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Spin relaxation and linear-in-electric-field frequency shift in an arbitrary, time-independent magnetic field

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Abstract

A method is presented to calculate the spin relaxation times T_1, T_2 due to a non-uniform magnetic field, and the linear-in-electric-field precession frequency shift $\delta\omega_E$ when an electric field is present, in the diffusion approximation for spins confined to a rectangular cell. It is found that the rectangular cell geometry admits of a general result for T_1, T_2, and $\delta\omega_E$ in terms of the spatial cosine-transform components of the magnetic field.

Keywords:

1. Introduction

An experiment to measure the neutron electric dipole moment (nEDM), to be installed at the FnPB beamline at Oak Ridge National Laboratory, will utilize a helium-3 comagnetometer in the central, superfluid-helium-filled measurement cell [1][2]. The helium-3 polarization must remain high over the entire measurement period, ~1000 seconds, as the helium atoms precess in the holding field and diffuse within a rectangular cell. Also, as there is a strong electric field $\vec{E}$ applied across the cell, a subtle effect, in which the interplay of the motional $\vec{v} \times \vec{E}$ field with gradients in the static magnetic field cause the precession frequency to shift linearly with $\vec{E}$ [3], must be well-understood or shown to be negligible. Design optimization of the experimental apparatus includes calculating the helium-3 spin relaxation times T_1, T_2, and linear-in-electric-field frequency shift $\delta\omega_E$ due to given magnetic field non-uniformities.

In this article, a method is shown to calculate these quantities in the diffusion approximation in a rectangular cell and for an arbitrary magnetic field. The starting point for the relaxation times is the Redfield theory of spin relaxation [4]. In second order perturbation theory these can be written, for a

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holding field in the z direction, as [5]

$$\frac{1}{T_1} = \gamma^2 \left(k_{xx}(\omega_0) + k_{yy}(\omega_0)\right), \tag{1}$$

$$\frac{1}{T_2} = \frac{1}{2T_1} + \gamma^2 k_{zz}(0), \tag{2}$$

where the spectral density is given in terms of magnetic field perturbations $h_i(t)$,

$$k_{ij}(\omega) = \frac{1}{2}\int_{-\infty}^{\infty} \langle h_i(t) h_j(t+\tau)\rangle \cos\omega\tau d\tau \tag{3}$$

Here, the total field in each direction i is $H_i(t) = \langle H_i(t)\rangle + h_i(t)$, such that average perturbation $\langle h_i(t)\rangle = 0$, and $\omega_0 = \gamma\langle H_z(t)\rangle$ is the average spin precession frequency.

McGregor [6] calculated the ensemble average correlation of the field perturbations seen by a diffusing particle in the case of a time-independent, uniform gradient of H_z in the x-direction,

$$\langle h_z(t) h_z(t+\tau)\rangle = \left(\frac{\partial H_z}{\partial x}\right)^2 \langle x(t) x(t+\tau)\rangle, \tag{4}$$

resulting in an analytic expression for T_2 in a rectangular cell,

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{\gamma^2 L^4}{120D}\left[\frac{\partial H_z}{\partial x}\right]^2. \tag{5}$$

We relax the requirement of uniform gradient and find, in the case of a rectangular prism cell, that T_1 and T_2 can be written in terms of the components of the 3D cosine transform of $h_q(\vec{r})$ over the cell volume. The same technique is applied to dressed spins [7], with uniform holding field and non-uniform dressing field, by mapping non-uniformities in the dressing field to equivalent non-uniformities in the holding field. In Section 3, a variation of the technique is used for the linear-in-electric-field frequency shift. Finally, as an example, in Section 4 the relaxation times and linear-in-electric-field frequency shift due to a superconducting rod near the cell are computed.

