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INVESTIGATION OF THE LONGITUDINAL RELAXATION PROCESSES IN THE HYPERFINE STATES OF THE OPTICALLY PUMPED ALKALI METAL VAPOUR

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The longitudinal relaxation process of alkali atoms in the hyperfine states was investigated by means of a modification of the method observing transient effects in magnetic resonance. The contribution of processes like Zeeman relaxation, hyperfine relaxation, spin exchange and diffusion were measured. It is shown for the first time that it is possible to observe one single diffusion mode.

1. Introduction

Previous methods of investigating the relaxation processes of alkali atoms in the ground state consisted in observing the decays of the observables S_z and $\vec{S}\vec{I}$ which totally describe the relaxation process [1], [2]. This leads to the development of different techniques of measurements [3–6] as well as methods of pumping alkali atoms in different hyperfine states [7, 8].

In the paper we will concern ourselves with the longitudinal relaxation processes of atoms in hyperfine states and we will describe an experiment that allows us to determine different contributions to the relaxation of the longitudinal electron polarization $\langle S_z(t)\rangle_+$ and $\langle S_z(t)\rangle_-$ in the two hyperfine states $F_+ = I+1/2$ and $F_- = I-1/2$.

Investigation of the relaxation processes of an optically pumped alkali metal vapour for atoms in a given hyperfine state has become possible through a modification of the method of observing transiet effects in magnetic resonance [9].

The investigation of the decay of the orientation in darkness of the hyperfine states allows us to obtain information concerning the contributions of different processes (like Zeeman relaxation, hyperfine relaxation, and spin exchange processes) to the relaxation. The modified method will be presented in Section 2. In Section 3 of this work equations will be given that describe the time evolution of the longitudinal electron polarization for

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a small initial orientation of the system. The interactions between the alkali atoms and buffer gas atoms as well as exchange interactions are described in a way analogous to that of Gibbs [10]. These equations also contain the relaxation processes at the walls of the bulb. Such a process in the presence of a buffer gas is connected with the diffusion of optically oriented atoms to the walls. Assuming a similar space distribution of optically oriented cesium atoms in both hyperfine ground states, analytical solutions of the equations are obtained in the form of two infinite series of exponential functions. Choosing the proper geometry of the detection beam, according to our previous theoretical considerations [11], the evolution of the relaxation process can be well approximated by a two-exponential decay curve, which can be taken as the first diffusion mode. The decay constants of the polarization of the hyperfine states can be expressed as linear combinations of the relaxation times: nuclear relaxation time T_n, hyperfine relaxation time T_h and exchange relaxation time T_{ex} as well as the diffusion coefficient.

The present work is the first experimental verification (for cesium with neon as a buffer gas in a spherical cell) that it is really possible to observe only one diffusion mode as was predicted theoretically [11]. Results of these experiments as well as of investigations of the relaxation processes for cesium atoms in the two hyperfine states are shown in Section 4. Although the qualitative agreement of the experimental results with theoretical prediction is good, further investigation is required to verify the influence of other relaxation processes as well as the assumed character of the signal.

2. *Experiment*

Investigations of the longitudinal relaxation process for alkali atoms in the hyperfine states have been carried out according to the method described previously [9]. The main idea of this method was to separate Zeeman resonances in the two hyperfine states even in weak magnetic fields $\vec{H}_0$. This separation is possible because the gyromagnetic ratio of the levels has different signs and values (for example for Cs the difference in frequencies is about 1100 Hz/Oe):

$$\vec{\omega}_0 = \gamma_{F_\pm} \vec{H}_0 \approx \pm \frac{\gamma S}{2I+1} \vec{H}_0. \tag{1}$$

For this reason a proper rotating rf-field induces resonance effects only in one hyperfine state.

To explain the method of investigation let us introduce a spin system which can be described by the Bell and Bloom equations [12]. Thus we study a spin system in which the magnetic moment in the direction of the constant magnetic field has been induced by the optical pumping process. This system is interacting with a pulse of the rotating rf-field of resonance frequency $\vec{\omega}_0 = \gamma_{F_\pm} \vec{H}_0$. In the frame rotating with the frequency $\vec{\omega}_0$ around the z-axis ($X'Y'Z$; $\vec{\omega}_0$), the magnetic moment of the atomic system will start to rotate in the $Y'Z$ plane at the moment the rf pulse is switched on (Fig. 1). The rotating frame was choosen in such a way that the z-axis was defined by the direction of the constant magnetic field $\vec{H}_0$ and the rotation coincided with the vector $\vec{H}_1$.

At the same time in the laboratory frame $\vec{M}$ will precess about $\vec{H}_0$ with the frequency $\vec{\omega}_0$. At resonance, for $|\gamma_{F_\pm}|H_1T_1 = \omega_1 T_1 \ll 1$ and $\omega_1 T_2 \ll 1$ the projection of $\vec{M}$ on the direction perpendicular to the z-axis is given by the formula [13]:

$$M_y = M_{z0} \exp\{-t/\tau^*\} \sin \omega_1 t \cos \omega_0 t, \tag{2}$$

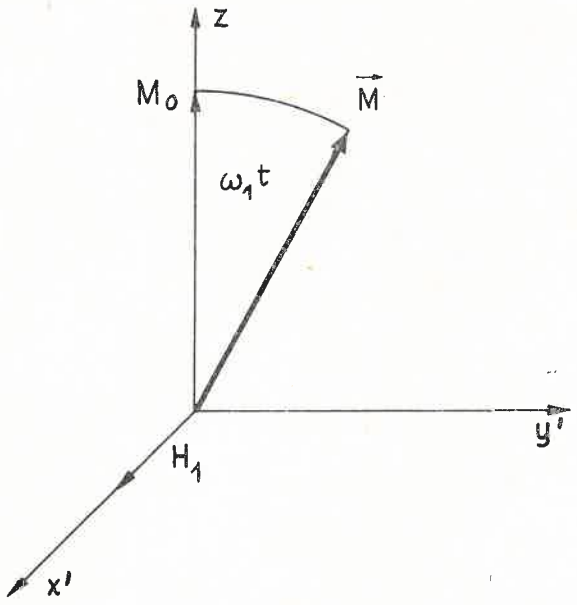


Fig. 1

where

$$\frac{1}{\tau^*} = \tfrac{1}{2}\left(\frac{1}{T_1} + \frac{1}{T_2}\right). \tag{3}$$

