Suppression of relaxation of higher polarization moments of alkali atoms in superfrequent spin-exchange collisions

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Abstract. Consideration of nonlinear spin-exchange equations showed that, in addition to the well-known suppression of the relaxation of the transverse orientation of atoms in a low magnetic field, relaxation of higher polarization moments (alignment, octupole, hexadecapole, etc) are also suppressed. Such suppression of relaxation is caused by the conservation of the transverse angular momentum of atoms in collisions.

1. Introduction

The suppression of the relaxation of transverse orientation in the case of longitudinal optical polarization in a weak magnetic field (the collision frequency is much higher than Larmor frequency) is well known [1-3]. It has found application in magnetometry [4] and is called SERF (spin exchange relaxation free). Another method for suppressing such relaxation was realized by a strong increase in polarization [5], which leads to the accumulation of atoms in the “stretched” state. Spin exchange transitions from such a state to other states are forbidden by the law of conservation of angular momentum.

In this work, by solving nonlinear spin exchange equations, it is shown that in SERF mode in the presence of longitudinal polarization, transverse relaxation is suppressed, in addition to orientation, for all higher polarization moments (PMs). As a result, the transverse higher PMs become comparable in amplitude with the transverse orientation. In zero magnetic field, the transverse relaxation of all PMs becomes zero. That is, all transverse PMs are preserved in time.

Thus, considering the nonlinearity of the spin exchange equations significantly complements the picture of the PMs evolution compared to linear equations [3], which in the SERF mode predict a strong relaxation of the higher PMs, therefore, their vanishingly small value compared to the PM of rank 1 (orientation).

The solution of nonlinear equations of spin exchange was carried out earlier by Runge-Kutta method [2] and by the method of perturbation theory [6]. For the atomic density matrix in these equations, the usual FM representation was used, which made it difficult to analyze the evolution of PMs in collisions. Therefore, the effect of suppressing the relaxation of higher PMs was not revealed in [2, 6].

2. Statement of the problem

Collisions of atoms of an alkali metal in the ground $^2S_{1/2}$ state are considered. Atoms have nuclear spin $I$ and can be located in two hyperfine structure sublevels with moments $F^\pm = I \pm 1/2$. The evolution of the density matrix of these atoms due to collisions is described by the following equation [7] (for $L>0$):
\[
\frac{\partial}{\partial t} f^L_q(F) \bigg|_{L>0} = -\gamma_\text{ex} \sum_{L_1} \left\{ W^F_{l_1}(L) f^L_q(F_1) + \sum_{L_2} R^L_{l_1,l_2}(F_1,F_2) \left\{ f^{(L_1)}(F_1) \otimes f^{(L_2)}(F_2) \right\}_q \right\}
\]

(1)

Here the matrices \( W(L) \) and \( R \) are given by the following expressions:

\[
W^F_{l_1}(L) = \sum_{L_2} \left[ R^L_{l_1,l_2}(F_1,F_2) + \delta_{L,1} R^L_{0,l_2}(F_1,F_2) \right] \frac{\Pi_{F_2}}{2 \Pi_l}
\]

(2)

\[
R^L_{l_1,l_2}(F_1,F_2) = \left[ 1 + (-1)^{L_1+L_2} \right] \left( 1.5 \frac{I}{\Pi \Pi} \right) \Pi_{l_1}^2 \frac{\Pi_{l_2}^2}{2} \left[ \begin{array}{c} 1/2 \ 1/2 \\ 1/2 \ 1/2 \end{array} \right] \left[ \begin{array}{ccc} I & F_2 \\ L & L_2 \end{array} \right] \left( \begin{array}{c} 1/2 \ 1/2 \\ 1 \ 1 \end{array} \right) \left( \delta_{k,0} \delta_{L_2,0} - \delta_{k,0} \delta_{L_2,1} \right)
\]

Here \( \Pi_{ab...} = \left[ (2a+1)(2b+1)... \right]^{0.5} \).