2. Correlation functions in the diffusion limit

The correlation function of h_i can be expressed as integrals over the cell volume weighted by the probability density $p(\vec{r}_0, t)$ that the particle is at $\vec{r}_0$ at the initial time t_0, and the joint probability density $p(\vec{r}, t|\vec{r}_0, t_0)$ that a particle at $\vec{r}_0$ at time t_0 will be at $\vec{r}$ at time t. Thus, [6]

$$\langle h_i(t_0) h_i(t_0+\tau)\rangle = \int_V d\vec{r}_0 h_i(\vec{r}_0) p(\vec{r}_0, t_0) \int_V d\vec{r} h_i(\vec{r}) p(\vec{r}, t_0+\tau|\vec{r}_0, t_0). \tag{6}$$

The particle density will be taken as uniform in the cell, $p(\vec{r},t) = 1/V$. The joint probability is the solution to the diffusion equation,

$$\frac{\partial}{\partial t} p(\vec{r},t|\vec{r}_0,t_0) = D\nabla^2 p(\vec{r},t|\vec{r}_0,t_0), \tag{7}$$

subject to reflecting boundary conditions at the walls,

$$\nabla p(\vec{r}_S,t|\vec{r}_0,t_0)\cdot\hat{n} = 0, \tag{8}$$

where $\vec{r}_S \in S$ and $\hat{n}$ is normal to the wall. In a rotated coordinate system (x',y',z') aligned with the cell walls, the solution for a $L_x \times L_y \times L_z$ box with walls at $x' = \pm L_x/2$, $y' = \pm L_y/2$ and $z' = \pm L_z/2$ is

$$p(\vec{r}',t|\vec{r}_0',t_0) = p(x',t|x_0',t_0;L_x)p(y',t|y_0',t_0;L_y)p(z',t|z_0',t_0;L_z), \tag{9}$$

with the 1D solution dependent on the time difference $\tau = t - t_0$, [6]

$$\begin{aligned} p(x',t|x_0',t_0;L_x) &= \frac{1}{L_x} + \frac{2}{L_x}\sum_{n=1,3,\ldots}^{\infty} e^{-n^2\pi^2 D\tau/L_x^2}\sin\frac{n\pi x'}{L_x}\sin\frac{n\pi x_0'}{L_x} \\ &\quad + \frac{2}{L_x}\sum_{n=2,4,\ldots}^{\infty} e^{-n^2\pi^2 D\tau/L_x^2}\cos\frac{n\pi x'}{L_x}\cos\frac{n\pi x_0'}{L_x}. \end{aligned} \tag{10}$$

It will be convenient to recognize the following:

$$p(x'-L_x/2,t|x_0'-L_x/2,t_0;L_x) = \frac{1}{L_x} + \frac{2}{L_x}\sum_{n=1,2,3,\ldots}^{\infty} e^{-n^2\pi^2 D\tau/L_x^2}\cos\frac{n\pi x'}{L_x}\cos\frac{n\pi x_0'}{L_x}. \tag{11}$$

Putting Eq. 9 and $p(\vec{r}_0',t_0) = 1/V$ into Eq. 6, changing the limits of integration to $0 \le q_i' \le L_i$ for each dimension q_i' and using Eq. 11, we have

$$\begin{aligned} g_{ii}(\tau) &= \langle h_i(t_0)h_i(t_0+\tau)\rangle \\ &= \sum_{n_x,n_y,n_z=0}^{\infty} e^{-\pi^2 D(n_x^2/L_x^2+n_y^2/L_y^2+n_z^2/L_z^2)\tau} \\ &\quad \times\frac{1}{V}\int_{V'} dx_0'\,dy_0'\,dz_0'\,h_i(x_0'-L_x/2,y_0'-L_y/2,z_0'-L_z/2) \\ &\quad C_{n_x}C_{n_y}C_{n_z}\cos\frac{n_x\pi x_0'}{L_x}\cos\frac{n_y\pi y_0'}{L_y}\cos\frac{n_z\pi z_0'}{L_z} \\ &\quad \times\frac{1}{V}\int_{V'} dx'\,dy'\,dz'\,h_i(x'-L_x/2,y'-L_y/2,z'-L_z/2) \\ &\quad C_{n_x}C_{n_y}C_{n_z}\cos\frac{n_x\pi x'}{L_x}\cos\frac{n_y\pi y'}{L_y}\cos\frac{n_z\pi z'}{L_z} \end{aligned} \tag{12}$$

in which the factor C_n has been introduced,

$$C_n = \begin{cases} 1 & \text{if } n = 0 \\ \sqrt{2} & \text{otherwise.} \end{cases} \tag{13}$$

