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Experimental Cesium Line Shapes⁺

LEWIS AGNEW AND C. SUMMERS
University of California
Los Alamos Scientific Laboratory
Los Alamos, New Mexico

ABSTRACT

The widths of some of the spectral lines emitted from a dense plasma are sensitive to the ion density but insensitive to the electron temperature. In partially-ionized cesium plasmas above 10^{13} ions-cm⁻³ these plasma-broadened lines are easily measured and can be used as a simple diagnostic method for the determination of ion density, once the width versus ion density relationship is established either theoretically or experimentally. Here we present measured line widths and compare results with theoretical widths. The profiles of various lines of the sharp, diffuse, and fundamental series lines of cesium were measured at known ion densities with a scanning monochromator on a hot-cathode parallel-geometry cesium diode. For each case, the ion density and the electron temperature were determined by measuring the intensity of the radiative recombination continuum. Line intensities gave independent measurements of temperature that were in agreement with electron temperatures, showing the existence of local thermodynamic equilibrium and providing a confirmation of ion density through the application of the Saha equation. Detailed results of line width measurements

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are presented for a range of plasma density from 3.2×10^{13} to 1.6×10^{15} ions-cm $^{-3}$ and for temperatures between 2000° and 6000°K. The results, valid in the vicinity of $N_e = 10^{15}$ ions-cm $^{-3}$, are summarized in the following table:

<u>Series</u>	<u>Transition</u>	<u>Electron temp. (°K)</u>	<u>Line width w(A)</u>
sharp	6P-nS, $9 \leq n \leq 11$	6000	$w = 2^{(n-9)} \times 0.30 N_e \times 10^{-15}$
diffuse	6P-nD, $7 \leq n \leq 10$	6000	$w = 2^{(n-7)} \times 0.35 N_e \times 10^{-15}$
fundamental	5D-nF, $5 \leq n \leq 8$	$2000 \leq T_e \leq 10,000$	$w = 2^{(n-5)} \times 1.5 N_e \times 10^{-15}$

In the table w is the full line width at half maximum and has an accuracy of 20%. The widths of the fundamental series lines are essentially independent of electron temperature, but temperature independence can not be claimed for the sharp and diffuse series lines.

I. Introduction

Spectral lines emitted from dense plasmas are broadened by effects due to the ions and electrons. Over a wide range of plasma conditions, this plasma broadening far exceeds other (i.e. doppler, natural, etc.) contributions to the broadening. For some, but not all, isolated lines the width is nearly proportional to ion density and nearly independent of the temperature. Thus line width measurements can be used to obtain information about the ion density.

Certain lines emitted from cesium plasmas at ion densities above 10^{13} cm^{-3} are broad enough to be easily measured. Their widths are simply related to ion density regardless of thermal equilibrium considerations. In this paper we present measured cesium line widths over a range in ion density from $3.2 \times 10^{13} \text{ cm}^{-3}$ to $1.6 \times 10^{15} \text{ cm}^{-3}$ at temperatures between 2000°K and 6000°K , and we describe some simple apparatus for making observations. We also present a comparison of measured results with theoretical cesium line broadening calculated by Stone¹ and by Griem². Finally we show how to test for reabsorption and how to correct for this effect when lines are only slightly reabsorbed.

II. Apparatus

Emission lines as broad as one to twenty angstroms can be measured readily with apparatus that might ordinarily be termed spectroscopically crude. Conventional spectrographs of quite modest resolution will suffice although they must be supplemented with some densitometry apparatus. A feature of the conventional spectrograph is the ability to make an exposure of many lines

simultaneously. By choice of film speed and optical arrangement exposure times can be cut short, obviating the need for careful control of the plasma over long periods. The need for a study of many lines, however, is unlikely except when reabsorption effects are difficult to estimate from other considerations. An undesirable feature is the non-linear response of film and hence the need for calibration.