For the density matrix \( \hat{f} \) in this work we use the representation of the polarization moments \( f^L_q(F) \) associated with the usual FM representation using the Clebsh-Gordan coefficients:

\[
f^L_q(L) = (\Pi_F / \Pi_L) \sum_{M} \sum_{q} (-1)^{F+M+L+q} C^L_{q FM} f^{FM}_{F,M'} f^{FM}_{FM'}
\]

Equation (1) contains the irreducible tensor product of PMs

\[
\left\{ f^{(L_1)}(F_1) \otimes f^{(L_2)}(F_2) \right\}_q = \sum_{q_1} \sum_{q_2} C^L_{q_1 q_2 l_1 l_2} f^{L_1}_{q_1}(F_1) f^{L_2}_{q_2}(F_2)
\]

and the frequency of spin-exchange collisions \( \gamma_\text{ex} = \nu \sigma_\text{ex} N_A \). Here \( \nu = \left( 16 k_B T / \pi m_A \right) \) is the average relative velocity of colliding atoms, \( k_B \) - the Boltzmann constant, \( T \) – temperature, \( m_A \) – the mass of an atom, \( \sigma_\text{ex} \) - the spin-exchange cross section and \( N_A \) is the concentration of atoms.

Equation (1) is a system of nonlinear differential equations. The first term in equation (1) characterizes its linear part and the second term is nonlinear, since it contains the product of two PMs of ranks \( L_1 \) and \( 1 \).

In this work, the following problem is solved. Alkali metal vapor is polarized by circularly polarized light propagating along Z axis. The constant magnetic field \( H_0 \) is also directed along Z axis. It is known [8] that in this case the density matrix is characterized by spin temperature parameter \( \beta \) and is given by the formula \( \hat{f} \sim \exp(\beta \hat{F}_Z) \). It gives the following expression for the PMs longitudinal components:

\[
f^L_0(F, \beta) = (\Pi_F / \Pi_L) \sum_{M} (-1)^{F+M+L} C^L_{0 FM} \exp(\beta M) \left[ \sum_{M'} \sum_{M'} \exp(\beta M') \right]^{-1}
\]

(3)

It follows from this expression that \( f^L_0(F, \beta) = 0 \) at \( \beta = 0 \).

It is required to find out how the transverse components of the PMs \( f^L_q(F) \bigg|_{q \neq 0} \) evolve in the field \( H_0 \) after the end of the light pulse that excited them.

The problem will be solved by the method of expanding the PMs in a perturbation theory series. The zero-order approximation is given by the longitudinal PMs of formula (3). The first-order approximation considers the contribution of transverse PMs. Accurate to first-order terms, the density matrix is written in the form:

\[
\frac{\partial}{\partial t} f^L_q(F) \bigg|_{q \neq 0} = ...
\]
Substituting this expression into equation (1), discarding the terms of zero and second orders and considering of the field $H_0$, we obtain the following first-order equation for the evolution of transverse PMs:

$$\frac{d}{dt} f_q^L (F) = \delta_{q,q_0} f_0^L (F, \beta) + \left(1 - \delta_{q,0}\right) f_q^L (F)$$

(4)

Here $g \left( F^+ \right) = \mp 2/(2I + 1)$ are the g-factors of $F^\pm$-states, $\mu_0 = \mu_B / \hbar$, $\mu_B$ is the Bohr magneton,

$$N_{F_1}^{LF} (q, \beta) = \sum_{F_2 \neq 0} \left[ R_{L_1(F_1),L(F_2)}^{LF} C_{Lq\rightarrow L_0}^{iq} f_0^L (F_2, \beta) + \delta_{L_1,\beta} \sum_{L_2 > 0} R_{L_1(F_1),L(F_2)}^{LF} C_{Lq\rightarrow L_0}^{iq} f_0^L (F_2, \beta) \right]$$

(5)

Thus, instead of nonlinear equation (1), linear equation (4) is obtained, which considers the longitudinal polarization of colliding atoms.

An important property of equation (4) is that it connects the PMs of different ranks $L$, but does not connect the PMs with different values of the projection $q$.

3. Calculation for alkali atoms with nuclear spin $I=3/2$

For such atoms, Eq. (4) includes PMs with ranks $L=1, 2, 3$ and 4 in a state with moment $F^+ = 2$ and PMs with ranks $L=1$ and 2 in a state with moment $F^- = 1$. For each value of $L$, the projection $q$ can take values $q = \pm 1, \pm 2, \pm 3$. In calculation, one may restrict oneself to calculating the PMs with only positive values of $q$, since PMs have the property

$$f_q^L (F) = (-1)^q \left[ f_q^L (F) \right]^*$$

The calculation will be carried out for the spin exchange of potassium atoms $K^{39}$ (I=3/2) using the cross section $\sigma_{ex} = 1.8 \cdot 10^{-14}$ cm$^2$ [9] and the dependence of $N_\lambda$ on $T$ from the reference book [10].