The maximum amplitude of the envelope of this curve has a value

$$M_y(T/4) = M_{z0} \exp\{-T/4\tau^*\}, \tag{4}$$

where $T = 2\pi/\omega_1$. So when $T \ll \tau^*$, $M_y = M_{z0}$. By measuring the y-component of the magnetization $\vec{M}$ we can determine at any time the value of the longitudinal magnetization.

In general, an atomic system possessing hyperfine structure cannot be described as simply as above. The quantum-mechanical theory predicts that for a spin system interacting with a rf-field the output signal is determined by the expectation value of a monitoring operator Q [16] given by

$$\langle Q \rangle = \mathrm{Tr}(\varrho Q), \tag{5}$$

where ϱ satisfies the equation

$$\frac{d\varrho}{dt} = -\frac{i}{\hbar}[\mathscr{H}, \varrho]. \tag{6}$$

$\mathscr{H}$ being the Hamiltonian of the spin system. Only for spin 1/2 are the equations for the expectation value identical with the Bloch equations. For $J > 1/2$ we have to assume homogeneous relaxation processes and apply an approximation in which expressions will only include nondiagonal terms, involving oscillations at frequencies ω_0, to get Bloch type equations.

Hence for an optically pumped atomic vapour with hyperfine structure, relaxation processes are not homogeneous and Bell-Bloom-Bloch equations cannot be applied. However, since the choosen detection technique is characterized by an observation time T, much shorter than the relaxation times, the types of the relaxation processes are unimportant. Then our atomic system can be treated as two sets of different atoms having different angular momentum F_+ and F_- and different resonance frequencies. Therefore, we assume that the evolution of the two sets of atoms is described by two systems of Bell-Bloom-Bloch equations (in the presence of the rf field) only during the observation time T, which is much shorter than the relaxation time of the system.

The way of using this method to investigate the relaxation process for atoms in the hyperfine states is shown in Fig. 2. We study the time evolution of the atomic system after

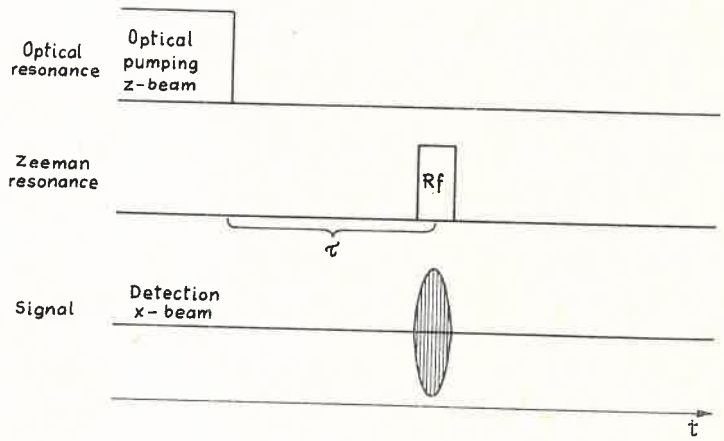


Fig.2. Double resonance sequence of events

the pumping light is turned off, at $t = 0$. After a variable time interval τ an rf pulse, in resonance with the investigated hyperfine Zeeman levels, is applied to the system and we observe the signal of a second weak detection beam perpendicular to $\vec{H}_0$. The maximum value of the amplitude of the signal is proportional to the actual value of the magnetic moment of the investigated hyperfine state. Repeating this procedure for different values of τ one can get a decay curve of the longitudinal polarization of the atomic system in the tested hyperfine state.

In an experiment thus carried out (besides the conditions mentioned previously and conditions which must be satisfied by the detection beam [4]) one has to be very careful, that the initial conditions from which the atomic system starts to relax in the darkness are always the same. For the observation of the resonances in two groups of atoms in different

hyperfine states whose frequencies lie near each other, the separation of the signals coming from both groups wil be most important.

A block diagram of our pulsed magnetooptical double-resonance apparatus is presented in Fig. 3. The constant magnetic field $\vec{H}_0$ was produced by a set of two pairs of coils. This assured a homogeneity of the order of 10^{-5} in a volume 0.5 dm^3. The earths magnetic field in the perpendicular directions were compensated very accurately by similar sets of coils. Two lamps of the Varian type [26] were used as sources of resonance radiation L_p

