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THE INFLUENCE OF FOREIGN GASES ON THE RESONANT AND SENSITIZED HANLE CURVES IN THE SODIUM D LINES

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Synopsis

Earlier measurements of the Hanle effect in the sensitized fluorescence of sodium vapour have been resumed with improved experimental geometry and over an extended range of the foreign-gas pressure. The results shed some more light upon the interactions which interfere in the production of this effect. The most noteworthy of these interactions appears to be collisional quenching of hyperfine interaction which seldom has been so clearly observed before. The broadening of the resonant D_1 Hanle signal is also measured and hence cross sections for the desorientation of the state $3^2P_{1/2}$ are derived. The values of this cross section are in $Å^2$: 146, 137, 260, 306, 356 for the noble gases He, Ne, Ar, Kr and Xe, respectively.

1. *Introduction.* Orientation transfer may be observed as a concomitant of excitation transfer as was reported by numerous authors$^{1-6}$). It is likewise present, however, in the collisional transformation of states of the same atom. The number of papers which devoted to the latter effect is much more limited^{7,8}). There are only few instances where this effect has been studied, mainly excited sodium and potassium atoms colliding with noble-gas atoms. Nevertheless, the effect has attracted the interest of theoreticians^{9,10,15}) and, therefore, we thought it worthwhile to repeat part of the experiments in order to provide results which are more accurate and easier to interpret.

An ensemble of atoms excited to a certain state and oriented spatially emit unequal amounts of left- and right-handed circularly-polarized light when undergoing spontaneous decay. The rates of left- and right-handed light quanta differ the most when observed parallel or antiparallel to the axis of orientation. Perpendicular to this axis, no difference of these rates exists. If we denote the axis of orientation by x, the difference of the counting rates of right- and left-handed light quanta, $I_r - I_l$, provides a measure for the electronic orientation $\langle J_x \rangle_J$ (*cf.*, for instance refs. 7 and 8). (J is the electronic angular momentum of the state in question.) The electronic orientation can be created through excitation with circularly polarized light or, as in the case considered here, through orientation transfer from another oriented state which during a collision is eventually transformed into that state. In this latter case the orientation may be found so weak that it may be hard to discriminate the signal $I_r - I_l$ from reflections and natural asymmetries of the circular filter settings. If so, discrimination is yet made possible by magnetic depolarization which is commonly known as the Hanle effect. Applying a magnetic field to the oriented ensemble parallel to the z axis makes the orientation precess in the xy plane. When the excitation is steady in time, the signal $I_r - I_l$ will decrease with rising field according to a lorentzian-shaped curve provided the fluorescent beam is directed along the x axis or is located at least in the xz plane. If, however, the fluorescent beam is detected in the y direction or in the yz plane, the signal turns out to be shaped like a dispersion curve. Any position of the detector in the xy plane yields more or less asymmetric curves. This shortcoming which was inherent in earlier measurements[7,8]), was avoided during this work.

It was intriguing to note that the signal obtained in these circumstances is lorentzian-shaped only when the observation is made with resonant fluorescent light, that is to say if excitation and detection are performed with the same atomic state. As soon as more than one state gets involved in the process of excitation and reemission, the signal does not drop monotonically with rising field but intersects the abscissa and even gets inverted before vanishing[+]. This behaviour seems quite commonplace in all the "sensitized" Hanle curves which have been reported to date. The reasons, however, which are held responsible for such a behaviour, are of a greater variety. Baylis[9]) explains the inversion of the signal by considering a hertzian dipole which precesses in a magnetic field while decaying. The speed of the precession and the decay constant at first pertain to the initial atomic state. After a certain time a collision happens and the state suddenly is transformed into another state with different magnetic properties and a different decay constant. The hertzian dipole, however, during the transformation maintains its instantaneous position. Then it continues its motion according to the modified atomic constants. On averaging the angular distribution of the radiation emitted from the final dipole with respect to the time of its creation, Baylis[9]) arrives at a formula which explains the inverted shape of the Hanle signal. This equation [(2) in Baylis's paper] clearly shows that the inversion always occurs if either the decay constants of both states differ or the magnetic properties (g_J-values) differ or both. Thus, his formula comprises earlier results[4,7]) obtained on the ground of one of these assumptions alone.