We identify the 3D cosine transform of $h_i(\vec{r}')$ within Eq. 12,

$$\begin{aligned}\mathcal{A}^{\vec{n}}\{h_i\} &\equiv \frac{1}{V}\int_{V'} dx'\,dy'\,dz'\,h_i(x'-L_x/2, y'-L_y/2, z'-L_z/2) \\ &\quad \times \cos\frac{n_x\pi x'}{L_x}\cos\frac{n_y\pi y'}{L_y}\cos\frac{n_z\pi z'}{L_z},\end{aligned} \tag{14}$$

(where the integral is over the range $x' \in [0, L_x]$, $y' \in [0, L_y]$, $z' \in [0, L_z]$), giving finally

$$g_{ii}(\tau) = \sum_{n_x,n_y,n_z=0}^{\infty} e^{-\tau/\tau_c^{\vec{n}}} C_{n_x}^2 C_{n_y}^2 C_{n_z}^2 \left(\mathcal{A}^{\vec{n}}\{h_i\}\right)^2, \tag{15}$$

with the characteristic time τ_c for a given spatial mode defined by

$$\frac{1}{\tau_c^{\vec{n}}} = \pi^2 D\left(\frac{n_x^2}{L_x^2} + \frac{n_y^2}{L_y^2} + \frac{n_z^2}{L_z^2}\right) \tag{16}$$

Putting this expression into Eq. 3 and performing the integral over τ gives

$$k_{ii}(\omega) = \sum_{n_x,n_y,n_z=0}^{\infty} C_{n_x}^2 C_{n_y}^2 C_{n_z}^2 \frac{\tau_c^{\vec{n}}}{1+\omega^2(\tau_c^{\vec{n}})^2}\left(\mathcal{A}^{\vec{n}}\{h_i\}\right)^2, \tag{17}$$

Substitution into Eqs. 1 and 2 results in complete expressions for the longitudinal and transverse relaxation times.

For arbitrary fields including field maps, the cosine-transform amplitudes $\mathcal{A}$ can be numerically calculated by fast discrete cosine transform (DCT) over $h_q(\vec{r})$ sampled at a sufficient number of points throughout the cell volume. In this case the summation over n_q is truncated accordingly, and accuracy is checked by increasing the number of sample points and comparing results.

2.1. Extension to dressed spins with non-uniform dressing field

An RF magnetic field with amplitude B_1 applied transverse to the holding field B_0 modifies the effective precession frequency of a particle. In terms of dimensionless "dressing parameters" [8]

$$X = \frac{\gamma B_1}{\omega_{\mathrm{RF}}},\ Y = \frac{\gamma B_0}{\omega_{\mathrm{RF}}}, \tag{18}$$

in the limit $Y \ll 1$ the effective gyromagnetic ratio becomes [7]

$$\gamma_{\mathrm{eff}} = \gamma J_0(X). \tag{19}$$

Thus for a dressing field with spatially varying amplitude $B_1(\vec{r}) = \langle B_1\rangle + \delta B_1(\vec{r})$, the equivalent variation $\delta\tilde{B}_0$ in the holding field B_0 is given by [1]

$$\gamma_{\mathrm{eff}}\delta\tilde{B}_0 \leftrightarrow \delta\gamma_{\mathrm{eff}}B_0 = B_0\frac{\partial\gamma_{\mathrm{eff}}}{\partial X}\delta X = B_0\gamma J_1(X)\delta X, \tag{20}$$

The expressions from the previous sections can be used to calculate T_2 for the dressed spin with a non-uniform dressing field by setting

$$\gamma \rightarrow \gamma_{\rm eff}, \tag{21}$$

$$\omega_0 \rightarrow \gamma_{\rm eff} B_0, \tag{22}$$

$$h_z(\vec{r}) \rightarrow Y \frac{J_1(X)}{J_0(X)} \delta B_1(\vec{r}) + \delta B_0(\vec{r}), \tag{23}$$

where the variation δB_0 in the holding field itself has been included in h_z.