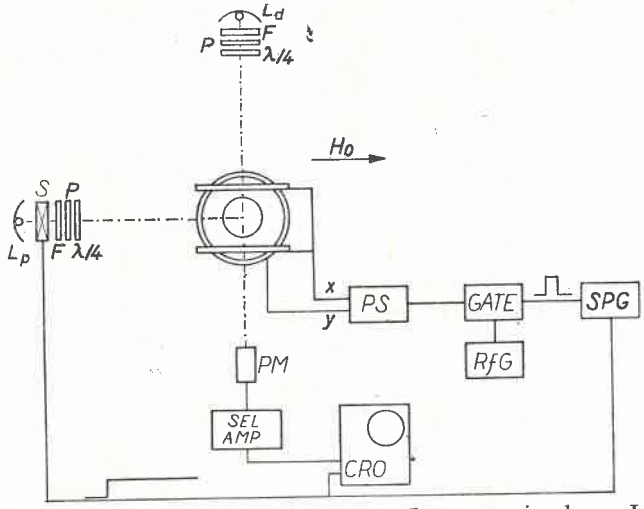


Fig. 3. The block diagram of the experimental apparatus. L_p — pumping lamp, L_d — detection lamp, S — mechanical shutter, F — interference filter, $\lambda/4$ — quarter wave plate, P — polarizer, PM — photomultiplier, RfG — radio frequency generator, PS — phase shifter, SPG — square pulse generator, CRO — oscilloscope

and L_d. The detection system contained a photomultiplier PM type M12 F35 "Zeiss" connected to a selective amplifier tuned in a wide band of frequencies. The signal was displayed on an oscilloscope screen and photographed. A mechanical shutter was operated periodically by an electric motor. The closing time of the shutter was less than 10 ms. The rf pulses with an amplitude of the order of a few mOe were supplied by a gated continuously operating rf generator. The square pulse generator of variable width was supplemented with means for variably delayed triggering. The oscilloscope as well as the pulse generator was triggered by the mechanical shutter. The apparatus was arranged in such a way that after a sequence of about 10 pulses for a given time interval τ, the delay time of the pulse was changed automatically to a new value. This has been achieved by employing a synchronous motor to drive the delay potentiometer in the square pulse generator. The repetition of the pulses for the same time interval τ was used to average different types of fluctuations. The rotating rf-field which was applied to separate completely the studied resonances, was formed by a phase shift system PS combined with an amplifier. This system converted the rf pulse into two pulses of the same amplitude shifted by $\pi/2$ in phase. These pulses supplied two pairs of coils perpendicular to each other. The observed signals are shown in Fig. 4.

3. *Rate equations for relaxation processes*

In this section a crude theory is used to derive expressions describing the time evolution of the longitudinal electron polarization for atoms in both hyperfine states when they relax in darkness. In the investigated atomic system, relaxation usually involves the following mechanisms: it is induced by collisions with rare gas atoms as well as with alkali atoms and by collisions with the walls after diffusion through the buffer gas.

3.1. Relaxation processes induced by alkali-atom-buffer-gas-atom

The analysis of relaxation caused by collisions with rare gas atoms suggest that this process is due to a perturbation that varies randomly in time [1, 17, 21]. The rate equations which are used describe the evolution of the ground state electronic spin polarizations $\langle S_z\rangle_+$ and $\langle S_z\rangle_-$ of an alkali-metal vapour subject to relaxation processes induced by the alkali-atom-rare-gas-atom collisions are derived in a straight forward way [10],

$$\langle \dot{S}_z\rangle_+^R = \frac{1}{T_1}\langle S_z\rangle_+ - \frac{2I^2+I+1}{(2I+1)^2}\frac{1}{T_1'}\langle S_z\rangle_+ - \frac{2(I+1)(2I+3)}{(2I+1)^2}\frac{1}{T_1'}\langle S_z\rangle_-$$

$$\langle S_z\rangle_-^R = -\frac{1}{T_1}\langle S_z\rangle_- - \frac{I(2I-1)}{(2I+1)^2}\frac{1}{T_1'}\langle S_z\rangle_- - \frac{2I^2+3I+2}{(2I+1)^2}\frac{1}{T_1'}\langle S_z\rangle_+, \tag{7}$$

where

$$\frac{1}{T_1} = \frac{J(\omega_F)-J(\Delta W)}{(2I+1)^2}, \qquad \frac{1}{T_1'} = J(\Delta W). \tag{8}$$

ω_F and ΔW are, respectively, the Zeeman and hyperfine energy level separations of the alkali atom in the ground state. $J(\omega_F)$ and $J(\Delta W)$ are the spectral density functions of the randomly oriented magnetic field which are the Fourier transforms of the correlation functions [22]. I is the nuclear spin.

If the interaction can be characterized by a single correlation time τ_c, the spectral density function is of the form

$$J(\omega) \sim \frac{2\tau_c}{1+\omega^2\tau_c^2}. \tag{9}$$

The usually introduced time constants T_e and T_n [1, 4, 18] which characterize the longitudinal relaxation process are related to T_1 and T_1' by

$$\frac{1}{T_e} = \frac{1}{T_1} + \frac{1}{T_1'}, \qquad \frac{1}{T_n} = \frac{1}{T_1} + \frac{2}{(2I+1)^2}\frac{1}{T_1'}. \tag{10}$$

For convenience the following relaxation times are introduced:

$$\frac{1}{T_Z} = \frac{J(\omega_F)}{(2I+1)^2},$$

$$\frac{1}{T_h} = J(\Delta W), \tag{11}$$

which we will call the Zeeman and hyperfine relaxation times, respectively.

3.2. Spin exchange between identical atoms

The effect of spin exchange collisions between alkali atoms which play the most important role in optical pumping experiments, have been studied by many authors [23—25]. However in this case the time evolution of the electronic-spin polarization of alkali atoms in hyperfine levels of the ground state cannot be described by a single exponential function only. In the limit of small pumping light intensity one can get an closed system of rate equations. The small pumping light limit is achieved when the probability for an atom to be in the state $|FM\rangle$ is slightly different from that in the thermal equilibrium,

$$p_{FM} = \frac{1}{2(2I+1)} + \delta_{FM}, \tag{12}$$

where δ_{FM} is a small deviation. The deviations should be so small that one can neglect all but the linear terms in δ_{FM}. This leads to the following rate equation for the spin exchange [10]:

$$\langle \dot{S}_z \rangle_{\pm}^{\mathrm{ex}} = -\frac{2I(2I-1)}{3(2I+1)^2}\frac{1}{T_{\mathrm{ex}}}\langle S_z \rangle_{\pm} - \frac{(2I+2)(2I+3)}{3(2I+1)^2}\frac{1}{T_{\mathrm{ex}}}\langle S_z \rangle_{\mp}, \tag{13}$$

where T_{ex} is the spin exchange relaxation time.

