

COLLISIONAL DEPOLARIZATION OF EXCITED SODIUM ATOMS

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Synopsis

Sodium vapour is excited by D_2 radiation. The linear polarization of the fluorescent D_2 radiation emitted at right angles to the incident-light beam is measured as a function of the pressure of an added noble gas. A magnetic field of 270 Oe which points parallel to the incident-light beam, serves to decouple the nuclear spin in the excited state. From the decrease of the polarization observed at rising pressure, cross sections for the disalignment of the electron in the excited state are derived. They are 167, 174, 308, 341, 376 Å² for the noble gases He, Ne, Ar, Kr, Xe at 383 K. These and earlier results on sodium depolarization are summarized and compared with theoretical results by Masnou *et al.*, Reid, and Lewis *et al.* Satisfactory agreement is obtained.

1. *Introduction.* This article pursues a twofold aim. Firstly, to present the method and results of a new attempt to determine the relaxation of an alignment created in the $3^2P_{3/2}$ state of sodium subsequently subjected to collisions with noble-gas atoms. Secondly, to summarize the results of earlier investigations on relaxation and transfer of polarizations in the sodium 3p doublet. The latter purpose has become desirable since a series of theoretical studies on sodium depolarization has appeared during the recent months^{1,2,3}).

The authors of those investigations occasionally attempted to compare their results with those of the experiment. But in doing so they frequently overlooked that at least part of the experimental results are obtained with a hyperfine structure being present in the sodium 2P states. The influence of hyperfine coupling on relaxation and transfer of polarization is significant. It tends to slow down relaxation^{4,5,6}) and may even invert polarization transfer⁷). It has been therefore suggested⁸) to carry out experiments on relaxation and polarization transfer in magnetic fields which are high enough to warrant sufficient decoupling of the nuclear spin. A series of

experiments have been performed on sodium in high fields^{8,9,10}) which will now be concluded and summarized in the present article. The results of high-field measurements may directly be compared with the results of any theory which disregards nuclear spin.

2. *Experimental.* It has been shown elsewhere that the difference of the intensities of fluorescent σ^+ and σ^- light, $I_{\sigma^+}^0 - I_{\sigma^-}^0$ *, emitted from any radiatively decaying atomic state provides a measure for the z component of the orientation, $\langle J_z \rangle_j$, existing in that state. It is even more obvious that the combination

$$\frac{1}{2}(I_{\sigma^+}^0 + I_{\sigma^-}^0) - I_{\pi}^0$$

provides a measure for the z component of the alignment, $\langle 3J_z^2 - J^2 \rangle_j$, existing in this state. To this end, let us consider this combination in terms of dipolar transition matrix elements and density matrix elements, $\langle ifm_f | \rho | ifm_f \rangle$, of the initial state. Capital quantum numbers pertain to the final state, JFM_F . n_j stands for the total number of atoms in the initial state.

$$\begin{aligned} & \frac{1}{2}(I_{\sigma^+}^0 + I_{\sigma^-}^0) - I_{\pi}^0 \propto n_j \sum_{fm_f} \sum_{FM_F} \langle ifm_f | \rho | ifm_f \rangle \\ & \times \{ \frac{1}{2} [\langle ifm_f | (x + iy) / \sqrt{2} | JFM_F \rangle \langle JFM_F | (x - iy) / \sqrt{2} | ifm_f \rangle \\ & + \langle ifm_f | (x - iy) / \sqrt{2} | JFM_F \rangle \langle JFM_F | (x + iy) / \sqrt{2} | ifm_f \rangle] \\ & - \langle ifm_f | z | JFM_F \rangle \langle JFM_F | z | ifm_f \rangle \}. \end{aligned} \quad (1)$$

When considering that the summation over the final states F, M_F includes all the substates of an eigenstate of J^2 we may substitute for the coordinates x, y, z their related components of the angular momentum: J_x, J_y, J_z , to find the desired relation

$$\begin{aligned} & \frac{1}{2}(I_{\sigma^+}^0 + I_{\sigma^-}^0) - I_{\pi}^0 \\ & \propto n_j \sum_{fm_f} \langle ifm_f | \rho | ifm_f \rangle \langle ifm_f | \frac{1}{2}(J_x^2 + J_y^2) - J_z^2 | ifm_f \rangle \\ & \propto n_j \langle 3J_z^2 - J^2 \rangle_j \equiv n_j \langle \mathcal{J}^{\textcircled{2}} \rangle_j. \end{aligned} \quad (2)$$

