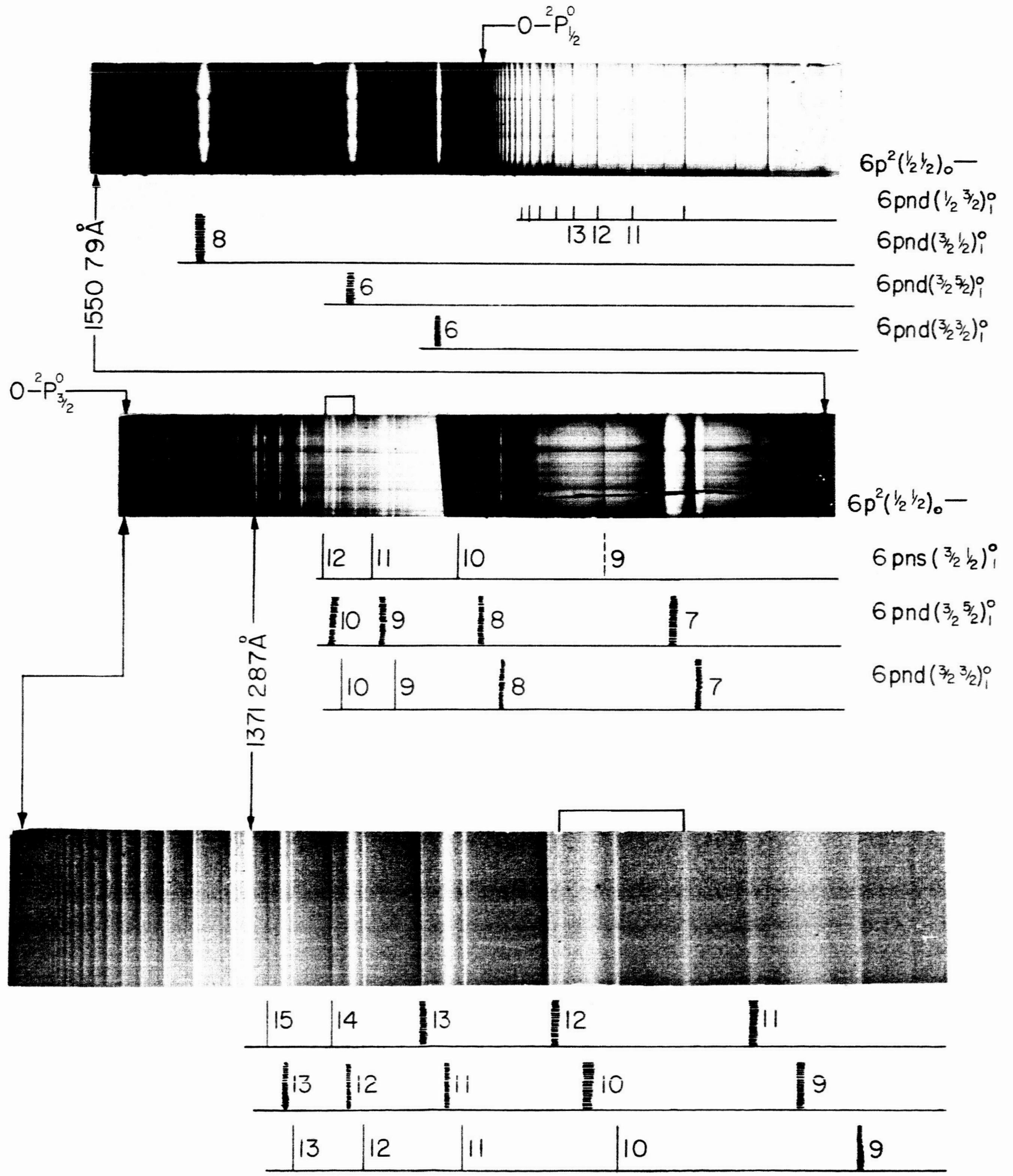
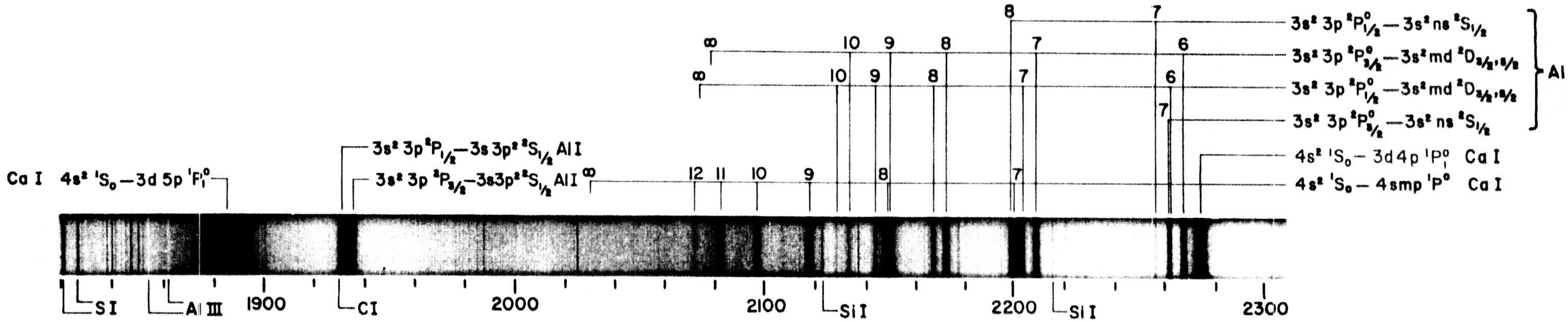


Plate 4. Pb I absorption spectrum, showing series, including Beutler-Fano resonances, converging on the ground-doublet of Pb II.



ABSORPTION SPECTRUM OF SHOCK-HEATED $CaS + Al_2S_3$

Plate 5. Absorption spectrum of shock-heated plasma containing Al and Ca in known ratio.

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13. ABSTRACT

Autoionization effects present in the ultraviolet absorption spectra of a number of atomic vapours (e.g. of the alkaline earths Ca-Ba, the Group III A elements Al-Tl and those of Group IV B, Si-Pb), are described, and some of the detail is interpreted qualitatively in the framework of the theory of line-shapes ("Beutler-Fano profiles"), recently put forward by Fano.

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