INVESTIGATION OF TRANSVERSE RELAXATION PROCESSES OF OPTICALLY PUMPED CESIUM VAPOUR IN THE PRESENCE OF NEON BY MEANS OF THE SPIN ECHO METHOD

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The transverse relaxation times of optically pumped cesium vapour in the presence of neon were measured by Carr-Purcell's spin echo method. The resonance cell was placed in constant magnetic field H_0 with a given gradient. Measurements were carried out using rotating rf field ω_F^0 at resonance with a chosen hyperfine state F. The evaluated cross section for spin exchange is $\sigma_{ex} = (2.28 \pm 0.1) \times 10^{-14}$ cm^2, the cross section for disorientation collisions with foreign gas atoms $\sigma_g = (2.4 \pm 0.3) \times 10^{-23}$ cm^2 and the ratio of transverse relaxation times $T_2^{F-}/T_2^{F+} = 0.7 \pm 0.1$ (at temperature 301 K and pressure $p_{Ne} = 60$ Tr). Besides temperature dependence of the diffusion coefficient $D_0(T) = 0.198 \times (T/273$ K$)^{2.3}$ cm^2 s^{-1} was determined.

1. Introduction

The aim of this work was to determine the relaxation times T_2^F of the expectation values $\langle S_x(t) \rangle_F$ of an optically orientated system of cesium Cs^{133} atoms in the ground state in the presence of buffer gas. The Carr-Purcell spin echo method [1, 2] is particularly useful here for it eliminates the influence of the inhomogeneity of the magnetic field upon decay constants, and it allows one to make the exact measurements of the diffusion coefficient D_0. Previous measurements of D_0 in optically pumped gaseous systems were mostly based on the dependence of the longitudinal relaxation time on the buffer gas pressure. The method of the multiple spin echoes enables us to evaluate the diffusion coefficient at a given buffer gas pressure employing a constant magnetic field with the exactly known gradient constant through the sample $\vec{H} = \vec{H}_0 + [(\vec{r}\nabla)\vec{H}]_0 = \vec{k}[H_0 + zG]$. Choosing appropriate detection method the envelope of the spin echoes can be described as follows

$$S_{F\pm}(t) = S_{F\pm}(0) \exp\left\{-\left[\frac{1}{T_2^{F\pm}} + \frac{D\gamma_{F\pm}^2 G^2}{12f^2}\right]t\right\}, \qquad (1)$$

where D is the diffusion coefficient, $\gamma_{F\pm}$ — giromagnetic ratio, $G = \partial H/\partial z$ — field gradient, f — frequency of spin echo occurrence.

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A strong dependence of the transverse relaxation times upon the strength of the magnetic field was observed when measurements were carried out by means of the spin echo method — Fig. 1. This dependence has not been taken into account in previous papers [3–7].
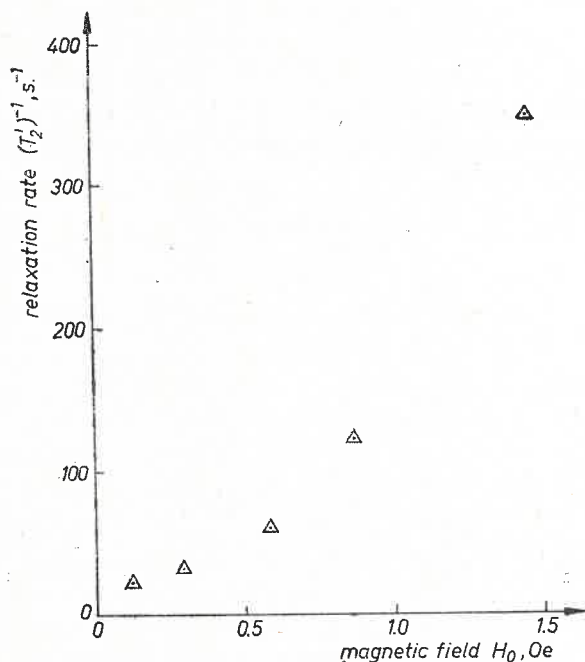

Fig. 1. Dependence of decay rate of Carr-Purcell's spin echoes upon intensity of the constant magnetic field H_0. $p_{Ne} = 60$ Tr, $T = 308$ K, $F^+ = 4$

