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THE INTERPRETATION OF CESIUM SPECTRA*

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ABSTRACT

A review of the methods of plasma diagnosis by the interpretation of the cesium spectrum is given. Since accurate analysis of spectroscopic results depends on knowledge of the fundamental radiative properties of cesium, we call attention to recent experimental results for oscillator strengths and radiative recombination. Accurate analysis depends also on knowledge of the equilibrium properties of the plasma, which is discussed in some detail, especially the concept of local thermodynamic equilibrium. Molecular effects are also discussed and molecular absorption results are given. Certain refinements in experimental method and analysis are mentioned.

I. INTRODUCTION

Spectroscopy is a powerful tool that has already contributed much to our knowledge of plasma conditions within the cesium diode.¹ Early cesium spectroscopy yielded only qualitative and semi-quantitative results due mainly to the absence of adequate basic knowledge needed for spectral interpretation. But recent advances both in theory²⁻⁴ and experiment^{5,6} allow more precise measurements. Much can be gained also by the adoption of refined experimental techniques and more penetrating analyses.

The purpose of this paper is to provide a general review of the methods of plasma diagnosis by the interpretation of the cesium spectrum. The accurate analysis of spectroscopic results depends on knowledge of the equilibrium properties of the plasma in addition to knowledge of the radiative properties. We therefore discuss in detail the concept of local thermodynamic equilibrium as well as call attention to recent experimental results for absolute oscillator strengths and cross sections for radiative recombination. In addition the cesium spectrum is described, the role of trapped resonance radiation is discussed, molecular effects are considered, cesium molecular absorption is shown, and refinements in experimental method and analysis are mentioned.

Rigid space limitations prevent full treatment for many of the topics discussed here. We have only indicated methods in some cases, and have often omitted definitions for common symbols and notation. The reader is referred to "Plasma Spectroscopy" by H. Griem⁷ for an up-to-date general reference.

II. INTERPRETING THE CESIUM SPECTRUM

An example of a cesium spectrum emitted from a thermionic converter is shown in Fig. 1. Shown on the spectrum are a number of details, including at (1) the higher principle series lines approaching the series limit; at (2) the sharp and diffuse series limit plus the associated recombination continuum extending toward the blue; at (3) the fundamental series limit plus continuum; at (4) the strong molecular absorption near 7600 Å due to Cs₂ concentra-

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tion in the vapor surrounding the plasma region, and at (5) and (6) the strong resonance lines connecting the first excited state (doublet: $6P_{1/2}$ and $6P_{3/2}$) to the ground state ($6S_{1/2}$), shown here with the line cores completely reversed due to cool vapor in the observation path. Not detectable here are additional molecular absorption bands at 4800 Å, 5700 Å, and 6200 Å (see Section IV).

In the following paragraphs we list the commonly measured quantities and indicate the plasma properties obtainable under the various equilibrium conditions. These conditions, especially local thermodynamic equilibrium (LTE), are discussed in Section III.

Lines. Relative intensity measurements of subordinate series lines are readily used to determine the electron temperature by means of a Boltzmann plot⁸ when the emitting states fulfill the conditions for partial LTE. Furthermore, at a known pressure and under any reasonable assumption about the heavy particle temperature, the absolute intensity of a single line will give the electron temperature to good accuracy (5 - 10%) when complete LTE holds.

Implied in the preceding paragraph is the evaluation of line intensity data using the usual relationship for spontaneous emission.⁹ For this one needs to know the appropriate oscillator strengths. Stone's calculated values² are the accepted standard for cesium. Experimental values have been recently reported⁶ for 50 subordinate series lines and a summary is given below.

Cesium oscillator strengths

<u>Series</u>	<u>Experiment⁶/Stone's theory²</u>
Sharp (6P - nS)	1.35 ± 0.15
Diffuse (6P - nD)	1.12 ± 0.15
Fundamental (5D - nF)	1.12 ± 0.15

The experimental result is 35% higher than Stone's calculated values for the sharp series. It has been found that the use of the experimental oscillator strengths will remove discrepancies involving sharp series lines in Boltzmann plots.¹⁰

Line shapes. Plasma broadening by Stark effect enables the determination of ion density through measurement of line widths. The method is especially convenient for cesium.^{3,5} The effect is insensitive to the degree of equilibrium and almost independent of temperature. Two easily met requirements are (1) remaining within the range of validity of the isolated line theory, and (2) avoiding plasma conditions where other broadening mechanisms compete with the Stark effect.

