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Detection of ^{3}He spins with ultra-low field nuclear magnetic resonance employing SQUIDs for application to a neutron electric dipole moment experiment

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ABSTRACT

The precession of ^{3}He spins is detected with ultra-low field NMR. The absolute strength of the NMR signal is accurately measured and agrees closely with theoretical calculations. The sensitivity is analyzed for applications to a neutron electric dipole moment (nEDM) fundamental symmetry experiment under development.

1. Introduction

A novel experiment has been proposed to measure the neutron electric dipole moment (EDM). By making a 50–100 times improvement over the current experimental limit, $d_n < 2.9 \times 10^{-26}$ e cm [1], the experiment will search for physics beyond the Standard Model. If the neutron has an EDM, it violates the parity and time-reversal symmetries. The proposed experiment takes place in a superfluid ^{4}He bath and will expose spin-polarized ultra-cold neutrons (UCNs) to parallel and anti-parallel weak magnetic and strong electric fields. The signal for a non-zero EDM would be a slight shift in the Larmor precession frequency of the UCN between the parallel and anti-parallel field orientations. The precession signal from the UCN is not directly measurable ($\sim 10^{3}$ neutrons/cm^{3}), but by doping the ^{4}He with a small amount ($\sim 10^{12}$ atoms/cm^{3}) of ^{3}He, the precession-dependent scintillation light from the ^{3}He(n, p)t reaction makes the beat frequency between the UCN and ^{3}He observable. SQUID sensors will be used to directly measure the nuclear magnetic resonance (NMR) signal from ^{3}He. Their precession frequency will provide a direct measure of magnetic field as well as the polarization of the ^{3}He nuclei. These measurements are needed to reduce systematic errors caused by spatial and temporal magnetic field fluctuations. A complete description of the physics motivation and experimental concept are provided elsewhere [2].

In the neutron EDM (nEDM) experiment described above, SQUID sensors are proposed as the detectors to measure the precession signal from highly spin-polarized ^{3}He dissolved in superfluid ^{4}He. From a practical standpoint, the SQUIDs are simply measuring NMR free induction decay (FID) from the spin-polarized ^{3}He with very long T_1 and T_2 NMR relaxation times [2]. As the measurement magnetic fields are very low (μT), the technique is applied in the ultra-low field (ULF) regime. Previous work indicates that SQUID-based ULF NMR is a very rich technique with applications to many problems [3–7] from medical imaging to fundamental physics. However, in the nEDM experiment, the signal is very small because the fractional density of the nearly 100%-polarized ^{3}He atoms in the superfluid ^{4}He will be only 10^{-10} (2×10^{12} atoms/cm^{3}) [2].

This paper describes an experiment in which the precession of the spins in a gas of ^{3}He atoms at 4 K was measured with a SQUID gradiometer via ULF NMR methods. The data and the extrapolation of the measurements of sensitivity to the nEDM experiment are discussed. A model of the ^{3}He NMR signal in a proposed gradiometer design is presented. The sensitivity data are applied to a very large gradiometer, such as the one proposed for use in the nEDM experiment, to evaluate whether the required sensitivity and signal-to-noise will be achieved.

2. Theoretical calculations of the strength of NMR signal

For the nEDM experiment, theoretical modeling of the detection sensitivity is essential. One purpose of the modeling is the optimization of the expected ^{3}He NMR signal for a given geometry of the helium cell and gradiometer-coil configuration.

The NMR signal to be detected is directly proportional to the magnetic flux through the pick-up coils of the SQUID sensor:

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$$\Phi = \int_P \mathbf{B_M} \cdot \mathrm{d}\mathbf{S}, \tag{1}$$

where Φ is the flux through the pick-up coils of the sensor due to the magnetic field $\mathbf{B_M}$ generated by the ^{3}He magnetization $\mathbf{M}$ and P is the area of the pick-up coils. Since the field is created by a volume sample, integration over the sample volume is necessary to obtain the field values. It is often convenient and numerically efficient to use an alternative equation based on the principle of reciprocity (see for example [8])

$$\Phi = \int_{\text{sample}} \mathrm{d}^3\mathbf{r} \left(\frac{\mathbf{B}_{\text{coil}}}{\mathbf{I}} \right) \cdot \mathbf{M}. \tag{2}$$

Here $\mathbf{B}_{\text{coil}}$ is the field created by the pickup coils due to a current $\mathbf{I}$, and the integration is over the sample volume.