Baylis expressly exempts from his consideration the case where a nuclear spin is present. A nuclear spin usually gives rise to hyperfine splitting of both the atomic levels. Consequently, from each level various hertzian dipoles contribute

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[+] As this fluorescence is "sensitized" by collisions we shall call the signal due to transferred orientation a "sensitized" Hanle signal.

to the signal which pertain to the various hyperfine sublevels and, accordingly, precess with different speed. The contributions of the various dipoles are usually indiscernible and hence extraordinary theoretical difficulties arise. Also the strength at which the various dipoles are collisionally coupled, depends on the interaction. Hence points of view enter the discussion which are not yet satisfactorily mastered. However, the experimental results themselves show unambiguously which aspects are decisive in the orientation transfer in the presence of hyperfine interaction. It will be outlined subsequently that magnetic breaking of hyperfine coupling and also collisional hyperfine quenching are the dominant aspects in this connection. Hyperfine quenching in particular is a phenomenon which is well-known in connection with perturbed angular correlation. The reader is referred, for instance, to the survey on hyperfine interactions of fast recoil nuclei in a gas given by Sprouse^{11}). In atomic physics, hyperfine quenching has seldom become as apparent as in the experiment reported below.

2. *Experimental.* We used a conventional setup for the measurement of $\Delta m = 1$ Hanle signals. As displayed in fig. 1, it consists of a cell with sodium vapour placed inside a set of Helmholtz coils, whereupon circularly polarized light from a conventional sodium lamp is directed. This creates an orientation $\langle J_x \rangle$ along the x axis in the state $^2P_{1/2}$ or $^2P_{3/2}$ depending on the spectral line

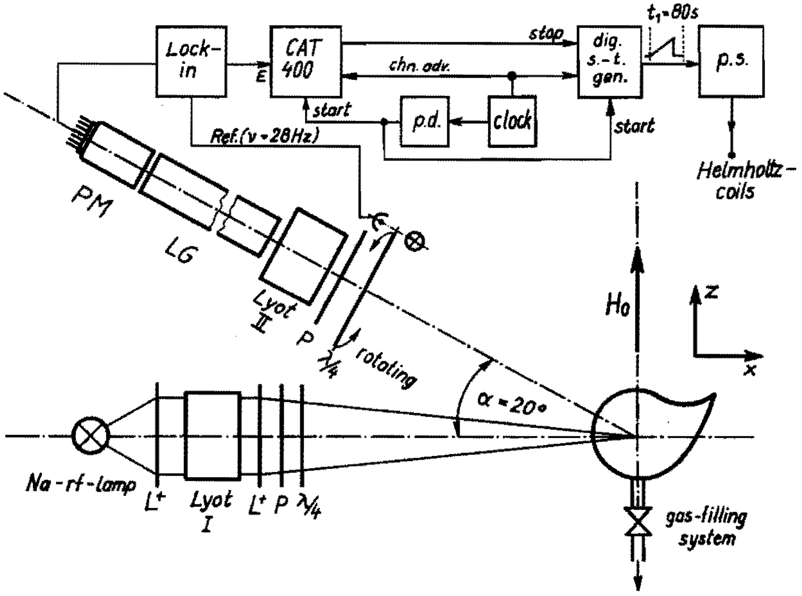


Fig. 1. Arrangement of the apparatus. L, lenses; P, linear polarizers; $\lambda/4$, quarter wave plates; PM, photo multiplier; LG, light guide; pd, presettable divider; chn.adv., channel advance; dig.s.-t.gen., digital saw-tooth generator; p.s., programmable power supply.

