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Pressure Effects of Foreign Gases on the Absorption Lines of Cesium. VI. Intensity Measurements of the Cesium Resonance Lines and Their Associated Satellites in the Presence of Various Foreign Gases*

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The intensity contours of the pressure-broadened Cs resonance lines and their associated satellites in the presence of He, Ne, Ar, Kr, Xe, and CF_4 are studied. The total integrated intensity of the lines and satellites in the presence of these gases is found to decrease exponentially with the relative density (rd) of foreign gas. For the range of foreign-gas pressures studied (0 to ~50 rd), the ratio of the integrated intensity of the $^2P_{3/2}$ component to that of the $^2P_{1/2}$ component is found to be close to the theoretical value of 2 for all rd's only when the integrated intensity of the satellites is included with the parent lines. These ratios (1.90 ± 0.03 for Ar, 2.00 ± 0.03 for Kr, and 2.06 ± 0.04 for Xe) indicate that satellites are part of the lines. The integrated intensity of the violet satellites associated with the $^2P_{3/2}$ component in the presence of these gases is found to be proportional to the cube of the low-energy collision diameter of the particular foreign gas. A violet satellite associated with the $^2P_{1/2}$ component is observed in the presence of CF_4.

I. INTRODUCTION

When an optical atom is placed in a foreign gas, not only are the frequencies and widths of its spectral lines modified, but satellite bands usually appear in the neighborhood of the first few members of the spectral line series.[1] For alkali atoms, such as Cs, red satellites appear on the red side of each component of the first few principal series doublets if the foreign gas used produces a red shift of the spectral lines. Also, a satellite usually appears on the violet side of the $^2P_{3/2}$ component of the resonance lines (the first doublet of the absorption series) for all foreign gases. The separation between the violet satellite and the reso-

nance line is much greater than that for the red satellites, but the corresponding separation for the case of the second doublet decreases so rapidly that the violet satellite is resolvable only for light gases.

Although several hundred satellites of this nature have been observed, there was no adequate intensity data. With the addition of a dual-beam system employing photomultiplier output and phase-sensitive detection along with electronic log conversion to our 35′ grating spectrograph, it is possible to obtain, as direct output, a plot of the absorption coefficient as a function of wavelength. The background intensity is flat over a wide range in wavelengths, so intensity informa-

tion, particularly in the line wings, is no longer in doubt.

This sixth article of the series[2] deals with intensity measurements of the cesium resonance lines and their associated satellites.

II. RESULTS AND DISCUSSION

A. Red Satellites of Cs (1)/Xe

The separations of the red satellites from the $^2P_{1/2}$ and $^2P_{3/2}$ components of the cesium resonance lines in the presence of Xe at 0.5 relative density (rd) are 12.6 and 36.5 cm^{-1}, respectively. Figure 1 illustrates the evolution of these red satellites in Cs(1) caused by Xe [hereafter denoted by Cs(1)/Xe] as the rd of Xe is changed from 0.15 to 1.10. As the rd is increased further, the intensity of the red satellite will become comparable to that of the line, finally blending into one unresolvable contour, causing a discontinuity in the graphs of shift and half-widths versus rd.[3]

Because of the close proximity of the red satellite to the line, quantitative intensity measurements of the satellites are not accurate. Attempts at decomposition were made in order to obtain a rough estimation of the relative intensities of the two red satellites. The result indicates that, for Cs(1)/Xe at rd = 0.02 and $T = 442$ °K, the $^2P_{1/2}$ red satellite is approximately twice as intense as the $^2P_{3/2}$ red satellite.