3.3. Relaxation on the walls after diffusion in the buffer gas

Because of the process of diffusion in the buffer gas, the expectation value of an observable S_z of the investigated atom is actually a function of time and position of the atom in the cell $\langle S_z(\vec{r}, t)\rangle$. The average is taken over a volume large enough to contain a large number of atoms and small enough to assure that the expectation value is constant over that volume. In this case one gets the diffusion equation [20]

$$\langle \dot{S}_z(\vec{r}, t)\rangle_{\pm}^{D} = D\Delta\langle S_z(\vec{r}, t)\rangle_{\pm} \tag{14}$$

with boundary conditions which depend on the nature of the walls. In our experiment the walls of the cell were uncoated and we assume that every collision disorients the alkali atom spin completly:

$$\langle S_z(r = R, t)\rangle_{+} = \langle S_z(r = R, t)\rangle_{-} = 0, \tag{15}$$

where R is the radius of the spherical resonance cell.

3.4. Relaxation under simultaneous effects

Since the above mentioned processes are independent, the corresponding relaxation rates simply add. Therefore the time evolution of the longitudinal electronic spin polarization of the hyperfine states are as follows:

$$\begin{aligned} \langle \dot{S}_z \rangle_{+} &= D\Delta\langle S_z \rangle_{+} - \alpha_{+}\langle S_z \rangle_{+} - \beta_{+}\langle S_z \rangle_{-} \\ \langle \dot{S}_z \rangle_{-} &= D\Delta\langle S_z \rangle_{-} - \alpha_{-}\langle S_z \rangle_{-} - \beta_{-}\langle S_z \rangle_{+}, \end{aligned} \tag{16}$$

where

$$\alpha_+ = \frac{1}{T_Z} + \frac{I}{2I+1}\frac{1}{T_h} + \frac{2I(2I-1)}{3(2I+1)^2}\frac{1}{T_{ex}}, \tag{17}$$

$$\beta_+ = \frac{(I+1)(2I+3)}{(2I+1)^2}\left(\frac{1}{T_h} + \tfrac{2}{3}\frac{1}{T_{ex}}\right),$$

$$\alpha_- = \frac{1}{T_Z} + \frac{2I^2+3I+1}{(2I+1)^2}\frac{1}{T_h} + \frac{2(I+1)(2I+3)}{3(2I+1)^2}\frac{1}{T_{ex}}$$

$$\beta_- = \frac{(2I-1)I}{(2I+1)^2}\left(\frac{1}{T_h} + \tfrac{2}{3}\frac{1}{T_{ex}}\right),$$

T_Z and T_h are defined by Eq. (11).

This set of rate equations with the above mentioned boundary condition can be solved by using the separation-of-variables technique if we assume that the spatial distribu-

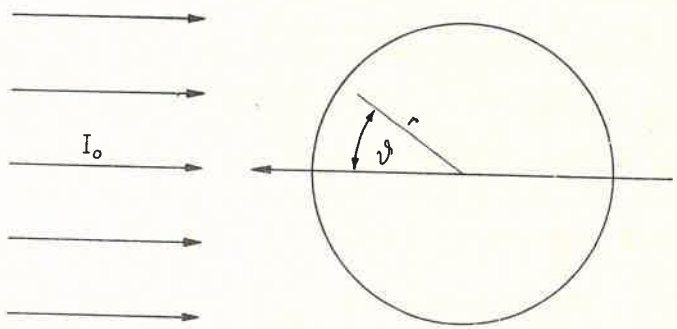


Fig. 5

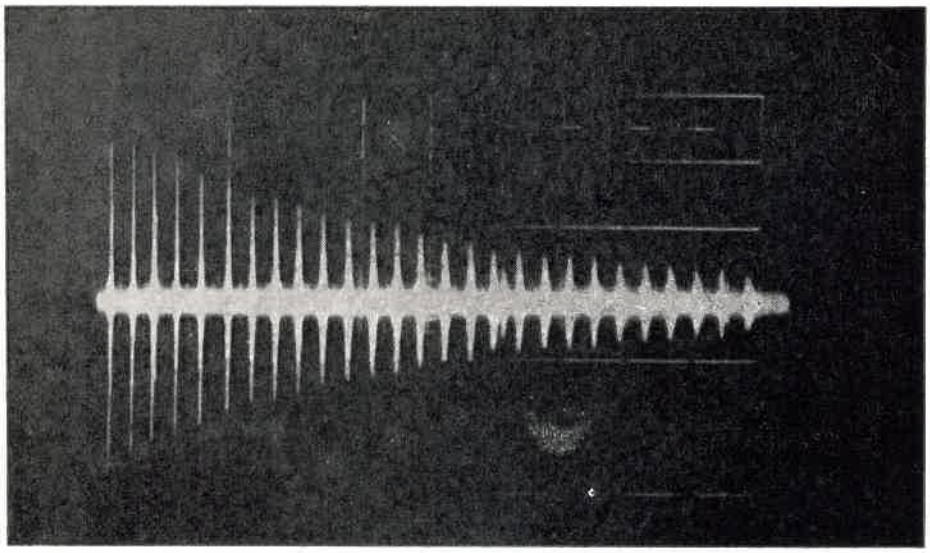
tions for atoms in both hyperfine states are proportional to each other. Taking into account the symmetry introduced to the system by the pumping process one can write the most general solutions (in spherical coordinates — Fig. 5) as:

