PHYSICAL REVIEW A VOLUME 12, NUMBER 4 OCTOBER 1975

Doublet line-strength ratios for the principal series of cesium*

D. Popescu
Institute of Physics, Bucharest, Romania

Iovitzu Popescu and J. Maurer
University of Bucharest, Bucharest, Romania

C. B. Collins and B. W. Johnson
University of Texas at Dallas, Dallas, Texas 75230

(Received 6 May 1974; revised manuscript received 16 June 1975)

Space-charge amplification of the indirect ionization resulting from photoabsorption in cesium vapor has been shown to offer to conventional techniques of absorption spectroscopy a useful complement of highest sensitivity. It was used in this work together with a xenon continuum source filtered to a linewidth of 0.14 Å to measure the relative absorption probabilities for transitions of the cesium-principal-series ($6\,^2S$-$n\,^2P_{1/2,3/2}$) doublets. Anomalous pressure effects on the resulting line-strength ratios, resulting in part from polarization molecules of Cs_2*, were examined over a range of cesium-atom concentration varying from 1.2×10^{14} to 1.5×10^{15} cm^{-3}. From the corrected doublet line-strength ratios for $11\leq n\leq 16$, the segment of the curve of Fano X parameters corresponding to discrete energies was constructed. Interpolation between these results and those from electron-polarization experiments served to locate the pole in the principal-series, doublet line-strength ratio near $19\,^2P$.

I. INTRODUCTION

Recent studies[1] of the photoionization of spin-oriented alkali-metal atoms, notably, cesium, have pointed the way toward the development of practicable sources of polarized electrons.[2] These same results, together with complementary studies[3,4] of the ionization of unoriented atoms by circularly polarized light, have focused attention upon the importance of adequate characterization of the spin-orbit coupling responsible for these effects. Following Fano's suggestion,[5] the coupling strength has been generally described by the X-parameter,

$$X(E)=(2R_3+R_1)/(R_3-R_1), \qquad (1)$$

where R_1 and R_3 are the radial matrix elements corresponding to the $6\,^2S_{1/2}$-$^2P_j(E)$ transitions to $j=\frac{1}{2}$ and $\frac{3}{2}$, respectively, E is the photon energy and $P_j(E)$ can represent either discrete or continuum P states.

In cesium spin-orbit coupling is strongest and it is the accurate determination of this X parameter as a function of excitation energy which has been the object of recent concern.[6-8] Calculations and experimental measurements reported to date have not converged to the extent desired and the principal experimental measurements[1,3] of $X(E)$ in cesium appear to agree only on the energy of the zero in X at $E_0=4.52$ eV. The corresponding computations have generally[6,8,9] made use of model potentials containing several free parameters and agreement with experiment has been dependent upon the choice of parameter values.

A number of important spin-orbit effects, in addition to polarized electron yields, are critically dependent upon the X parameter and its functional dependence upon excitation energy. These include the shape and depth of the minimum in the photoionization cross-section curve[10] and doublet line-strength ratios for the discrete principal-series transitions in absorption.[11] In most cases[6,8,9] it has been from these spectroscopically observed spin-orbit effects that model-potential parameters have been taken. It appears that the most successful agreement has been obtained with the model potential of Weisheit[6] used to explain the experimental results of Baum *et al.*,[1] but necessarily only for the range of energies above the photoionization threshold.

Perhaps the most sensitive test of such theories, however, is the perturbation ΔR in the matrix element caused by the spin-orbit effects, particularly for values of energy in the neighborhood of the zero in R_1. As can be seen from Eq. (1), the photon energy E_p for which $R_1(E_p)=0$ gives $X(E_p)=2$, and corresponds to a pole in the oscillator line-strength ratio ρ, where

