Magnetic field dependence of cross sections for collisional disorientation of $6\,^2P_{1/2}$ cesium atoms*

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The magnetic field dependence of the cross sections for the disorientation of $6\,^2P_{1/2}$ cesium atoms, induced in Cs-He collisions, has been studied using Zeeman scanning techniques. A Cs-He mixture, contained in a fluorescence cell which was located in a magnetic field variable from 0 to 10 kG, was irradiated with the circularly polarized 8943-Å component of cesium resonance radiation in a direction parallel to the field, and the polarization of the resonance fluorescence was monitored in the backward direction in relation to the helium pressure and to the magnetic field strength. The cross sections, which are in good agreement with the results obtained previously at high and low magnetic fields, exhibit a variation with the magnetic field, which is in accordance with theoretical predictions.

I. INTRODUCTION

Disorientation and disalignment of ^2P alkali-metal atoms by collisions with noble-gas atoms has been the subject of many theoretical and experimental investigations. General theoretical treatments of excited-state relaxation were developed by D'yakonov and Perel,[1] Omont,[2] and Wang and Tomlinson,[3] while Franz *et al.*[4,5] and Elbel and Naumann[6] derived the $m_J \not\leftrightarrow -m_J$ selection rule for collisional transfer between Zeeman substates which, however, was not substantiated by more recent theoretical studies. The extensive experimental results have been obtained in several laboratories engaged in studies of collisional depolarization of atomic resonance fluorescence. The Zeeman-scanning method was used by Krause and co-workers, who studied collisional disorientation of the $4\,^2P$ states of potassium[7,8] and the $^2P_{1/2}$ state of cesium,[9] while Gallagher[10] employed the Hanle-effect method to investigate the $5\,^2P$ states of rubidium and the $6\,^2P_{1/2}$ state of cesium. Subsequently, Bulos and Happer[11] showed that Hanle signals could be considerably distorted by nuclear spin effects and that the collisional relaxation rates obtained from such signals lead to values of disorientation cross sections for the $^2P_{1/2}$ states, which have been decreased by a factor of at least 3, as the result of the "nuclear flywheel" effect.

Franz and Sooriamoorthi[12] used white-light optical-pumping techniques in conjunction with the nuclear-spin-decoupling approximation, and determined the cross sections for disorientation of $6\,^2P_{1/2}$ cesium atoms, induced in collisions with helium and neon atoms, corrected for nuclear spin and at essentially zero magnetic field. Unfortunately, there is considerable disparity between their results and those yielded by Hanle-effect measurements (corrected for nuclear spin), even though there is reasonable agreement between the optical-pumping data and those obtained from Zeeman-scanning measurements performed at kilogauss fields (9.8 kG).

The lack of agreement between the two sets of disorientation cross sections measured at zero field is perplexing, especially since both had been subjected to corrections for the effects of nuclear spin. It is also possible that the agreement between the zero-field cross sections obtained from optical pumping and those measured at 10 kG is fortuitous, because it might be expected that the influence of the magnetic field on the disorientation process would manifest itself as an increase in the disorientation cross sections. It has been suggested that such an effect might exist and might be indirectly due to fine-structure mixing between the ^2P states, induced by the magnetic field.[13]

In order to test this hypothesis and to resolve the existing discrepancy between the cross sections, it was decided to determine cross sections for the disorientation of cesium $6\,^2P_{1/2}$ atoms induced in collisions with He, over a range of magnetic fields from 0 to 8 kG, by monitoring the depolarization of cesium resonance fluorescence in relation to He pressure, as described in the subsequent paragraphs.