The system of equation (4) can be written in matrix form:

$$\frac{d}{dt} Y(q) = M (q, \beta, \gamma_{ex}, \omega_0) Y(q)$$

(6)

$$M (q, \beta, \gamma_{ex}, \omega_0) = -\gamma_{ex} N (q, \beta) - i \omega_0 q D (q)$$

$$N (q, \beta = V + N (q, \beta))$$

$$Y_{L_1F_1}^{LF} = \delta_{L_1,\beta} \left[ (1 - \delta_{L_2F_2 = -1}) \left(1 - \delta_{L_2F_2 = 1}\right) + \left(\delta_{L_2F_2 = -1} + \delta_{L_2F_2 = 1}\right) \delta_{F,F'} \delta_{F,F'} \right] W_{F_1}^{F} (L)$$

Here $\omega_0 = 2 \pi \gamma \gamma H_0$ is the Larmor frequency, $\gamma_k = 700$ kHz/Oe.

The solution of equations (6) requires finding the $n$ pieces of eigenvalues $\Lambda_k$ of the matrix $M$ having dimension $n \times n$. The quantities $\Lambda_k$ are $n$ roots of the equation

$$\left| M (q, \beta, \gamma_{ex}, \omega_0) - \Lambda_k [1]_n \right| = 0$$

(7)

Here $[1]_n$ is the unit matrix with dimension $n \times n$.

The dimension $n$ of the column of unknowns $Y(q)$ and the dimension $n \times n$ of matrices $N (q, \beta)$ and $D(q)$ in equation (6) depend on the magnitude of the projection $q$, since the inequality $L \geq q$ must hold. For values $q=1, 2, 3$ and 4 number $n$ equal 6, 4, 2 and 1, respectively.

Let us first consider the evolution of transverse PMs with projection $q=1$. With $q=1$ the number $n=6$. The matrix $D(1)$ and transposed column $Y(1)$ look like this:
To solve equation (6), it is necessary for each eigenvalue \( \Lambda_k \) of the matrix \( M \) to find the eigenvector \( R_{k,n} \) by solving the equation

\[
\begin{bmatrix}
M - \Lambda_k [1]_n
\end{bmatrix} R_{k,n} = (0)_n.
\]

Here \((0)_n \) is the zero column with dimension \( n \).

The solution of equation (6) for initial value of the column \( Y_{in} \) of unknowns at \( t=0 \) gives the following formula [11]

\[
Y_{in}(t) = \sum_{k=1}^{n} Z_{m,k} C_i \exp(\Lambda_i t)
\]

Here \( Z = \begin{bmatrix} R_{1,1} & \ldots & R_{1,n} \\ \vdots & \ddots & \vdots \\ R_{n,1} & \ldots & R_{n,n} \end{bmatrix} \) and \( C = Z^{-1} Y_{in} \).

Figure 1 shows the dependences of the quantities \( \Lambda_k = \Gamma_k + i\Omega_k \) on the field \( H_0 \) at \( T=473 \) K and on the temperature \( T \) at \( H_0=1 \) mOe. The calculations of these quantities were carried out by solving equation (7) at \( q=1 \) and \( \beta=1 \).

Figure 1. Parts (a) and (b) are values \( -\Gamma_k /2\pi \) [Hz]. Part (c)

value \( \Omega_k /2\pi \) [Hz]. Bold line \((k=1)\), thin line \((k=2)\), dotted
\( (k=3)\), dots \((k=4)\), circles \((k=5)\) and diamonds \((k=6)\).

Figures 1a and 1b show that the first relaxation mode \( \Gamma_1 \) of the PMs differ greatly in magnitude from the other five relaxation modes of the PMs. A sharp decrease in this mode occurs either when the \( H_0 \) decreases to less than 10 mOe (at \( T=473 \) K) or as the temperature \( T \) rises above 420 K (in the field \( H_0=1 \) mOe). In both cases, the inequality \( \chi = \gamma_{ex} (T)/\omega_0 \gg 1 \) was fulfilled.

Such a sharp suppression of relaxation at \( \chi \gg 1 \) is well known and observed [1-4] for orientation (it is called SERF). However, in our case, equation (6) takes into account the evolution not only the
orientation \((L=1)\), but also of all higher PMs with ranks \(L=2, 3\) and \(4\). And in accordance with formula (8), a sharp decrease of \(\Gamma_1\) should lead to suppression of relaxation as well higher PMs with a projection \(q=1\). In addition, the predominant contribution of the relaxation mode \(\Gamma_1\) to the amplitudes of all PMs will lead to their oscillations with one frequency \(\Omega_1\), shown in figure 1c by the bold line.