$$\rho=f_{3/2}/f_{1/2}\simeq2(X+1)^2/(X-2)^2. \qquad (2)$$

Interpreted in these terms, the two principal experimental values from electron polarization measurements have given contradictory values indicating either $E_p-E_i=0.6$ eV (Refs. 3 and 4) or $E_p-E_i<0$ (Refs. 1 and 2), where E_i represents the threshold energy for ionization. Corresponding theory has indicated $E_p \simeq E(16\ ^2P)$ (i.e., $E_p - E_i < 0$) with a model potential normalized principally[6] by the experimental value of $E_0 = 4.52$ eV and, conversely, $E_p - E_i \simeq 0.01$ eV when normalized[8] principally by the experimentally observed term values, $E(6\ ^2P_{1/2})$ and $E(6\ ^2P_{3/2})$. As reviewed recently by Norcross,[8] direct spectroscopic measurements of line-strength ratios reported in the literature[12,13] do not tend to resolve this discrepancy, nor even indicate whether the pole lies above or below the photoionization limit. Experimental difficulties in obtaining both adequate resolution and sensitivity, together with an anomalous dependence of line-strength ratio on buffer-gas pressure in the vapor cell,[13] limited measurements of $\rho(E)$ to $E < E(12\ ^2P_{3/2})$ in experiments reported before 1973. These results were strongly divergent at the higher of those energies, with one investigation[12] indicating $\rho(10\ ^2P)$ to be a sharp maximum and the other[13] indicating a function monotonically increasing with energy. For $12\ ^2P$ the discrepancy in ρ between the two measurements is an order of magnitude.

As mentioned above, the recent emphasis thrown on this topic by the interest in polarized electron effects has led to three important remeasurements of the line-strength ratio in cesium. First, the measurement of Fullop and Stroke[14] tended to confirm the early Sambursky[12] results with the report of a maximum at relatively low n, in this case $n = 11\ ^2P$. Second, the more recent measurements of Pichler,[15] among the most carefully done, were the first to report a continuously increasing ratio to $n = 18$ in qualitative agreement with theory.[6,8] Though these measurements encompassed observations over a wide range of particle densities, sensitivity was low and measurement at the higher values of n could only be done at pressures sufficient to merge components of the $n = 19$ doublet by pressure broadening. Finally, a third measurement[16] to $n = 15$ conducted in a multipass vapor cell containing a very large (100 Torr) pressure of buffer gas again gave qualitative agreement with the theoretical trend, but with ratios generally reduced by 20% from those of Pichler.

The paper reports the direct reexamination of the doublet line-strength ratio for the principal series of cesium. Using the space-charge diode detection scheme, recently shown to have adequate sensitivity and resolution to record the two-photon $6\ ^2S - n\ ^2D$ absorption spectrum of cesium,[17] the relative absorption probabilities for the $6\ ^2S_{1/2} - n\ ^2P_{1/2}$ and $6\ ^2S_{1/2} - n\ ^2P_{3/2}$ transitions were measured to $n = 16$ and detected to $n = 18$. It is a particular characteristic of this technique that sensitivity, and thus accuracy, increases with increasing n. Anomalous pressure effects were observed and corrected and the portion of the curve of $X(E)$ corresponding to discrete energies was constructed. It was found to smoothly join that obtained from the Weisheit theory[6] for $E > E_i$ and the Baum *et al.*[1] experimental results. Interpolation served to locate the pole in the line-strength ratio at $E_p \simeq E(19\ ^2P)$.

II. EXPERIMENTAL METHOD

The apparatus of this experiment was essentially that used earlier[18] for the measurement of spectroscopic constants of cesium $n\ ^2P$ and $n\ ^2D$ states in the limit of large n, but further refined to optimize sensitivity and resolution. In contrast to the techniques of standard absorption spectroscopy, which are based on the measurement of small attenuations of large signals, the method used in these works is based on the detection of the positive ions produced directly or indirectly from the photon absorptions. These ions then enhance the output signal of a thermionic diode containing the absorbing vapor by partially neutralizing the negative space charge surrounding a heated diode filament adjacent to the absorbing volume. The relatively longer time which the positive ions require to cross the potential well of the space-charge region provides for the release of $10^4 - 10^6$ electrons per ion from that region.[19,20] Essentially, this technique, when coupled with light modulation and synchronous detection, represents a modernization of the early experiments on photoionization and now forms a valuable complement to conventional absorption spectroscopy. As currently implemented it is sufficient to allow detection of ion signals as small as a few ions per second.