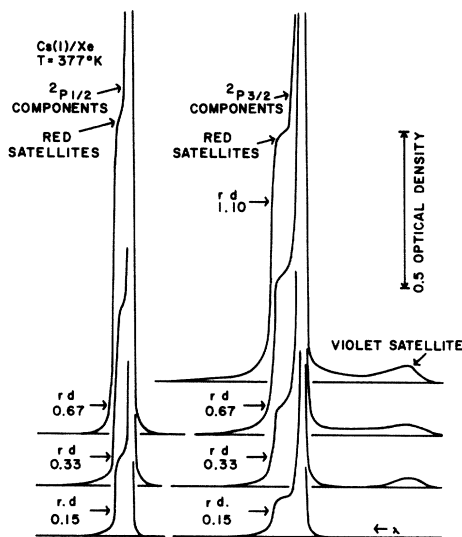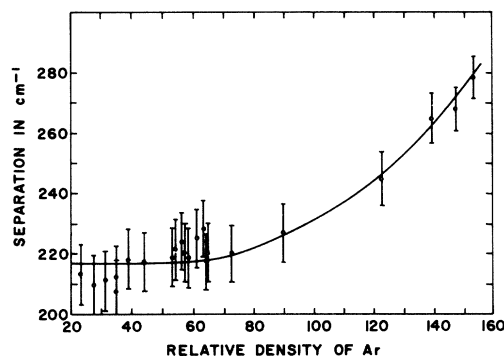FIG. 2. The separation of the VS (in cm^{-1}) from the peak of the $^2P_{3/2}$ component of Cs(1) versus the relative density of argon.

B. Violet Satellite of Cs (1) in the Presence of Various Foreign Gases

1. Satellite Separations

The separation of the peak of the violet satellite (VS) from that of the $^2P_{3/2}$ component of Cs(1) for rd < 2 is 367 ± 15 (387 ± 9), 236 ± 7 (253 ± 7) 217 ± 7 (223 ± 7), 153 ± 7 (156 ± 7), 250 ± 15 (253 ± 7), and 138 ± 7 (140 ± 7) cm^{-1} for He, Ne, Ar, Kr, CF$_4$, and Xe, respectively. The first figure indicates the separation before the satellite and line are decomposed, and the second figure, within the parenthesis, indicates the separation of the satellite after decomposition. The superposition of the line wing and satellite causes the apparent satellite peak to appear closer to the line. The discrepancy of the present data with previous data[4] is related to the conditions under which the measurements were made. Previous measurements were made at higher temperatures, causing the satellite peak to appear closer to the line due to the influence of a more intense violet line wing.

As the rd of foreign gas is increased, the line and the satellite shift together exhibiting no change in separation until the rd is higher than about 70 for Ar. After this point, the line is shifted more rapidly than the satellite and the separation increases, as shown in Fig. 2.

2. Relative Peak Intensity

If I_p (VS) stands for the "peak" intensity of the violet satellite and $I_p(^2P_{3/2})$ stands for that of the $^2P_{3/2}$ component line including any unresolved red satellite, the ratio $I_p(VS)/I_p(^2P_{3/2})$, as a function of the rd of foreign gases is shown in Fig. 3. As the pressure of foreign gas is increased, the peak intensity of the violet satellite grows rapidly with respect to the parent line. At any given rd, the ratio depends upon the particular foreign gas used,
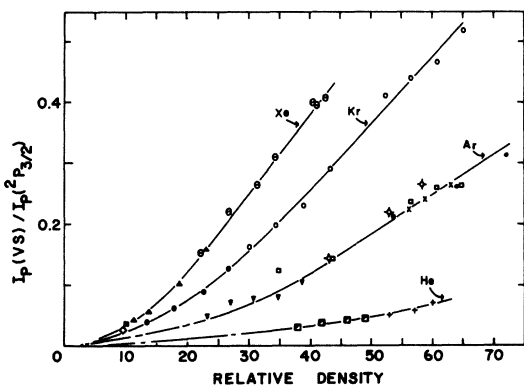
FIG. 1. Traces illustrating low-pressure evolution of the red satellite for Cs(1)/Xe.