II. THEORETICAL

A mixture of cesium vapor at low pressure mixed with helium, contained in a fluorescence cell, and located in a variable magnetic field. was irradiated with circularly polarized 8943-Å cesium resonance radiation in a direction parallel to the field. The resonance fluorescence was observed in the backward direction, and its degree of circular polarization was monitored in relation to the helium pressure at various field strengths. When the cesium atoms are being continuously excited to the $6\,^2P_{1/2}$ state and undergo spontaneous decay and collisional disorientation, the quasi-equilibrium value for their orientation, $\langle J_z\rangle_e^{eq}$, is given by the following expression which does not include the effects of nuclear spin:

$$\langle J_z\rangle_e^{eq} = \frac{1}{2}\frac{1}{1+Z\tau} \quad . \tag{1}$$

Equation (1) is equivalent to the Stern-Volmer equation[14]

$$P = P_0 \frac{1}{1+Z\tau}, \tag{2}$$

where P_0 is the polarization at zero He density, P is the polarization corresponding to helium density N, τ is the lifetime of the Cs $^2P_{1/2}$ state (3.4×10^{-8} sec), and Z is the collision number (frequency of disorienting collisions per $^2P_{1/2}$ atom):

$$Z = NQv_r . \tag{3}$$

v_r is the average relative speed of the colliding atoms, and Q is the disorientation cross section. A disorientation cross section may be obtained by fitting Eq. (2) to an experimental plot of the degree of circular polarization of the resonance fluorescence against helium pressure.

Equation (1) makes no provision for the effect of the nuclear spin on the relaxation process, which can be important at low magnetic fields. It is well known that, when the experimental determination is based on the Hanle effect, failure to consider nuclear spin effects may lead to differences of several hundred percent between the measured and true cross sections. However, the effect of nuclear spin on depolarization experiments is much less pronounced, and differences between the actual and measured values amount to less than 25%.

Franz and Sooriamoorthi[15] derived quasi-equilibrium values for $\langle J_z\rangle_e^{eq}$, corresponding to various values of I, the nuclear-spin quantum number. For $^2P_{1/2}$ cesium atoms ($I=\frac{7}{2}$),

$$\langle J_z\rangle_e^{eq} = \tfrac{1}{2}(11+Z\tau)[(1+Z\tau)(32+Z\tau)]^{-1}. \tag{4}$$

It is assumed in this treatment, which applies to the case of zero magnetic field, that the optical pumping is weak and that the collision frequencies are lower than the hyperfine frequency. These criteria were fulfilled in the present investigation by using a low-power spectral lamp and restricting He pressures to less than 10 Torr.

Under single-collision conditions which exist at helium pressures of less than 10 Torr, the disorientation cross sections for Na and K, which have $I=\frac{3}{2}$, are affected by nuclear spin to the extent of 20%, but in the cesium cross section ($I=\frac{7}{2}$) the effect does not exceed 5%. Thus, in the present investigation, the Stern-Volmer equation could still be employed in the analysis of the depolarization data, even at zero magnetic field. However, it was considered preferable to use Eq. (4) for the analysis of the zero-field results, but for the results obtained at high magnetic fields, the Stern-Volmer equation was considered entirely satisfactory. A more detailed analysis of the various processes involving the populations of specific Zeeman sublevels would be rather tedious. In view of the complexities of such an analysis and the established validity of the nuclear-decoupling approximation which forms part of the basis of Eq. (4), such a detailed analysis was deemed unnecessary.

III. EXPERIMENTAL

The arrangement of the apparatus used in this investigation is shown in Fig. 1. Cesium resonance radiation emitted from a spectral lamp located in a magnetic field[16] was passed through an interference filter and circular polarizer, and the resulting 8943-Å, σ^+ component was brought to a focus in a fluorescence cell containing cesium vapor at a pressure of 10^{-6} Torr, mixed with helium as a buffer gas. The cell was mounted between the poles of a 12-in. electromagnet which generated a magnetic field parallel to the exciting light beam. The resonance fluorescence was observed along the direction of the magnetic field through an aperture in the pole-piece of the magnet, and was detected with a refrigerated ITT