Details of the experimental arrangement are found in previous reports of the related work[18] and only the general outline of the experimental method will be repeated here. The absorption cell itself consisted of a Pyrex glass envelope with quartz windows and an included detecting diode composed of a 0.15-mm-diam tungsten-wire filament and a silver-disk anode, both activated with cesium and separated by 1.5 mm. The exact position of the illuminated volume within the diode structure was not particularly critical provided the diode elements were not directly illuminated. The cell was filled with cesium vapor and the pressure was controlled by a two-chamber oven according to standard techniques. The working pressure range corresponded to saturated vapor at temperatures selected from the 140 to 220 °C interval. Optimal filament temperature was around 1100 °K.

The light source employed was a 450-W high-pressure xenon arc lamp, the continuum from which was filtered with a 1-m Czerny-Turner monochromator with a 1200-line/mm grating used in second order. Slits were selected to give 0.14-Å resolution.

III. RESULTS AND DISCUSSION

A typical direct recording of ion signal as a function of wavelength over the region containing the principal-series ($6\,^2S_{1/2}$-$n\,^2P_{1/2,3/2}$) limit is shown for a relatively low vapor pressure of 0.17 Torr in Fig. 1. System resolution can be verified by the observation of discrete lines for different principal quantum numbers to $n = 58$. Data for the lower principal quantum numbers are reproduced in Figs. 2(a) and 2(b), which clearly show the doublet line-strength ratio to be a strongly increasing function of increasing n for $n \leq 16$. Figure 2(b) shows the data for transitions to $n = 17$, 18, and 19, recorded with a sensitivity increased by a factor of about 30 over that of Fig. 2(a). Although the resolution was still adequate to separate the doublet structure to $n = 19$, the rapid decrease with increasing n of intensity of the transition to $n\,^2P_{1/2}$ prevented the accurate measurement of intensity for $n > 17$. However, from the qualitative inspection of Fig. 2, it is clear that a pole in the doublet line-strength ratio may not be expected before $n = 19$, if not $n = 20$.

The quantitative interpretation of the data of Fig. 2 was complicated by a pressure dependence of the line-strength ratio similar to the dependence on buffer-gas pressure reported previously by Agnew.[13] For example, the ratio of detected ion currents observed in this work to result from absorption to $12\,^2P_{3/2}$ and $12\,^2P_{1/2}$ levels were found to vary from 2.5 to 25 as the cesium-atom number density varied from 1.5×10^{15} to 1.2×10^{14} cm^{-3}. Such a pronounced pressure dependence suggested the need to first reexamine the detection sequence.

It is believed that the ionization actually detected resulted from the photoabsorption according to one of the mechanisms suggested by Lee and Mahan[21,22]:

$$\mathrm{Cs}^*(n\,^2P) + \mathrm{Cs} \rightarrow \mathrm{Cs}_2^{**}, \tag{3a}$$

followed by either

$$\mathrm{Cs}_2^{**} \rightarrow \mathrm{Cs}^+ + \mathrm{Cs}^- \quad (\text{for } n \geq 12) \tag{3b}$$

or

$$\mathrm{Cs}_2^{**} \rightarrow \mathrm{Cs}_2^+ + e \quad (\text{for } n \leq 12), \tag{3c}$$

where Cs_2^{**} is a highly excited molecular state. Since Cs_2^{**} represents the particular associative state formed in collisions with ground-state atoms and $n\,^2P_{1/2}$ or $n\,^2P_{3/2}$, it would necessarily differ in the two cases, with possibly different branching ratios between (3b) and (3c). As the over-all detector sensitivity was, itself, a function of pressure, it was necessary to consider possibly different functional dependences on pressure of the sensitivities for the detection of the products of the two branches, (3b) and (3c).

Figure 3 shows as a function of temperature of the saturated vapor the normalized sensitivity of the space-charge diode and associated electronics for the detection of Cs_2^+ and Cs^+ ions. Plotted is the ratio of detected current to absorber number density for the signal resulting from the direct photoionization of the ambient Cs_2 concentration according to

