Direct measurement of the $\gamma_{\text{He}}/\gamma_{\text{Xe}}$ ratio at ultralow magnetic field

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Abstract. A co-located $^3$He and $^{129}$Xe nuclear spin free precession measurement at sub-$\mu$T magnetic field was carried out in a magnetically shielded environment. The uncorrected quotient of the gyromagnetic ratios between neutral $^3$He and $^{129}$Xe atoms is determined to be 2.754 082 81(07), accounting for only statistical error. Our measurement shows that this ratio has a stability of $1.4 \times 10^{-5}/\sqrt{\tau}$, demonstrating the ability to reach the current precision limit of the quotient in a 10000 s of averaging time $\tau$. This precision is enough for the next-generation EDM search in neutral $^{129}$Xe atoms based on a similar comagnetometer scheme.

1. Introduction

The interaction of the nuclear electric dipole moment ($\vec{d}$) with an external electric field ($\vec{E}$) creates a measurable energy $\vec{d} \cdot \vec{E}$ for an atom or molecule despite the Schiff’s screening effect [1]. In the nuclear spin free precession setting, this $\vec{d} \cdot \vec{E}$ term corresponds to an additional shift to the Larmor frequency. We are currently involved in a collaboration in the search of non-vanishing electric dipole moment (EDM) in neutral $^{129}$Xe atoms [2]. The experiment will be based on the free spin precession in the gaseous phase at ultralow magnetic field ($\approx \mu$T) in a heavily shielded environment such as the ones located in Berlin [3] and Munich [4].

The expression for determining the atomic electric dipole moment of $^{129}$Xe is given as follows:

$$d_{\text{Xe}} = \frac{h}{4E} \left[ (f_{\text{Xe}}^{\uparrow \uparrow} - f_{\text{Xe}}^{\downarrow \downarrow}) - \gamma_{\text{Xe}} \Delta B_0(t) \right],$$

where the arrows $\uparrow \uparrow$ and $\downarrow \downarrow$ indicate the relative orientations of the externally applied electric and magnetic fields, $\Delta B_0(t)$ is the temporal drift of the magnetic field during measurements, and $E$ is the applied electric field that is stable in time. The signature of an EDM would be a change in the $^{129}$Xe precession frequencies ($f_{\text{Xe}}$) that is correlated to the inversion of the relative orientation of the electric and magnetic fields. However, a similar signature (i.e. a fake EDM signal) can also result if the magnetic field term drifts in time.

To cancel out the common magnetic systematic and other noise sources, simultaneous precessions of two atomic species located in the same sample space are used (e.g. see the previous EDM search in $^{129}$Xe which dealt with frequencies in the microwave regime [5].) Based on the Zeeman effect ($f_{\text{Larmor}} = \gamma \cdot B_0$), the issue of magnetic field instability is alleviated by
Table 1. Gyromagnetic ratios for $^3$He and $^{129}$Xe derived from measurements in the literature. All values are stated without shielding corrections.

| Species | Latest | Source |
|---------|--------|--------|
| $^3$He  | 42.576 3866(10) MHz/T | Ref [9] |
| $^{129}$Xe | 11.776 7392(10) MHz/T | Combine Ref [7] and [9] |
| $^3$He/$^{129}$Xe | 3.754 081 35(22) | Ref [6] |
| $^{129}$Xe/$^3$He | 3.754 081 60(31) | Ref. [7], [8] via $\gamma_p$ |
| $^3$He/$^{129}$Xe | 3.754 081 60(18) | Ref. [7], [8] via Eqn. 5 |

It is evident from Eqn. 2 that a crucial parameter is the quotient of the gyromagnetic ratios $\gamma_{^3$He}/$\gamma_{^{129}$Xe}$, or equivalently $\gamma_{^3$He}/$\gamma_{^{129}$Xe}$. In this report, we present data and analysis of a direct measurement of the $\gamma_{^3$He}/$\gamma_{^{129}$Xe}$ ratio at 0.4 $\mu$T and discuss the impact of its precision on the EDM search.

