Nuclear-Spin Comagnetometer Based on a Liquid of Identical Molecules

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Nuclear-spin comagnetometer based on a liquid of identical molecules

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Atomic comagnetometers are used in searches for anomalous spin-dependent interactions. Magnetic field gradients are one of the major sources of systematic errors in such experiments. Here we describe a comagnetometer based on the nuclear spins within an ensemble of identical molecules. The dependence of the measured spin-precession frequency ratio on the first-order magnetic field gradient is suppressed by over an order of magnitude compared to a comagnetometer based on overlapping ensembles of different molecules. Our single-species comagnetometer is capable of measuring the hypothetical spin-dependent gravitational energy of nuclei at the 10^{-17} eV level, comparable to the most stringent existing constraints. Combined with techniques for enhancing the signal such as parahydrogen-induced polarization, this method of comagnetometry offers the potential to improve constraints on spin-gravity coupling of nucleons by several orders of magnitude.

Atomic comagnetometers typically consist of overlapping ensembles of at least two different species of atomic spins [1–4]. Practically, it is the ratio of the spin-precession frequencies of the different species under the influence of a bias magnetic field that is measured. The ratio is relatively insensitive to changes in the magnetic field, but retains sensitivity to Zeeman-like nonmagnetic spin interactions. Comagnetometers have been widely used for fundamental physics experiments [5], such as measurements of permanent electric dipole moments (EDMs) [6–10], tests of CPT and Lorentz invariance [11–16], and searches for exotic spin-dependent interactions mediated by hypothetical bosonic fields [17–24]. Comagnetometers also find practical applications as sensitive gyroscopes [25, 26].

In fundamental-physics experiments using comagnetometers based on overlapping ensembles of different species, one of the major systematic effects reducing accuracy is due to uncontrolled magnetic field gradients [8, 24, 27]. Previous work demonstrates that there exists some spatial separation between the ensemble-averaged position of different spin species due to nonuniform polarization [27], gravity [8], and/or thermodiffusion effects [28]. In the presence of a magnetic field gradient, the average magnetic field sensed by different spin species is different. The ratio of spin-precession frequencies acquires a magnetic-field-gradient dependence that can add noise and is difficult to distinguish from other sources of nonmagnetic torques on spins. Thus, complex arrangements are needed to monitor and reduce the magnetic field gradient for each cycle of measurement [4, 24, 29].

In contrast to comagnetometers which utilize overlapping ensembles of different atomic or molecular species, here we introduce and demonstrate a new comagnetometer configuration based on an ensemble of identical molecules. In this single-species comagnetometer, different nuclear spins are probed within the same molecule. Thus, the spatial sampling of the field by the different nuclear spins is made nearly identical and systematic errors related to field gradients are highly suppressed. By taking advantage of the techniques of ultralow-field nuclear magnetic resonance (NMR) and sensitive atomic magnetometry, the J-coupling (indirect spin-spin coupling) spectrum of a liquid-state ensemble of acetonitrile-2-^{13}C molecules can be measured with sub-mHz precision with a single scan (10 s measurement time). Under the influence of a bias magnetic field, the J-coupling resonance lines at different frequencies split into separate peaks. The frequency separation between the split peaks for each J-coupling resonance has distinct linear coefficients with respect to the magnetic field. Measurements of these splittings can be employed as a comagnetometer. We experimentally demonstrate that in the presence of a temperature gradient, such a comagnetometer is insensitive to first-order magnetic field gradients within experimental uncertainty. We analyze a possible application of this new kind of comagnetometer for measurement of a coupling between nuclear spins and gravitational fields.