$$\mathrm{Cs}_2 + h\nu \rightarrow \mathrm{Cs}_2^+ + e, \tag{4}$$

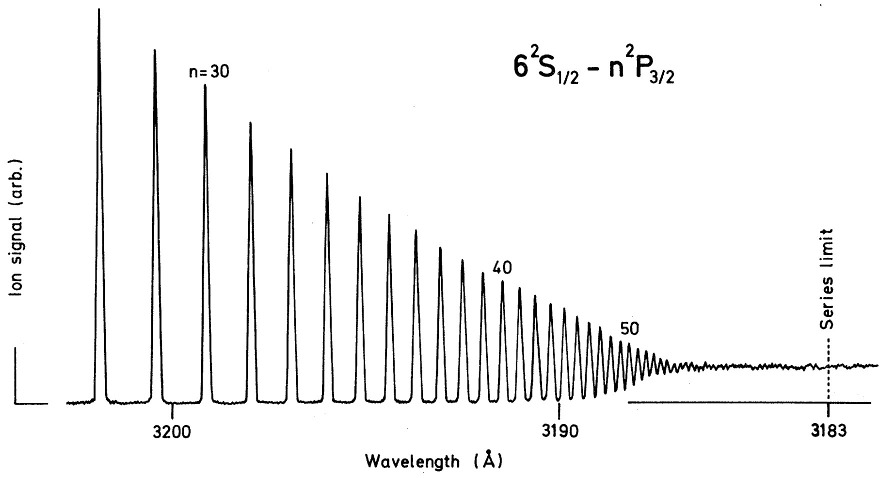

FIG. 1. Typical ion signal spectrum showing the cesium principal series ($6\,^2S \rightarrow n\,^2P$) near the series limit. Clearly resolved lines can be seen for $n \leq 58$ with resolution being limited by the 0.14-Å linewidth of the light source. Pressure of the saturated vapor in the absorption cell was 0.17 Torr.

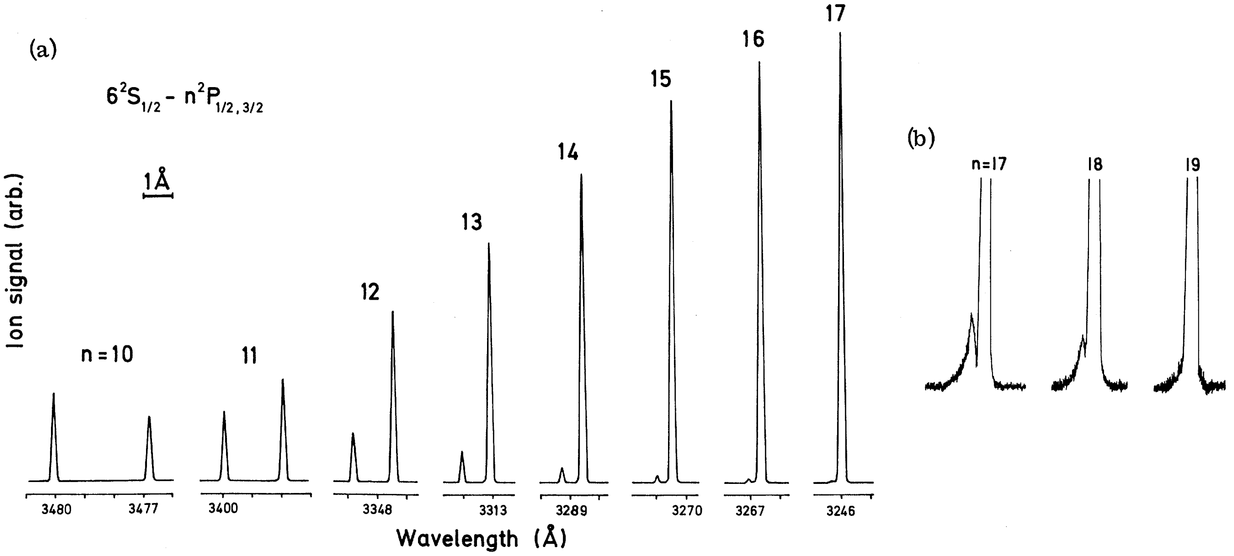

FIG. 2. (a) Ion signal spectrum of the resolved doublets of the principal series for transitions to $n\,^2P_{1/2}$ and $n\,^2P_{3/2}$, where $10 \le n \le 17$, as marked above each pair. The wavelength scale is in Å and is not continuous between doublets of different n. Pressure of the cesium vapor was 0.046 Torr. (b) Ion signal spectrum of the resolved doublets of the principal series for $n=17$, 18, and 19, as marked. Recording sensitivity was 30 times greater than that used to obtain Fig. 2(a).