FIG. 3. The ratio of the peak intensity of the VS, I_P(VS), to the peak intensity of the $^2P_{3/2}$ component of CS(1), $I_P(^2P_{3/2})$, versus the relative density of He, Ar, Kr, and Xe. The identification of data points is as follows: +, He 443 °K; ▨, He 431 °K; □, Ar 424 °K; ✦, Ar 427 °K; ✕, Ar 436 °K; ▼, Ar 477 °K; ●, Ar 498 °K; ⬤, Kr 417 °K; O, Kr 439 °K; ◇, Kr390 °K; ■, Xe 301 °K; ▲, Xe 418 °K; ⊖, Xe 444 °K.

and increases as the atomic number of the foreign gas is increased. The data in Fig. 3 shows that

$$I_p(\text{VS})/I_p(^2P_{3/2}) = \text{const.} \times (\text{rd})^{1.7} \qquad (1)$$

for all foreign gases used, i.e., He, Ar, Kr, and Xe.

3. Integrated Intensity with Respect to that of the Parent Line

Integrated intensity is defined by

$$I_A = \text{const.} \times \int \alpha(\nu) d\nu , \qquad (2)$$

where $\alpha(\nu)$ is the absorption coefficient as a function of ν in cm^{-1}. Figure 4 shows the rd dependence of the ratio of the integrated intensity of the violet satellite I_A(VS), to that of the $^2P_{3/2}$ component of Cs(1), $I_A(^2P_{3/2})$, for various foreign gases. To obtain these intensity values it is necessary to carry out a decomposition of the line and the satellite. A typical profile illustrating decomposition is shown in Fig. 5. The definition of satellites provided by this method of decomposition is regarded as a way of analysis (good for whatever might be learned from it) rather than an assumption as to the true nature of a separate line and satellite. The decomposition process is based upon experience obtained from observing the shape of the line at lower pressures (with no satellite in evidence) as well as the general evolution of the profiles with pressure. The data in Fig. 4 reveals that
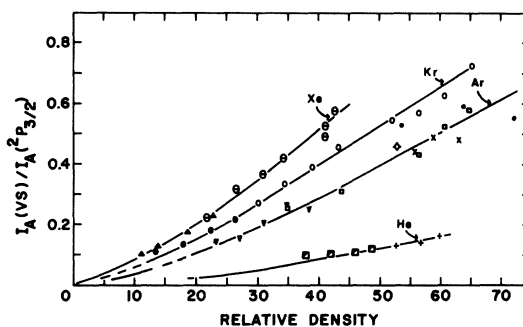

FIG. 4. The ratio of the integrated intensity of the VS I_A (VS), to that of the $^2P_{3/2}$), versus the relative density of He, Ar, Kr, and Xe. The identification of data points is as follows: +, He 443 °K; ▨, H 431 °K; □, Ar 424 °K; ✦, Ar 427 °K; ✕, Ar 436 °K; ▼, Ar 477 °K; ●, Ar 498 °K; ⬤, Kr 417 °K; O, Kr 439 °K; ▲, Xe 418 °K; ⊖, Xe 444 °K.

$$I_A(\text{VS})/I_A(^2P_{3/2}) = \text{const.} \times (\text{rd})^b \qquad (3)$$

where b is 1.1 ± 0.1, 1.3 ± 0.1, 1.3 ± 0.1, and 1.2 ± 0.1 for He, Ar, Kr, and Xe, respectively.

4. Integrated Intensity versus rd of Foreign Gases

An attempt was made to observe the effects of foreign-gas rd on the integrated intensity of the decomposed violet satellite I_A(VS). In order to reduce the effects of changes in cesium vapor concentration, the temperature was held constant (442 °K) and the foreign-gas pressure used was low (rd < 2). This temperature was selected such that the most intense satellites [Cs(1)/Xe] were not overabsorbed at the highest pressures and
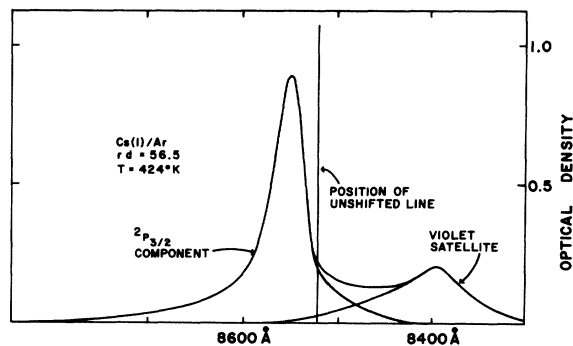