The device is based on a zero- to ultralow- field NMR configuration and the experimental setup is described in detail in Refs. [28, 30]. The spin ensemble we use to realize the comagnetometer is liquid-state acetonitrile-2-^{13}C ($^{13}CH_3CN$, from Sigma-Aldrich, 100 μL), which is flame-sealed under vacuum in a standard 5 mm NMR tube. The sample is initially polarized in a 1.8 T Halbach magnet for 30 s, and then pneumatically shuttled down into a four-layer magnetic shield (Twinleaf MS-1F). During the transit, a $\sim$ 30 μT magnetic field is applied with a solenoid to guide the initial spin magnetization along the vertical direction (y). After the sample drops into the detection region ($\sim$ 1 mm above a rubidium vapor cell), the guiding field is turned off within 10 μs. The initial spin magnetization then evolves under the J-coupling interaction between ^{13}C and the three ^{1}H protons, which generates an oscillating magnetization signal and is detected with a rubidium atomic magnetometer (sensitivity $\approx$ 10

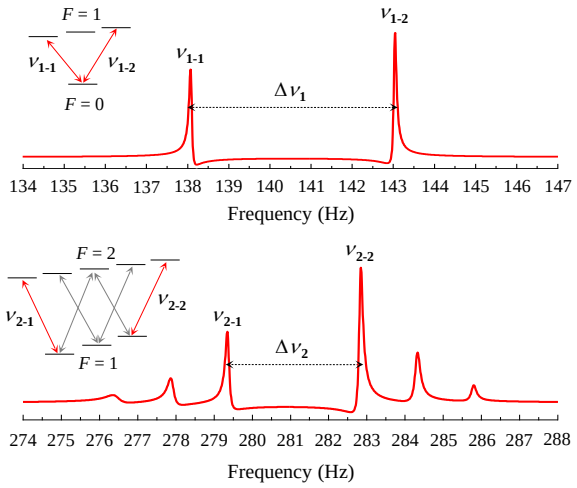

FIG. 1. (color online). Experimentally measured J-coupling spectrum of acetonitrile-2-^{13}C ($^{13}CH_3CN$) in a 80 nT bias field along z. The top and bottom traces show the split spectrum at J_{CH} and $2J_{CH}$, respectively. The related transitions used for comagnetometry are shown with solid red arrows.

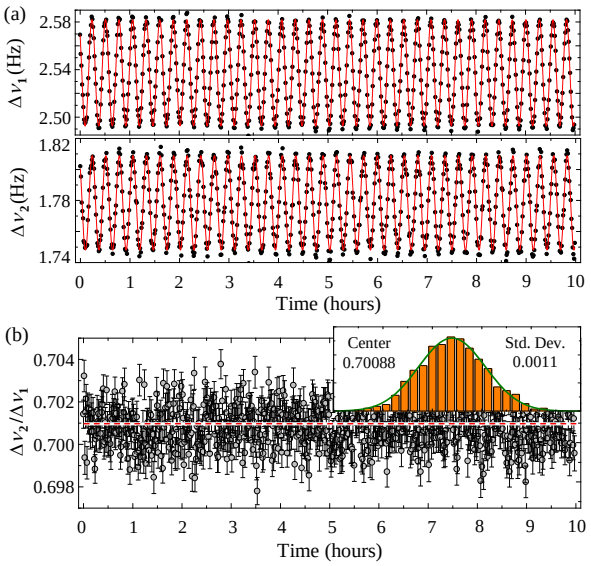

FIG. 2. (color online). Experimental demonstration of comagnetometry with an ensemble of identical molecules. (a) The measurements were taken while an oscillating magnetic field was applied along z (see text). The red solid lines are the fitted curves. (b) The calculated $\Delta\nu_2/\Delta\nu_1$ based on (a) with a reduced χ^2 of 1.09. The average value of $\Delta\nu_2/\Delta\nu_1$ (red dashed line) is 0.70088(4). The inset shows the histogram of $\Delta\nu_2/\Delta\nu_1$ which follows a Gaussian distribution.

fT/Hz$^{1/2}$). A small bias field along z is applied by coils within the innermost shield layer, and can be regarded as a small perturbation to the dominant J-coupling interaction, which in turn splits the J-coupling spectrum.