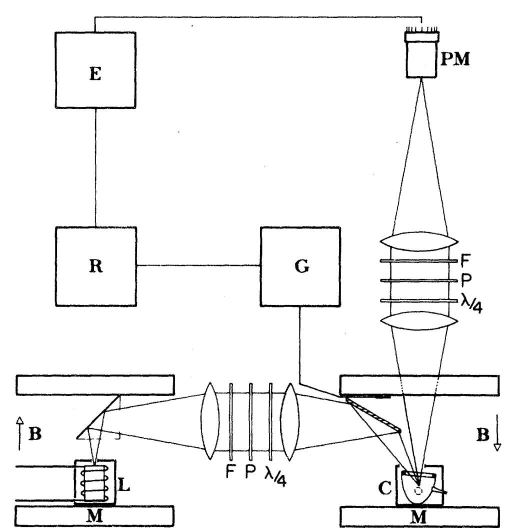

FIG. 1. Arrangement of the apparatus. L, spectral lamp; C, fluorescence cell; both are in magnetic fields of electromagnets M. F, interference filters; P, polaroids; λ/4, quarter-wave plates; G, Gaussmeter; PM, photomultiplier; E, electrometer; R, x–y plotter.

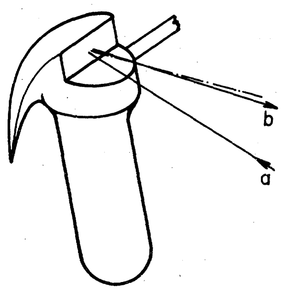

FIG. 2. Sketch of fluorescence cell, showing the window, side-arm, and connecting tube. The directions a and b indicate excitation and observation, respectively, and the dashed line corresponds to the direction of the magnetic field.

FW118 photomultiplier tube whose output, after amplification, was applied to the y-axis of an x-y plotter, the x-axis of which was connected to the output of the magnetometer. The 8943-Å component of the cesium resonance doublet was isolated in the exciting and fluorescent light with a spectral purity of 0.1% by means of interference filters. The inclusion of circular polarizers in the exciting and fluorescent light beams permitted the excitation of the $m_J = +\frac{1}{2}$ Zeeman substate of the $6\,^2P_{1/2}$ cesium atoms by means of σ^+ light, and the separate observation of the fluorescent σ^+ and σ^- components resulting from the decay of the $m_J = +\frac{1}{2}$ and $m_J = -\frac{1}{2}$ Zeeman substates, respectively; the relative intensities of the σ^+ and σ^- components could be recorded directly in relation to the magnetic field strength.

The Pyrex fluorescence cell, which is depicted in Fig. 2, included a "Wood's horn" to decrease the intensity of stray light. The axis of the cell window was tilted at about 5° to the direction of the exciting light beam to reduce reflections from surfaces. The region of fluorescence was restricted to a small volume (and thus small optical depth) by the configuration of the window and the steeply angled Wood's horn. Such construction of the cell, together with the very small cesium vapor density, rendered negligible the effects due to imprisonment of cesium resonance radiation. The fluorescence tube, together with its side-arm which contained liquid cesium, was mounted in a small two-compartment oven which maintained the cell and side-arm at preset temperatures to within 1 and 0.3°C, respectively, over long periods of time. The body of the cell was connected by a 4-mm Pyrex tube to a vacuum and a gas-filling system from which helium could be admitted as required. Gas pressures were measured with a CVC GM 100A trapped McLeod gauge.

Considerable attention was given to ensure that the cesium and helium samples should be free of contamination. Cesium metal of 99.95% purity, supplied by the A.D. McKay Co. of New York, was subjected to prolonged heating under vacuum before the start of the experimental runs. The helium (research grade) was obtained from the Linde Co. and was subjected to gettering with hot cesium vapor for several days before being admitted to the fluorescence cell.