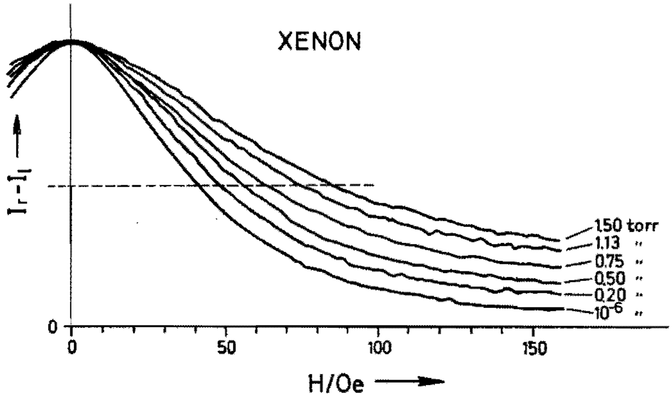
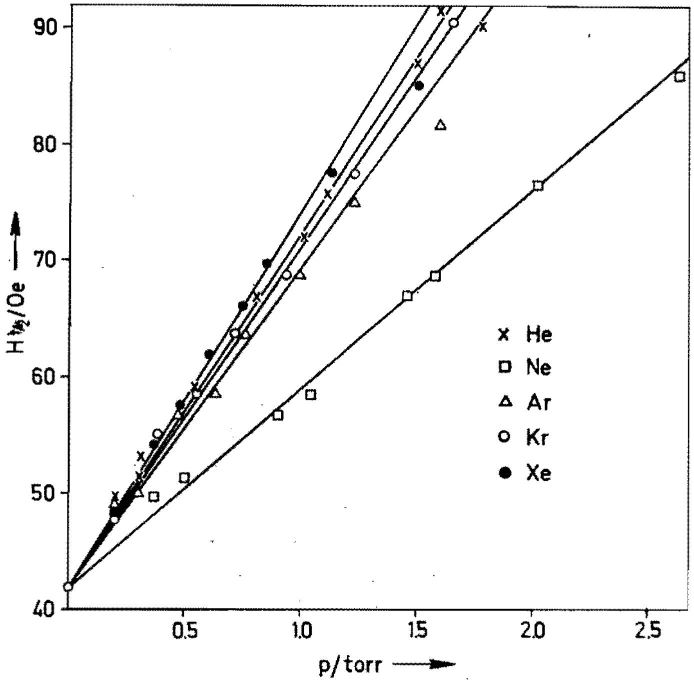


Fig. 2. a) Resonant Hanle curves obtained in sodium D_1 fluorescent light. $I_r - I_l$ denotes the difference of the intensities of right-handed and left-handed circularly polarized light, respectively. The pressure of xenon which was used as a foreign gas here, serves as a parameter. The dotted horizontal line indicates the field strengths $H_{\frac{1}{2}}$ at which the signal dropped to half the zero-field value.



b) A plot of $H_{\frac{1}{2}}$ *versus* the foreign-gas pressure for various noble gases. Straight lines are yielded if the contributions of the hyperfine sublevel $F = I - \frac{1}{2}$ can be neglected compared with the contributions of the level $F = I + \frac{1}{2}$.

D_1 or D_2 which is used for excitation[+]. The existence of an orientation in these states is revealed by unequal amounts of left-handed and right-handed circularly polarized quanta which appear in the fluorescent light. The difference of these amounts is detected by means of a rotating circular analyzer which alternately transmits left-handed and right-handed circularly polarized light. Lock-in techniques then permit the subtraction of the respective photomultiplier signals and the differential signal is then fed into a signal averager of type CAT-400 whose channels are advanced by a clock. The same clock, on the other hand, advances the magnetizing current which is fed into the Helmholtz coils. The latter provide a magnetic field which points into the z direction. Each run of the signal averager lasted 80 s, typically 500 runs constituted one curve. The sodium cell was connected to a vacuum and gas-filling system. The five noble gases were used as sensitizing gases. Gas pressures were measured by means of an aneroid barometer. The gas was cleaned before use by baking it over night in the presence of cesium vapour.

Fig. 2 shows a sequence of resonant Hanle curves obtained with D_1 light used for both excitation and detection. The extreme pressure broadening of the Hanle signal becomes apparent. From this broadening the cross section for desorientation of the electronic angular momentum in the state ${}^2P_{1/2}$ can be derived as outlined in the subsequent section. A similar programme with the state ${}^2P_{3/2}$ must fail because magnetic decoupling of the nuclear spin occurs within the range of the Hanle effect. The curves of the resonant Hanle effect in D_2 light are therefore omitted here.