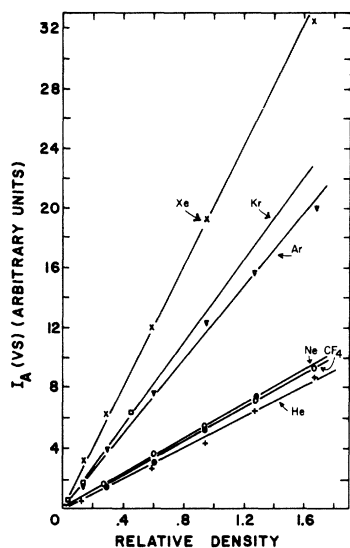FIG 5. Typical high-pressure profile illustrating decomposition.

FIG. 6. Integerated intensity of VS I_A (VS) in arbitrary units versus the relative density of foreign gases (442 °K). The identification of data points is as follows: +, He; O, Ne; ●, CF$_4$; Δ, Ar; □, Kr; X, Xe.

such that the least intense satellites [Cs(1)/He] were sufficiently strong to allow accurate measurements to be made. At this temperature and with the low rd of foreign gas used, the peak of the parent line was greatly overabsorbed.

I_A(VS), in arbitrary units, of the decomposed VS as a function of rd is shown in Fig. 6. The data points exhibit a linear relationship with rd. The slopes are given in the second column of Table I. These slopes will change if the range of rd is extended. Figure 7 shows a typical plot of I_A (VS) for an extended rd range at constant temperatures.

Several theories relate the origin of the violet satellite to the repulsive part of the interaction potential. A log-log plot of I_A(VS)/rd against σ, the low-energy collision diameter in the Lennard-

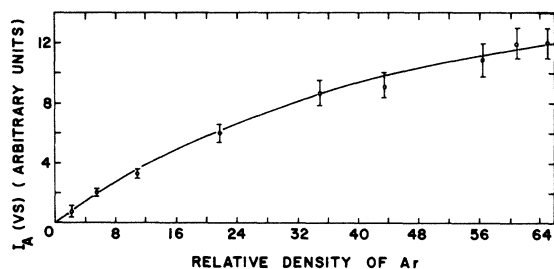FIG. 7. The integrated intensity of VS I_A(VS) at a constant temperature of 442 °K versus relative density of argon.

TABLE I. Slopes of I_A (VS)-versus-rd plots, and the low-energy collision diameters.

Foreign gas	Slopes of Fig. 6	σ (Å)
He	5.0	2.58
Ne	5.8	2.79
Ar	12.4	3.42
Kr	13.8	3.61
Xe	20.2	4.06
CF$_4$	5.6	4.70

Jones (6-12) potential,[5,6] is shown in Fig. 8. This plot suggests that

$$I_A (VS) = \text{const.} \times \sigma^3 \qquad (4)$$

for all gases used except CF$_4$.

CF$_4$, as a heavy large spherical molecule, with a mass and polarizability between Kr and Xe, would be expected to act like a heavy rare gas. This is indeed the case for shift and broadening.[2] However, it does not fit the heavy-rare-gas model for the separation of the peak of the vs from that of the parent line, the intensity I_A (VS) as a function of rd (Fig. 6), and the plot of I_A(VS)/rd versus σ³ (Fig. 8). This may be expected with the following considerations: Since CF$_4$ is composed of a tetrahedral arrangement of fluorine atoms about a carbon atom, during close collisions the Cs atom only sees the repulsive fluorine core and not the CF$_4$ molecule as a whole. Thus, the violet satellite of Cs(1)/CF$_4$ should be similar to that of Cs(1)/Ne. Whereas, when the shift and broadening of Cs(1)/CF$_4$ are considered, the interatomic potential at greater ranges is involved, and the Cs atom is influenced by the whole CF$_4$ molecule.