This relation holds as long as the summation includes all the substates of the J multiplet independently of the coupling scheme which classifies the

*The superscript ⁰ denotes the total intensity integrated over the solid angle.

substates. It is therefore still valid when the nuclear spin is being decoupled, *e.g.*, in the Paschen–Back region of hfs.

Our measurements were carried out in this latter region (*cf.* fig. 1) Sodium vapour was kept in a Pyrex vessel which had two plane windows attached to it at right angles. The density of sodium atoms corresponded to the temperature of the side arm where a small deposit of sodium metal was kept at 100°C . A beam of sodium D_2 light was incident upon the vapour entering the vessel perpendicularly to one of the windows. The fluorescent D_2 light was observed perpendicularly to the second window. It was at first transmitted through a linear polarizer and then detected by a photomultiplier. The linear polarizer permitted the selection of σ and π fluorescent light*. A static magnetic field parallel to the incident-light beam was applied on the vapour. It served for the decoupling of the nuclear spin

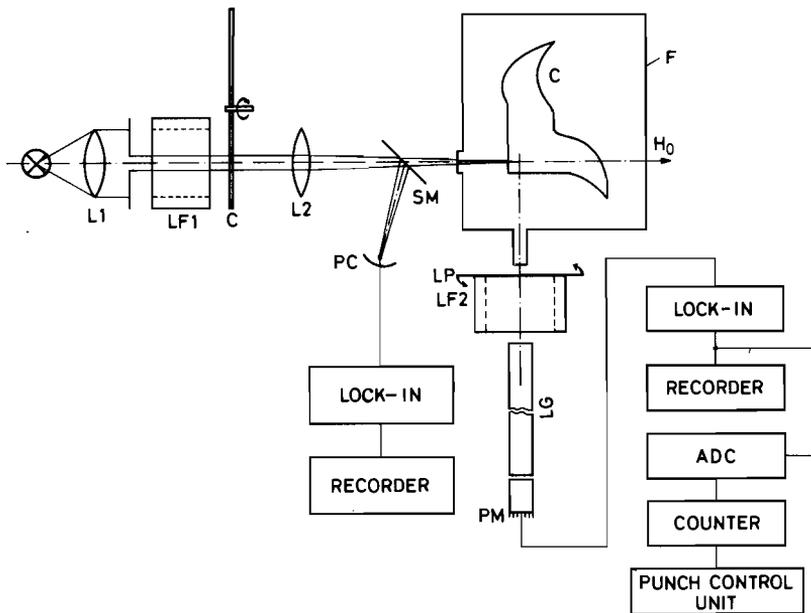


Fig. 1. Experimental arrangement. L_1 , L_2 , lenses; LF_1 , LF_2 , Lyot filters; C , chopper wheel; SM , semireflecting mirror; LP , linear polarizer; PC , photocell; PM , photomultiplier; LG , light guide; F , furnace; V , vessel.

*As the intensities $I_{\sigma} \equiv I_{\sigma^+} + I_{\sigma^-}$ and I_{π} are measured at right angles to the z axis, the following useful relation to the overall intensities I_{σ}^0 and I_{π}^0 should be borne in mind: $(I_{\sigma} - I_{\pi})_{90^{\circ}} \propto \frac{1}{2}(I_{\sigma^+}^0 + I_{\sigma^-}^0) - I_{\pi}^0$.

in the excited $3^2P_{3/2}$ state. From the hfs splitting constants and the g_j value of this level a critical field strength of 27 Oe was derived. Indeed, when the magnetic field was increased from zero to a multiple of this critical field strength, e.g., 100 Oe, then $I_\sigma - I_\pi$, the difference of the σ and π light fluorescent intensities, increased to almost the threefold value (*cf.*, fig. 2). This phenomenon can be understood when considering that the alignment which in the absorption process is imparted to the electronic angular momentum alone, is redistributed at low field strengths through hfs interaction among electronic and nuclear angular momentum, thus leading to a reduced signal $I_\sigma - I_\pi$. This redistribution would be inhibited at high field strengths when hfs coupling is eliminated, thus leading to an enhanced signal $I_\sigma - I_\pi$. Our measurements of depolarization were carried out at 270 Oe which we felt was sufficient to provide rather perfect decoupling conditions.