If D_1 light is used for excitation and D_2 light is detected, the sensitized Hanle signal can be discerned provided the gas pressure in the cell is not too low. A sequence of sensitized Hanle curves is displayed in fig. 3. These sensitized curves are markedly different from the resonant ones. They consist of a positive central peak with negative wings attached to it. Also their behaviour under varying pressure is dissimilar to ordinary Hanle curves. If the foreign-gas pressure is increased the central peak is not broadened. Instead, the central peak decreases much faster than the negative wings until, at a pressure of about 18 torr helium it completely disappears so that the signal looks then like an ordinary, though inverted, Hanle curve. Similar curves are obtained with the other noble gases.

3. *Theoretical.* 3.1. The sensitized Hanle curves. We cannot attempt to provide more than a crude understanding of the experimental findings because of the complexity of the problem. First we state that the ${}^2P_{3/2}$ level has an extremely narrow hyperfine structure, which is only a few tens of megacycles wide. Accordingly a field strength of a few tens of oersteds which we need for efficiently de-

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[+] The separation of the D lines is accomplished by means of a pair of Lyot filters each of which had a spectral resolving power of better than 2 in 10^3.

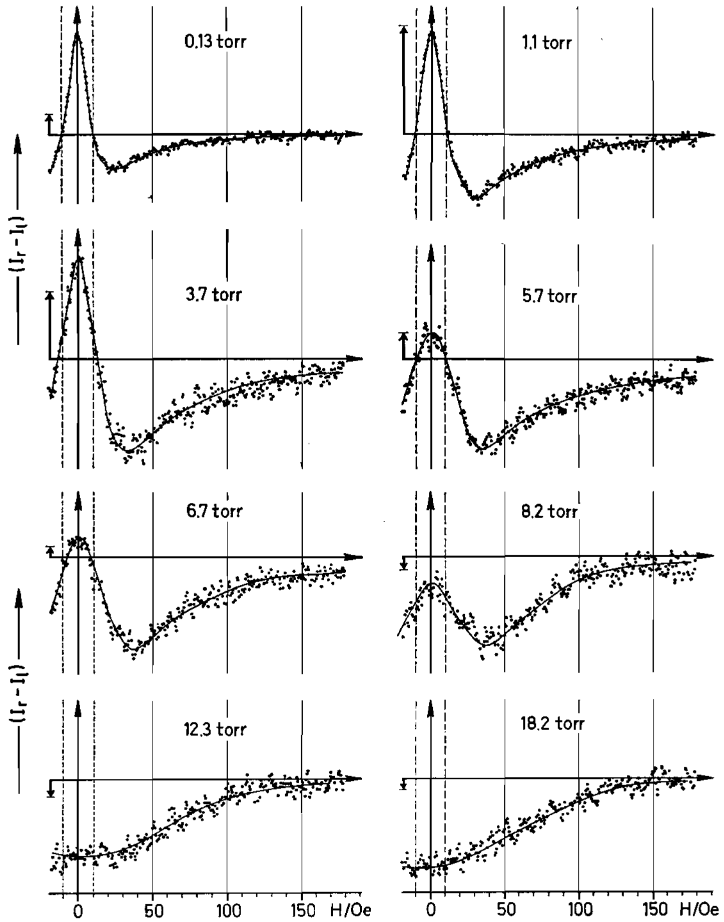

Fig. 3. Sensitized Hanle curves obtained in D_2 fluorescent light. D_1 σ^+ light was used for excitation. Right-handed and left-handed circularly polarized D_2 light was detected. The difference of the respective intensities is plotted *versus* the magnetic field for various pressures of the foreign gas. Helium was used as the foreign gas. The curves are scaled to approximately equal height. The length of the arrow at the left of the centre provides an absolute measure for the zero-field value.

polarizing a state as short-lived as the one in question (1.6×10^{-8} s[12])), suffices also for breaking hyperfine coupling. Hence we find hyperfine coupling essentially unaffected within the range of the central peak, whereas in the range of the wings it proves to be essentially broken.