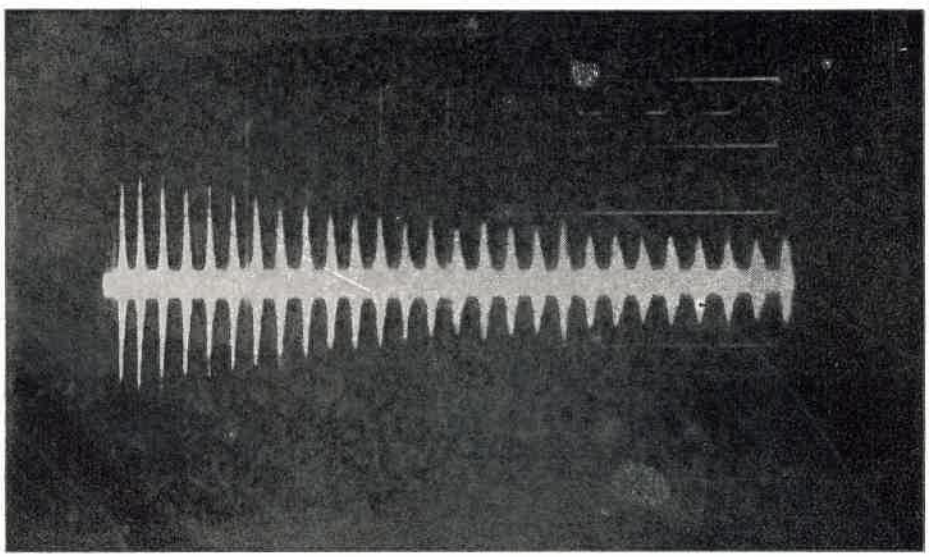
$$\langle S_z(r, \vartheta, t)\rangle_\pm = \sum_{k,m=0}^{\infty} J_{k+\frac{1}{2}}\left(\frac{\mu_m^k r}{R}\right)\frac{1}{\sqrt{r}} P_k(\cos\vartheta)$$

$$\times [a_\pm^{km} \exp(-Z_1^{km} t) + b_\pm^{km} \exp(-Z_2^{km} t)], \tag{18}$$

where: $P_k(\cos\vartheta)$ is the Legendre polynomial of order k, $J_{k+\frac{1}{2}}$ is the Bessel function of the order $k+\frac{1}{2}$, $a_\pm^{km}$ and $b_\pm^{km}$ are determined by the initial conditions, Z_1^{km} and Z_2^{km} are decay constants given by

$$Z_{1,2}^{km} = D\left(\frac{\mu_m^k}{R}\right)^2 + \frac{1}{T_Z} + \frac{1}{2T_h} + \tfrac{4}{3}\frac{I(I+1)+3}{3(2I+1)^2}\frac{1}{T_{ex}} \pm \frac{4I(I+1)}{2(2I+1)^2}\left(\frac{1}{T_h} + \tfrac{2}{3}\frac{1}{T_{ex}}\right)\eta, \tag{19}$$





b

Fig. 4. Typical signals for cesium atoms in neon: a — decay of signals for the F_+ (4) hyperfine state, b — decay of signals for the F_- (3) hyperfine state

where

$$\eta = \left[1 + \frac{24I(I+1)+9}{[4I(I+1)]^2} - \frac{8(2I+1)^2(1+T_h/T_{ex})}{[4I(I+1)]^2(1+2T_h/3T_{ex})^2}\right]^{1/2} \tag{20}$$

and μ_m^k are the roots of $J_{k+\frac{1}{2}}(\mu) = 0$. If we note that the signal is proportional to

$$S_\pm \sim \int_V \langle S_z(r, \vartheta, t)\rangle_\pm dv \tag{21}$$

where V is this part of the cell through which the detection beam passes, then in accordance with our previous considerations [11] we need to take into account only the first few terms for $k = 0,1$.

However, when the spatial distribution of the oriented atoms is symmetrical in relation to the plane $\vartheta = \pi/2$ only the $k = 0$ terms will appear in the series and the shape of the signal will be given by

$$S_\pm \sim \sum_{m=0}^{\infty} A_\pm^m \exp(-Z_1^m t) + B_\pm^m \exp(-Z_2^m t). \tag{22}$$

The coefficients $A_\pm^m$ and $B_\pm^m$ are determined by the initial conditions at $t = 0$ and are proportional to

$$\int_V J_{\pm\frac{1}{2}}\left(\frac{\mu_m^0 r}{R}\right)\frac{1}{\sqrt{r}}\,dv \tag{23}$$

hence they change similarly and depend on the method of detection. This problem has been thoroughly discussed in paper [11].

When the monitoring process is carried out with a low intensity beam travelling through the central part of the cell (the radius of the beam $r = 0.55\ R$) the signal can be well approximated by the first term $m = 0$.

Choosing a proper method of observation we will be able to describe the relaxation process by a two-exponential decay curve. The decay constants in the discussed case can be expressed as follows:

$$\begin{aligned} Z_1^0 &= D\left(\frac{\pi}{R}\right)^2 + \frac{1}{T_Z} + \lambda_1\frac{1}{T_h} + \mu_1\frac{1}{T_{ex}}, \\ Z_2^0 &= D\left(\frac{\pi}{R}\right)^2 + \frac{1}{T_Z} + \lambda_2\frac{1}{T_h} + \mu_2\frac{1}{T_{ex}}, \end{aligned} \tag{24}$$

where

$$\begin{aligned} \lambda_{1,2} &= \frac{4I(I+1)(1\mp\eta)+1}{2(2I+1)^2}, \\ \mu_{1,2} &= \frac{4I(I+1)(1\mp\eta)+3}{3(2I+1)^2}. \end{aligned} \tag{25}$$

The sign − refers to index 1, and sign + to index 2. Since η in a slowly varying function of the ratio T_h/T_{ex}, so are factors $\lambda_{1,2}$ and $\mu_{1,2}$ too.