Acetonitrile-2-^{13}C is a $^{13}CH_3$ system with three equivalent protons. The resulting zero-field J-coupling spectrum consists of two resonance lines, with one at J_{CH} and the other at $2J_{CH}$ [31, 32]. The measured J-coupling frequency for acetonitrile-2-^{13}C in our experiment is 140.55002(3) Hz, which is a function of the sample temperature ($\sim$ -125 μHz/K). With a small bias magnetic field ($\sim$ 80 nT), the two lines split into different patterns of peaks, see Fig. 1. The spectrum around J_{CH} splits into two peaks, while the spectrum around $2J_{CH}$ splits into six. All the peaks are from a linear combination of magnetization signals along y and x, which are measured simultaneously with the magnetometer in the presence of a bias field along z [28]. The asymmetry within the peaks is thus due to the different responses of the magnetometer to magnetic fields along y and x, which can, in principle, be eliminated using decoupling techniques [4]. Within the $2J_{CH}$ multiplet, we focus on the central two peaks, as they have the highest signal-to-noise ratio compared to the others. Neglecting all other nonmagnetic spin interactions, the frequencies for the two splittings $\Delta\nu_{1,2}$ are $\Delta\nu_1 = (\gamma_h+\gamma_c)B_z$, $\Delta\nu_2 = \frac{1}{2}(\gamma_h+3\gamma_c)B_z$, where $\gamma_{h,c}$ are the gyromagnetic ratios for ^{1}H and ^{13}C, respectively, and B_z is the bias magnetic field [31, 33]. There are no contributions from the second-order Zeeman effect on $\Delta\nu_{1,2}$, and the third-order Zeeman effect is an order of magnitude smaller than the current experimental uncertainty (see the Supplemental Material). Since $\Delta\nu_1$ and $\Delta\nu_2$ are both proportional to B_z but with different linear coefficients, we can employ them to realize a comagnetometer based on an ensemble of identical molecules.

Comagnetometers should be able to suppress the variations in the bias magnetic field. As a demonstration, we apply a slowly varying magnetic field along the same direction (z) as the bias field, with 1 mHz frequency and 0.5 nT amplitude. Since the total acquisition time for each scan is 10 s, the oscillating magnetic field is effectively DC within this sampling window. Figure 2(a) shows the measured frequencies $\Delta\nu_{1,2}$, both of which display an evident 1 mHz modulation. The ratio between $\Delta\nu_{1,2}$ is calculated and shown in Fig. 2(b). Compared with Fig. 2(a), there is no apparent modulation of the frequency ratio. We perform Fast Fourier Transform to $\Delta\nu_{1,2}$ and $\Delta\nu_2/\Delta\nu_1$. The amplitude of $\Delta\nu_2/\Delta\nu_1$ at 1 mHz is around 3×10^{-5}, consistent with the background noise, which corresponds to a magnetic field noise suppression factor of greater than 750. Based on measurements over 10 hours, the averaged value of $\Delta\nu_2/\Delta\nu_1$ is 0.70088(4). By using $\gamma_h = 42.5775$ MHz/T [34] and $\gamma_c = 10.7077$ MHz/T [35], and taking into account the shielding factors of acetonitrile-2-^{13}C, i.e., $\sigma(^1\text{H}) = 31$ ppm, $\sigma(^{13}\text{C}) = 185$ ppm [35, 36], the theoretical value is 0.70092. Besides this, the third-order Zeeman effect modifies the frequency ratio at the level of 10^{-6} based on the current experimental parameters (see the Supplemental Material). However, systematic effects related to such a difference can be suppressed by employing field-reversal methods [24].