It was found convenient to carry out the experiments at low and high magnetic fields under somewhat different conditions and in slightly different ways. In the range 0-6.3 kG, the spectral lamp was kept in a field of 0.40 kG and the photomultiplier tube was used in a dc mode, with the output being registered with a Keithley picoammeter equipped with a "zero-suppression" feature. When measurements were carried out at higher field strengths, the field surrounding the lamp was increased to 8 kG to produce coincidence between the Zeeman components in emission and absorption and increase the efficiency of excitation. The fluorescent light was chopped and the photomultiplier output was connected to a lock-in amplifier. The wheel of the chopper was fitted with four pairs of polaroid sheets of alternatingly crossed orientations, so that the output of the lock-in amplifier was proportional to the intensity sum $I_{\sigma^+} + I_{\sigma^-}$ when monitored at the chopping frequency, and to the difference $I_{\sigma^+} - I_{\sigma^-}$ at one-half the chopping frequency.

To eliminate the effect of stray magnetic fields on the photomultiplier, the tube was shielded by two netic-conetic shields. Before each experimental run, the photomultiplier signal was carefully scrutinized as a large magnet was moved to various positions in the immediate vicinity of the photomultiplier. On at least one occasion, one of the shields had to be replaced as its effectiveness had diminished demonstrably.

IV. RESULTS AND DISCUSSION

To verify the degree to which imprisonment of resonance radiation affected the experimental results, a preliminary experiment was carried out, in which the intensity and degree of polarization of the resonance fluorescence in pure cesium vapor were monitored in relation to the vapor pressure. Effects of radiation trapping were evident above about 2×10^{-6} Torr at low magnetic fields, but at high magnetic fields no such effects could be detected at even higher vapor pressures. Consequently, all the experimental data used to calculate the depolarization cross sections at low magnetic fields were obtained at a cesium pressure of 1.7×10^{-6} Torr. Some measurements were also carried out at 2.6×10^{-6} Torr and yielded

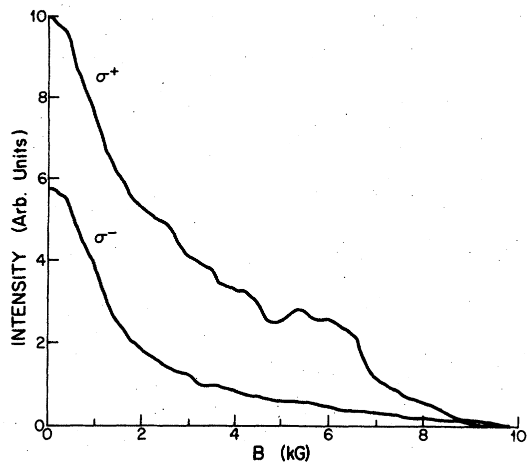

FIG. 3. Traces of magnetic-field scans of the fluorescence observed parallel to the magnetic field. The spectral lamp was located in a 0.4-kG field.

cross sections which agreed with the lower pressure values within experimental error; the experiments at high fields were carried out at 4.6×10^{-6} Torr.

In the course of the experiments, the relative fluorescent intensities $I_{\sigma+}$ and $I_{\sigma-}$, or their differences, were recorded in repeated magnetic-field scans of the fluorescent spectrum at various helium pressures in the range 0–10 Torr. The resulting degrees of polarization $P=(I_{\sigma+}-I_{\sigma-})/(I_{\sigma+}+I_{\sigma-})$ were fitted by the method of least squares, either to Eq. (1) at high magnetic fields or to Eq. (4) at low fields. The least-squares analyses which also involved Eq. (3) yielded the degrees of polarization P_0, corresponding to zero He pressures, the disorientation cross sections Q and their rms deviations. In determining each cross section, at least 25 measurements of the degrees of polarization P were carried out, together with the corresponding He pressures, and the data were acquired during many experimental runs to ensure reproducibility.