and the corresponding direct photoionization at shorter wavelength of primarily atomic cesium,

$$\mathrm{Cs} + h\nu \rightarrow \mathrm{Cs}^+ + e. \qquad (5)$$

The similarity of the resulting curves is in agreement with the more qualitative indication of Marr and Wherrett that the diode amplification factor in such devices is independent of ion mobility.[23] The line-strength ratio can then be expressed from (2) in terms of experimentally measured ion signals S_1 and S_3 for absorption to $n\,^2P_{1/2}$ and $n\,^2P_{3/2}$, respectively, as

$$\rho = \left[\frac{I_1 P_i(\frac{1}{2})}{I_3 P_i(\frac{3}{2})}\right]\frac{S_3}{S_1}, \qquad (6)$$

where I_1 and I_3 are the intensities of the illumination and $P_i(\frac{1}{2})$ and $P_i(\frac{3}{2})$, the probabilities of ionization of the resulting P states, for $n\,^2P_{1/2}$ and $n\,^2P_{3/2}$, respectively. This implies that the pressure dependence arises from the terms in brackets. In expression (6) the difference in energy of the two transitions has been neglected in writing the equal sign.

At least three mechanisms might be expected to contribute to the observed pressure dependence. First the intensities of illumination at the two wavelengths of the transitions, although approximately equal at the source, are attenuated by passage through the absorbing layer of cesium vapor surrounding the sensitive space-charge region of the diode. Since $f_{3/2} > f_{1/2}$ over the range of n of interest, the first ratio in brackets in Eq. (6) must have an exponential dependence increasing with pressure, at a rate depending on the difference in f-numbers and the thickness and density of the absorbing layer. The second ratio in

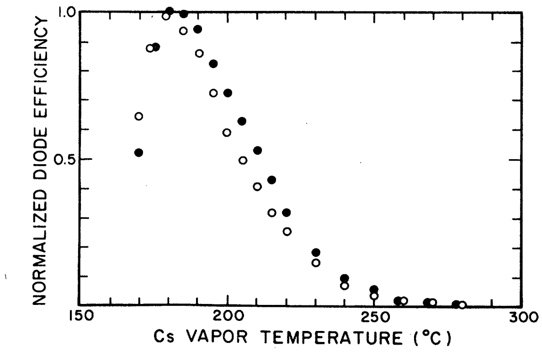

FIG. 3. Normalized curve showing, as a function of the temperature of the saturated vapor, the sensitivity of the detecting diode and associated electronics for the detection of atomic Cs^+ (○), and molecular Cs_2^+ (●) ions. For each curve the ordinate represents the ratio of diode signal to atomic and molecular cesium absorber density, respectively, when illuminated by light of wavelength 3164 and 3280 Å, respectively.

brackets concerns the relative ionization probabilities for $n\,^2P_{1/2}$ and $n\,^2P_{3/2}$. However, the magnitude of spin-exchange cross sections, even for lower excited states,[24] suggests that the populations of $n\,^2P_{1/2}$ and $n\,^2P_{3/2}$ will be mixed by collision within a lifetime for radiative decay and, therefore, that a large difference in $P_i(\frac{1}{2})$ and $P_i(\frac{3}{2})$ could not occur. To assume the contrary requires the unlikely case of the state of much lower P_i having a cross section for collisional deexcitation to a nonionizing quantum level greatly in excess of both the spin-exchange cross section between $\frac{1}{2}$ and $\frac{3}{2}$ for the same n and the cross section for collisional ionization. This, together with the fact that near $X=2$ changes in ρ of even a factor of 2 have negligible effect on the consequent value of X, apparently makes $P_i(\frac{1}{2})=P_i(\frac{3}{2})$ a reasonable approximation from which to derive values of X. Further, the accuracy of this approximation can be expected to increase with n, particularly at values of $n>12$, for which the branching ratio between (3b) and (3c) is less ambiguous.