To reveal the mechanism of suppression of the relaxation of higher PMs, let us consider the dependence of the mode \(\Gamma_1\) on the field \(H_0\) in figure 1a. It follows from this figure that at \(H_0 \leq 10\) mOe the value of \(\Gamma_1\) quadratically depend on the field: \(\Gamma_1 \sim (H_0)^2\). So, at \(H_0=0\) \(\Gamma_1 = \Lambda_1 = 0\). Substituting this result into equation (7), we obtain the identity
\[
M (1, \beta, \gamma, 0) = -\gamma \epsilon [N(1, \beta)] = 0
\] (9)
The check has shown that such an identity is indeed satisfied. It turned out that it is the consequence of the conservation of the transverse angular momentum of atoms \(\hat{F} = S p \left( \hat{F}, \hat{J} \right)\) in the absence of a magnetic field. To prove this, consider the expression for derivative of this moment with respect to time
\[
\frac{d}{dt} \left( \hat{F}_1 \right) = - \left[ F(F+1) \right]^{0.5} \frac{d}{dt} f_1^1 (F) = -\gamma \epsilon \sum_{m=1}^{6} [6^{0.5} N(1, \beta)_{1,m} + 2^{0.5} N(1, \beta)_{2,m}] Y_m = 0
\] (10)
This expression is obtained using equation (6) for derivatives of two elements \(Y_1 = f_1^1 (F^+)\) and \(Y_2 = f_1^1 (F^-)\) of column \(Y\). Equality to zero is obtained due to the fact
\[
[6^{0.5} N(1, \beta)_{1,m} + 2^{0.5} N(1, \beta)_{2,m}] = 0
\] (11)
with \(m = 1 \div 6\). This was shown by the calculation. Identity (10) means the preservation in time of the moment \(\hat{F}_1\).

We now consider the determinant of matrix \(N\) using the well-known properties of the determinants and identity (11):
\[
\begin{vmatrix}
N_{1,1} & \ldots & N_{1,6} \\
\ldots & \ldots & \ldots \\
N_{6,1} & \ldots & N_{6,6}
\end{vmatrix} = \begin{vmatrix}
6^{0.5} N_{1,1} + 2^{0.5} N_{2,1} & \ldots & 6^{0.5} N_{1,6} + 2^{0.5} N_{2,6} \\
\ldots & \ldots & \ldots \\
N_{6,1} & \ldots & N_{6,6}
\end{vmatrix} = 0
\] (12)
Thus, identity (10) for the considered case \(q=1\) is proved.

In addition to suppressing the relaxation of higher PMs, it is of interest to elucidate how these PMs appear and change over time.

Figure 2 shows the time dependence of the PMs after excitation at \(t=0\) of orientation \(f_1^1 (2) = 0.3\) at \(T=473\) K and \(\beta=1\). The calculation was carried out according to the formula (8).

The figure 2a shows the evolution of the PMs at \(H_0=0\). In the course of this evolution, first, in a time \(\Delta t = 5 \cdot 10^{-5}\) sec, there is a rapid increase in the magnitude of all PMs, equal to 0 at \(t=0\), up to some stationary values. Then the growth of all PMs stops. In this case, there are no changes in time of the total moment \(\left( \hat{F}_1 \right) = 0.3 \cdot 6^{0.5}\). The increase in PMs over time \(\Delta t\) is caused by the contribution of relaxation modes \(\left( -\Gamma_{k,1} \right) \approx \gamma \epsilon = 2 \cdot 10^5\) sec\(^{-1}\).

Figure 2b shows the evolution of all PMs at \(H_0=1\) mOe. This evolution is slow, as it is due to the main suppressed mode \(\Gamma_1 = -70\) sec\(^{-1}\). It is the same for all PMs.