Figure 3 shows the reproduction of recorder traces obtained in a magnetic-field scan of the relative fluorescent intensities $I_{\sigma+}$ and $I_{\sigma-}$, at zero He pressure and with the spectral lamp in a 0.4-kG field. The fluorescence cell was at a temperature of 316°K, and the cesium vapor pressure was 1.7×10^{-6} Torr. A series of such scans was carried out at various helium pressures up to 10 Torr. The spectral intensities were recorded with the aid of the electrometer amplifier. This method was useful only at magnetic fields below 6.5 kG because, as may be seen, the fluorescent intensities decreased significantly at higher fields. Figure 4 shows magnetic-field scans of the intensity differences $I_{\sigma+}-I_{\sigma-}$ at two helium pressures, obtained at a temperature of 338°K and cesium vapor pressure of 4.6×10^{-6} Torr. The spectral lamp was located in an 8-kG field. Several such traces were obtained at various helium pressures, and were used to determine the disorientation cross sections at magnetic fields above 5 kG, at which there is already considerable Zeeman split-

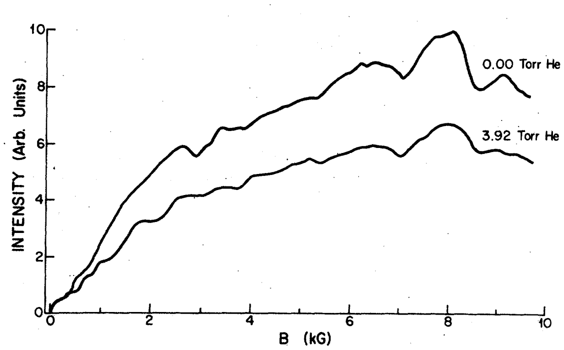

FIG. 4. Traces of magnetic-field scans of the fluorescence signal $I_{\sigma+}-I_{\sigma-}$ at 0- and 3.92-Torr He. The spectral lamp was located in a 8.0-kG field.

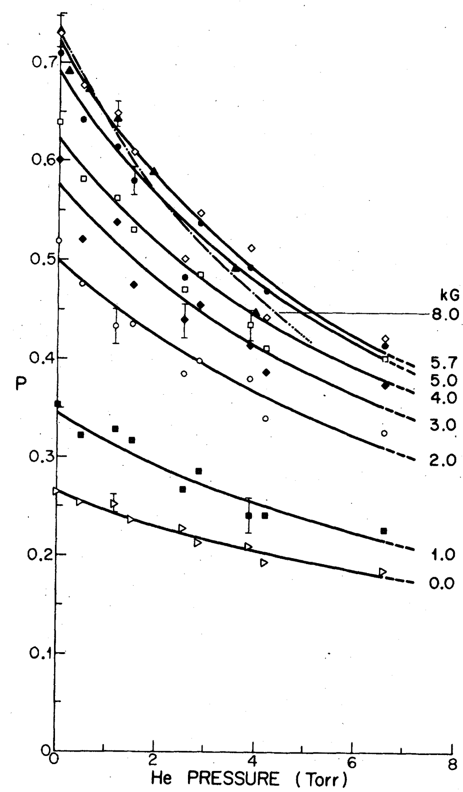

FIG. 5. Variation of the degree of polarization with helium pressure at various magnetic fields as indicated, in kG. The error bars are typical of each set of results, and the curves represent fits of Eq. (4) to the experimental data obtained at fields below 6.3 kG, and of Eq. (1) at stronger fields.

TABLE I. Cross sections for disorientation of $6\,^2P_{1/2}$ Cs atoms, induced in Cs-He collisions.

Source	B (kG)	Cross section (Å^2)
This investigation	0–8	6.0 ± 0.4–11.1 ± 1
Guiry and Krause (Ref. 9)	9.8	11.8 ± 0.5
Bulos and Happer (Ref. 11)	0	6.1
Franz and Sooriamoorthi (Ref. 12)	0	12.5 ± 4
Gordeev *et al.* (Ref. 17)	0	9.0 (theoretical)

ting of the absorption line. The most efficient excitation of the resonance fluorescence would be expected when the lamp and cell are located in magnetic fields of equal strengths, to produce coincidence between the Zeeman components in the exciting light and in absorption. The field of 8 kG was found experimentally to provide the best compromise in this respect, for the purpose of the experiments at high fields.