2. Current status of $\frac{\gamma_{^3$He}}{\gamma_{^{129}$Xe}}$

The most precise value available for the $^{129}$Xe nuclear magnetic moment was derived from the ratio of Larmor frequencies of $^{129}$Xe and $^1$H at 1.5 T [7]. The $^1$H reference signal was obtained by replacing the 0.98 bar Xenon sample with a demineralized H$_2$O sample of supposedly the same spherical shape. The frequency ratio was determined to be:

$$\frac{f_{^{129}$Xe}(\text{gas, sphere, 22.5} \degree \text{C})}{f_{^1$H}(\text{H}_2\text{O, sphere, 22.5} \degree \text{C})} = 0.276 \ 602 \ 600(17).$$  \hspace{1cm} (3)

For $^3$He and $^1$H, an even more precise value of the Larmour frequency ratio was derived at 0.1 T [8]. The internal field difference caused by the cell shape differences was eliminated by replenishing the 4 mbar of $^3$He with H$_2$O in the same cell consecutively. Both $^3$He and H$_2$O signals were compared to a common water NMR reference standard. This ratio was determined to be:

$$\frac{f_{^3$He}(\text{gas, sphere, 25} \degree \text{C})}{f_{^1$H}(\text{H}_2\text{O, sphere, 25} \degree \text{C})} = 0.761 \ 786 \ 131(33).$$  \hspace{1cm} (4)

Equation 4 leads to the most precise value for the helion magnetic moment and is accepted in the 2010 evaluation of Fundamental Physical Constants [9]. Dividing it by Eqn. 3 yields the quotient of interest for the EDM search in $^{129}$Xe:

$$\frac{f_{^3$He}/f_{^1$H}}{f_{^{129}$Xe}/f_{^1$H}} = \frac{\gamma_{^3$He}}{\gamma_{^{129}$Xe}} = 2.754 \ 081 \ 60(18).$$  \hspace{1cm} (5)

It should be noted that the uncertainty of $6.5 \times 10^{-8}$ is dominated by the uncertainty of the gyromagnetic ratio of $^{129}$Xe. The value for the quotient of gyromagnetic ratios can also be deduced from the absolute numbers of the respective gyromagnetic ratios provided that the shielded proton gyromagnetic ratio is known.

Values of gyromagnetic ratios from various measurements are summarized in Table 1. The uncertainty for the quotient, however, will be slightly bigger for the value derived from the proton than the value shown in Eqn. 5 due to the contribution of the uncertainty of the shielded...
proton gyromagnetic ratio. In Ref. [6], a high-field NMR measurement at 11.7586 T was done separately for $^3$He and $^{129}$Xe with the same set-up to derive a $\frac{2\mu}{\gamma_{\text{Xe}}}$ value of 2.754 081 35(22), which is in agreement with Eqn. 5 with a comparable precision.

3. Experiment

The measurement was carried out in the magnetically shielded room in Berlin (BMSR-2) [3]. The nuclear spins of $^3$He were polarized by means of metastability exchange optical pumping, whereas the nuclear spins of $^{129}$Xe were polarized by means of spin exchange optical pumping. $^3$He and $^{129}$Xe atoms were mixed in a single cylindrical cell (diameter=58 mm, length=60 mm) with the following partial pressure conditions $^3$He: 3.6 mbar, $^{129}$Xe: 5.3 mbar, and $N_2$: 26.3 mbar. The nitrogen buffer gas pressure was chosen [11] based on the trade-offs between (a) reducing relaxation caused by formation of Van der Waals molecules (need high $N_2$ concentration) and (b) reducing relaxation caused by magnetic field gradients (need low total gas pressure).

![Figure 1](image)

Figure 1. (Colour Online) (a) The software gradiometer signal. The non-zero offset is a derivative of the working point of the SQUID locking circuit. (b) The data and the fit at the beginning of the measurement. (c) The data and the fit near the end of the measurement. The open dots are the measured values. The red lines represent the model-regressed curves.