The dependence of η on T_h/T_{ex} for different values of the nuclear spin is shown in Table I. It is easy to see that if we take $\eta = 1$ we will get a fairly good approximation.

TABLE I

T_h/T_{ex} \ J	7/2	5/2	3/2
0	0.98413	0.97143	0.93333
1	1.00231	1,00474	1.01509
∞	1.04762	1.08571	1.20000

The solution obtained above is not the most general one, but it seems to us to be better than the previous approximations although there are certain complications which arise due to the form of the decay constants (24). Here we have in mind the more troublesome mathematical procedure which leads to the determination of the cross-sections for different processes. We define these cross-sections as follows:

$$\sigma_Z = (T_Z v_r' n)^{-1}, \quad \sigma_h = (T_h v_r' n)^{-1}, \quad \sigma_{ex} = (T_{ex} v_r N)^{-1}, \tag{26}$$

where n and N are the numbers of the atoms of the buffer gas and alkali atoms in 1 cm^3, v_r' and v_r are the relative mean velocities of the interacting atoms respectively.

4. Experimental results

4.1. The contributions of different diffusion modes

From the study in the preceding section, we see that under certain experimental conditions we can observe only one term of the expansion (22). To verify this conclusion we have investigated the dependence of the decay curve as a function of the radius of the detection beam. The direction of the monitoring beam coincidences with the axis of symmetry of the system. When we restrict the analysis of the signal to only two diffusion modes the decay curve will be described by four constants Z_1^m and Z_2^m for $m = 0.1$.

The least squares method modified by Newton–Raphson has been used to approximate the decay curves by a series of exponential functions. The decomposition of the decay curve into four exponential functions was not unique. For studying the relaxation processes for atoms in the F_+ hyperfine state the coefficients B_+^m in the series expansion were much smaller than A_+^m and the decay constants Z_2^m much larger than Z_1^m. So the best representation of the experimental data is given by a two term expansion (22).

Using the experimental technique discussed in the first section of this paper we have made measurements for optically pumped Cs in Ne buffer at the temperature 25.5°C. Experimental data from the double-exponential fit are presented in Fig. 6 and 7. The evaluated values of the diffusion coefficient from these measurements is $D_0 = 0.22$ cm^2 s^{-1}

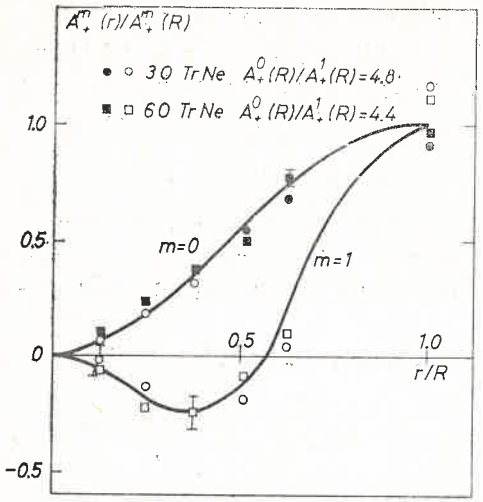


Fig. 6. The dependence of the amplitude of different diffusion modes as a function of the radius of the detection beam. The amplitudes have been normalized to one for the radius of the detection beam equal to R. The solid line is the predicted curve

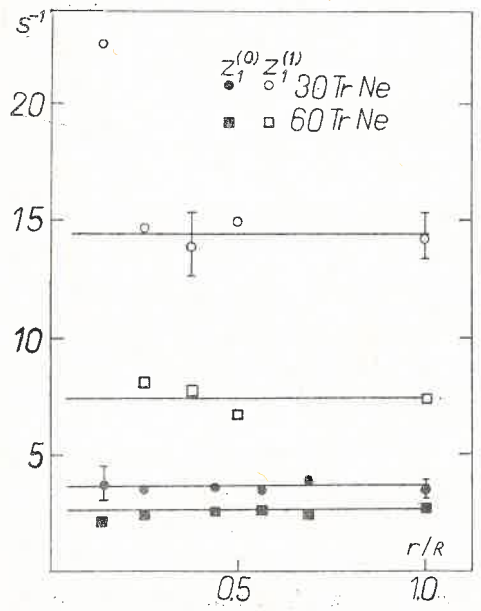


Fig. 7. Decay constant Z_1 for the F_+ state plotted against the radius of the detection beam

for 30 Tr pressure of the buffer gas and $D_0 = 0.20$ cm^2 s^{-1} for 60 Tr are in good agreement with previous valuations. However, the present experiment does not ensure that the precision of the measurement of the diffusion coefficient is high enough because of the smallness of the expansion constant of the term decaying with Z_1^1 and of the incompleteness of the series used for the description of the decay curve.

In our experiment we observed single exponential decay for the detection beam of the 1.2 R diameter which is slightly bigger than the calculated value. We believe that this discrepancy can be caused by the scattered radiation from the walls of the bulb. In further experiments the diameter of the detection beam was set to 1.2 R.