Radiative recombination. The intensity distribution of the radiative recombination continuum enables the determination of the electron temperature by relative intensity measurements and the electron density by absolute intensity measurements when the electron velocity distribution is Maxwellian. In cesium, the continua beyond the first excited state (6P) and the second excited state (5D) are both accessible for easy measurement (see Fig. 1). The intensity distribution for radiative recombination into the j^{th} state is given by:

$$Q_j(\lambda)d\lambda = N_e^2 \int v \sigma_j(v) F(v)dv, \quad (1)$$

where $Q_j(\lambda)$ is the rate of photon emission in the interval $d\lambda$ corresponding to electron velocity in the range dv , $\sigma_j(v)$ is the cross section for an electron of velocity v , N_e is the electron density, and $F(v)dv$ is the fraction of electrons in the range dv . The electron

temperature, T_e , enters as a parameter in $F(v)dv$ when the distribution is Maxwellian. New values have recently been reported⁶ for the cross-sections:

$$\sigma_{6P}(v) = 3.74 \times 10^{-6}/v^2 \text{ cm}^2,$$

$$\text{and } \sigma_{5D}(v) = 8.3 \times 10^{-6}/v^2 \text{ cm}^2$$

when v is in cm/sec. These values differ both in magnitude and velocity dependence from earlier measurements by Mohler.¹¹ Electron temperatures determined using the new cross sections are about 15% lower than when Mohler's cross section is used, and measured ion densities (rarely done because line broadening is so much easier) are about 35% lower.

The new cross sections are in good agreement with preliminary results of a theoretical calculation by Norcross and Stone.⁴

III. LOCAL THERMODYNAMIC EQUILIBRIUM

Definition. Following the arguments of Spitzer¹² and Griem,¹³ certain concepts can be applied to characterize a steady-state cesium plasma. The most useful of these for spectroscopy is the concept of local thermodynamic equilibrium (LTE), which can occur in optically thin plasmas that are dominated by electron collisions.

LTE is most readily defined for a region of plasma that is removed from boundaries and for cases where transport or transient effects are of secondary importance. Complete LTE is said to exist when all the states, both free and bound (including the ground state), have Boltzmann-Saha relative populations characterized by the electron temperature. When the plasma is in complete LTE the absolute populations of all the states can be known if, for example, the ground state population is known. A less stringent (but often useful) related concept is partial LTE, wherein some of the states have specific populations with respect to a limited range of neighboring states. Partial LTE is said to exist for those states which are related (1) to immediately neighboring states by Boltzmann factors and/or (2) to continuum states by the Saha relation. Partial LTE usually exists for a given state and higher states only; it is lower states that lag behind in the establishment of LTE.

No requirement is placed on the kinetic temperature of the atoms and ions under LTE. The heavy particles may have a temperature (T_H) distinct from the electron temperature and absolute densities are fixed by the relationship $P_{\text{total}} = \sum N_i k T_i$.

Conditions. A necessary condition for the establishment of LTE is the existence of a Maxwellized electron velocity distribution. Both electron-ion and electron-neutral collisions contribute to the isotropy of the electron velocity distribution, but it is the electron-electron collisions that are effective in thermalization. Spitzer¹² gives a "self-collision" or thermalization time, t_c , for electrons:

$$t_c = 0.265 T_e^{3/2} / (N_e \ln \Lambda) \text{ sec} \quad (2)$$

where $\ln \Lambda$ is a slowly changing function of T_e and N_e related to the Coulomb cut-off distance. For the thermionic diode $T_e \sim 3000^\circ\text{K}$, $N_e \sim 10^{14}$, $\ln \Lambda$ is about 5, and t_c becomes $\sim 10^{-10}$ sec. This corresponds to an equilibration distance of $\sim 10^{-3}$ cm. In this paper we will restrict our attention to interelectrode gaps of 10^{-2} cm or more, for which the conditions for electron Maxwellization are fulfilled.