The spin precession was initiated by a non-adiabatic 90° change of the direction of the guiding field. The guiding field ($B_0 = 350$ nT) was induced by two sets of crossed Helmholtz coils. The signal of the precessing spins was detected with a multichannel vector magnetometer system, containing directly coupled superconducting quantum interferometer devices (dc-SQUID) [10]. More details of the experimental setup are described in Ref. [11] and Ref. [12].

In this measurement, 33 SQUID magnetometer channels were recorded, differing in their positions with respect to the cell. Thus, far-field common-mode backgrounds and mechanical-vibration modes, originating from relative motions of the detectors and the cell, could be suppressed by a software gradiometer. Specifically, signals from two magnetometer channels were combined during the post-processing to form one gradiometer signal. One of the combining channels should be close to the cell to maximize the sensitivity of the spin precession. The other combining channel should be sufficiently far away from the cell such that hardly any precession signal is detected which would otherwise reduce the signal-to-noise ratio (SNR) of the calculated gradiometer signal.

Fig. 1-a shows the superposition of the two decaying sinusoidal signals of $^3$He and $^{129}$Xe. The measurement contains 10,501,760 data points in total. The time interval between subsequent data points is 1/250 s as shown by the open dots in Fig. 1-b and Fig. 1-c. The signal, and
consequently the SNR, is decreasing with time due to the dephasing of the nuclear spins with a transversal relaxation time $T_2^*$. In this measurement, more than 59 hours and 5 hours of $T_2^*$ times were obtained for $^3$He and $^{129}$Xe, respectively.

4. Frequency Extraction by Regression Model

Precession frequencies ($\omega = 2\pi f$) of $^3$He and $^{129}$Xe need to be extracted to derive $\frac{2\pi f}{\gamma}$, Common methods to extract the frequency estimator from a time-domain precession signal are (a) the zero-crossing counting, (b) the frequency mixing and demodulation, and (c) the time-series regression. Methods (a) and (b) typically involve pre-stage and post-stage filtering processes whereby deducing the impacts on the final uncertainty budget is cumbersome. Therefore, method (c) is the preferred analysis tool. However, the pre-requisite of the time-series regression is that the measurement needs to be partitioned into blocks so that the frequency shift in a block is negligible in comparison to the estimation uncertainty.

A regression model in its most general form can be expressed as $\sum_{j=1}^{d} b_j t^j + \sum_{k=1}^{\beta} [c_{jk} \cos(\omega_k t) + s_{jk} \sin(\omega_k t)] + \epsilon$, where $\epsilon$ is the noise, $[b_j, c_{jk}, s_{jk}]$ are parameters to be fitted, $[d, \beta]$ are ordering parameters for the regression model, and $\beta$ is the number of frequency components to be regressed. Setting the ordering parameters $d$ and $\lambda$ to 1 and allowing the existence of two frequencies, the model to fit is obtained as:

$$\mu(t) = b_0 + b_1 t + \sum_{k=1}^{2} [c_k \cos(\omega_k t) + s_k \sin(\omega_k t)] + \epsilon.$$  \hspace{1cm} (6)

Equation 6 is a linear model \(^1\) in the sense that the response $\mu$ is a linear combination of predictors $[t, \cos(\omega t), \sin(\omega t)]$ scaled by corresponding coefficients $[b_1, c_1, s_1]$. For signals within a block, assumptions implicitly made by the model are: (a) the drift in the signal offset is approximated to be linear as represented by the first two terms in Eqn. 6, (b) the amplitude decay is neglected by the fit \(^2\), (c) the noise $\epsilon$ is stationary with a covariance that does not depend on the time shift, and (d) the frequency is constant.

Equation 6 can be re-expressed in the matrix form as:

$$\mathbf{U} = \begin{bmatrix} 1 & t \cos(\omega_1 t) \sin(\omega_1 t) \cos(\omega_2 t) \sin(\omega_2 t) \end{bmatrix} \cdot \begin{bmatrix} b_0 \\ b_1 \\ c_1 \\ s_1 \\ c_2 \\ s_2 \end{bmatrix} + \epsilon$$ \hspace{1cm} (7)