Let us consider now what effects the nuclear spin has in collision-induced orientation transfer. To this end, let us denote all the initial sublevels of the $^2P_{1/2}$ state with letters J, F, M_F. The final sublevels shall be denoted by j, f, m_f or j, m_j, m_I

depending on the coupling scheme which is realized. By excitation in the initial state a density matrix is created whose elements read $\langle JFM_F \pm 1|\varrho|JFM_F\rangle$. At the cost of these density-matrix elements, the density matrix in the final state is built up from collisional interaction:

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle jfm_f \pm 1|\varrho|jfm_f\rangle = \frac{1}{T}\sum_{F,M_F}\langle jfm_f \pm 1|T^{+}(\boldsymbol{k},\boldsymbol{k}')|JFM_F \pm 1\rangle \times \langle JFM_F \pm 1|\varrho|JFM_F\rangle\langle JFM_F|T(\boldsymbol{k},\boldsymbol{k}')|jfm_f\rangle\big|_{\text{av. over } k,k'} . \tag{1}$$

Here $T(\boldsymbol{k},\boldsymbol{k}')$ stands for the transition operator, T denotes the mean collision time, k, k' denote the relative momenta before and after the collision. On the other hand, with hyperfine coupling broken in the final state, we have

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle jm_j \pm 1\, m_I|\varrho|jm_jm_I\rangle = \frac{1}{T}\sum_{F,M_F}\langle jm_j \pm 1\, m_I|T^{+}(\boldsymbol{k},\boldsymbol{k}')|JFM_F \pm 1\rangle \times \langle JFM_F \pm 1|\varrho|JFM_F\rangle\langle JFM_F|T(\boldsymbol{k},\boldsymbol{k}')|j\, m_jm_I\rangle\big|_{\text{av. over } k,k'} . \tag{2}$$

Multiplying both sides with $\langle jm_jm_I|J_x|jm_j \pm 1\, m_I\rangle$ and tracing over m_I, m_j yields the rate at which $\langle J_x\rangle$ is transferred to the $^2P_{3/2}$ state. Making for $T(\boldsymbol{k},\boldsymbol{k}')$ the easiest assumption possible, namely that it randomizes $\boldsymbol{L}$ without affecting $\boldsymbol{S}$ and $\boldsymbol{I}$ ($\boldsymbol{L}$-randomization model[16]), we find that the rate $\mathrm{d}(\langle J_x\rangle_{J=3/2,\, I \text{ coupled}})/\mathrm{d}t$ is positive and opposite, but equal in magnitude to $\mathrm{d}(\langle J_x\rangle_{J=3/2,\, I \text{ decoupled}})/\mathrm{d}t$, which means

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle J_x\rangle_{J=3/2,\, I \text{ coupled}} = -\frac{\mathrm{d}}{\mathrm{d}t}\langle J_x\rangle_{J=3/2,\, I \text{ decoupled}} . \tag{3}$$

Eq. (3) may well explain why the orientation $\langle J_x\rangle$ transferred to the $^2P_{3/2}$ state is parallel to the one in the initial $^2P_{1/2}$ state, so long as the hyperfine coupling exists, and is inverted as soon as hyperfine coupling is broken. The rate of transfer for I coupled provides a value of the sensitized Hanle curve for $H = 0$, the rate of transfer for I decoupled first must be subjected to magnetic depolarization according to $g_J(^2P_{3/2}) = 4/3$ in order to yield the asymptotic sensitized Hanle curve. In fig. 4 both these limiting cases are drawn. Between them a transition must occur which has been drawn in arbitrarily for the sake of demonstration. We must mention, however, that we have consequently omitted magnetic depolarization in the $^2P_{1/2}$ state whose g_F-values are smaller by a factor of eight than the g_J-value of the state $^2P_{3/2}$.