3. Linear electric field frequency shift

A spin moving though an electric field experiences a motional magnetic field that may, in conjunction with gradients of the magnetic field, produce a shift in the precession frequency dependent on the electric field direction and magnitude. [3] Of particular concern in searches for electric dipole moments are effects that are linearly proportional to the electric field E. These may mimic effects expected for an electric dipole moment, thereby creating a "false EDM."

As shown by Lamoreaux and Golub [9], the linear-in-electric-field frequency shift for spins in a confined volume is given by the expression

$$\delta\omega_E = -\frac{1}{2}\int_0^t d\tau \, \cos\omega_0\tau \{\langle \omega_x(t)\omega_y(t-\tau)\rangle - \langle \omega_x(t-\tau)\omega_y(t)\rangle\}, \tag{24}$$

where the perturbations can be written more generally as

$$\omega_x(t) = \gamma h_x(t) + \gamma \frac{E}{c} v_y(t), \quad \omega_y(t) = \gamma h_y(t) - \gamma \frac{E}{c} v_x(t). \tag{25}$$

Expanding the expression for $\delta\omega_E$ and keeping only terms linear in E results in

$$\begin{aligned} \delta\omega_E &= \frac{\gamma^2 E}{2c}\int_0^t d\tau \, \cos\omega_0\tau \{\langle h_y(t)v_y(t-\tau)\rangle + \langle h_x(t)v_x(t-\tau)\rangle \\ &\quad -\langle h_y(t-\tau)v_y(t)\rangle - \langle h_x(t-\tau)v_x(t)\rangle\}. \end{aligned} \tag{26}$$

The cosine-transform method developed in the present work can be used to compute Eq. 26 in the diffusion limit for the nEDM cell geometry. While the expressions,

$$y(t) = y_0 + \int_0^t v_y(t')dt', \quad y(t-\tau) = y_0 + \int_0^{t-\tau} v_y(t')dt', \tag{27}$$

were used in Ref. [9] to eliminate y in favor of an expression with v_y, here we remove the velocity components from the correlation functions and use instead the Fundmental Theorem of Calculus and the above expression for $y(t-\tau)$. The correlation functions can then be written

$$\langle h_y(t)v_y(t-\tau)\rangle = -\frac{\partial}{\partial\tau}\langle h_y(t)y(t-\tau)\rangle, \tag{28}$$

$$\langle h_y(t-\tau)v_y(t)\rangle = \langle h_y(t)v_y(t+\tau)\rangle = \frac{\partial}{\partial\tau}\langle h_y(t)y(t+\tau)\rangle. \tag{29}$$

In the latter expression, averages are assumed to be independent of the overall time offset t, as appropriate for a stationary problem.

The expressions in Section 2 are modified to give the correlation function in the diffusion limit in terms of the cosine-transform components of $h_y(\vec{r})$ and $y(\vec{r})$,

$$\langle h_y(t)y(t-\tau)\rangle = \sum_{n_x,n_y,n_z} e^{-\tau/\tau_c^{\vec{n}}} C_{n_x}^2 C_{n_y}^2 C_{n_z}^2 \mathcal{A}^{\vec{n}}\{h_y\}\,\mathcal{A}^{\vec{n}}\{y\}. \tag{30}$$

$$\langle h_y(t)v_y(t-\tau)\rangle = \sum_{n_x,n_y,n_z} \frac{1}{\tau_c^{\vec{n}}} e^{-\tau/\tau_c^{\vec{n}}} C_{n_x}^2 C_{n_y}^2 C_{n_z}^2 \mathcal{A}^{\vec{n}}\{h_y\}\,\mathcal{A}^{\vec{n}}\{y\} \tag{31}$$