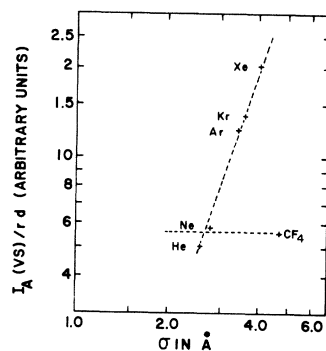In addition to the VS on the $^2P_{3/2}$ component for

FIG. 8. The integrated intensity of the VS/rd; I_A(VS)/ rd, versus the Lenard-Jones (6-12) low-energy collision diameter σ.

$Cs(1)/CF_4$, a weak, nearly unresolvable VS is seen on the $^2P_{1/2}$ component.

C. Ratio of the Integrated Intensity of the $^2P_{3/2}$ Component and Satellites to that of the $^2P_{1/2}$ Component and Satellites

Let $\sum I_A (^2P_3/2 + \text{satellites})$ be the sum of the integrated intensity of the broadened $^2P_{3/2}$ component of the cesium resonance lines and that of the associated (red and violet) satellites, and $\sum I_A (^2P_1/2 + \text{satellites})$ be the corresponding sum of integrated intensity for the $^2P_{1/2}$ component and satellites. The ratios of the former to the latter for various rd's of Ar, Kr, and Xe are plotted in Fig. 9. The arithmetic means for the ratios are 1.90, 2.00, and 2.06 for Ar, Kr, and Xe, respectively. The probable error of the arithmetic mean is ±0.03, and the probable error of a single point is ±0.14 [±0.04 and ±0.15, respectively, for Cs(1)/Xe]. It should be noted that the satellite area is a major portion of the total, especially at higher rd's.

It is striking that this ratio is maintained constant only when the satellites are included. This oscillator strength information strongly supports the idea that the satellites must be considered as part of the parent line and not due to any separate mechanism such as transitions between mixed states.

D. Total Integrated Absorption Intensity of the Cs Resonance Lines and All Associated Satellite Lines under Various Pressures of Foreign Gases

A plot of the total integrated intensity, I_A in arbitrary units, of both components of the resonance lines and all associated satellites versus
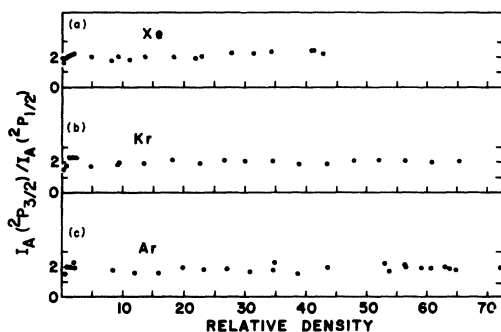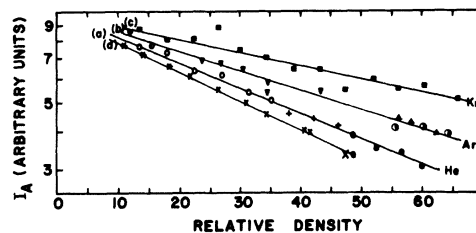

FIG. 10. The total integrated intensity I_A of the doublets of CS(1), including satellites versus relative density of various rare gases. The identification of data points is as follows: O, He 403 °K; +, He 431 °K; ●, He 443 °K; ▲, Ar 436 °K; ◐, AR 464 °K; ▼, Ar 477 °K; ▽, Ar 424 °K; ◑, 424 °K; □, Kr 417 °K; ■, Kr 439 °K; ✳, Xe 418 °K; ×, 444 °K.

rd of various foreign gases, as shown in Fig. 10 reveals that

$$I_A = \text{const.} \times e^{-a(\text{rd})} \qquad (5)$$

for all gases studied. Table II gives the values of the constant a for various gases as well as the ranges of rd. Since it was necessary to change the temperature of the absorption cell at various rd intervals, all data were normalized to the temperatures shown in the last column of Table II.