It has been demonstrated that, for a dual-species comagnetometer, the spin-precession frequency ratio is a function of the magnetic field gradient. A thorough investigation of magnetic-field-induced systematic effects

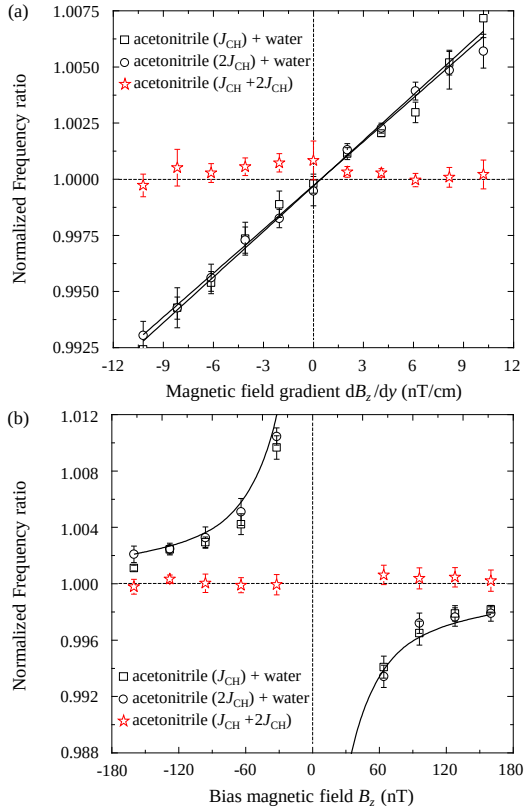

FIG. 3. (color online). Comparison of the normalized frequency ratios between a single-species comagnetometer (red star, $\Delta\nu_2/\Delta\nu_1$) and two dual-species comagnetometers (black square, $\nu_h/\Delta\nu_1$, black circle, $\nu_h/\Delta\nu_2$, discussed in the text). All the data are taken with the same acetonitrile-2-^{13}C (with $\sim$ 1% water) and are normalized with the theoretical values at zero magnetic field gradient. (a) The normalized frequency ratio as a function of the gradient dB_z/dy, with constant bias magnetic field B_z = 80 nT. The solid lines are the linearly fitted curves. (b) The normalized frequency ratios as a function of bias magnetic fields B_z, with constant gradient dB_z/dy = -3 nT/cm. The solid line is the fitted curve based on B_z^{-1}. Each point is an average of eight measurements.

can be found in Ref. [27]. Although their analysis is based on a gas-phase comagnetometer, many of the conclusions are also valid for a liquid-state comagnetometer. Here, we focus on the shifts in the spin-precession frequency ratio due to the first-order gradient, typically, the gradient of the bias magnetic field along the vertical direction (y), i.e., dB_z/dy. If there exist temperature gradients, different spin ensembles can experience different thermodiffusion rates, which causes gradients in the concentration of the ensembles. A first-order magnetic field gradient thus introduces an additional component in the frequency ratio, which has the form $G_1\Delta/B_z$, where G_1 is the first-order magnetic field gradient, Δ is the separation of the centers of the ensemble-averaged position of the spins. Besides thermodiffusion, concentration gradients may also arise due to the different densities under the influence of gravity, a process known as barodiffusion [28, 37]. Previous work demonstrates that the concentration gradients induced by thermodiffusion and barodiffusion are comparable if the thermal gradient is of a few mK/cm. Since our sample is placed closely ($\sim$ 1 mm) above the rubidium vapor cell, which is heated to $\sim$ 170°C, the temperature gradient is large along the vertical direction ($\sim$ 25 K/cm). Thus, the barodiffusion effect is negligible under the conditions of our experiment.

In order to determine the sensitivity to magnetic field gradients, we compare our single-species comagnetometer to two dual-species reference comagnetometers. The dual-species comagnetometers are based on the same acetonitrile-2-^{13}C, but use one of the splittings, $\Delta\nu_1$ or $\Delta\nu_2$, together with the precession frequency of ^{1}H in residual water present in the sample ($\sim$ 1%). The precession frequency of ^{1}H can be written as $\nu_h = \gamma_h B_z$. Therefore, for the two reference comagnetometers, the measured spin-precession frequency ratios are $\nu_h/\Delta\nu_1 = \gamma_h/(\gamma_h+\gamma_c)$ and $\nu_h/\Delta\nu_2 = 2\gamma_h/(\gamma_h+3\gamma_c)$, respectively.