Performing the integral in Eq. 26, we have

$$\int_0^\infty d\tau\, \cos\omega_0\tau\langle h_y(t)v_y(t-\tau)\rangle = \sum_{n_x,n_y,n_z} \frac{1}{1+(\omega_0\tau_c^{\vec{n}})^2} C_{n_x}^2 C_{n_y}^2 C_{n_z}^2 \mathcal{A}^{\vec{n}}\{h_y\}\,\mathcal{A}^{\vec{n}}\{y\}, \tag{32}$$

leading to an expression for the frequency shift,

$$\delta\omega_E = \frac{\gamma^2 E}{c} \sum_{n_x,n_y,n_z} \frac{1}{1+(\omega_0\tau_c^{\vec{n}})^2} C_{n_x}^2 C_{n_y}^2 C_{n_z}^2 \left[\mathcal{A}^{\vec{n}}\{h_y\}\,\mathcal{A}^{\vec{n}}\{y\} + \mathcal{A}^{\vec{n}}\{h_x\}\,\mathcal{A}^{\vec{n}}\{x\}\right]. \tag{33}$$

The summation can be reduced by computing the cosine transform components of $x(\vec{r})$ and $y(\vec{r})$ analytically,

$$\mathcal{A}^{\vec{n}}\{x\} = \begin{cases} -\frac{2L_x}{n_x^2\pi^2} & \text{if } n_x = 1,3,\ldots; n_y = n_z = 0 \\ 0 & \text{otherwise.} \end{cases} \tag{34}$$

The result for the frequency shift in the diffusion approximation is

$$\delta\omega_E = \frac{4\gamma^2 E}{c}\left[\sum_{n_y=1,3,\ldots} \frac{L_y}{n_y^2\pi^2}\frac{\mathcal{A}^{(0,n_y,0)}\{h_y\}}{1+(\omega_0\tau_c^{(0,n_y,0)})^2} + \sum_{n_x=1,3,\ldots} \frac{L_x}{n_x^2\pi^2}\frac{\mathcal{A}^{(n_x,0,0)}\{h_x\}}{1+(\omega_0\tau_c^{(n_x,0,0)})^2}\right]. \tag{35}$$

4. Example application employing the discrete cosine transform

The bulk of the computational effort required for practical application of the present technique is in finding the cosine transform amplitudes $\mathcal{A}^{\vec{n}}$ of the field non-uniformities. The form of Eq. 14 is amenable to numerical computation with Fast Fourier Transform software libraries. The example below uses the multidimensional discrete cosine transform (DCT) feature of the freely-available software library FFTW3 [10]. Input to the DCT for each field component is an array of field perturbations $h_q(\vec{r})$ sampled over the cell volume at $N_xN_yN_z$ grid points, and the output is an array of DCT amplitudes which, after scaling

by $1/(8N_xN_yN_z)$, correspond to the desired amplitudes $\mathcal{A}^{\vec{n}}$. The summations in Eqs. 17 and 35 are truncated according to the amplitudes available from the DCT. Accuracy of the result can be checked by increasing the number of sample points and repeating the computation.

We calculate the effect of a superconducting rod placed near the cell in an otherwise uniform holding field. For an infinite-length rod along the z axis and through the origin in a magnetic field B_0 applied along the x axis, the net field around the rod is [11]

$$B_\rho = (1 - \frac{a^2}{\rho^2})B_0 \cos\phi, \tag{36}$$

$$B_\phi = (1 + \frac{a^2}{\rho^2})B_0 \sin\phi, \tag{37}$$

where a is the rod radius and (ρ, ϕ) are polar coordinates in the xy plane. While the cosine transforms of B_x, B_y derived from these equations (appropriately translated to the desired location of the superconducting rod) could perhaps be calculated analytically, here the equations are used to generate a field map that is subsequently run through the machinery to produce values for T_1, T_2, and $\delta\omega_E$. Results are shown in Figure 1, with physical parameters given in the caption.