A scanning monochromator equipped with photomultiplier attachment can also be used to measure line shapes. For example, the data reported below in Section III were taken using a Jarrell-Ash 50 cm Ebert monochromator with a 15,000 line-per-inch grating having a response from 2000 Å to 16,000 Å. This wide spectral response was useful and the poor resolution (32 Å/mm reciprocal linear dispersion) was partially compensated by having all of the visible spectrum available in second order. Photomultiplier signals from a tube mounted at the exit slit of the monochromator were amplified and displayed on a recording potentiometer. This system was linear with intensity of light input, thus the profiles could be interpreted quickly and easily. A linear response is not difficult to achieve because the change in photomultiplier spectral response is insignificant over the wavelength interval concerned in the broadening of a single line, and because saturation effects, including both local fatigue of the photo-cathode and overdriving the anode current, are easy to avoid. An undesirable feature of this system is the need to maintain uniform conditions within the plasma over the length of time necessary to complete the scan, usually a period of several minutes. Fluctuations or drifts

in temperature and pressure may cause distortion of line profiles.

Cesium vapor is known to attack both quartz and pyrex, but sapphire windows are not affected at temperatures at least as high as 600°K. Sapphire windows of good quality are available commercially in a variety of sizes. Sapphire can be vacuum brazed to several metals, among them are tantalum, zirconium, and Ni-Fe alloy.

It is important to arrange the plasma cell and the external optical elements so that a homogeneous region of plasma can be viewed. Non-homogeneous plasmas are probably impossible to diagnose by analysis of line profiles without some independent knowledge of plasma conditions. We have found it adequate to use an external optical system consisting only of a single lens (magnification of one or more) and a stop with a quite large f-number. This combination gives a good depth of focus and allows the sampling of very small regions of the plasma. An alternate arrangement might be the use of collimating devices without lenses.

The plasma cell that was used to obtain data reported here will not be described in detail because similar apparatus is described elsewhere³. The lines were emitted from the inter-electrode region of a plane-parallel hot-cathode cesium diode. The cathode was heated by electron bombardment and the anode cooled to about 600°K by circulating silicone oil. Most of the data reported here were taken with a UC:ZrC emitter at temperatures above 2000°K. The electrodes were 16 mm in

diameter and spaced 1.4 mm apart. The cesium vapor pressure was held at 2.2 mm Hg by maintaining equilibrium with cesium liquid at a condensation temperature of 580°K. Some of the data were taken at slightly lower vapor pressures in a wider-spaced cell having a tantalum emitter. Observations were made at currents ranging from zero to 30 amp/cm², the latter flowing at an applied voltage of 4 v dc. The measurements, all made in a plane parallel to the electrodes, showed homogeneous emission from the interior region with insignificant emission from edge regions except for resonance lines.

III. Results and Discussion

The width w (full width at half maximum in angstroms) is chosen as the definitive parameter of the line shape because it is easily obtained by scanning and because it is almost directly proportional to ion density for plasmas considered here. The broadening effects due to electrons in the plasma dominate the core (which includes the half-maximum point) of the line while the effects due to the ions dominate the wings and also are responsible for asymmetries. The width due to electrons alone is strictly proportional to electron density. For all plasma discussed here, the electron density is equal to the ion density, and both are referred to as ion density.

The line widths are less sensitive to temperature than to ion density. While experimental evidence of this is

inconclusive, we can cite some theoretical predictions by both Stone and Griem⁴. Both make calculations that are based on the same general theory and which are extensions of the recent work done by Baranger and by Kolb and Griem⁵. This work has already shown excellent agreement with experiment for hydrogen and helium⁶.

Griem's calculations of line widths for the sharp and diffuse series show a temperature dependence that is as large as $T^{1/3}$ or $T^{1/2}$, which is quite contrary to the case for the fundamental series lines. The latter have been calculated by both Griem and Stone, whose over-all agreement is very good but not precise. Both predict that fundamental series line widths change less than 10% in the range from 2000°K to 10,000°K when ion density is held constant. In spite of the fact that a specific test is lacking, we feel that the over-all excellent agreement between theory and experiment warrants acceptance of the calculated temperature dependence.

The line broadening is due, in a simplified picture, to the Stark broadening of the upper level by the ions, and also to impact perturbations by electrons during the emission process. The effects attributable to perturbations of the lower level are much less important than those attributable to the upper level. Then one would expect that lines emitted from the same initial state but having different final states should have the same shapes. Thus the sharp series doublets, $6P_{1/2}-nS_{1/2}$ and $6P_{3/2}-nS_{1/2}$, should have the same profiles when plotted versus frequency, but widths measured in wavelength units should differ by the ratio of the square of the wavelengths.