When buffer gas was admitted to the cell, the signal $I_\sigma - I_\pi$ was seen to decrease with rising pressure. As this quantity provides a measure for the alignment

$$n_j \langle \mathcal{J}_0^{(2)} \rangle_j = n_j \sum_{m_j} \langle jm_j | 3J_z^2 - J^2 | jm_j \rangle \rho_{m_j} \quad (3)$$

and as $n_j \langle \mathcal{J}_0^{(2)} \rangle_j$ evolves in time according to the rate equation

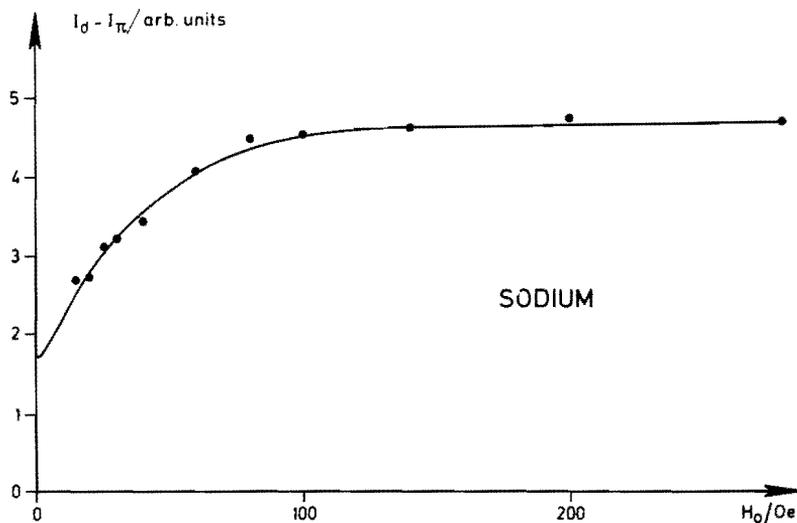


Fig. 2. The difference of the fluorescent σ and π intensities, $I_\sigma - I_\pi$ as a function of the strength of the longitudinal field H_0 , which acts on the sodium vapour.

$$\frac{d}{dt} n_j \langle \mathcal{J}_0^{(2)} \rangle_j = C - \left(\frac{1}{\tau} + N v_r \sigma_j^{(2)} \right) n_j \langle \mathcal{J}_0^{(2)} \rangle_j, \quad (4)$$

whose stationary solution

$$n_j \langle \mathcal{J}_0^{(2)} \rangle_j = C / [(1/\tau) + N v_r \sigma_j^{(2)}], \quad (5)$$

applies to our experimental conditions, one would expect

$$\frac{1}{I_\sigma - I_\pi} \propto \frac{1}{\langle \mathcal{J}_0^{(2)} \rangle_j n_j} \propto \frac{1}{\tau} + N v_r \sigma_j^{(2)}. \quad (6)$$

Here C denotes the rate at which $n_j \langle \mathcal{J}_0^{(2)} \rangle_j$ is created due to optical excitation, τ denotes the radiative life time, N stands for the density of noble-gas atoms and v_r for the mean relative velocity between noble-gas atoms and sodium atoms. One hence concludes that a plot of $1/(I_\sigma - I_\pi)$ versus N or versus the pressure in the vessel would yield a straight line, whose slope provides a measure of $\sigma_j^{(2)}$. In particular, the ratio of the slope and the ordinate section equals the ratio of the collisional and radiative decay rate of $n_j \langle \mathcal{J}_0^{(2)} \rangle_j$. As the radiative lifetime is known to an accuracy of a few percent [$\tau(3^2P_{3/2}) = 1.62 \times 10^{-8} \text{ s}^{11}$] $\sigma_j^{(2)}$ is easy to determine from this ratio. A linear dependance of $1/(I_\sigma - I_\pi)$ on N was indeed found for each noble gas. Fig. 3 gives two examples. The values of the cross sections $\sigma_j^{(2)}$ which we derived according to the outlined programme, are given in table I. The accuracy is 10%. It must be emphasized however that the validity of the method depends on the constancy of C when the pressure changes. A slight dependence of C on N would be conceivable when the absorption line is pressure-broadened, so that the