4.2. Measurements of the relaxation processes

Systematic measurements of relaxation processes for cesium atoms with neon as the buffer gas were carried out under such experimental conditions that only one diffusion mode was observed. The experimental data obtained for the buffer gas at pressures ranging

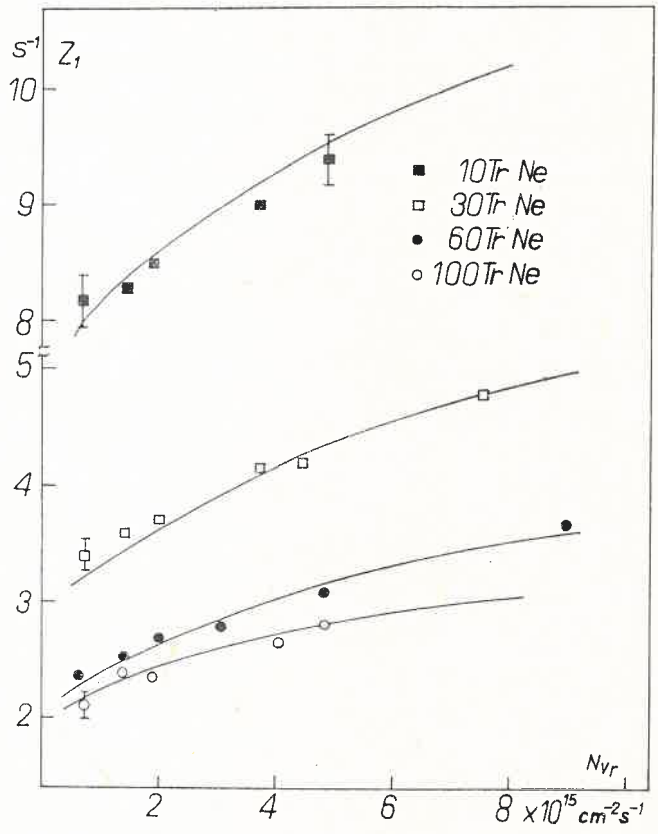


Fig. 8. Decay constant Z_1 plotted against Nv_r for F_+ state

from 10 to 100 Tr neon and temperatures from 18°C to 40°C. The constant magnetic field $\vec{H}_0$ about 1.5 Oe was high enough to separate the two Zeeman hyperfine resonances even for an oscillating rf field. For smaller fields $\vec{H}_0$ one has to use rotating rf fields. Our measurements are summarized in Fig. 8, 9 and 10. The decay curve for the F_+ hyperfine state, was close to a single-exponential. However, the decay curve for the F_- hyperfine state could be well fitted by two exponential functions. One of them, with a larger coefficient,

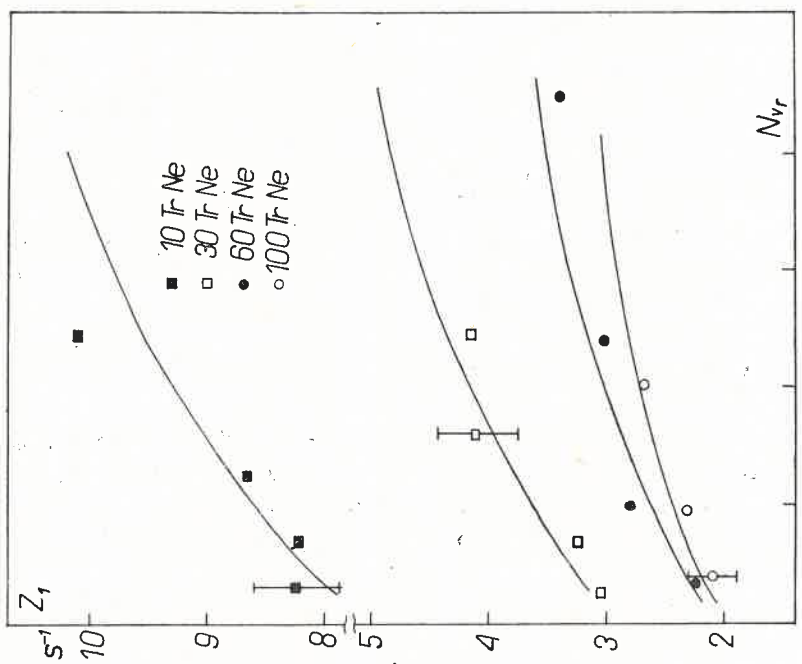


Fig. 9

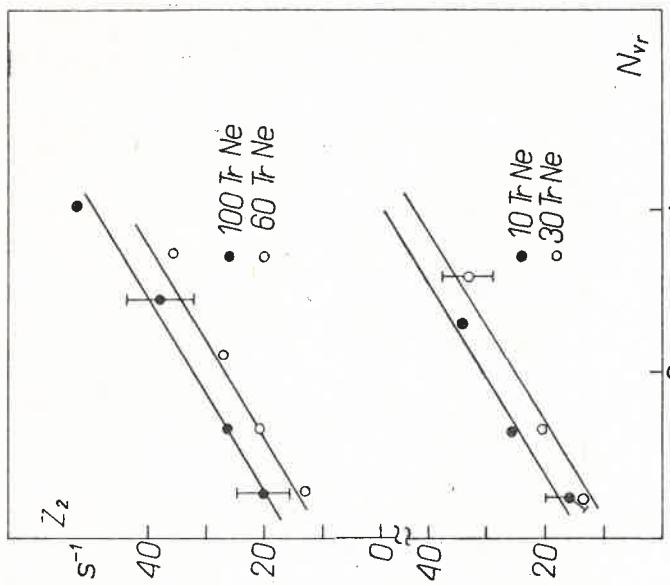


Fig. 10

Fig. 9. Plot of Z_1 vs Nv_r for the F_- hyperfine state
Fig. 10. Plot of Z_2 vs Nv_r for the F_- hyperfine state

is within the limits of error, the same as for F_+ hyperfine state. The other one has a coefficient with opposite sign. When we take into account only the first two terms of the expansion (22) the signal can be described by

$$S_\pm = A_\pm \exp(-Z_1 t) + B_\pm \exp(-Z_2 t), \tag{27}$$

where the index $m = 0$ has been left out.