Also the F-levels of the cesium atom have an unresolved spin-orbit splitting and hence fundamental series lines ($5D_{3/2}-nF$ and $5D_{5/2}-nF$) appear as doublets. The same argument may be applied to these lines that was applied to the sharp series doublets and even carried further here. The wavelengths of the two members of the doublet never differ by as much as 1%, which is not important even when the ratio is squared. Hence fundamental series lines originating in the same level are not considered separately in this paper, but are averaged together instead.

Line profiles have been measured for 6 lines of the sharp series, 13 lines of the diffuse series, and 14 lines of the fundamental series (where 14 lines are considered as only 8 separate transitions for reasons already stated). The essential results are presented in Tables 1 and 2, where values of w are given for a variety of plasma conditions. For each set of data the ion density, N_e , and temperature were determined by absolute intensity measurements of both continuum and lines, as mentioned in the next paragraph.

The shape of the radiative recombination continuum beyond the first excited states of cesium (6P) is related to the electron temperature (T_e) and to the cross section, $\sigma(v)$, for recombination of electrons of velocity v into the 6P state. The ion density is related to the height of the continuum. We have used Mohler's⁷ experimental determination of $\sigma(v)$ along with measurements in the continuum to get values of N_e and T_e . We have also measured line intensities and, using Stone's⁸

oscillator strengths for cesium, we have determined excitation temperatures (T) assuming thermal equilibrium. A detailed discussion of both methods will be found in reference 3. For all cases we found agreement within experimental error between T_e and T and also between measured values of ion density and the value calculated on the basis of Saha equilibrium at temperature T . Of course, the observation of equilibrium properties in these dense plasmas is expected but the agreement between the two independent measurements of T and N_e is nevertheless reassuring.

Measured values of line width are subject to experimental error as indicated in Tables 1 and 2. The resolution of the monochromator limits the accuracy of the width measurements for the narrower lines but is unimportant for most of the data. Lower series members are sometimes partially reabsorbed. When this occurs the observed line profile appears unusually broad. A discussion of this effect is given in Section IV below. Large errors have been assigned to widths that have been significantly corrected for reabsorption (see for example the lowest members of the diffuse series). Reproducibility in measured line widths was only 8% upon day-to-day repetition of seemingly identical conditions. This deviation is assumed to be due to undetected differences in ion density. The broadest lines are difficult to measure accurately because they merge into the continuum or nearby lines. All of the above factors were considered in assigning the rather conservative line width errors in Tables 1 and 2.

Also shown in Table 1 are calculated line widths due to Stone⁹ and to Griem¹⁰. It will be noted that comparison of theory and experiment shows agreement within the experimental error.

A set of simple empirical equations has been derived to fit some of the experimental data. They are,

for the sharp series 6P-nS,

$$w = 2^{(n-9)} \times 0.30 N_e \times 10^{-15} \quad 9 \leq n \leq 11, T_e = 6060^\circ\text{K} \quad (1)$$

for the diffuse series, 6P-nD,

$$w = 2^{(n-7)} \times 0.35 N_e \times 10^{-15} \quad 7 \leq n \leq 10, T_e = 6000^\circ\text{K} \quad (2)$$

and for the fundamental series, 5D-nF,

$$w = 2^{(n-5)} \times 1.5 \times N_e \times 10^{-15} \quad 5 \leq n \leq 8, 2000 \leq T_e \leq 10,000^\circ\text{K} \quad (3)$$

where the widths are given to about 20% accuracy and the equations are valid in the vicinity of 10^{15} cm^{-3} .

In Eqs. (1) and (2) sharp and diffuse series line widths are stated for $T_e = 6000^\circ\text{K}$. According to Griem's calculations, the sharp series lines are expected to behave about like $w \propto T_e^{1/2}$ and the diffuse series lines about like $w \propto T_e^{1/3}$. The use of the electron temperature, T_e , is important here. This is because the theory is applied to cases where the ions are slow enough to obey quasi-static theory, but the electron effects are averaged over the velocity

distribution of the electrons, which is taken to be Maxwellian. Thus the temperature dependence enters through the electron velocity distribution which is characterized by the parameter T_e . This point is important experimentally too, because it is quite possible to generate plasmas that do not exhibit kinetic equilibrium, but have instead a mixture of relatively cold, heavy particles and a relatively hot Maxwellized electron gas.