When comparing these theoretical predictions with the measured curves in fig. 3, the rapid vanishing of the central peak with rising pressure remains unexplained. The reason for that can be seen in a collisional decoupling of I and J, which occurs depending on the coupling scheme which is realized. By excitation in the initial state a density matrix is created whose elements read $\langle JFM_F \pm 1 | \varrho | JFM_F \rangle$. At the cost of these density-matrix elements, the density matrix in the final state is built up from collisional interaction:

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle jfm_f \pm 1 | \varrho | jfm_f \rangle = \frac{1}{T} \sum_{F, M_F} \langle jfm_f \pm 1 | T^+ (\boldsymbol{k}, \boldsymbol{k}') | JFM_F \pm 1 \rangle$$

$$\times \langle JFM_F \pm 1 | \varrho | JFM_F \rangle \langle JFM_F | T(\boldsymbol{k}, \boldsymbol{k}') | jfm_f \rangle |_{\text{av. over } k, k'} . \qquad (1)$$

Here $T(\boldsymbol{k}, \boldsymbol{k}')$ stands for the transition operator, T denotes the mean collision time, k, k' denote the relative momenta before and after the collision. On the other hand, with hyperfine coupling broken in the final state, we have

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle jm_j \pm 1\, m_I | \varrho | jm_j m_I \rangle = \frac{1}{T} \sum_{F, M_F} \langle jm_j \pm 1\, m_I | T^+ (\boldsymbol{k}, \boldsymbol{k}') | JFM_F \pm 1 \rangle$$

$$\times \langle JFM_F \pm 1 | \varrho | JFM_F \rangle \langle JFM_F | T(\boldsymbol{k}, \boldsymbol{k}') | j\, m_j m_I \rangle |_{\text{av. over } k, k'} . \qquad (2)$$

Multiplying both sides with $\langle jm_j m_I | J_x | jm_j \pm 1\, m_I \rangle$ and tracing over m_I, m_j yields the rate at which $\langle J_x \rangle$ is transferred to the $^2P_{3/2}$ state. Making for $T(\boldsymbol{k}, \boldsymbol{k}')$ the easiest assumption possible, namely that it randomizes $\boldsymbol{L}$ without affecting $\boldsymbol{S}$ and $\boldsymbol{I}$ ($\boldsymbol{L}$-randomization model[16]), we find that the rate $\mathrm{d}(\langle J_x \rangle_{J=3/2,\, I\ \text{coupled}})/\mathrm{d}t$ is positive and opposite, but equal in magnitude to $\mathrm{d}(\langle J_x \rangle_{J=3/2,\, I\ \text{decoupled}})/\mathrm{d}t$, which means

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle J_x \rangle_{J=3/2,\, I\ \text{coupled}} = -\frac{\mathrm{d}}{\mathrm{d}t} \langle J_x \rangle_{J=3/2,\, I\ \text{decoupled}} . \qquad (3)$$

Eq. (3) may well explain why the orientation $\langle J_x \rangle$ transferred to the $^2P_{3/2}$ state is parallel to the one in the initial $^2P_{1/2}$ state, so long as the hyperfine coupling exists, and is inverted as soon as hyperfine coupling is broken. The rate of transfer for I coupled provides a value of the sensitized Hanle curve for $H = 0$, the rate of transfer for I decoupled first must be subjected to magnetic depolarization according to $g_J(^2P_{3/2}) = 4/3$ in order to yield the asymptotic sensitized Hanle curve. In fig. 4 both these limiting cases are drawn. Between them a transition must occur which has been drawn in arbitrarily for the sake of demonstration. We must mention, however, that we have consequently omitted magnetic depolarization in the $^2P_{1/2}$ state whose g_F-values are smaller by a factor of eight than the g_J-value of the state $^2P_{3/2}$.

When comparing these theoretical predictions with the measured curves in fig. 3, the rapid vanishing of the central peak with rising pressure remains unexplained. The reason for that can be seen in a collisional decoupling of I and J, which occurs at very high pressures (hyperfine quenching). At very high pressures the collisions of any excited sodium atom follow one another so rapidly that hyperfine coupling has no time to get established during the very short intervals between successive collisions. Then, finally, the nuclear spin does not feel more from the electronic angular momentum than a fast sequence of random torques to which it is too inert to react. Thus we have collisional quenching of hyperfine coupling in the $^2P_{3/2}$ state over the entire magnetic range and, as a consequence, the sensitized Hanle curve takes on an ordinary Hanle shape.