In the nEDM experiment, the ^{3}He polarization will be close to 100%. Thus the number density of polarized nuclei N_{p} will be approximately equal to that of ^{3}He in the cell, which is assumed to be $\sim 2 \times 10^{12}$/cc. The magnetization density is

$$M = \mu_{\text{I}} N_{\text{p}}, \tag{3}$$

where μ_{I} is the magnetic moment of the nucleus; $\mu_{\text{I}} = -1.07 \times 10^{-26}$ J/T in the case of ^{3}He. For comparison, the proton's magnetic moment $\mu_{\text{I}} = 1.41 \times 10^{-26}$ J/T.

For the tests described here, a method of thermal pre-polarization was used, and the value of equilibrium magnetization density is given by

$$M = \frac{\mu_{\text{I}}^2 B}{k_{\text{B}} T} n, \tag{4}$$

where $k_{\text{B}} = 1.38 \times 10^{-23}$ J/K, n is the number density of spins, T is the temperature and B is the strength of the magnetic field applied to polarize the sample. In the case of a sphere of a radius a, the magnetic field B_{sph} created by the sample along the direction of the magnetization at some distance R is

$$B_{\text{sph}} = \frac{2\mu_0}{3} \frac{a^3}{R^3} M, \tag{5}$$

where μ_0 is vacuum permeability. From Eq. (1) for a coil of area A far away from the cell

$$\Phi = A \frac{2\mu_0}{3} \frac{a^3}{R^3} M. \tag{6}$$

When the radius of the pick-up coil r_{c} is comparable to R, a more accurate equation is

$$\Phi = A \frac{2\mu_0}{3} \frac{a^3}{(R^2 + r_{\text{c}}^2)^{3/2}} M. \tag{7}$$

For the case of a spherical sample, a numerical code based on Eq. (2) was compared to the simple analytical result from Eq. (7). Then the numerical simulation was compared with experiment in the well-controlled geometrical arrangement described in the following section.

3. Experimental setup and calibration

The experiments were designed to provide a means to validate the calculation of the NMR signals, test the sensitivity of the currently available SQUID gradiometer, and project this sensitivity to the nEDM experiment. The ULF NMR system [3] (Fig. 1) was used inside a magnetically shielded room to measure the signal from a thermally polarized ^{3}He sample. A review of ULF NMR and MRI methods is given in Ref. [9]. A standard protocol [3] was employed: first, a strong field B_{p} is applied for a sufficiently long time to polarize the nuclei; then the field is switched off and a measurement field B_{m} is applied that causes the nuclear spins to precess at a well-defined frequency; finally, after a short delay during which transients in the SQUID detector decrease to an acceptable level ($\sim$ms time), the signal from SQUID gradiometer is recorded. This sequence is repeated periodically; the data are averaged in the time domain to improve the signal-to-noise ratio.

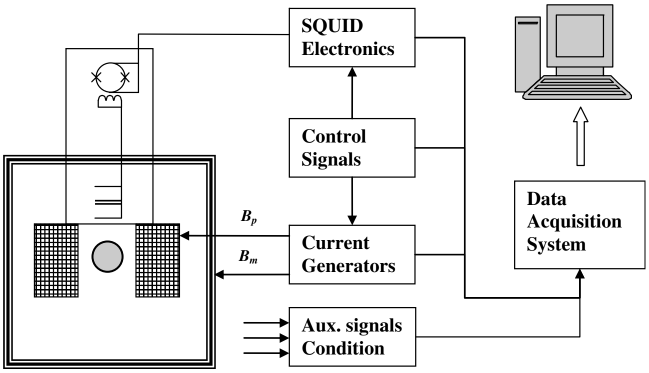

Fig. 1. Schematic diagram of the ULF NMR system. Current generators provide current to the magnetic field coils producing B_{p} and B_{m}. Control signals synchronize fields, the SQUID, and data acquisition. Auxiliary signals are also acquired during the experiment (for example sample temperature, etc.).