The data given above in (1), (2), and (3) and in Tables 1 and 2 may be extrapolated linearly to higher values of N_e so long as the lines can still be considered isolated. A line is no longer isolated when the line width (in terms of the energy smearing of the initial level) becomes comparable to the energy difference between the initial level and neighboring levels. Non-isolated lines have an ion-density dependence that is less strong than directly proportional. Some fundamental series lines become non-isolated as low as 10^{15} cm^{-3} and all of them are suspicious at densities above 10^{16} cm^{-3} .

The highest diffuse series lines (see Table 1) seem smaller than expected and perhaps should be used with some caution.

In Figs. 1 and 2 we present examples of complete line profiles. Figure 1 gives a fundamental series line (5D-6F) and Fig. 2 shows a sharp series line (6P-11S). Both show a theoretical profile for comparison. The wavelength scale is centered at the peaks of the lines (in this paper we ignore line shifts) and the peaks have been normalized to unity.

The proper method of normalization would be to match the areas under the theoretical and experimental curves. Our method is much easier and leads to insignificant differences in fit near the core of the line. An interesting aspect of the profile in Fig. 1 is the line 5D-6G clearly shown at about -10A. This line is normally forbidden but appears here because of state mixing due to plasma perturbations.

IV. Reabsorption

In the plasma cell used here, the hot plasma region (temperatures well above 2000°K) was surrounded by a region of much cooler plasmas ($\approx 600^\circ\text{K}$). This situation is difficult to avoid because the window cannot survive at high temperatures. Two experimental schemes that may avoid the above situation are (a) a plasma that is differentially pumped through a small slit and (b) magnetic confinement for highly ionized plasmas. However, the cooler plasma is not really important even though all radiation must pass through it before emerging from the cell. The cooler plasma is populated principally by Cs_2 molecules, which are dissociated in the hot region, and by atoms in the ground state. Cs_2 has certain absorption bands and data should be taken in these spectral regions with some caution. Ground state atoms absorb significantly only the resonance lines (principal series). However, these principal series lines are already strongly self-absorbed in the hot region and therefore of little diagnostic value. Lines of the sharp,

diffuse, and fundamental series, which are not reabsorbed in the cool plasma, can be partially self-absorbed in the hot region. This problem is discussed in detail in the following paragraphs, where a method of correcting for reabsorption effects is described.

Consider a homogeneous slab of plasma of breadth L cm. Let the light that is emitted be collimated and let the light intensity in the interval $d\lambda$ that arrives at an observation point be $I(L, \lambda)d\lambda$ erg/sec. Let the distance from the plasma to the observer be large compared to other dimensions.

Now the shape of a plasma broadened line for optically thin cases, ignoring line asymmetries, approximates a dispersion curve,

$$I(L, \lambda)d\lambda = \frac{J(L)}{2\pi} \frac{w d\lambda}{\lambda^2 + (w/2)^2} \quad (4)$$

where

$$J(L) = \int_{-\infty}^{\infty} I(L, \lambda)d\lambda \quad (5)$$

and where λ is the wavelength in angstroms measured from the line center, w is the full width at half maximum in angstroms, and $J(L)$ is the total intensity of the line. We now introduce the subscripts "c" and "o" denoting "corrected" and "observed" parameters. Let us consider lines that are only slightly reabsorbed. Then both the corrected profile $I_c(L, \lambda)d\lambda$ and the observed profile $I_o(L, \lambda)d\lambda$ have dispersion shapes with parameters w_c and w_o respectively. Now we define a number δ that is small compared to unity such that

$$J_c(L) = J_o(L) (1 + \delta) \quad (6)$$

Then it can be shown that

$$\delta = \frac{e^2 \lambda_o^2 L f(i,f) N(f)}{2 m c^2 w} \times 10^{-8} \quad (7)$$