Figure 3(a) shows the spin-precession frequency ratios for the three comagnetometers as a function of dB_z/dy. In order to compare the results at the same level, the measured spin-precession frequency ratios of each comagnetometer are normalized to the corresponding theoretical values at zero magnetic field gradient. For the two reference comagnetometers, the normalized frequency ratios are both linear in the magnetic field gradient, with slopes of $6.71(22)\times10^{-4}$ cm/nT ($\nu_h/\Delta\nu_1$, black squares) and $6.51(14)\times10^{-4}$ cm/nT ($\nu_h/\Delta\nu_2$, black circles), respectively. The slopes of the normalized frequency ratios for the two reference comagnetometers are nearly identical since they are based on the same sample. For the single-species comagnetometer, the results display a negligible linear dependence with the magnetic field gradient ($\Delta\nu_2/\Delta\nu_1$, red stars). Fitting the results with a linear function gives a slope of $-0.24(24)\times10^{-4}$ cm/nT, which is at least an order of magnitude smaller than the dual-species reference comagnetometers and, in fact, consistent with zero. The residual nonlinear dependence could be attributed to higher order effects of the gradient on the precession frequencies, which could introduce broadening and shift of the resonance lines [27].

Figure 3(b) shows the spin-precession frequency ratios as a function of the bias magnetic field. We apply a constant gradient dB_z/dy = -3 nT/cm. For the dual-species comagnetometer, the results are fit to the inverse of the bias magnetic field amplitude, B_z^{-1}, in agreement with the $G_1\Delta/B_z$ form of gradient dependence described above. Under the same condition, there is no apparent dependence of the frequency ratio on B_z^{-1} for the single-species comagnetometer.

We also apply first-order magnetic field gradients along x and z directions. Under these conditions, the frequency ratios measured with the reference comagnetometers similarly show no linear dependence on the first-order magnetic field gradient. Since there are negligible temperature gradients along x and z, the first-order gradient does not change the frequency ratio up to the second order of

the gradient strength, if the Larmor frequency is much larger than the diffusion rate across the cell (D/R^2, D is the diffusion constant, and R is the cell radius) [27, 38]. This situation is well satisfied in our experiment, considering that the diffusion constants for acetonitrile and water are both on the order of $10^{-5}\mathrm{cm}^2/\mathrm{s}$, the radius of the tube is ~ 0.2 cm, and the Larmor frequency is ~ 2 Hz. The results presented in Fig. 3 confirm that, in the presence of a temperature gradient, the first-order magnetic field gradients introduce systematic errors for a conventional dual-species comagnetometer, while they have a negligible effect on the single-species comagnetometer.

In addition to the first-order magnetic field gradient, higher order gradients can also introduce systematic effects. It is demonstrated that second-order gradients cause a shift in the frequency ratio proportional to the third power of the gradient strength [27]. Their analysis is based on the shift of the center of spin due to nonuniform polarization of the different spin ensembles. From this point of view, our single-species comagnetometer should also be free from this systematic effect.

This new kind of single-species liquid-state comagnetometer can be applied to tests of fundamental physics. One promising application is a search for a spin-gravity coupling. Detailed discussions of possible spin-gravity couplings can be found in [5, 17, 22–24] and references therein. Here we focus on the coupling of the nuclear spin to the gravitational field of the Earth. A possible spin-gravity coupling to the ^{13}C and ^{1}H nuclei of acetonitrile-2-^{13}C can be parameterized as modifications of the spin-precession frequencies [39]

$$\Delta\nu_1(\pm) = (\gamma_h + \gamma_c)B_z \pm (-\frac{1}{3}\chi_n + \chi_p)\frac{g\cos\phi}{\hbar}, \quad (1)$$

$$\Delta\nu_2(\pm) = \frac{(\gamma_h + 3\gamma_c)B_z}{2} \pm (-\frac{1}{2}\chi_n + \frac{1}{2}\chi_p)\frac{g\cos\phi}{\hbar}. \quad (2)$$