where the design matrix $\mathbf{D}$ is uniquely determined for a given set of frequency values. There are two stages in the frequency estimation process. In the first step, the initial guess of $\omega$’s is determined by the peak location in the Fourier spectrum of the whole time series \(^3\). In the second step, the parameter vector is found by $\mathbf{P} = (\mathbf{D}^\mathbf{T})^{-1} \mathbf{D}^\mathbf{T} \mathbf{U}$ in the linear least-square sense. The variable projection functional to be minimized is the difference between $\mathbf{U}$ and $\mathbf{D}\mathbf{P}$. The nonlinear least-square solver used is \texttt{lsqnonlin} package in Matlab\(^8\). This variable projection optimization technique for nonlinear least-square problems is well documented in Ref. [13].

Typical fits at the beginning and tail parts of the measurement are shown in Fig. 1-b,c. The apparent existence of a frequency beat note pattern in Fig. 1-b is a clear indicator that there are

\(^1\) A comparison can be made to the more familiar expression $A \sin(\omega t + \phi)$, which is nonlinear in this context since there are two variables $[\omega, \phi]$ inside one functional predictor (the sine function).

\(^2\) The shortest relaxation time of the signal is about 18,950 $s$, implying a decrease in the signal amplitude of 0.005% at most within one block length assuming an $\exp(-t/T_2^*)$ dependence.

\(^3\) The estimation error in this initial guess is no more than half of the frequency bin size $\approx 12$ $\mu$Hz.


multiple resonant frequencies. The disappearance of the frequency beat note pattern in Fig. 1-c is caused by the differences in the dephasing rates of the resonant species. A 100-s block length is chosen such that the averaging time is short enough to avoid the detrimental effect on the frequency stability caused by the magnetic field drift. This blocksize results in a total of 420 blocks. The lowest precession frequency of the signal is about 4.16 Hz, corresponding to 416 cycles per block. Each block is treated as an independent dataset for the regression.

The covariances of fitted parameters, hence of the fitting uncertainties, are derived from the first-order partial derivatives of the model (i.e. the Jacobian matrix \( J \)) as follows:

\[
\text{cov}(\hat{\theta}) = \frac{s^2}{\text{dof}} (J^T J)^{-1},
\]

where \( \hat{\theta} = (\hat{b}_0, \hat{b}_1, \hat{c}_1, \hat{s}_1, \hat{c}_2, \hat{s}_2, \hat{\omega}_1, \hat{\omega}_2) \) are the best-fit parameter values, \( s^2 \) is the sum of the residual squares, and \( \text{dof} \) is the degree of freedom (the number of data point subtracted by the number of parameters).

### 5. Results and Discussion

The extracted spin precession frequencies of the two nuclear species are shown in Fig. 2-a,b. Converting the frequency values to the \( B_0 \) field yields the expected 0.35 \( \mu \)T.

![Figure 2. (Colour online) The extracted precession frequencies of (a) \(^{129}\)Xe (blue curve) and (b) \(^{3}\)He (green curve). The insets are the amplitude curves of the respective nuclei. The ratio \( R \) of the two frequencies is shown in (c). The blocksize used is 100 s.](image)

Fig. 2-c depicts the quotient of gyromagnetic ratios \( R = \frac{f_{^{3}\text{He}}}{f_{^{129}\text{Xe}}} \). Using the uncertainties of the fitted frequencies, denoting \( \sigma_{f_{^{3}\text{He}}} \) and \( \sigma_{f_{^{129}\text{Xe}}} \), the block uncertainty of the ratio of the two frequencies and hence the quotient of the gyromagnetic ratios can be derived using the principles listed in GUM [14] as \( \sigma_R = \sqrt{(\sigma_{f_{^{3}\text{He}}}/f_{^{3}\text{He}})^2 + (f_{^{3}\text{He}}/f_{^{129}\text{Xe}}^2 \cdot \sigma_{f_{^{129}\text{Xe}}})^2} \). The uncertainty and the fluctuation of the ratio increases with the measurement time due to the decaying signal amplitude via the \( T_2^\ast \) relaxation mechanism of the nuclear spins.