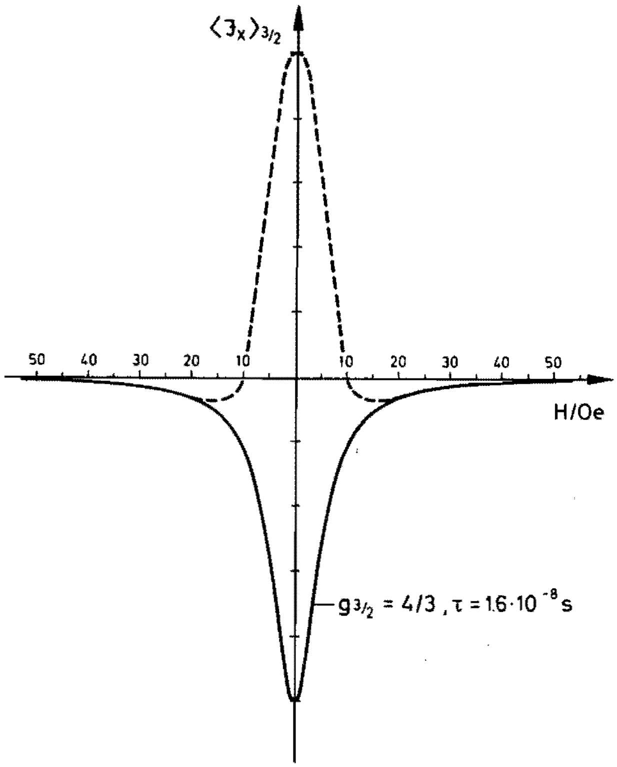

Fig. 4. The orientation $\langle J_x \rangle$ transferred from the $^2P_{1/2}$ state to the $^2P_{3/2}$ state and subjected to magnetic depolarization in the latter state (broken line). This curve is to explain the sensitized Hanle curves of fig. 3 at very low pressures. If no nuclear spin were present, the solid line would be valid over the entire range of *H*.

3.2. **The D_1 resonant Hanle curves.** Here magnetic decoupling of *I* is absent over the range of *H* where magnetic depolarization occurs. This is so, because the hyperfine splitting in the $^2P_{1/2}$ state is much wider than in the $^2P_{3/2}$ state. The signal, as before, measures

$$\langle J_x \rangle_{\frac{1}{2}} = (2I+1)^{-1} \left(\langle F_x \rangle_{F=I+\frac{1}{2}} - \langle F_x \rangle_{F=I-\frac{1}{2}} \right). \tag{4}$$

The equations which govern the motion of $\boldsymbol{F}$ in each of the states, read

$$\frac{\mathrm{d}\langle\boldsymbol{F}\rangle_F}{\mathrm{d}t} = \boldsymbol{\omega}_F \times \langle\boldsymbol{F}\rangle_F - \left(\frac{1}{\tau} + \frac{1}{T_F}\right)\langle\boldsymbol{F}\rangle_F + \alpha_{F'\to F}\langle\boldsymbol{F}\rangle_{F'} \tag{5}$$

$$\omega_F = g_F \frac{\mu_\mathrm{B} H}{\hbar}; \qquad g_F = \begin{bmatrix} +\,2/3\,(2I+1) & \text{for} & F = I + \frac{1}{2}, \\ -\,2/3\,(2I+1) & \text{for} & F = I - \frac{1}{2}. \end{bmatrix}$$

Here F, F' denote the F quantum numbers of the respective,, states. τ is the radiative life time, T_F the relaxation time pertinent to $\langle\boldsymbol{F}\rangle_F$. $\alpha_{F'\to F}$ denotes the rate of collisional transfer of $\langle\boldsymbol{F}\rangle$ from state F' to state F. The rigorous solution of the problem has been undertaken by Bulos and Happer^{13}) for the heavier alkalies where almost no collisional mixing between the fine-structure states occurs. This mixing, however, must be allowed for in the case of sodium.