TABLE I

Measured cross sections for the disalignment of the state $3p^2P_{3/2}$ of sodium: $\sigma_{3/2}^{(2)}$. Earlier measurements on the cross section $\sigma_{3/2}^{(1)}$ for disorientation of that state are given for comparison. The ratio of the two which had been predicted by various authors (*cf.* for instance ref. 14) is given in the third line.

Cross section (\AA^2)	He	Ne	Ar	Kr	Xe
$\sigma_{3/2}^{(2)}$	167	174	308	341	376
$\sigma_{3/2}^{(1)}$	128	107	205	243	281
$\sigma_{3/2}^{(2)} / \sigma_{3/2}^{(1)}$	1.30	1.62	1.50	1.40	1.34

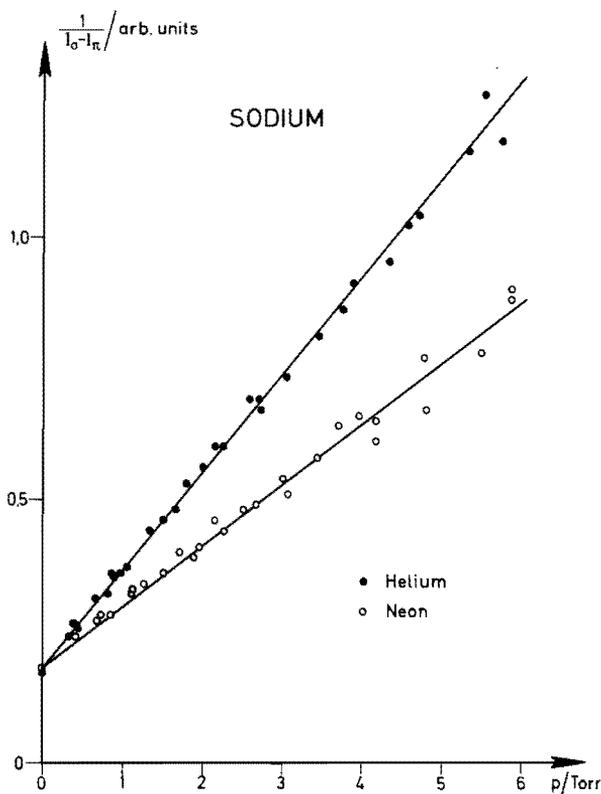


Fig. 3. The reciprocal of the difference of the fluorescent intensities, $1/(I_0 - I_\pi)$, as a function of the pressure of added noble gases.

absorption rate would change appreciably. It is important to consider in this respect that the vapour pressure in the vessel is kept low to ensure that the optical depth is large compared with the dimensions of the fluorescent region in the cell. Then line broadening does not give rise to any change of the excitation rate. Also the range of the pressure in our experiment was chosen small in comparison with pressure values which might broaden the absorption line beyond the Doppler width (\approx several atm). The neglect of pressure broadening therefore appears justified.

3. *Theoretical.* Overleaf we will give a summary on all the preceding measurements on sodium 2P depolarization. To this end we recall the definitions of irreducible cross sections in terms of relaxation and transfer of tensorial polarizations. Concerning the latter, we use the following definitions which deviate from Fano's¹²).

TABLE II

Comparison of theoretical and experimental cross sections for relaxation and transfer in the $3p\ ^2P$ states of sodium induced in collisions with helium. a: Weak optical pumping by D_2 light at varying buffer gas pressures; b: linear polarization of fluorescent light emitted at right angles to the incident unpolarized light beam; c: broadening of D_1 resonant Hanle curves, corrected for the action of the nuclear spin; d: circular polarization of fluorescent light, emitted in backward direction towards the incident circularly polarized light beam. Methods a, b, d make use of magnetic decoupling of the nuclear spin in the excited state.