The overlapping Allan standard deviation of \( R \) is shown in Fig. 3. A slope of \( 1.4 \times 10^{-5}/\sqrt{T} \) implies that the common-mode drift in \( R \) appears to be successfully removed by the quotient operation. In an averaging time of 10000-s, the stability of \( R \) reaches \( 1.4 \times 10^{-7} \).

A weighting factor is assigned to each block according to \( \kappa_i = 1/\sigma_{R_i}^2 \), which leads to the corresponding weighted mean \( \bar{R} = \sum_i R_i \kappa_i / \sum_i \kappa_i \). The standard deviation of the mean is taken as \( \delta \bar{R} \). This generic weighted averaging procedure is the standard method when combining...
Figure 3. The stability plot of $\frac{\gamma_{\text{He}}}{\gamma_{\text{Xe}}}$ as a function of averaging time. The dashed line is a linear fit to the Allan deviation points from 100 s to 10000 s.

results from different clocks to compute the International Atomic Time [16]. Thus, the $\frac{\gamma_{\text{He}}}{\gamma_{\text{Xe}}}$ is determined to be 2.754 085 44(07), with a 2.5 × 10$^{-8}$ relative uncertainty exhibiting less than half the errors as given by Eqn. 5 and the value stated in Ref. [6].

The difference between our value and those from literature arises from the fact that various systematic effects have not been considered. We have to take into account that our measurement was done in a non-inertial frame of reference due to the constantly rotating surface of the Earth. The precession frequencies measured in our laboratory frame were thus modified. The sense of spin rotation can be deduced from the Bloch equations. The rate of the earth rotation fluctuates from day to day. On the day when the measurement was taken, the rotational speed of the earth was 11.605 761 7234(87) µHz [17]. The projection of the earth rotation to the lab frame can be expressed by $sid = \Omega \cdot \cos(\phi) \cdot \cos(\rho)$, where $\Omega$ is the angular velocity of the rotating earth, $\phi$ is the latitude of the setup, and $\rho$ is the angle between $B_0$ and the North-South axis. The uncertainty of $sid$ can be found by $\sigma_{sid} = \sqrt{(\sigma_\Omega \cos(\phi) \cos(\rho))^2 + (\sigma_\rho \Omega \cos(\phi) \sin(\rho))^2}$, where the $\sigma$’s are the uncertainties in the respective parameters. The latitude of the setup is 52.5164(1) assuming a perfect spherical model for the Earth. The angle between $B_0$ and the North-South axis is 28.0(5)$^\circ$. Therefore, the systematic correction due to the earth rotation is $sid = 6.236(29)$ µHz, with the uncertainty mainly dominated by the accuracy of the $B_0$ orientation. Considering the sidereal correction, the ratio $R$ is determined to be 2.754 082 81(07).

There are other systematic effects which could arise due to the spin-spin interaction among and between the nuclei. An asymmetry in the spherical shape of the sample cell in the presence of a $B_0$ field inhomogeneity could cause an additional shift in the precession frequency. Typical known effects include the geometric phase [18], the chemical shift [19], the geometric shift [15], the gravitational shift and the Ramsey-Bloch-Siegert shift [20, 21]. Careful examinations are needed in all of these aspects to derive a corrected $R$ value.

The sensitivity coefficient of $^{129}$Xe EDM to $\frac{\gamma_{\text{He}}}{\gamma_{\text{Xe}}}$ is determined to be 4.14 × 10$^{-46}$ Jm/V under a 4-kV electric field and a 1-µT magnetic field. Therefore, our current $\frac{\gamma_{\text{He}}}{\gamma_{\text{Xe}}}$ instability contributes $\lesssim 10^{-32}$ e-cm to the $^{129}$Xe EDM. This precision is well below the $10^{-28}$ e-cm target value.

6. Conclusion
The direct determination of $\frac{\gamma_{\text{He}}}{\gamma_{\text{Xe}}}$ has been carried out in a comagnetometer setup under an ultralow ambient magnetic field ($< \mu$T). The resultant precision is higher than the current literature value and will be sufficient for the next generation search of the EDM in $^{129}$Xe. Careful and thorough investigations on the systematic effects are underway to understand the remaining deviations from the literature value.

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