5. Conclusion

A method to calculate spin relaxation times and the linear-in-E frequency shift in the diffusion approximation was presented. The technique is based on the observation that, for a particle diffusing in a rectangular cell, the correlation function of a position-dependent field is the weighted sum over the squared cosine-transform components of the field (see Eq. 15).[3]

As the formulation is intended for practical computation in a rectagular cell, the result is for the complete 3-dimensional geometry. It could also be applied to a cylindrical cell if the field variation in coordinates transverse to the cylinder axis can be neglected. Actual computation is done using fast discrete cosine transforms of the field components. This method could be used for example in magnet coil design optimization: given a field map, T_1, T_2, and $\delta\omega_E$ can be quickly evalulated and combined into a figure of merit.

6. Acknowledgements

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[3]During preparation of this manuscript, independent work was published based on essentially the same observation. These authors point out that their result may be used as a probe for possible unknown spin interactions and applied it to improve the limits on axion-like interactions with the cell walls [13].

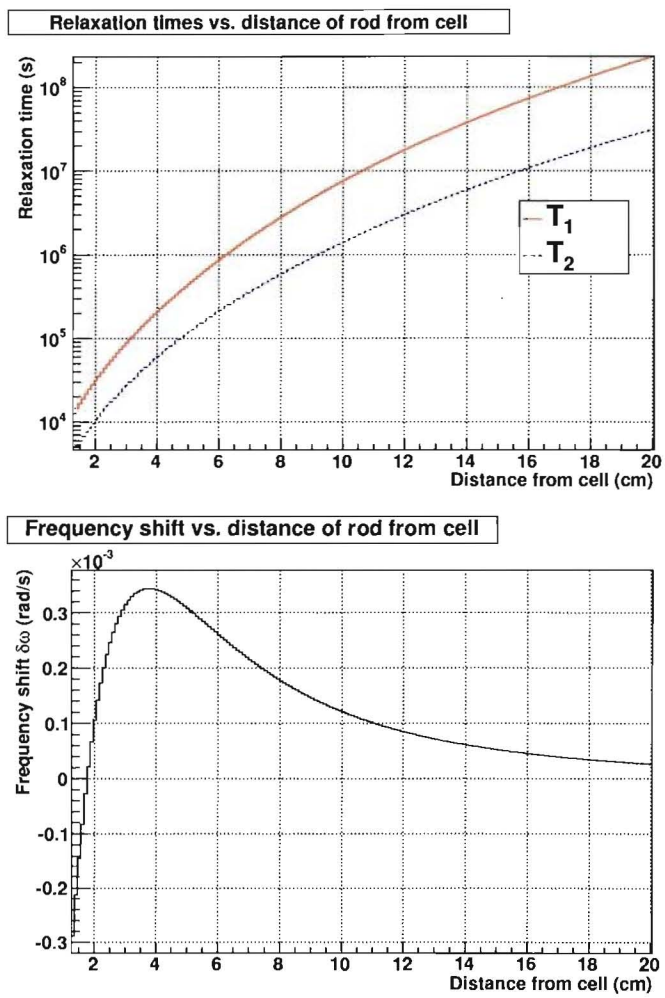

Figure 1: Calculated (a) relaxation times $T1$, $T2$ and (b) frequency shift $\delta\omega_E$ due to the magnetic field distortion from a 3-mm-diameter, infinitely-long superconducting rod parallel to the z-axis and placed near the cell. The parameters are $\gamma = -20393.963$ rad $\mathrm{G}^{-1}\mathrm{s}^{-1}$ (gyromagnetic ratio for ^{3}He), $D = 428$ cm^2/s (corresponding to a temperature of 450 mK [12]), $B_0 = 10$ mG (applied parallel to the x-axis), $E = 100$ kV/cm (parallel to B_0), cell dimensions $L_x = 7.8$ cm, $L_y = 10$ cm, $L_z = 40$ cm.

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