Finally, a third and more serious source of pressure dependence can be expected near the node in R_1 from the contribution to S_1 of absorptive transitions from van der Waals molecules of cesium. A weak potential minimum in the Cs_2 $(^3\Sigma_u^+)$ state has been shown[25,26] to provide for a significant population of molecules in kinetic equilibrium with the atomic cesium, and transitions from this state to other similar states appear as satellite lines of molecular origin superimposed on the normal atomic absorption lines. Such satellites are neither necessarily displaced from the parent line nor affected by the node in the matrix element R_1, descriptive of the atom, and so could add a significant pressure-dependent component to the otherwise vanishing S_1. Because of this, expression (6) for the line-strength ratio should contain an additional multiplicative correction term $(1-r)^{-1}$, where r is the fraction of S_1 contributed by molecular satellites. Since r approaches 1.0 asymptotically with increasing pressure, this correction factor also becomes an increasing function of pressure and for small r is approximately e^r.

Of the three correction factors needed to relate the measured ratios of ion currents to line strengths as discussed above, the second is negligible for $n>12$, and the first and third suggest the empirical procedure of logarithmically extrapolating ion-current ratios measured as a function of pressure to obtain zero-pressure values, for which correction factors are unity. Some indication of procedural consistency can be obtained from the range of linearity covered by data and the corresponding fractional extent of the extrapolation. Figure 4 presents as a function of number density of cesium atoms the measured values of ion-signal ratios S_3/S_1, plotted logarithmically together with the corresponding extrapolations to zero density. Data can be seen to be most satisfactory in terms of linearity for $12 \leqslant n \leqslant 15$. In the case of $n=12$ the range of linearity spans a decade in ordinate and a factor of 7 in abscissa, with the extrapolation to zero representing only a 15% extension of the linear range. For 13, 14, and 15 the range is more restricted, but the extrapolation, nevertheless, represents less than a 30% extension. The data for $n=16$ suffers from insufficient sensitivity to detect the signal at the lower densities of absorber and the extrapolation to zero indicated in Fig. 4 appears defensible only because of the relatively similar slopes of the linear regions for the higher n values.

Finally, from the inversion of Eq. (2) and the zero intercepts of Fig. 4, the Fano X parameter can be obtained as a function of principal quantum number, and, hence, energy of the upper $n\,^2P$ state. Results are plotted in Fig. 5, together with

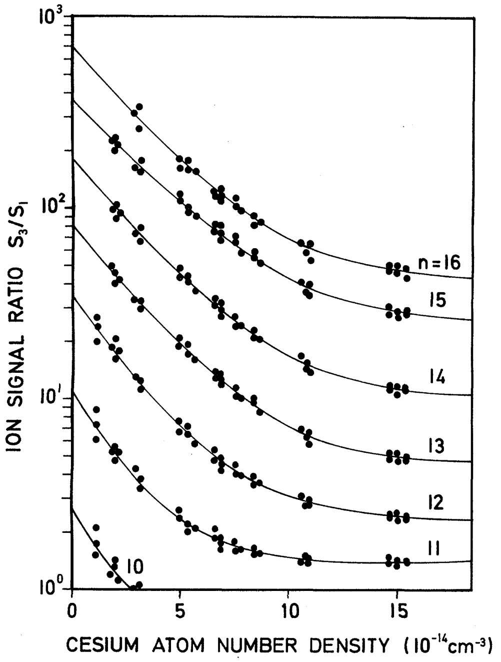

FIG. 4. Logarithmic plot of the ratio S_3/S_1 of diode signal currents S_3 and S_1, corresponding to principal-series transitions to $n\,^2P_{3/2}$ and $n\,^2P_{1/2}$, respectively. Ratios are plotted as functions of cesium-atom number density for $11 \leq n \leq 16$, as marked. Extrapolations to intercept values characteristic of zero density are indicated by solid lines.