Similar measurements in the past have not been conclusive. Ch'en[7] reported a decrease in I_A for the Rb resonance lines for increasing pressures of Ar and He. Füchtbauer et al.[8] also reported a decrease in I_A for the 2537 line of Hg in Ar, but Michels and DeKluiver[9] reported an increase in I_A for the same Hg line with Ar, and a decrease in intensity for increasing He pressures. Ch'en and Smith[10] reported an increase in I_A for the $(5p\text{-}6s)$ line of I_n ($\lambda 4101$ and 4511) in Ar and a constant value for these lines in He.

Interpretation of these results is complicated by the fact that the Cs vapor pressure cannot be assumed to be constant, at a fixed temperature, as the foreign-gas pressure changes. From thermodynamic arguments, the vapor pressure of Cs should increase as it is subjected to an increasing foreign-gas pressure. On the other hand, collision-enforced transition in the presence of foreign gas would tend to cause a reduction in

FIG. 9. The ratio of the integrated intensity of the $^2P_{3/2}$ component and its satellites to that of the $^2P_{1/2}$ component and its satellites versus relative density of Ar, Kr, and Xe.

TABLE II. Values of a in Eq. (5).

Foreign gas	a	Ranges of rd	Temperature (°K)
He	0.0192 ± 0.0034	13—60	443
Ar	0.0150 ± 0.0048	12—65	436
Kr	0.0102 ± 0.0014	13—65	439
Xe	0.0222 ± 0.0012	11—43	444

I_A at increased foreign-gas pressures. Preston[11] observed a collision-enforced emission band associated with a forbidden transition in Hg. Lapp[12] observed absorption bands associated with forbidden transitions in Cs induced by Xe at atmospheric pressures. The results reported here might indicate that the effects of increased vapor pressure, when the rd is increased, may be overridden by quenching.

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Transition Probabilities and Mean Lives of the $3s_2$ Laser Level in Neon I†

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The mean life of the $3s_2$ (Paschen notation) laser level in Ne I has been measured for a series of pressures in the range of 3–7 μm Hg using a method of delayed coincidence. The lifetime values, obtained by means of the $3s_2 \to 2p_4$ (6328 Å) transition, show an increase in magnitude with increasing pressure. This pressure dependence was interpreted as being due to the imprisonment of the 600.04 Å radiation emitted in the transition of the $3s_2$ level to the ground state. The Holstein theory of the imprisonment of resonance radiation was applied to the analysis of the lifetime-versus-pressure data to yield the following results: $A_{600} = 2.45 \pm 0.20 \times 10^7$ sec^{-1}, $f_{600} = 0.0040 \pm 0.0003$, $\sum 'A = (\sum A - A_{600}) = 1.61 \pm 0.12 \times 10^7$ sec^{-1}, and lifetimes of 25 ± 2 and 62 ± 5 nsec for the low-pressure and high-pressure limits, respectively. The error limits were derived from the standard deviations in the measured lifetimes and the measured pressures. Experimental results of other workers along with theoretical values calculated under various assumptions are presented for comparison with the results of the present work.

INTRODUCTION

Because of the importance of the transition emitting the 6328 Å spectral line in the functioning of the helium-neon laser, measurements have been made of the mean life of the upper level of this transition, the $3s_2$ level (Paschen notation) in Ne I.[1] These measurements were carried out by means of a previously described method of delayed coincidence[2,3] utilizing a pulsed beam of low-energy electrons for excitation and appropriate fast electronics to measure and record the decay times. The lifetimes were obtained by observing the decay of the level in question by means of the $3s_2 \to 2p_4$ (6328 Å) transition. A dependence of lifetime on pressure was detected for pressures