Let us now consider the case of evolution of PMs with a projection \(q>1\). All determinants \(N(q, \beta)\)
Figure 2. Bold line - $|f_{i1}^1(2)|$, dots - $|f_{i1}^3(1)|$, dashed line - $|f_{i1}^2(2)|$, line with rhombuses – $|f_{i1}^3(1)|$, squares - $|f_{i1}^2(2)|$, line with circles - $|f_{i1}^1(2)|$ and triangles – $\left|\langle F_{i1} \rangle \right|=6^{0.5}f_{i1}^1(2)+2^{0.5}f_{i1}^1(1)$.

with such values of $q$ are not equal to zero. So, none of the modes $\Gamma(q)_\chi$ goes to 0 at $H_0 \to 0$. Consequently, there is no suppression of the relaxation of transverse PMs $f_{i1}^1(F)|q|=1$. In fact, the relaxation rates of all these PMs approximately are equal to $\gamma_{e\chi}$ and in SERF regime when $\chi \gg 1$ they are several orders of magnitude larger than PMs with $q=1$ (at $\beta > 1$).

It is interesting to consider the dependence of the relaxation of transverse PMs on the value of parameter $\beta$. Figure 3 shows the temperature dependence of the suppressed mode $\Gamma_1$ of relaxation and oscillation $\Omega_1$ for several values of $\beta$ at $H_0=1$ mOe.

Figure 3a shows that the value of $\beta$ has a significant effect on the value of $\Gamma_1$. It decreases by two orders of magnitude as $\beta$ changes from 1 to 5, and this decrease in relaxation occurs over a wide temperature range. The reason for this is the predominant population of the “stretched” state sublevel $M = F^+$ [5]. The transition from it to other sublevels is prohibited by the law of conservation of angular momentum.

The temperature dependence of the frequency $\Omega_1$ shown in figure 3b is well known [2]. The dependence on $\beta$ at $\chi \gg 1$ for atoms with a nuclear spin $I=3/2$ is given by the formula

$$\Omega_1(\beta) = \omega_0 2 \cosh(\beta) \sqrt{1 + 2 \cosh(\beta)}$$

According to this formula at $T=500$ K, it turns out that $\Omega_1/2\pi = 467, 520, 618, 667$ and 695 Hz.
at $\beta=0$, 1, 2, 3 and 5, respectively. This result exactly matches the data in figure 3b.

![Figure 3](image)

**Figure 3.** Temperature dependence of the values $(-\Gamma_i/2\pi)$ (a) and $\Omega_i/2\pi$ (b) [Hz] at the values of the parameter $\beta$: 0 (bold line), 1 (dashed line), 2 (dots), 3 (rhombuses) and 5 (circles). A feature of figure 3 is that the relaxation and oscillation modes for each nonzero value of $\beta$ refer to all 6 types of PMs with projection $q=1$. But the relaxation and oscillation mode calculated at $\beta=0$ refers only to two PMs with $L=1$: $f_{\pm 1}^0 (F^\pm)$. This is due to the fact that the structure of the spin exchange equations for $\beta=0$ radically differs from that for $\beta>0$.

In the case "$\beta=0$" the second term in equation (4) is absent and resulting equations does not connect PMs with different ranks. So, in this case we obtain two matrix equations for the PMs with ranks $L=1$ and 2

$$\frac{d}{dt} Y(L,q)\bigg|_{L=1,2} = \left[ \begin{array}{c} -\gamma_\omega W(L) - i\omega, q [d] \end{array} \right] Y(L,q)$$

$$Y(L,q)\bigg|_{L=1,2} = \left[ \begin{array}{c} f_{\pm}^L (F^+) \ f_{\pm}^L (F^-) \end{array} \right]$$

and two simple equations:

$$\frac{d}{dt} f_{\pm}^L (F^+)\bigg|_{L=3,4} = \left[ -\gamma_\omega W^F (L) - i\omega, q \right] f_{\pm}^L (F^+)$$

The calculation showed that $|W(1)|=0$, $|W(2)|=0.094$, $W^2(3)=0.375$ and $W^2(4)=0.5$. Consequently, of all PMs, relaxation will be suppressed at $\beta=0$ only for two orientation components $f_{\pm 1}^0 (F^\pm)$. As for the higher PMs $f_{\pm}^{L+1} (F^\pm)$, their relaxation will not be suppressed.

Thus, in the present work, the effect of suppressing the relaxation of the transverse components $f_{\pm}^{L+1} (F^\pm)$ of higher PMs was revealed at $\beta>0$ and the absence of such an effect at $\beta=0$. This conclusion can be verified in an experiment by observing the relaxation of the transverse alignment $f_{\pm}^{L+1} (F^\pm)$ in the absorption of linearly polarized light.

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