Cross section (\AA^2)	Nomenclature by Schneider ⁶	Masnou and Roueff ²	Reid ³	Lewis and McNamara ¹	Experiment	Exp. method and ref.
$\sigma_{\frac{1}{2}}^{(1)}$	$\sigma_{\text{relax}}(\frac{3}{2})$	109	86	94	128	a, 9
$\sigma_{\frac{3}{2}}^{(2)}$	—	155	123	122	167	this work, b
$\sigma_{\frac{1}{2}}^{(1)}$	$\sigma_{\text{relax}}(\frac{1}{2})$	129	99	114	146	c, 10
$\sigma_{\frac{1}{2} \rightarrow \frac{3}{2}}^{(0)}$	$\sigma_{\text{sens}}(\frac{1}{2} \rightarrow \frac{3}{2})$	100	79	90	89	d, 8
$\sigma_{\frac{3}{2} \rightarrow \frac{1}{2}}^{(0)}$	$\sigma_{\text{sens}}(\frac{3}{2} \rightarrow \frac{1}{2})$	50	40	—	44	d, 8
$\sigma_{\frac{1}{2} \rightarrow \frac{3}{2}}^{(1)}$	$\sigma_{\text{poltrans}}(\frac{1}{2} \rightarrow \frac{3}{2})$	-58	-72	—	-64, -72	d, 8
$\sigma_{\frac{3}{2} \rightarrow \frac{1}{2}}^{(1)}$	$\sigma_{\text{poltrans}}(\frac{3}{2} \rightarrow \frac{1}{2})$	-5.8	-7.2	—	-6.6, -7.6	d, 8
$\sigma(\frac{1}{2} \frac{1}{2} \rightarrow \frac{1}{2} - \frac{1}{2})$	$\sigma_{\text{depol}}(\frac{1}{2})$	14.5	11	—	16.7	d, 8

$$\begin{aligned} \mathcal{J}_0^{(0)} &= 1, & \mathcal{J}_0^{(1)} &= J_z, \\ \mathcal{J}_0^{(2)} &= 3 J_z^2 - J^2, & \mathcal{J}_0^{(3)} &= 5 J_z^3 - (3 J^2 - 1) J_z. \end{aligned} \quad (7)$$

In 1965 it was shown in two pioneering papers^{13,14}) that tensorial polarizations of different k , q are not coupled by collisions whose collisional planes are oriented at random. The proof of this statement can be easily extended to the case where tensorial polarizations are being transferred during collisions from an initial state j' to a final state j . Then the tensorial polarization which is built up in the state j on account of the one in state j' , pertains to the same k , q values as the one in state j' .

Bearing in mind these rules, we may now define cross sections $\sigma_j^{(k)}$ for relaxation and $\sigma_{j \rightarrow j'}^{(k)}$ for transfer of polarization by writing down the corresponding rate equation

$$\begin{aligned} \frac{d}{dt} n_j \langle \mathcal{J}_q^{(k)} \rangle_j &= -N v_r [\sigma_j^{(k)} n_j \langle \mathcal{J}_q^{(k)} \rangle_j \\ &\quad - \sigma_{j \rightarrow j'}^{(k)} n_{j'} \langle \mathcal{J}_q^{(k)} \rangle_{j'}]. \end{aligned} \quad (8)$$

Here N denotes the density of foreign-gas atoms, v_r the mean relative velocity of the colliding atoms, while $n_{j'}$, n_j are the total densities of atoms in the indicated states. We emphasize that in our understanding it is the "macroscopic" polarizations $n_j \langle \mathcal{J}_q^{(k)} \rangle_j$ which undergo relaxation and transfer, rather than the polarizations themselves. The occasional omission of n_j , $n_{j'}$ in eq. (8) in one instance has led to a misunderstanding in the literature¹⁵). We must admit, however, that cross sections may be defined for the relaxation of the proper polarizations as well. To this end we carry out the differentiation of the "macroscopic" polarization to find

$$\frac{d}{dt} n_j \langle \mathcal{J}_q^{(k)} \rangle_j = n_j \frac{d}{dt} \langle \mathcal{J}_q^{(k)} \rangle_j + \langle \mathcal{J}_q^{(k)} \rangle_j \frac{d}{dt} n_j.$$