2. Transverse relaxation of cesium in the presence of neon

Let us describe the physical quantity of interest like S_x, S_y, S_z generally by the observable Q and the corresponding expectation value $\langle Q \rangle$. When several independent processes take place in the investigated system, the time evolution of the expectation value $\langle Q \rangle$ can be described by a set of differential equations of the form

$$\frac{d\langle Q \rangle}{dt} = \frac{d^{(1)}\langle Q \rangle}{dt} + \frac{d^{(2)}\langle Q \rangle}{dt} + \frac{d^{(3)}\langle Q \rangle}{dt} + \frac{d^{(4)}\langle Q \rangle}{dt} + \frac{d^{(5)}\langle Q \rangle}{dt} \tag{2}$$

and it is possible to treat each relaxation process separately. $d^{(1)}\langle Q \rangle/dt$ describes the evolution of $\langle Q \rangle$ influenced by the interaction of the spin system with internal and external magnetic fields. The radio frequency pulses are not taken into account since they do not disturb the relaxation processes. They are only introduced to eliminate the influence of the inhomogeneity of the constant magnetic field [1]. Consequently, it will be assumed that H_0 is homogeneous. The term $d^{(2)}\langle Q \rangle/dt$ describes the effect of the relaxation caused by collisions of the alkali atoms with the walls after diffusion through

the buffer gas. The term $d^{(3)}\langle Q\rangle/dt$ describes the relaxation caused by the Cs-gas atoms collisions, $d^{(4)}\langle Q\rangle/dt$-relaxation caused by the spin exchange collisions. The term $d^{(5)}\langle Q\rangle/dt$ describes the interaction with the pumping and detection beams. However, this last term can be omitted for observations carried out in "darkness", using very weak detection beam.

The ground state of cesium is composed of two hyperfine sublevels $F^{\pm} = I \pm 1/2$ separated by the energy difference ΔW. The giromagnetic ratios of these sublevels are of different signs. For further considerations let us assume that the coherence between Zeeman sublevels of the hyperfine state F^+ and F^- may be excluded.

The expectation value $\langle Q\rangle$ may be represented as $\langle Q\rangle = \sum_i Q_i$ and the relaxation process for each Q_i can be described by one decay time [9]. According to the theoretical considerations of Bouchiat [10, 11] and Grossetête [12, 13], in the case the condition

$$(\omega_{F,m_F;F,m_F-r} - \omega_{F,m_{F'};F,m_{F'}-r})T_2 \ll 1 \tag{3}$$

is fulfilled, the decay of $\langle S_x\rangle^*_{F\pm}$ is described by a single exponential function. $\omega_{F,m_F;F,m_F-r}$ — energy difference between Zeeman sublevels $|F, m_F\rangle$ and $|F, m_F-r\rangle$ in the frequency scale, T_2 — the transversal relaxation time, $\langle S_x\rangle^*_F$ — transversal component of electronic polarization in the frame rotating with frequency $\vec{\omega}_F$ (mean resonance frequency). In case when condition (3) is not fulfilled, the problem becomes more complicated and $\langle Q\rangle$ has to be represented by $4 \times I$ values Q_i having different decay constants. In this case the evolution of the elements of the density matrix depends upon F and m_F. When condition (3) is fulfilled the case becomes much more interesting because of simple interpretation which can be given to the experimental results and possibilities of obtaining very interesting corollaries. It turns out that $\langle S_x\rangle^*_F$ have different decay constants, different for F^+ and F^- states. Hence, if one wants to observe signals from one hyperfine state, the second must be necessarily eliminated. Using a rotating rf field $\vec{H}_1(t)$ it is possible to separate both components.

Theoretical considerations of various types of interactions lead to different relations between the transversal relaxation time T_2, and the longitudinal relaxation time T_1 of spin $S = 1/2$. Considering the spin exchange collision processes (as examined in [14], under the condition $\omega_F T_{ex} \gg 1$) between alkali atoms we obtain

$$\frac{d^{(4)}\langle S_x\rangle^*_{F^+}}{dt} = -\frac{1}{T_{ex}}\left[\frac{2I(2I-1)}{3(2I+1)^2}\right]\langle S_x\rangle^*_{F^+}, \tag{4a}$$

$$\frac{d^{(4)}\langle S_x\rangle_{F^-}}{dt} = -\frac{1}{T_{ex}}\left[\frac{2(2I^2+5I+3)}{3(2I+1)^2}\right]\langle S_x\rangle^*_{F^-}, \tag{4b}$$