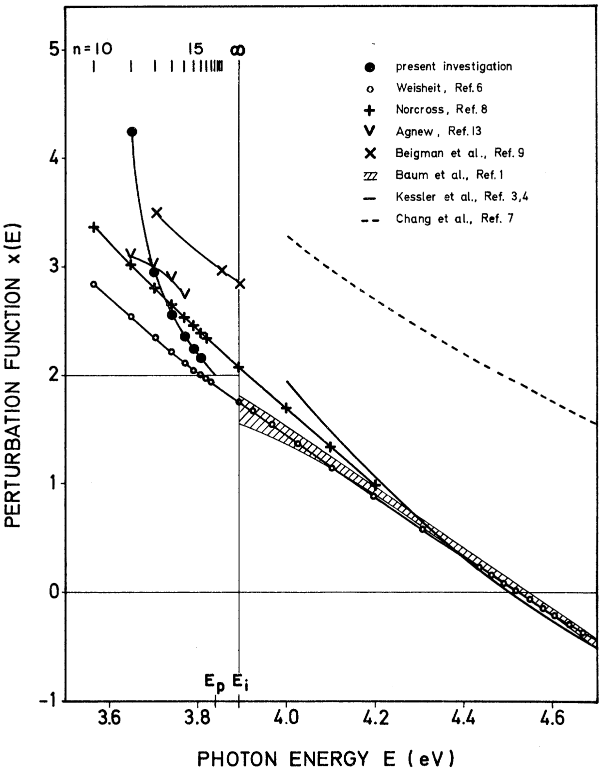

FIG. 5. Graph of Fano X parameter [Eq. (1)] as a function of excitation energy above the ground $6\,^2S_{1/2}$ state. The ionization limit is marked by the vertical line at $E = 3.893$ eV. Results of this work are given by the filled circles and values from the literature are from the sources indicated. The experimental work from Refs. 15 and 16 cannot be resolved on this scale from the theoretical data of Norcross and should be considered represented by the symbols for Ref. 8.

other values from the literature mentioned above. On this scale differences in the results of Norcross, Pichler, and Exton cannot be distinguished and the symbol for the Norcross theory should be considered representative of each.

IV. CONCLUSIONS

It appears from Fig. 5 that the present results are incompatible with the experimental results of Kessler *et al.*[3,4] and the theoretical models of Norcross[8] and Chang and Kelly.[7] Moreover, a closer agreement with either those experiments or theory would require much smaller line-strength ratios, a fact which appears even more difficult to reconcile with the data of Fig. 4. Within the range of linearity for most n, values of S_3/S_1 were measured which give larger ρ and smaller X, even before correction for pressure-dependent effects, than that expected from these other works. This is illustrated in Table I, where line-strength ratios obtained from this work are compared with the more recent experimental values and theory. Disagreement between the values of this work and other experimental measurements though disturbing, *a priori*, is significantly less than the disagreement between theoretical values. In particular, it should be noted that the reliability of the values in this work should be expected to increase with principal quantum number for both empirical and theoretical reasons. From Fig. 4, it can be seen that data for both $n = 10$ and 11 suffer from an inadequate characterization of the low-pressure region from which extrapolation must be performed. This, together with the changing detection sequence from reaction (3b) to

TABLE I. Comparison of the most recent measurements of the oscillator-strength ratio ρ with theoretical calculations.

Principal quantum number	Theory		Experiment			
	Norcross (Ref. 8)	Weisheit (Ref. 6)	Fulop (Ref. 14)	Pichler (Ref. 15)	Exton (Ref. 16)	This work
10	20	42	19	20	20	3
11	31	84	27	30	28	10
12	45	173	18	40	41	33
13	64	408		62	54	80
14	88	1168		77	63	180
15	116	4288		114	91	360
16	149	99 140		142		700
17	188	30 933		186		
18		4645		238		

(3c), makes the anomalously low values obtained for $\rho(10)$ and $\rho(11)$ of little significance. Conversely, as mentioned above, data for $\rho(15)$, for example, is particularly well defined and can be seen to have risen to 220 at the lowest pressure examined. This strongly suggests that the value of $\rho(15) > 220$ and the extrapolation to the value given in Table I is clearly indicated.

Though not agreeing with the Weisheit calculation[6] in the region $E < E_i$, the results of this work extrapolate to join smoothly those calculations for energies above the ionization limit. As mentioned previously, in that region occurs the best agreement between theory[6] and previous experiment. It therefore appears warranted to interpolate between the Weisheit theory and the present results to locate the energy E_p of the pole in the line-strength ration.