We now consider that dn_j/dt means the change of the total population due to transfer to and from another state j' . Hence we have

$$\frac{d}{dt} n_j = N v_r (n_{j'} \sigma_{j \rightarrow j'}^{(0)} - n_j \sigma_{j \rightarrow j'}^{(0)}).$$

As a consequence, the cross sections for relaxation of the "macroscopic" polarizations according to eq. (8) must be diminished by $\sigma_{j \rightarrow j'}^{(0)}$ to yield the respective cross sections for the proper polarizations.

Surprisingly, theoretical physicists have preferred to give the results of their quantitative calculations on sodium depolarization in terms of cross sections for transitions between Zeeman sublevels: $\sigma(jm_j \leftrightarrow j'm_{j'})$. The meaning of the latter ensues from the rate equations:

$$\dot{n}_{j,m_j} \equiv \frac{d}{dt} n_{j,m_j} = -N v_r \left(n_{j,m_j} \sum_{j',m_{j'}} \sigma(jm_j \leftrightarrow j'm_{j'}) - \sum_{j',m_{j'}} n_{j',m_{j'}} \sigma(jm_j \leftrightarrow j'm_{j'}) \right). \quad (9)$$

Here n_{j,m_j} denotes the occupation number of the Zeeman sublevel. The irreducible cross sections $\sigma_j^{(k)}$, $\sigma_{j \rightarrow j'}^{(k)}$ can be expanded in terms of the $\sigma(jm_j - j'm_{j'})$. To find these expansions one simply has to multiply any equation for \dot{n}_{j,m_j} by the corresponding matrix element of

$$\langle j m_j | \mathcal{J}_0^{(k)} | j m_j \rangle. \quad (10)$$

After carrying out this procedure on both sides of eq. (9) one has to sum up all the resultant equations with respect to m_j . On so doing one arrives at an equation of type (8) whence the desired expansions of the irreducible cross sections become apparent. The results are expressed in eqs. (11) and (12).

Cross sections for relaxation:

$$\begin{aligned} \sigma_{3/2}^{(1)} &= \frac{2}{3} \sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, \frac{1}{2}\right) + \frac{4}{3} \sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, -\frac{1}{2}\right) + 2\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, -\frac{3}{2}\right) + \sigma_{\frac{3}{2} \rightarrow \frac{1}{2}}^{(0)} \\ &= -2\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, \frac{1}{2}\right) + 2\sigma\left(\frac{3}{2}, \frac{1}{2} \rightarrow \frac{3}{2}, -\frac{1}{2}\right) + 4\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, -\frac{1}{2}\right) + \sigma_{\frac{3}{2} \rightarrow \frac{1}{2}}^{(0)}, \\ \sigma_{3/2}^{(2)} &= 2\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, \frac{1}{2}\right) + 2\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, -\frac{1}{2}\right) + \sigma_{\frac{3}{2} \rightarrow \frac{1}{2}}^{(0)}, \\ \sigma_{3/2}^{(3)} &= 4\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, \frac{1}{2}\right) - 2\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, -\frac{1}{2}\right) + 2\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, -\frac{3}{2}\right) + \sigma_{\frac{3}{2} \rightarrow \frac{1}{2}}^{(0)} \\ &= \frac{4}{3}\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, \frac{1}{2}\right) + \frac{2}{3}\sigma\left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{3}{2}, -\frac{1}{2}\right) + 2\sigma\left(\frac{3}{2}, \frac{1}{2} \rightarrow \frac{3}{2}, -\frac{1}{2}\right) + \sigma_{\frac{3}{2} \rightarrow \frac{1}{2}}^{(0)}, \\ \sigma_{1/2}^{(1)} &= 2\sigma\left(\frac{1}{2}, \frac{1}{2} \rightarrow \frac{1}{2}, -\frac{1}{2}\right) + \sigma_{\frac{1}{2} \rightarrow \frac{3}{2}}^{(0)}. \end{aligned} \quad (11)$$

The cross sections $\sigma_{3/2}^{(1)}$ and $\sigma_{3/2}^{(3)}$ apparently are represented in a twofold way. These twofold representations are consequences of general conservation laws which the transition operator for depolarizing collision has to satisfy. They can be used for checking sets of $\sigma(j'm_{j'} \leftrightarrow jm_j)$, which have been published by various authors. This check was successfully applied to the sets given by Masnou and Roueff²⁾, Reid³⁾ and Gordeyev *et al.*¹⁶⁾.