where e , m , and c have their usual meaning (in esu), λ_o is the wavelength of the line in angstroms, $f(i,f)$ is the oscillator strength for the transition $i \rightarrow f$ in the notation of Condon and Shortley¹¹, and $N(f)$ is the population (in cm^{-3}) of the final state. Oscillator strengths for over a hundred transitions in cesium have been calculated by Stone¹². The familiar Boltzmann equation obtains for the equilibrium cases,

$$N(f) = N(0) \frac{g_f}{g_o} e^{-E_f/kT} \quad (8)$$

where $N(0)$ is the population of the ground state, g_f and g_o are the multiplicities, and E_f is the energy of the state f . Finally, the corrected width is given simply by

$$w_c = w_o (1 - \delta) \quad (9)$$

In order to determine δ a value of $N(f)$ must be known. Probably the best way to determine $N(f)$ is to determine the populations of several nearby states from the absolute intensities of lines originating in these states. Even in plasmas far from equilibrium such nearby states should approach a smooth Boltzmann-like distribution in relative population. Gross departures from a smooth distribution can be attributed

to reabsorption effects, while if no gross departures are seen, the value of $N(f)$ can be determined by interpolation. Such a procedure requires a calibration for absolute intensity measurements that is not always easy to accomplish. In cases where absolute intensities are difficult to measure, a value of T can be obtained from the relative intensities of several lines through the usual Boltzmann plot. Less confidence should be placed in a value of T obtained from relative intensities because anomalous populations are commonly observed in plasmas that are not in equilibrium.

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Figure Captions

Fig. 1. Profile of the 5D-6F line at 1.6×10^{15} ions-cm⁻³.

The normally forbidden 5D-6G transition is clearly seen in the experimental data curve at approximately -10A. The dashed line is a calculated profile due to Stone.

Fig. 2. Profile of the 6P-11S line at 1.6×10^{15} ions-cm⁻³.

The dashed line is a calculated profile due to Griem.

81

FIG 1

19

Fig 2

TABLE 1^a

Values of full line width at half-maximum in angstroms

Series	Transition	λ (Å)	Line Width		
			$N_e = 1.3 \pm 0.3 \times 10^{15} \text{ cm}^{-3}$	$N_e = 1.6 \pm 0.3 \times 10^{15} \text{ cm}^{-3}$	
			$T_e = 6000^\circ\text{K}$ $w(\text{meas})$	$T_e = 6000^\circ\text{K}$ $w(\text{meas})$	$w(\text{calc})^b$
sharp	$6P_{1/2}-9S_{1/2}$	6355	. . .	0.5 ± 0.2	0.4
	$6P_{3/2}-9S_{1/2}$	6587	. . .	0.6 ± 0.2	0.5
	$6P_{1/2}-10S_{1/2}$	5839	. . .	1.0 ± 0.2	0.9
	$6P_{3/2}-10S_{1/2}$	6034	. . .	1.1 ± 0.2	0.9
	$6P_{1/2}-11S_{1/2}$	5568	. . .	1.8 ± 0.2	1.9
	$6P_{3/2}-11S_{1/2}$	5746	. . .	1.7 ± 0.2	2.0
diffuse	$6P_{1/2}-7D_{3/2}$	6723	. . .	0.5 ± 0.2 - 0.4	0.5
	$6P_{3/2}-7D_{3/2}$	6983	. . .	0.6 ± 0.2 - 0.4	0.5
	$6P_{3/2}-7D_{5/2}$	6973	. . .	0.8 ± 0.3 - 0.5	0.6
	$6P_{1/2}-8D_{3/2}$	6010	0.9 ± 0.2	1.3 ± 0.3	1.1
	$6P_{3/2}-8D_{3/2}$	6218	. . .	1.3 ± 0.2	1.1
	$6P_{3/2}-8D_{5/2}$	6213	1.2 ± 0.3	1.5 ± 0.2 - 0.3	1.3
	$6P_{1/2}-9D_{3/2}$	5663	2.0 ± 0.2	2.1 ± 0.2	2.2
	$6P_{3/2}-9D_{5/2}$	5845	2.3 ± 0.3	2.8 ± 0.3	2.7
	$6P_{1/2}-10D_{3/2}$	5466	3.2 ± 0.5	. . .	...
	$6P_{3/2}-10D_{5/2}$	5635	3.9 ± 0.5	. . .	...
	$6P_{1/2}-11D_{3/2}$	5340	5.3 ± 0.7	. . .	...
	$6P_{3/2}-11D_{5/2}$	5503	5.1 ± 0.5	. . .	...
fundamental	5D-5F	8018, 8081	2.0 ± 0.3 - 0.4	2.3 ± 0.3 - 0.4	1.9
	5D-6F	7228, 7280	4.1 ± 0.6	5.4 ± 0.7	5.7
	5D-7F	6826, 6870	9.0 ± 2.0	12.2 ± 2.0	11.4
	5D-8F	6629	14.0 ± 3.0	20.5 ± 4.0	21.9