where T_{ex}^{-1} is the spin exchange rate. Following dependencies arise for the interactions of cesium atoms with buffer gas [10]

$$\frac{d^{(3)}\langle S_x\rangle^*_{F^+}}{dt} = -\frac{1}{T_r}\left[1 - \frac{I(2I+3)}{(2I+1)^2}\right]\langle S_x\rangle^*_{F^+}, \tag{5a}$$

$$\frac{d^{(3)}\langle S_x\rangle^*_{F^-}}{dt} = -\frac{1}{T_r}\left[1 - \frac{(I+1)(2I-1)}{(2I+1)^2}\right]\langle S_x\rangle^*_{F^-}, \tag{5b}$$

where T_r is an electron randomization relaxation time [15].

Under certain condition outlined in [9] one can write down the following diffusion equation for the expectation value of $\langle S_x \rangle_F^*$

$$\frac{\partial}{\partial t} \langle S_x(\vec{r}, t) \rangle_F^* = D\nabla^2 \langle S_x(\vec{r}, t) \rangle_F^*,$$

with the boundary condition

$$\langle S_x(\vec{r}, t) \rangle_F^* \big|_{r=R} = 0. \tag{6}$$

Hence, there arises the differential equation for the local mean quantity $\langle S_x(\vec{r}, t) \rangle_F^*$, describing all relaxation effects within the resonance cell

$$\frac{\partial}{\partial t} \langle S_x(\vec{r}, t) \rangle_{F\pm}^* = D\nabla^2 \langle S_x(\vec{r}, t) \rangle_{F\pm}^* - \frac{1}{T_2^{F\pm}} \langle S_x(\vec{r}, t) \rangle_{F\pm}^*, \tag{7}$$

where $T_2^{F\pm}$ describes the effects of spin exchange collisions and cesium-buffer gas interactions. The spatial boundary condition (6) implies that the general solution of (7) in terms of exponential function, averaged over the whole resonance cell, is

$$\langle S_x(t) \rangle_{F\pm}^* = \langle S_x(0) \rangle_{F\pm}^* \sum_{\nu=1}^{\infty} A_\nu \exp\left\{-\left[D_0 \frac{p_0}{p} \frac{\nu^2\pi^2}{R^2} + \frac{1}{T_2^{F\pm}}\right]t\right\}, \tag{8}$$

where D_0 denotes diffusion constant, p — buffer gas pressure, $p_0 = 760$ Tr, R — radius of a spherical resonance cell. The coefficients A_ν are determined by the initial conditions at $t = 0$. Choosing the appropriate geometry of the detection beam we can obtain a signal S_F for which the contributions of modes higher than one are insignificant and can be omitted [16]. We obtain a single exponential decay

$$S_{F+} = S_{F+}(0) \exp\left\{-\left[D_0 \frac{p_0}{p} \frac{\pi^2}{R^2} + \frac{1}{T_r}\left(1 - \frac{I(2I+3)}{(2I+1)^2}\right)\right.\right.$$

$$\left.\left. + \frac{1}{T_{ex}} \frac{2I(2I-1)}{3(2I+1)^2} + \frac{D_0 p_0 \gamma_{F+}^2 G^2}{12 f^2 p}\right]t\right\}, \tag{9a}$$

$$S_{F-} = S_{F-}(0) \exp\left\{-\left[D_0 \frac{p_0}{p} \frac{\pi^2}{R^2} + \frac{1}{T_r}\left(1 - \frac{(I+1)(2I-1)}{(2I+1)^2}\right)\right.\right.$$

$$\left.\left. + \frac{1}{T_{ex}} \frac{2(2I^2+5I+3)}{3(2I+1)^2} + \frac{D_0 p_0 \gamma_{F-}^2 G^2}{12 f^2 p}\right]t\right\}, \tag{9b}$$

where the influence of the field gradient is included.