The system was calibrated initially via measurements with a spherical water phantom (a ping-pong ball). The water was at room temperature. While the expected proton NMR signal can be predicted theoretically with high accuracy, there are some effects that can reduce the NMR signal in experiments. For example, the excitation of NMR precession in the ULF setup is based on rapidly switching off of the pre-polarization field and turning on the measurement field within 1 ms. Non-zero residual fields in the system will result in non-collinear B_{p} field removal and hence in the excitation of transverse polarization with reduction of longitudinal polarization. In addition, if the ambient and applied measurement fields fluctuate, the time averaging of several scans used to improve the signal-to-noise ratio (SNR) can lead to a decrease in the signal due to de-phasing amongst different scans.

The specific geometry of the experiment is shown in Fig. 2. The water sample is a sphere of 38.9-mm diameter at room temperature. Measurements were made with a second-order axial gradiometer

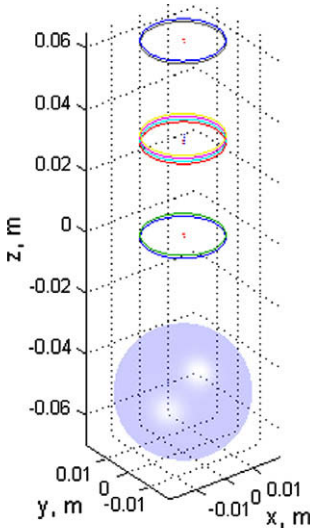

Fig. 2. Geometrical arrangement of the water phantom: the phantom is a sphere of diameter 38.9 mm with the center positioned at (0, 0, −50 mm); the second-order gradiometer has pick-up loops of 24-mm diameter and a baseline of 32 mm.

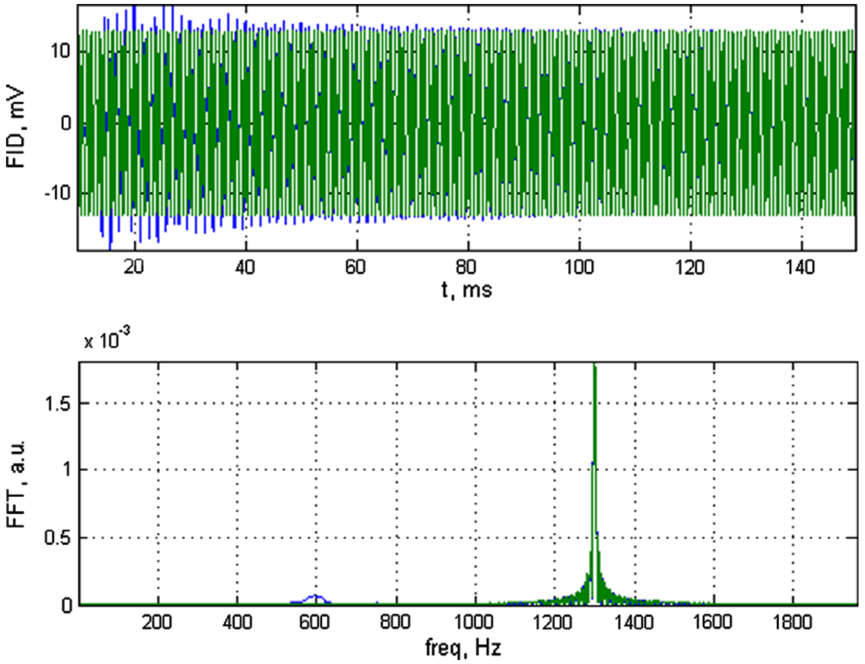

Fig. 3. The NMR signal from the water phantom.