a. The fundamental series experimental data appearing here have been reported previously by Stone and Agnew (reference 1).

b. Calculations for the sharp and diffuse series are due to Griem (reference 2) and calculations for the fundamental series are due to Stone (reference 1).

Table 2^a

Values of full line width at half-maximum in angstroms

Series	Transition	$\lambda(\text{\AA})$	Conditions	w (meas)
diffuse	$6P_{3/2}-8D_{5/2}$	6213	b	0.5 ± 0.2
	$6P_{3/2}-8D_{5/2}$	6213	c	0.4 ± 0.2
	$6P_{3/2}-8D_{3/2}$	6218	c	0.4 ± 0.2
	$6P_{3/2}-9D_{5/2}$	5845	b	0.6 ± 0.2
	$6P_{3/2}-9D_{3/2}$	5847	b	0.5 ± 0.2
fundamental	5D-5F	8018, 8081	b	0.6 ± 0.2
	5D-6F	7228, 7280	b	1.1 ± 0.2
	5D-7F	6826, 6870	b	2.5 ± 0.3
	5D-7F	6826, 6870	c	1.7 ± 0.2
	5D-8F	6629	b	3.7 ± 0.7
	5D-8F	6629	c	2.8 ± 0.3
	5D-8F	6629	f	1.6 ± 0.3
	5D-9F	6432, 6473	d	6.3 ± 0.6
	5D-9F	6432, 6473	e	4.0 ± 0.4
	5D-9F	6432, 6473	f	2.6 ± 0.3
	5D-10F	6326, 6366	d	10.0 ± 2.0
	5D-10F	6326, 6366	e	6.2 ± 0.6
	5D-10F	6326, 6366	f	4.2 ± 0.4
	5D-11F	6250, 6289	e	7.8 ± 1.0
	5D-11F	6250, 6289	f	5.5 ± 0.6
	5D-11F	6250, 6289	g	1.8 ± 0.4
	5D-12F	6232	g	2.4 ± 0.4
	5D-12F	6232	h	1.4 ± 0.5

$$b. \quad N_E = 3.9 \pm 0.9 \times 10^{14} \text{ cm}^{-3}, \quad T_E = 2400^\circ\text{K}$$

$$c. \quad N_E = 2.8 \pm 0.8 \times 10^{14} \text{ cm}^{-3}, \quad T_E = 2360^\circ\text{K}$$

$$d. \quad N_E = 3.3 \pm 0.8 \times 10^{14} \text{ cm}^{-3}, \quad T_E = 2670^\circ\text{K}$$

$$e. \quad N_E = 1.4 \pm 0.4 \times 10^{14} \text{ cm}^{-3}, \quad T_E = 2570^\circ\text{K}$$

$$f. \quad N_E = 1.1 \pm 0.3 \times 10^{14} \text{ cm}^{-3}, \quad T_E = 2500^\circ\text{K}$$

$$g. \quad N_E = 4.4 \pm 1.0 \times 10^{13} \text{ cm}^{-3}, \quad T_E = 2200^\circ\text{K}$$

$$h. \quad N_E = 3.2 \pm 0.8 \times 10^{13} \text{ cm}^{-3}, \quad T_E = 2150^\circ\text{K}$$

a. The fundamental series data appearing here have been reported previously by Stone and Agnew (reference 1).