One such interpolative curve fitting the present results for $11 \leqslant n \leqslant 16$, the Weisheit theory for $E \geqslant E_i$, and consistent with the experiment of Baum *et al.*[1] is shown in Fig. 5. The intercept with the line, $X = 2$, locating the pole can be seen to occur between 3.835 and 3.845 eV, giving $E_p \cong 3.840$ eV. The closest discrete energy levels are the $19\,^2P$ levels at 3.838 eV, so that the pole in line-strength ratio can be concluded to occur at an energy $E_p \cong E(19\,^2P)$. Since the energy of this pole in the line-stength ratio is one of the spin-orbit effects most sensitive to assumed theoretical parameters, it can be reasonably assumed that the experimental location of the pole near the $19\,^2P$ levels now provides an additional parameter useful for the further refinement of theoretical model potentials.

ACKNOWLEDGMENT

The authors gratefully acknowledge the advice and encouragement of Dr. M. V. McCusker.

---

[*] This work was conducted as part of the U. S.–Romania Cooperative Program in Science and Technology in association between The University of Texas at Dallas and the Institute of Physics of Bucharest. Financial support was provided in part by the U. S. National Science Foundation under Grant No. GF-443 and in part by the Comitetul de Stat pentru Energia Nucleara and the Consiliul National pentru Stiinta si Technologie of Romania.

[1] C. Baum, M. S. Lubell, and W. Raith, Phys. Rev. Lett. 25, 267 (1970).

[2] V. W. Hughes, R. L. Long, Jr., M. S. Lubell, M. Posner, and W. Raith, Phys. Rev. A 5, 195 (1972).

[3] J. Kessler and J. Lorenz, Phys. Rev. Lett. 24, 87 (1970).

[4] U. Heinzmann, J. Kessler, and J. Lorenz, Z. Phys. 240, 42 (1970).

[5] U. Fano, Phys. Rev. 178, 131 (1969).

[6] J. C. Weisheit, Phys. Rev. A 5, 1621 (1972).

[7] J. J. Chang and H. P. Kelly, Phys. Rev. A 5, 1713 (1972).

[8] D. W. Norcross, Phys. Rev. A 7, 606 (1973).

[9] I. L. Beigman, L. A. Vainshtein, and V. P. Shevelko, Opt. Spectrosc. 28, 229 (1970).

[10] M. J. Seaton, Proc. R. Soc. A 208, 418 (1951).

[11] E. Fermi, Z. Phys. 59, 680 (1930).

[12] S. Sambursky, Z. Phys. 49, 731 (1928).

[13] L. Agnew, Bull. Am. Phys. Soc. 11, 327 (1966).

[14] G. F. Fulop and H. H. Stroke, in *Atomic Physics III, Proceedings of the Third International Conference on Atomic Physics, Boulder 1972* (Plenum, New York, 1973).

[15] G. Pichler, Seventh Yugoslav Symposium on Physics of Ionized Gases, Rovinj, Yugoslavia, 1974, pp. 261–263 (unpublished).

[16] R. J. Exton, J. Quant. Spectrosc. Radiat. Transfer (to be published).

[17] D. Popescu, C. B. Collins, B. W. Johnson, and Iovitzu Popescu, Phys. Rev. A 9, 1182 (1974).

[18] D. Popescu, M. L. Pascu, C. B. Collins, B. W. Johnson, and Iovitzu Popescu, Phys. Rev. A 8, 1666 (1973).

[19] K. H. Kingdon, Phys. Rev. 21, 408 (1923).

[20] G. Z. Hertz, Z. Phys. 18, 307 (1923).

[21] Yuan-Tseh Lee and B. H. Mahan, J. Chem. Phys. 42, 2893 (1965).

[22] Yuan-Tseh Lee and B. H. Mahan, J. Chem. Phys. 42, 2016 (1965).

[23] G. V. Marr and S. R. Wherrett, J. Phys. B 5, 1735 (1972).

[24] C. K. Chang and R. H. Walker, Phys. Rev. 178, 198 (1969).

[25] K. Niemax, Phys. Lett. 38A, 141 (1972).

[26] C. B. Collins, B. W. Johnson, M. Y. Mirza, D. Popescu, and Iovitzu Popescu, Phys. Rev. A 10, 813 (1974).