Two further prerequisites for LTE should be mentioned. The first is that electrons gain energy from local fields in amounts $< kT_e$ in a distance of one mean free path. The second is that lifetimes for diffusion must be large compared to collisional lifetimes for each state. These two conditions are also met in typical converter operation at a spacing of 10^{-2} cm.

Detailed balance. LTE can occur for a wide range of cesium plasmas that are called "collision dominated". The important collisions are the inelastic electron-neutral collisions. Excitation and de-excitation cross sections are related through detailed balance in such a manner that in the absence of any other mechanism for excitation or de-excitation, an isolated plasma consisting of electrons and atoms will arrive at LTE regardless of the temperature.

Calculating LTE. In a real plasma, radiative processes compete with collisional excitation and de-excitation. The onset of LTE is given by comparing collisional rates with radiative rates as can be seen from the following simplified argument: Consider an atom that has only two levels, i and j. If inelastic collisions and radiative processes both occur, we have the following rate equation:

$$\begin{aligned} dN_i/dt = & -N_i A(i,j) + N_j B(j,i)\rho(\nu) - N_i B(i,j)\rho(\nu) \\ & + \int N_j N_e v \sigma_{j,i}(\nu) F(\nu) d\nu - \int N_i N_e v \sigma_{i,j}(\nu) F(\nu) d\nu \\ = & -dN_j/dt = 0 \text{ in steady state.} \end{aligned} \quad (3)$$

The first three terms on the right account for the rate of spontaneous emission (R_{em}), absorption (R_{abs}), and induced emission respectively, while the two integral terms account for rate of excitation (R_{ex}) and de-excitation (R_{de}) by inelastic electron collisions. In a completely radiation-dominated optically-thick case, the integral terms can be ignored and the first three terms can be solved to produce the Boltzmann factor for N_i/N_j . Similarly when the first three terms are negligible compared to the integral terms, the Boltzmann factor for N_i/N_j is also obtained via detailed balance conditions on the σ 's. But when the spontaneous emission term is small and non-negligible, we get

$$\frac{N_i}{N_j} = \left(\frac{N_i}{N_j} \right)_{LTE} \left(1 - \frac{N_i A(i,j)}{\int N_i N_e v \sigma_{i,j}(\nu) F(\nu) d\nu} \right) \quad (4)$$

Or, approximately for a given state,

$$N/N_{LTE} = 1 - R_{em}/R_{de} \quad (5)$$

The extrapolation to a multi-level atom is straightforward and we find that small deviations from LTE should scale like the ratio of radiative and collisional rates.

Radiation. The thermionic converter is typically optically thin for radiation corresponding to transitions between higher states and optically thick for the lower resonance states. In the latter case, the absorption term must also be considered in calculating N/N_{LTE} and R_{em} in equation 5 is then replaced by $R_{em}(1 - R_{abs}/R_{em})$, where R_{abs}/R_{em} is the degree of radiation trapping.

For cesium the absorption term is strongest for the first resonance doublet and these characteristic red lines are heavily trapped. This radiation trapping helps to establish complete LTE by reducing the effective radiative decay rate of the first excited state. In the following paragraphs we will consider these resonance lines in detail and give a sample calculation of the radiation trapping.

In order to calculate the degree of trapping, it is essential to know the detailed line shape. For the purpose of our sample calculation we can combine the two lines of the first resonance doublet by adding the multiplicities (g) and oscillator strengths (f_{em}) to give a single line (6S-6P) at the weighted-average energy level. For assumed plasma conditions of $T_e = 3000^\circ\text{K}$, $P = 1$ Torr, and $T_H = 1500^\circ\text{K}$ we calculate at LTE that $N_{6S} = 4.2 \times 10^{15}$ and $N_e = 7.1 \times 10^{14} \text{ cm}^{-3}$. Then the line has the following parameters:

Natural width:⁹

$$\Gamma_N/2 = \text{width (half-half) of dispersion curve} \quad (6)$$

$$= 2\pi f_{em} (e^2/mc^2) (g_{6S}/g_{6P}) = 7 \times 10^{-13} \text{ cm.}$$

Stark width:⁷

$$\Gamma_S/2 = \text{width (half-half) of dispersion curve} \quad (7)$$

$$\sim 7.5 \times 10^{-26} \times N_e = 5 \times 10^{-11} \text{ cm.}$$

Resonance (pressure) width:¹⁴

$$\Gamma_R/2 = \text{width (half-half) of dispersion curve} \quad (8)$$

$$= (2.74/4\pi) (e^2/mc^2) \lambda^3 f_{em} N_{6S} (g_{6P}/g_{6S})^{1/2} = 3.4 \times 10^{-10} \text{ cm.}$$

Doppler width:

$$g_D = \text{width (half-e}^{th}\text{) of Gaussian curve} \quad (9)$$

$$= (\lambda/c) (2kT_H/M_{CS})^{1/2} = 1.3 \times 10^{-10} \text{ cm.}$$

The above contributions give, when properly folded together, the exact shape of an emission or absorption line that would be seen in an optically thin sample of the plasma under consideration. Resonance broadening and Doppler broadening are evidently the most important contributions to the core of the line. However the line wings, where the Doppler contribution is insignificant, are important in radiation trapping. Thus in the present case it will be sufficient to consider only the resonance broadening.

Following Holstein,¹⁵ and considering radiation which upon emission from the center of a 10^{-2} cm gap must traverse at least a distance $L = 5 \times 10^{-3}$ cm to escape, we find the degree of trapping is:

$$R_{abs}/R_{em} \cong 1 - (\pi k_d L)^{-1/2} = 0.95 \quad (10)$$

where

$$2\pi k_d = \lambda^2 N_{6S} (g_{6P}/g_{6S}) (\Gamma_N/\Gamma_R).$$

Due to the form of Γ_R , this result is independent of pressure and temperature so long as resonance broadening dominates. However, the trapping increases from 95% at 5×10^{-3} cm to 99.5% at 0.5 cm. The main effect of radiation trapping seems to be that it inhibits plasma decay, i.e., the radiation from strong lines that would provide a potent cooling mechanism in the optically thin case is instead largely retained in the plasma.

Radiation flow from the cathode to the plasma is unimportant since electron temperatures in the ignited mode are always observed higher than the brightness temperature of the cathode.

LTE results. Since radiative properties are known for cesium and absorption can be calculated as shown above, equation 5 can be evaluated for a given state upon the determina-

tion of the appropriate collisional rates. The collisional theory of Bates, Kingston, and McWhirter¹⁶ was adapted to cesium for this purpose. This adaptation involves simple procedures that relate cesium states to equivalent hydrogen states while retaining the cesium multiplicities, oscillator strengths, and energy levels. The fit is expected to be good, which in this context means that the theory is expected to hold within about a factor of two.

The results, which include the contribution of radiation trapping, are presented in Fig. 2, where curves of N/N_{LTE} for three different states are presented as a function of N_e at $T_e = 3000^\circ\text{K}$ for plasma 5×10^{-3} cm from a boundary (or for plasma at the mid-point of a 10^{-2} cm gap). Each curve is drawn with respect to Boltzmann factors connecting the specific state to the next lower state. Thus, while the two upper curves give information only about partial LTE, the lower (6P) curve specifies approach to complete LTE. The position of this lower curve is sensitive to the radiation trapping; for a larger plasma dimension it would be higher on the plot.

The results in Fig. 2 allow us to conclude that the states from about 10D and higher are already close to partial LTE for $N_e > 10^{11}$, and complete LTE is obtained in a gap of 10^{-2} cm for $N_e > 10^{13}$.

IV. MOLECULAR EFFECTS

There has been some speculation on the role of molecules and molecular ions in the cesium diode.¹⁷ However quantitative measurements of actual concentrations have not yet been reported. In the next few paragraphs we argue that in the regime considered here, i.e. plasma conditions at least approaching complete LTE, molecular effects are insignificant within the diode.