As one concludes from the g_F values, the $\langle\boldsymbol{F}\rangle$ vectors in both F states precess in opposite sense. Fortunately, the state $F = I - \frac{1}{2}$ contributes much less to the signal than the state $F = I + \frac{1}{2}$. Thus we may omit it from our consideration. From the broadening of the Hanle signal we then verify directly

$$1/T_{F=I+\frac{1}{2}} = n v_\mathrm{r} \sigma_\mathrm{relax}\,(\langle\boldsymbol{F}\rangle_{F=I+\frac{1}{2}}). \tag{6}$$

n is the density of the foreign-gas atoms, v_r the relative velocity. If $H_{\frac{1}{2}}$ is the magnetic field pertinent to the half-width of the Hanle signal then, by means of the well-known relation

$$\frac{2}{3\,(2I+1)}\,\frac{\mu_\mathrm{B} H_{\frac{1}{2}}}{\hbar} = \frac{1}{\tau} + \frac{1}{T_{F=I+\frac{1}{2}}}, \tag{7}$$

we got $\tau = 1.6 \times 10^{-8}$ s (a value which is already in the literature^{12})) and $\sigma_\mathrm{relax}(\langle\boldsymbol{F}\rangle_{F=I+\frac{1}{2}})$. The cross section pertains to the relaxation of $\langle\boldsymbol{F}\rangle_{F=I+\frac{1}{2}}$. Our interest is rather directed to the cross section which pertains to the relaxation of the electronic orientation itself $\langle\boldsymbol{J}\rangle_{J=\frac{1}{2}}$. The l-randomization model predicts the ratio of both σ's to be

$$\sigma_\mathrm{relax}(\langle\boldsymbol{F}\rangle_{F=I+\frac{1}{2}})/\sigma_\mathrm{relax}(\langle\boldsymbol{J}\rangle_{J=\frac{1}{2}}) = 43/52. \tag{8}$$

Likewise we obtain

$$\sigma_\mathrm{relax}(\langle\boldsymbol{F}\rangle_{F=I-\frac{1}{2}})/\sigma_\mathrm{relax}(\langle\boldsymbol{J}\rangle_{J=\frac{1}{2}}) = 47/52. \tag{9}$$

These ratios deviate little from unity. Hence they prove to be different from the respective ratios for heavy alkalis where Bulos and Happer^{13}) for $I = \frac{3}{2}$ find 0.375. The reason for this can be seen in the fact that for light alkalis population losses of the $^2P_{1/2}$ state are the main source of relaxation, whereas they are absent in the case of heavier alkalis. Their contribution to the numbers 43, 47, 52 in the ratios in eqs. (8) and (9) uniformly amounts to 36.

The cross sections derived from the D_1 resonant Hanle curves by means of eqs. (6), (7) and (8) are given in table I. For helium a theoretical value exists given by Masnou and Roueff[14]. The agreement with the experiment is satisfactory.

TABLE I

Cross sections for the relaxation of $\langle J \rangle$ in state $3^2P_{1/2}$ of sodium. They are derived from the broadening of the D_1 resonant Hanle curve and corrected for the influence of the nuclear spin.

Foreign gas	$\sigma_{\text{relax}}\langle J \rangle_{J=\frac{1}{2}}$ Experiment ($Å^2$)	Theory ($Å^2$)
Helium	146	129
Neon	137	
Argon	260	
Krypton	306	
Xenon	356	

4. *Conclusions.* A study of pressure influences on the Hanle curves of sodium 3^2P states is reported. Our main findings consist of a) collisional quenching of hyperfine structure as a dominant interaction in connection with orientation transfer between the 3^2P levels of sodium; and b) pressure broadening of the D_1 resonant Hanle curves as a method for determining the cross sections of the relaxation of $\langle J \rangle$ in the $3^2P_{1/2}$ state of sodium.

This work represents part of a series of investigations on collisional relaxation and orientation transfer in the sodium 3^2P states. Earlier results have been compared with the theory of Masnou and Roueff in ref. 14 to which the reader is referred for more information on this problem.

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