Cross sections for transfer:

$$\begin{aligned} \sigma_{1/2 \rightarrow 3/2}^{(0)} &= \sum_{m_j'} \sigma \left(\frac{1}{2} m_j \rightarrow \frac{3}{2} m_j' \right), & \sigma_{3/2 \rightarrow 1/2}^{(0)} &= \sum_{m_j'} \sigma \left(\frac{3}{2} m_j \rightarrow \frac{1}{2} m_j' \right), \\ \sigma_{1/2 \rightarrow 3/2}^{(1)} &= 3 \sigma \left(\frac{1}{2}, \frac{1}{2} \rightarrow \frac{3}{2}, \frac{3}{2} \right) + \sigma \left(\frac{1}{2}, \frac{1}{2} \rightarrow \frac{3}{2}, \frac{1}{2} \right) \\ &\quad - \sigma \left(\frac{1}{2}, \frac{1}{2} \rightarrow \frac{3}{2}, -\frac{1}{2} \right) - 3 \sigma \left(\frac{1}{2}, \frac{1}{2} \rightarrow \frac{3}{2}, -\frac{3}{2} \right), & (12) \\ \sigma_{3/2 \rightarrow 1/2}^{(1)} &= \frac{3}{10} \sigma \left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{1}{2}, \frac{1}{2} \right) - \frac{3}{10} \sigma \left(\frac{3}{2}, \frac{3}{2} \rightarrow \frac{1}{2}, -\frac{1}{2} \right) \\ &\quad + \frac{1}{10} \sigma \left(\frac{3}{2}, \frac{1}{2} \rightarrow \frac{1}{2}, \frac{1}{2} \right) - \frac{1}{10} \sigma \left(\frac{3}{2}, \frac{1}{2} \rightarrow \frac{1}{2}, -\frac{1}{2} \right). \end{aligned}$$

If the gap between the doublet levels can be considered small in comparison with the collisional energy the cross sections for inter-Zeeman transitions become symmetric under interchange of the initial and the final state. Then the following symmetries for the transfer cross sections hold

$$\sigma_{1/2 \rightarrow 3/2}^{(0)} = 2\sigma_{3/2 \rightarrow 1/2}^{(0)}, \quad (13)$$

$$\sigma_{1/2 \rightarrow 3/2}^{(1)} = 10\sigma_{3/2 \rightarrow 1/2}^{(1)}. \quad (14)$$

For sodium where this condition applies, these symmetries could be verified experimentally (*cf.* table II). If the tensor polarizations were defined according to Fano the transfer cross sections for both directions would be equal (*cf.* Niewitecka *et al.*¹⁵).

In table II we give a synopsis of experimentally determined cross sections for relaxation and transfer in He and of cross sections derived from sets of theoretical inter-Zeeman cross sections by virtue of eqs. (11) and (12). The theoretical cross sections pertain to an average kinetic energy corresponding to 400 K. Approximately the same temperature was maintained in our experiments (383 K).

4. Conclusions. It has been shown that the measurement of the polarization of fluorescent light emitted at right angles to the exciting light beam provides a convenient method for determining the cross section appropriate to disalignment. Moreover, the determined cross section can be attributed to the disalignment of the electron alone if provisions are made that no interactions of the electron and the nucleus occur in the excited state. The easiest provision consists in magnetic decoupling of electronic and nuclear spin. In the last part of the article some of the data obtained and earlier results are summarized and compared with theoretical

results published during the recent months. The agreement turns out to be satisfactory bearing in mind that the experimental error might easily amount to 10%.

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