The degrees of polarization P were obtained by combining experimental data such as are presented in Figs. 3 and 4 with the total fluorescent intensity $I_{\sigma^+} + I_{\sigma^-}$. The latter was monitored separately and, at any particular magnetic-field strength, was found to remain constant with He pressure in the range 0–10 Torr, indicating a negligible effect of pressure broadening on the measurements. Figure 5 shows several plots of the degree of polarization against He pressure, each corresponding to a particular value of the magnetic field. The density of the points, their scatter, and indicated deviations are typical of all the data which were taken off the recorder traces at 0.3-kG intervals.

The disorientation cross sections determined in the course of this investigation are listed in Table I, together with experimental and theoretical

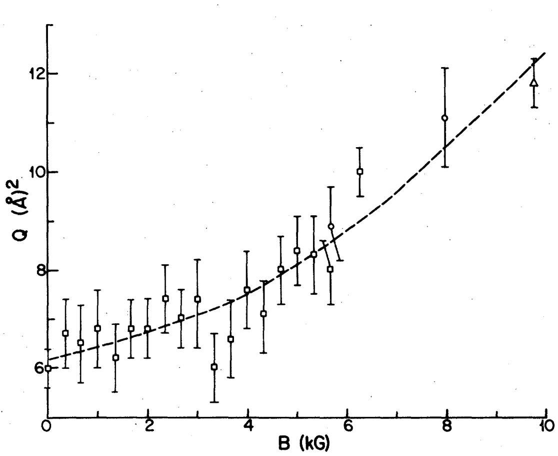

FIG. 6. Variation of the disorientation cross section with magnetic field: ○, spectral lamp at 8 kG; □, spectral lamp at 0.4 kG; △, value from Ref. 9.

values reported elsewhere. As may be seen in Fig. 6, our results indicate that the cross sections exhibit a magnetic-field dependence in the range 0–8 kG, and are in good agreement with a value determined previously at 9.8 kG.[9] The zero-field cross section agrees well with that quoted by Bulos and Happer,[11] who corrected Gallagher's[10] earlier values for the effect of nuclear spin, but is significantly lower than the value calculated by Gordeev *et al.*[17] We cannot offer an explanation for the discrepancy with the zero-field cross section found by Franz and Sooriamoorthi[12] which, though in agreement with high-field values, substantially exceeds the other low or zero-field cross sections. We hope that additional experiments on collisional depolarization of cesium resonance radiation, which are now in progress in this laboratory, will contribute to the elucidation of this problem.

It has been suggested that the magnetic-field dependence of disorientation cross sections might be caused by magnetic-field-induced $^2P_{1/2} - {}^2P_{3/2}$ mixing, which contributes to the depolarization mechanism.[13] The observed variation of the cross section with magnetic-field strength, as shown in Fig. 6, may be represented by the relation

$$Q_B = Q_0 + KB^{1.8}, \qquad (5)$$

where Q_B and Q_0 are the cross sections at field B and at zero field, respectively, and K is a constant. This behavior of the cross sections is in good agreement with theoretical predictions.[18]

There is, unfortunately, almost no other infor-

TABLE II. Cross sections for disorientation of $4\,^2P_{1/2}$ K atoms by collisions with noble gases.

Collision partners	Collision cross sections (Å^2) At zero field[a]	At 5 kG[b]
K-He	24 ± 4	46
K-Ne	21 ± 3	39
K-Ar	37 ± 5	52
K-Kr	51 ± 7	79
K-Xe	79 ± 9	108

[a]Reference 19.
[b]Reference 7.

mation available on the magnetic-field dependence of disorientation cross sections. Berdowski and Krause[7] used Zeeman scanning to determine the disorientation cross sections for the $4\,^2P_{1/2}$ state in potassium at 5 kG, which are presented in Table II together with the most recent values obtained at zero magnetic field.[19] It appears that, in the case of potassium, the high-field cross sections are also significantly larger than the zero-field values.

V. ACKNOWLEDGMENT

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