When strong electronic relaxation exists in the optically pumped (with $D_1\sigma_+$ resonance light) system, $\langle S_z(0)\rangle_+$ is positive and $\langle S_z(0)\rangle_-$ negative. In such a case

$$A_+ > B_+ > 0, \quad A_- > 0, \quad B_- < 0$$

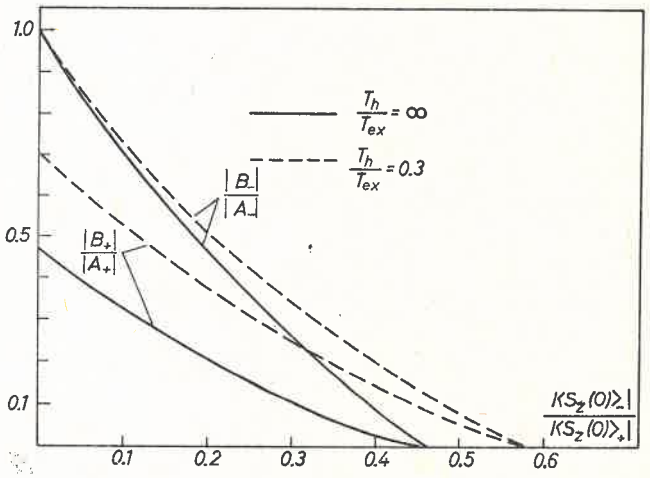


Fig. 11. Plot of the ratio of amplitudes of the exponential functions of the signal vs $|\langle S_z(0)\rangle_-|/|\langle S_z(0)\rangle_+$ for different values of T_h/T_{ex}

and the ratio of the coefficients depends on the initial conditions and on T_h/T_{ex} (Fig. 11). There is qualitative agreement of the experimental results with the assumed relaxation model. It is revealed in

a. equal values of the decay constants for both hyperfine states F_+ and F_-,

b. opposite signs of the coefficients of the "slow" and "fast" decay modes for F_-,

c. the linear dependence of the decay constant Z_2 on the density of Cs in the bulb.

For the F_+ state the observed decay curve is a single exponential and the coefficient of the fast decay mode could not be determined, because of the imperfections in the measuring system, especially due to the fairly long closing time of the mechanical shutter.

After determining the fast and slow decay components of the decay curve one can calculate the cross-sections σ_z, σ_h and σ_{ex} by making use of the least squares method with a simultaneous iteration procedure for the ratio T_h/T_{ex}. We have taken advantage of the single exponential approximation for the determination of the initial values of σ_h and σ_{ex} for the iteration procedure.

The pressure of cesium vapour has been determined using the formulas given by Taylor and Langmuir [28].

For the diffusion coefficient of cesium in neon buffer gas the following temperature dependence has been assumed

$$D_0 = AT^{1.8}. \tag{28}$$

The best fits to the experimental data were achieved when an additional term $N\sigma' v_r$, depending on the pressure of the cesium vapour was added to the rate constant Z_1. This is shown in Fig. 8, 9 and 10, where the solid lines give the best fits. Our numerical data together with earlier measurements are summarized in Table II. Our present measurements

TABLE I

D_0	0.15 ± 0.02 cm^{2}s^{-1}	0°C	this work
	0.15		Ernst, Strumia [7]
	0.24	26°C	Łęgowski [29]
	0.40	44°C	Franz, Lüscher [30]
	0.198		Bylicki, Łęgowski [31]
	0.153		Beverini et al. [33]
σ_z	$5.2 \pm 0.55 \times 10^{-24}$ cm^2		this work
σ_h	$7.5 \pm 1.0 \times 10^{-23}$ cm^2		this work
	9.0×10^{-23}		Ernst, Strumia [7]
σ_{ex}	$1.5 \pm 0.25 \times 10^{-14}$ cm^2		this work
	2.2×10^{-14} cm^2		Ernst, Strumia [7]
	2.4×10^{-14} cm^2		Bouchiat et al. [4]
	2.06×10^{-14} cm^2		Ressler et al. [32]
σ'	$2.3 \pm 1.2 \times 10^{-16}$ cm^2		this work
σ_n	$6.4 \pm 0.6 \times 10^{-24}$ cm^2		this work
	5.3×10^{-24}		Franz, Lüscher [30]
	8.4×10^{-24}		Łęgowski [29]
σ_e	$7.9 \pm 1.0 \times 10^{-23}$ cm^2		this work

of the diffusion coefficient agree quite well with the results obtained by Ernst, Strumia, Beverini, Minguzzi, Bylicki and Łęgowski. The remaining data are outside the limits of error and show a faster increase with temperature than the power 1.8.

The cross-sections σ_h and σ_n can be compared with the results of Ernst, Strumia, Franz Lüscher and Łęgowski. The agreement seems to be fairly good. However, for the fast collisions the ratio T_z/T_h should be of the order of $(2I+1)^2$ which is not observed in the present case. It does not seem likely that the formation of the Van der Waals molecules Cs–Ne contributes to the relaxation process because of the smallness of the dissociation energy of Cs–Ne molecule. It seems to us that it is rather the interaction of polarized Cs atoms with molecules Cs_2 that contributes to the measured relaxation rates. Here we have in mind the exchange processes of cesium atoms as well as the dissocia-

tion of Cs_2 molecules in collisions with buffer gas atoms [34]. All these are processes destroying the nuclear polarization of the system and changing mainly the nuclear relaxation time T_n.

Our present measurement of the exchange cross-section σ_{ex} differs from the values determined by other authors. The major disagreement lies in the determination of the pressure of cesium vapour. It has been observed that the vapour pressure depends very strongly on the buffer gas pressure [35] and decreases with an increase of the buffer gas pressure. Therefore the effective cross-section in the whole range of buffer gas pressure is lowered. Because of the low accuracy with which the relaxation rate T_{ex}^{-1} was determined, it was impossible to take into account the last effect.

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