with pick-up loops of 24-mm diameter and a baseline of 32 mm. A gradiometer was chosen for two reasons. The first is that it reduced the level of background noise by at least a factor of 1000. The second, and more important reason was that the gradiometer reduces the NMR fields in the SQUID and prevents flux-trapping in the SQUID. The relationship between the flux acquired by the gradiometer, Φ_{grad}, and the flux entering the SQUID, Φ_{SQUID}, is given by

$$\Phi_{\mathrm{SQUID}} = \Phi_{\mathrm{grad}}\left(\frac{M}{L_{\mathrm{grad}} + L_{\mathrm{input}}}\right), \tag{8}$$

where M is the mutual inductance between the input coil and the SQUID loop, L_{grad} is the inductance of the gradiometer pickup coils, and L_{input} is the inductance of the input coil. For this SQUID and coil configuration, $M = 7.4$ nH, $L_{\mathrm{grad}} = 1.52$ μH, and $L_{\mathrm{input}} = 0.42$ μH.

The resulting free induction decay (FID) and fast Fourier transform (FFT) are shown in Fig. 3 (blue[1]). An experimental fit (green[1]) is also superimposed to extract the signal level with better precision. From the FID fit, the signal strength of 12.9 mV is obtained. The SQUID detector sensitivity was determined to be 4.84 ± 0.18 V/nT (2.96 V/Φ_0 with noise of ~3.8 fT/Hz$^{1/2}$), by measuring the signal from a small movable coil of known geometry at various distances from the detector. The value of the pre-polarization field was determined to be 0.039 T by scaling measurements taken at a lower field with a fluxgate magnetometer. The pre-polarization time was 6 s, which is approximately twice the T_1 value for water. From Eq. (7) one would expect a total input flux at the SQUID of 1.17×10^{-17} Wb, which corresponds to the signal of 16.8 mV. The numerical model based on Eq. (2), which also accounts for the slight signal reduction due to a finite pre-polarization time, gives a signal in the range of 0.9×10^{-17} Wb, for an expected signal of 13.0 ± 0.5 mV. This result agrees with the experiment within error bar. Such close agreement provides confidence in the accuracy of both the measurements of absolute strength of NMR signal and the theoretical calculations. For the nEDM sensitivity tests, the absolute values of NMR signal are essential to determine the minimal number of detected polarized nuclei.

4. Experimental detection sensitivity of ^{3}He NMR with a SQUID

To estimate the sensitivity of detection of polarized neutrons with ^{3}He using ULF methods, the NMR signal was detected from 1.94×10^{19}/cm^3 of weakly spin-polarized ^{3}He gas in a glass cell. The cell, shown as the blue[2] object in Fig. 4, is a cylinder with a 60-mm length, a 25-mm diameter, and a rounded top in the shape of a hemisphere of the same diameter (volume 23.34 cm^3). Measurements were made with the second-order axial gradiometer used in the water phantom experiment. The experimental setup and the pulse sequence were also the same.

The cell was held at 4 K in a liquid helium bath in the tail of the cryostat. The ^{3}He atoms in the cell were pre-polarized in the field B_{p} of approximately 0.02 T. The NMR measurement field B_{m} was set to 30 μT, inducing the precession of helium nuclei at ~980 Hz. For the specific geometry of the cell and the experimental configuration, the numerical calculations based on Eq. (2) give a 223-fT field, if the flux entered only the lowest two turns of the gradiometer, a total gradiometer input magnetic flux of 1.98×10^{-16} Wb, corresponding to a SQUID magnetic flux of 7.55×10^{-19} Wb. Using the SQUID response factor 4.84 ± 0.18 V/nT (2.96 V/Φ_0), the expected NMR signal amplitude of 1.07 ± 0.03 mV is obtained. The observed amplitude of FID at $t = 0$ is 0.75 mV (Fig. 5) with a pre-polarization time of 48 s. The calculations are based on thermal polarization [Eq. (4)] and assume essentially infinite pre-polarization time. We experimentally observed a longitudinal relaxation time of $T_1 = 18 \pm 3$ s. After extrapolation of the experimental result to infinite pre-polarization time, the amplitude becomes 1.00 mV, in somewhat closer agreement with the theoretical prediction.