3. Experiment

Measurements of the transversal relaxation times $T_2^{F\pm}$ of cesium in neon as buffer gas have been carried out using a typical optical pumping apparatus — Fig. 2. Because a circularly polarized beam directed along the Ox axis (D_1 line without employing any special techniques of separation of the hyperfine components — assumption of a "broad line")
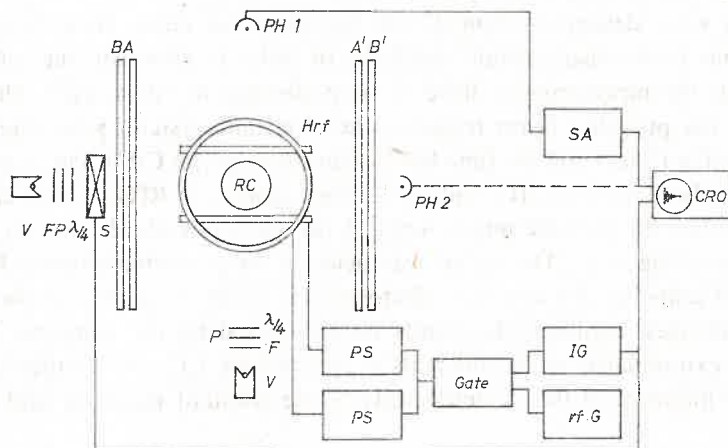

Fig. 2. A diagram of the used apparatus. AA', BB' — steady magnetic field coils, V — hf cesium lamp, P $\lambda/4$ F — optical system. Description within the text

was used for detection of the spin echoes, the obtained signal was proportional to $1/2 - \langle S_x \rangle$ [8]. Consequently, the amplitude $S_F(t) = S_F(n/f)$ of the echo signal after n-th π-pulse ($n = 0, 1, 2, ...$) will be expressed by the quantity proportional to $\langle S_x(t) \rangle_F^*$.

In order to fulfil (3), $H_0 < H_0^{crit}$ has to be used. Because of the dependence of decay rate $(T_2')^{-1}$ of the envelope of the spin echoes upon intensity of constant magnetic field, one can determine H_0^{crit} experimentally (assuming that the accuracy of the measurements is of the order of few per cent) — Fig. 1

$$\frac{1}{T_2'} = \frac{1}{T_2^{F\pm}} + \mu H_0^2,$$

where μ — coefficient depending on T_{ex} and T_r. H_0^{crit} corresponds to the intensity of magnetic field, for which

$$(\omega_{F,m_F;F,m_F-1} - \omega_{F,m_F';F,m_F'-1})T_2^{F\pm} \approx 1.$$

On the other hand the conditions $T \ll \Delta t_i \ll T_f < T_2^{F\pm}$ must be fulfilled, where $T = 2\pi/\omega_F$ is the period of vibrations of the rf field H_1, Δt_i — duration of the pulse $\pi/2$, T_f — decay time of the free precession. For the used experimental apparatus $T_2^{F+} \approx 50$ ms and $T_f \approx 3$ ms, hence the steady magnetic field should be $H_0 \approx 0.1$ Oe. A system of coils

placed on a surface of sphere with diameter 1.2 m was used to produce a constant magnetic field of high degree of homogeneity [17].

The rotating rf field in the region of resonance cell RC ($R = 4$ cm) was obtained by using two perpendicular Helmholtz coils Hrf with radius 13 cm, supplied by an rf generator RfG through phase shifter PS systems. Experimental measurements carried out using Meiboom-Gill spin echoes method [18] have confirmed high degree of homogeneity of rf-field in the region of the resonance bulb.

For very weak detection beam $I_0^d \approx 0$ the observed decay time does not depend on I_0^d (this has been experimentally verified). In order to eliminate the influence of the pumping light the measurements have to be performed in "darkness". The mechanical shutter S of the pumping beam triggered the electronic system, producing appropriate pulses of rotating rf field and the time basis of an oscillograph CRO. Pulses were produced by a set of pulse generators IG, radio-frequency generator RfG, gating circuit "Gate" and phase shifters PS. Suitable pulses supplied two pairs of coils Hrf, which produced the appropriate rotating field. The signal was detected by a photomultiplier PH1 followed by a selective amplifier SA and was photographed from the screen of the oscillograph CRO. The obtained oscillograph records were analyzed by the computer ODRA 1204. Results were extrapolated to the intervals of spin echoes $1/f \to 0$ in order to neglect the effects of the influence of the inhomogeneity of the constant magnetic field on the decay time $T_2^{F\pm}$.