Since the structure of the Cs_2 molecule and its radiative and collisional properties are not known adequately, it is not possible to develop for Cs_2 an LTE plot like Fig. 2. Qualitatively, however, for collision dominated plasmas the same sort of result is expected. Calculations for the molecular species at full thermal equilibrium have been made by Clifton.¹⁸ These calculations should hold within a factor of about 2 for the LTE case. Some of Clifton's results are shown in Fig. 3. Clifton finds that full thermal equilibrium concentrations of Cs_2 and Cs_2^+ are about four orders of magnitude below their atomic counterparts. Wilkins¹⁹ has considered this problem in detail. He concluded that the only region of important Cs_2^+ concentration occurs for $N_e < 10^{12}$, with the further restriction that the cathode temperature must be low. Here atomic ions are diffusion dominated and molecular ions are not.

In our laboratory we have often measured the absolute populations of the higher excited states of the atom through their emission intensities. We have also measured ion densities (for $N_e > 3 \times 10^{13}$ and $P > 0.3$ Torr) by the absolute intensity of the radiative recombination band, which is due to the species Cs^+ only. At the same time Stark broadening, which gives the total ion density but does not distinguish between Cs^+ and Cs_2^+ , was measured and found in agreement. Furthermore, all measurements agreed with LTE predictions within error, leaving scant possibility for Cs_2^+ except within errors of perhaps 20 to 30%.

Molecules can nevertheless be important in the spectroscopic analysis of the cesium diode because Cs_2 concentration in the observation path can absorb light emitted from the hot plasma region. In Fig. 1 the unmistakably strong band at $7600 \text{ \AA}$ was shown. This band has been discussed by Loomis and Kusch.²⁰ Additional bands of molecular absorption are spread throughout the visible spectrum. In Fig. 4, we present the results of a simple transmission experiment

wherein light was passed through cesium vapor maintained at known concentrations and temperatures. The band of absorption seen at about 6200 Å has already been described quantitatively by Lapp and Harris.²¹

Through reasonable knowledge of the vapor pressure and temperature conditions in the observation path, one can use the data in Figs. 3 and 4 to estimate the transmission in the various spectral regions. This is especially important during the measurement of radiative recombination at high cesium pressures.

V. REFINEMENTS

We conclude by presenting a partial list of refinements and corrections that we have found useful.

Optical thickness. For subordinate series lines, the degree of self-absorption can be estimated and corrections made for the less severe cases by the method given by Agnew and Summers.⁵ These corrections can be made not only for line intensity but also for the widths of Stark-broadened lines. Resonance lines should be treated by the methods given in reference 15.

Molecular absorption. See Section IV of this paper.

Resolution. Occasionally the widths of Stark-broadened lines are comparable to spectrometer resolution. In this regime the spectrometer slit function can be determined by observing known narrow lines. Von Planta²² has shown that the slit function is usually well approximated by a Gaussian. The true width of the observed line can be obtained by unfolding the resultant Voigt profile.²³

Plasma homogeneity. It is usually possible to make at least an approximate check (e.g. by lateral scanning) on the homogeneity of the plasma source volume. This can be an important point since emitted radiation usually scales exponentially with temperature and thus a hot region small in area can give erroneous implications. In the case of precise work, corrections must be made to account for sizable amounts of radiation from the edge regions where large gradients are known to exist. Each case must be treated separately since such corrections depend on the volume of homogeneous plasma viewed as well as the exact shape of the edge gradient.

Absorption methods. The methods of absorption spectroscopy, especially the line reversal method, can be extremely valuable both for direct measurement and for calibration purposes in absolute intensity measurements.

Plasma corrections. In very hot or very dense plasmas several plasma effects should be considered in the analysis of spectroscopic results. Notable among these is the lowering of the ionization potential²⁴ and the "burning out"²⁵ of the neutral atoms. Burning out occurs at higher temperatures as the neutral density decreases while the ion density increases. This is already an important effect at 3000°K and 1 Torr.

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Fig. 1. Cesium emission spectrum (uncorrected for variations in spectral response). Shown are resonance lines at 1, 5, and 6; radiative recombination continua at 2 and 3; and molecular absorption at 4. More details are given in text.

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Fig. 2. Degree of LTE for several cesium states vs electron density for $3000^\circ K$ plasma a distance of 0.005 cm from a boundary.

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Fig. 3. Fraction of Cs_2 in cesium vapor vs temperature calculated by Clifton¹⁸ for full thermal equilibrium.

Fig. 4. The transmission of light through cesium vapor as a function of the Cs_2 path length.