From the FFT data, the SNR is estimated to be ~14 in a 4-Hz bandwidth. From the FID fit of Fig. 5 (red[3] curve), the T_2^* of the sample was 0.25 s, much shorter than T_1. This reduced value can be attributed to magnetic field gradients in the measurement field. Due to experimental arrangement and necessity to insert the cell inside a Dewar, the sample cell was offset by 12.5 cm (the top of

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[1] For interpretation of color mentioned in Fig. 3 the reader is referred to the web version of the article.

[2] For interpretation of color mentioned in Fig. 4 the reader is referred to the web version of the article.

[3] For interpretation of color mentioned in Fig. 5 the reader is referred to the web version of the article.

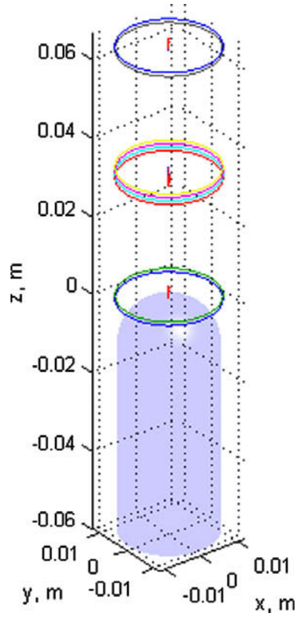

Fig. 4. Geometrical arrangement for the second-order gradiometer and the ^{3}He cell.

the cell, closest to the pick-up coil) from the center of the B_m coil. Such offset creates a gradient on the order of $0.001B_m$/cm, which is sufficient to shorten T_2 to the level observed in the experiment.

The number density of ^{3}He atoms in the cell was 2×10^{19}, and for the applied pre-polarization field of 0.02 T, $N_p = 0.8 \times 10^{14}$. Based on these results, the achieved sensitivity is $\sim 0.8 \times 10^{14}$ atoms/cc with a SNR of 14 in 0.25 s. The proposed measurement time for the nEDM experiment is 500 s (assuming T_1 and $T_2^* > 500$ s), which would improve the SNR by a factor of ~45, or give a sensitivity to $\sim 0.13 \times 10^{12}$ ^{3}He atoms/cm^3 with a SNR of 1. In the nEDM experiment, there will be $\sim 2 \times 10^{12}$ ^{3}He atoms/cm^3 in the nEDM cell. From the current experiment, even with its poorly optimized configuration, it can be concluded that it should be possible to detect the number of ^{3}He atoms proposed for use in the nEDM experiment with a SNR of 15.

5. Optimized sensitivity for the nEDM experiment

While the configuration considered in the previous section provides a sufficient sensitivity to detect the required number of ^{3}He nuclei in the nEDM experiment, further improvement in sensitivity can be achieved by optimization of the geometry of pick-up coils. One direction for such optimization is to increase the size of the pick-up coil. As shown in Eq. (6), the input flux scales quadratically with radius. However, as the gradiometer radius grows, the inductance L_{grad} grows linearly with radius. Thus, combining Eqs. (6) and (8), the sensitivity of a SQUID magnetometer or gradiometer grows proportionally to the radius of the pick-up coil. For example, using experimental data from a 9850 mm^2 axial gradiometer (112 mm diameter), with a predicted input flux of 5×10^{-17} Wb (an estimate of the expected flux from ^{3}He in the nEDM experiment), the peak-to-peak amplitude of the flux at the SQUID would be 12.5×10^{-20} Wb or 62.5 $\mu\Phi_0$. This flux translates to an RMS value of 22 $\mu\Phi_0$. (The experimental parameters are: $L_i = 2.5$ μH, $L_p = 0.42$ μH, $M = 7.4$ nH.) Using the dependence of SQUID noise on temperature [10], the expected RMS noise at 0.5 K is 0.5 $\mu\Phi_0/\text{Hz}^{1/2}$. Thus the SNR in 1 Hz band will be ~45. In practice an array of eight gradiometers of 30 cm^2 is proposed. While a factor of 3 in the signal will be lost due to reduced area, it will be compensated for by the array.