Here, $\pm$ refers to reversing the magnetic field direction, χ_n and χ_p are the gyrogravitational ratios of the neutron (from ^{13}C) and proton (from ^{1}H), respectively, g is acceleration due to gravity, and ϕ is the angle between the magnetic field and the Earth's gravitational field [40–42]. We construct the ratio $\mathcal{R}_\pm \equiv \Delta\nu_2(\pm)/\Delta\nu_1(\pm)$. The difference in the ratio obtained by field-reversal is

$$\Delta\mathcal{R} \equiv \mathcal{R}_- - \mathcal{R}_+ \approx \frac{\gamma_h + 3\gamma_c}{\gamma_h + \gamma_c}\left[\frac{(5\chi_p + 4\chi_n)g\cos\phi}{100\mu_N B_z}\right], \quad (3)$$

where μ_N is the nuclear magneton.

Due to the current system configuration, the angle ϕ is fixed at 90° and can not be changed. Thus, the contribution from the Earth's gravitational field is zero and can not be measured directly with our current system. However, we can still reverse the magnetic field direction, and record the variations in $\Delta\mathcal{R}$, which demonstrates the achievable sensitivity for a measurement of the spin-gravity coupling. Considering this, we reverse the magnetic field direction for each measurement scan. Each consecutive $\{+B_z, -B_z\}$ is taken as a group, for which $\Delta\mathcal{R}$ is calculated. We perform 1024 continuous measurements ($\sim$ 15 hours), which are divided into 512 groups of $\{+B_z, -B_z\}$. The measured frequency ratios $\mathcal{R}_\pm$ and the corresponding difference $\Delta\mathcal{R}$ are shown in the Supplemental Material. Based on these measurements, we find that $\Delta\mathcal{R} = (1 \pm 7_{\mathrm{stat}}) \times 10^{-5}$. This uncertainty level indicates that for the current system, $(5\chi_p + 4\chi_n)$ could be measured at a level of 10^{-32} g cm, which probes the spin-dependent gravitational energy of a linear combination of the proton and neutron at a level of 10^{-17} eV. This is comparable to the most stringent existing constraint on the spin-gravity coupling of protons [24].

The measurement uncertainty for the current system is statistics-limited based on the signal-to-noise ratio of a single scan ($\sim$ 100). If instead of thermal polarization using a permanent magnet, hyperpolarization methods such as PHIP (parahydrogen-induced polarization) are employed [43–45], it is possible to achieve more than a 10^4 enhancement of the polarization. This will enable a search for spin-gravity couplings of nuclei several orders of magnitude more sensitive than existing limits. We can also take advantage of high-sensitivity commercial atomic magnetometers, such as those from QuSpin Inc., which could make a new single-species comagnetometer more compact and easier to rotate. Another advantage for this comagnetometer is that, by using different kinds of molecular samples, one can realize comagnetometers to search for spin-gravity couplings using various combinations of protons and neutrons.

In conclusion, we have demonstrated a new single-species liquid-state nuclear spin comagnetometer. We have shown experimentally that the magnetic field gradient-induced systematic effects are significantly suppressed with a single-species comagnetometer as compared to a comagnetometer based on overlapping ensembles of different species. We have introduced a proof-of-principle experiment for a spin-gravity coupling measurement. Based on the current sensitivity, our system is already comparable to the most sensitive system for measuring the coupling of proton spins with Earth's gravitational field. We have outlined the next steps for improving our comagnetometer based on PHIP and compact atomic magnetometers. These improvements could facilitate the development of low-cost, high-precision, and robust table-top systems for long-term measurements of exotic spin-dependent interactions [5].

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