The obtained data for T_2 allow one to calculate the cross section for spin exchange σ_{ex}, cross section for disorientation of the spin in the Cs–Ne interactions σ_g, and the diffusion constant D_0. The cross section describing the cesium-cesium interactions was obtained from the expression

$$\sigma_{ex} = (T_{ex} N_{Cs} v_{rel}^{CsCs})^{-1}. \tag{10}$$

T_{ex} depends on temperature, because cesium vapour density markedly changes with the temperature. The cross section describing the cesium-neon interactions is determined by the following expression

$$\sigma_g = (T_r N_{Ne} v_{rel}^{CsNe})^{-1}. \tag{11}$$

T_r depends on the neon filling pressure and on the temperature of the resonance cell. The measurements were made for the $F^+ (= 4)$ state owing to a greater signal to noise ratio, and consequently greater accuracy of results. A series of measurements was also made for the $F^- (= 3)$ state. $T_2^{F^-}/T_2^{F^+} = 0.7 \pm 0.1$ was obtained for a cell filled with neon buffer gas at pressure 60 Tr and temperature 301 K. The measurements of $T_2^{F^+}$ were made for the neon pressure $p_{Ne} = 11.4 \div 400$ Tr and for the temperatures $290 \div 330$ K every 2 K. Some results are shown in Fig. 3.

To determine the temperature dependence of the diffusion constant independent measurements were carried out for the resonance cell placed in the magnetic field with the exactly known gradient (Eq. (1)). In order to eliminate stationary inhomogeneities of the magnetic field, measurements were made for two directions of the gradient and the
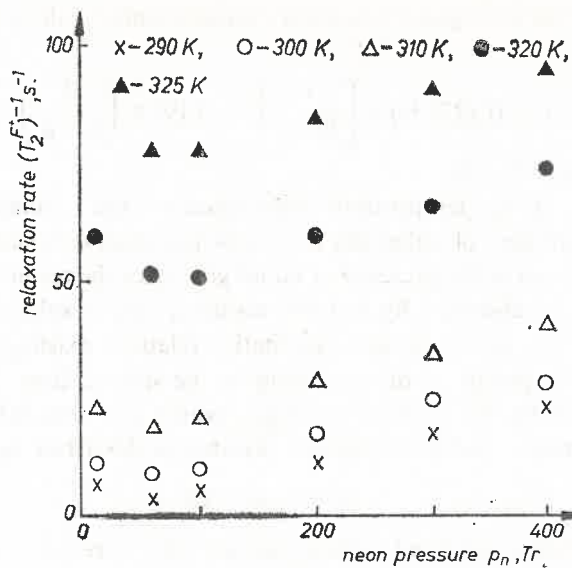
Fig. 3. Some extrapolated values of decay constants $(T_2^{F+})^{-1}$ for $H_0 = 0.1$ Oe. p_n — neon pressure in the resonance cell measured at the filling temperature 301 K
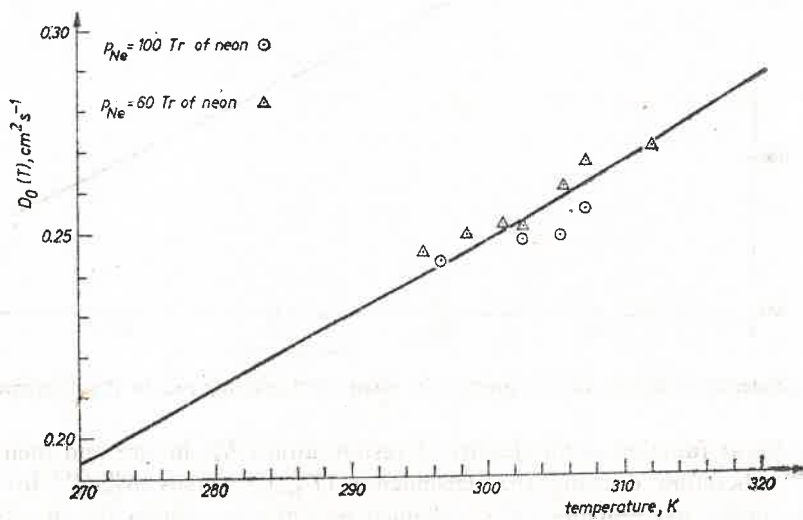

Fig. 4. Temperature dependence of the diffusion constant D_0

obtained results were averaged. From these measurements, made at different temperatures, the dependence

$$D_0(T) = D_0(273 \text{ K}) \times \left[\frac{T}{273 \text{ K}}\right]^\alpha = 0.198 \times \left(\frac{T}{273 \text{ K}}\right)^{2.3} \text{ cm}^2\text{s}^{-1} \tag{12}$$

was obtained [19] — Fig. 4.