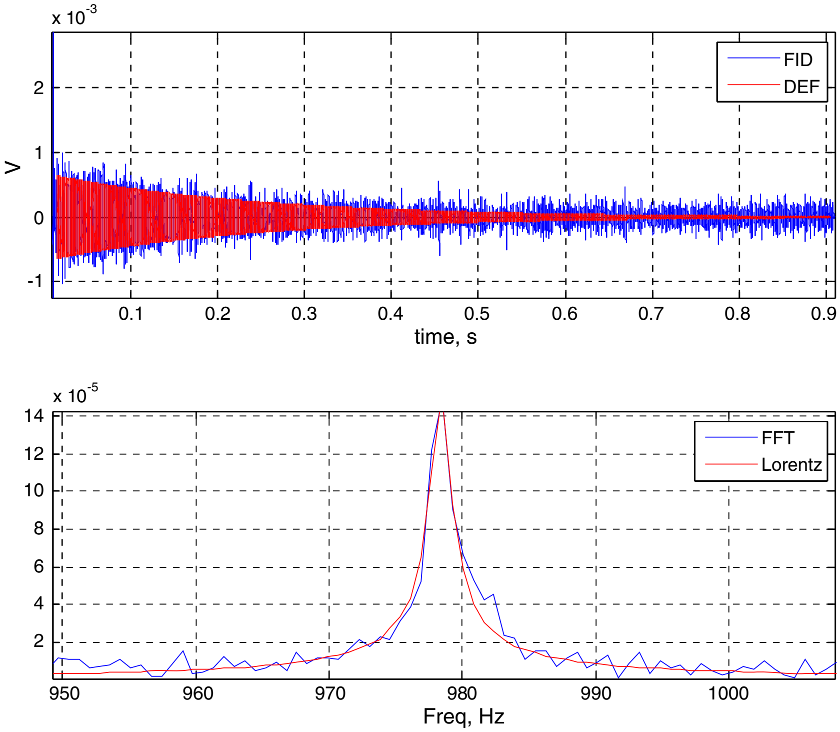

Fig. 5. (Top) The FID NMR signal (blue) in volts from thermally polarized ^{3}He sample. To convert to magnetic field units, the values have to be divided by 4.84 ± 0.18 V/nT. The fit with decaying exponent (red) gives $T_2^* = 250$ ms. (Bottom) FFT of the FID above. Noise density is ~10 $\mu\text{V/Hz}^{1/2}$ or ~2 $\text{fT/Hz}^{1/2}$ and SNR measured as the ratio of the peak value to noise density is ~14. (For interpretation of color mentioned in this figure the reader is referred to the web version of the article.)

From [11], the RMS frequency resolution is

$$(\delta f_3)^2 = \left(\frac{N}{A}\right)^2 \frac{3}{\pi^2} \frac{1}{T_{\mathrm{m}}^3}. \tag{9}$$

Using $N = 0.5\ \mu\Phi_0/\mathrm{Hz}^{1/2}$, $A = 22\ \mu\Phi_0$, and $T_m = 500$ s, the frequency resolution becomes $\delta f_3 \sim 1.2\ \mu\mathrm{Hz}$. The requirement of the nEDM experiment is 26 μHz. This calculation assumes the ^{3}He polarization does not decay significantly over the measurement period.

6. Conclusion

The experiments and theory discussed demonstrate the feasibility of SQUID detection for the concentration of polarized ^{3}He atoms proposed in the nEDM experiment even with the very simple setup presented here. Close agreement has been obtained between the predictions and the measurements of NMR signals from both protons and ^{3}He atoms, giving credence to the plans for the full nEDM experimental setup.

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