Examination of the temperature dependence of the relaxation caused by Cs–Cs collisions in the presence of buffer gas at various pressures may supply information about metal vapour pressure in the presence of buffer gas, since the resonance cell was filled with neon, which can be absorbed by metallic cesium giving a solution. However, the data obtained in that way do not render quantitative relations existing in the entire solution, but will give only a picture of those existing on the surface layer. It is caused by a neon adsorption process on the boundary surface, where gas concentration is greater. The assumption was made, that the observed pressure is described by the Raoult Law

$$p_i = p_{0i}(1 - \xi p_{Ne}), \tag{13}$$

where p_i is the cesium saturated vapour pressure, p_{0i} — cesium vapour pressure in the absence of the buffer gas, described by Taylor and Langmuir [20], ξ — coefficient.

Assuming that the neon solubility in cesium does not depend on temperature (which is a coarse approximation), we may eliminate from observed decay constants the diffusion terms and the contributions responsible for the Cs–Ne interaction. Because the component
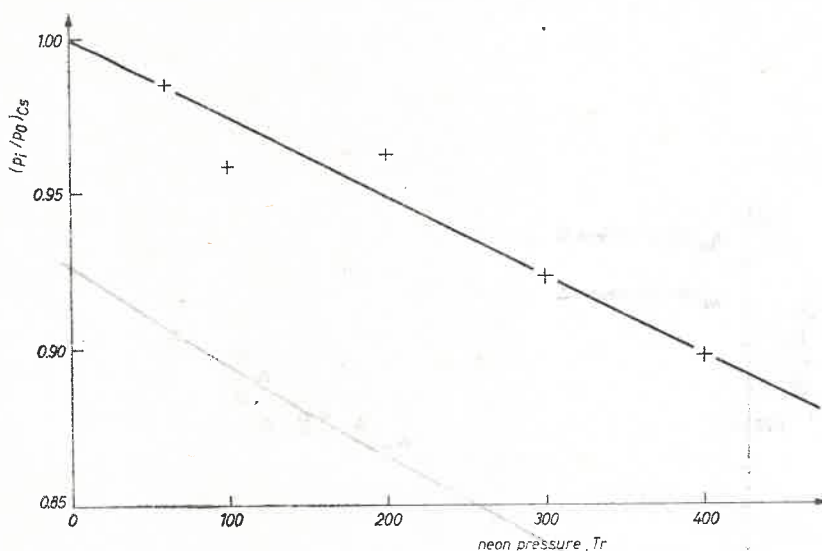

Fig. 5. Dependence of cesium vapour pressure p_i upon neon pressure p_{Ne} in the resonance cell

$(T_{2ex}^{F+})^{-1}$ is a linear function of the density of cesium atoms N_{Cs} in cm³ and their relative velocity v_{rel}^{CsCs}, therefore drawing the dependence $(T_{2ex}^{F+})^{-1}$ versus $N_{Cs}^0 v_{rel}^{CsCs}$ for cells of various neon buffer gas pressure (N_{Cs}^0 is defined by [20]), we obtain for the coefficient the value $\xi = (2.5 \pm 1) \times 10^{-4} \text{ Tr}^{-1}$ (from equation (13)) — Fig. 5.

TABLE

Quantity	Results	Authors
$\sigma_{ex}[cm^2]$	$(2.28 \pm 0.1) \times 10^{-14}$ 6.4×10^{-14} $(2.3 \pm 0.2) \times 10^{-14}$ 1.95×10^{-14}	This work [6] [7] [21]
$\sigma_g[cm^2]$	$(2.4 \pm 0.3) \times 10^{-23}$ 1.8×10^{-23} $(2.6 \pm 0.2) \times 10^{-23}$ 1.8×10^{-23}	This work [6] [7] [21]
$D_0(273 \text{ K}) \; [cm^2 s^{-1}]$	(0.198 ± 0.02) (0.12 ± 0.03) 0.19 0.23 0.12	This work, (12) This work, (8) [6] [7] [21]
α	(2.3 ± 0.5)	This work
$\xi[Tr^{-1}]$	$(2.5 \pm 1) \times 10^{-4}$ $(5 \pm 2) \times 10^{-4}$	This work [17]

Our results compared with those obtained by other authors for transverse relaxation processes are tabulated.

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