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Observation of the Nuclear Barnett Effect

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We have made the first observation of the nuclear Barnett effect. In the *electronic* Barnett effect, which was first observed in 1915 by Samuel Barnett, a ferromagnetic rod was spun about its long axis and a magnetization developed in the rod along the axis of rotation. This effect is caused by the coupling between the angular momentum of the electronic spins in the sample and the rotational motion of the rod. In our experiment, we measured the *nuclear* Barnett effect by rotating a sample of water at rotational speeds up to 13.5 kHz in a weak magnetic field and observed a change in the polarization of the protons in the sample that is proportional to the frequency of rotation. We measured this polarization by observing the change in the size of a nuclear magnetic resonance (NMR) signal. No NMR frequency shift was observed due to rotation, meaning that this magnetization was not produced by a real magnetic field.

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The coupling of mechanical rotation and magnetization was first proposed by Owen Richardson in 1907 [1] and was experimentally demonstrated by Einstein and de Haas in 1915 [2,3]. The Einstein-de Haas experiment showed that magnetizing a ferromagnetic object causes the transfer of mechanical orbital angular momentum to the object. Almost at the same time, Barnett discovered the reverse effect [4–6]. He found that rotating a ferromagnetic object at frequency $\vec{\omega}$ in the *absence* of a magnetic field produces the same magnetization as when the object is *not* rotating but in the presence of a magnetic field $\vec{B}_{\mathrm{eff}} = \vec{\omega}/\gamma$, where γ is the gyromagnetic ratio. (The electronic Barnett effect has been recently observed in paramagnetic materials [7]).

Both experiments measured the gyromagnetic ratio of the electron. The classical theory of the gyromagnetic effect, first presented by Richardson [1], predicts a value for the electron gyromagnetic ratio that is smaller than the experimental measurements by a factor of 2. We now know from precise measurements [8] and the Dirac equation that the g factor of the electron is approximately equal to 2.

In this Letter, we present an experimental measurement of the *nuclear* Barnett effect. By spinning a sample of water at rotation rates up to 13.5 kHz, we measured a change in the polarization of the hydrogen nuclei (i.e., protons) that is proportional to the rotation frequency. The size of this polarization was consistent with theoretical predictions.

Consider an object with moment of inertia I, rotating with angular velocity $\vec{\omega}$. The energy of the system is

$$E = \frac{1}{2} I\omega^2 = \frac{L^2}{2I}, \tag{1}$$

where $\vec{L}$ is the mechanical angular momentum of the object. The change in the energy of the system caused by a small change in the angular momentum is

$$\Delta E = \frac{L}{I}\Delta L = \omega \Delta L. \tag{2}$$

Now, consider a single spin-1/2 particle in the system ($s = \hbar/2$). Flipping the particle from a direction along $\vec{\omega}$ to a direction opposite to $\vec{\omega}$ results in a change in the spin of the particle equal to $-\hbar$. By conservation of angular momentum ($\Delta S + \Delta L = 0$), the mechanical angular momentum of the object changes by $+\hbar$, and consequently results in a change in the mechanical energy of the system by $+\hbar\omega$. If the system is thermally isolated, the internal energy of the system would change by the same value. Considering the object as a thermal reservoir at temperature T, we can then say that the relative probability of finding a spin along the direction of rotation vs opposite to the direction of rotation is

$$P_{\mathrm{along}}/P_{\mathrm{opposite}} = e^{\Delta E/k_B T} = e^{\hbar\omega/k_B T}. \tag{3}$$

Using the fact that $P_{\mathrm{along}} + P_{\mathrm{opposite}} = 1$, one finds that the average magnetization of a single particle is

$$\langle m \rangle = m_0 \tanh\left(\frac{\hbar\omega}{2k_B T}\right), \tag{4}$$

where m_0 is the magnetic dipole of a single particle. If the system contains N similar particles per unit volume, the total magnetization of the object is

$$M = N\langle m \rangle = M_0 \tanh\left(\frac{\hbar\omega}{2k_B T}\right), \tag{5}$$

where M_0 is the magnetization of the bulk object if all the magnetic dipoles were aligned. At room temperature and with realistic values of rotational frequency ($f = \omega/2\pi < 100$ kHz), $\hbar\omega/2k_B T$ is much smaller than one, and the Barnett magnetization is

$$M = M_0 \frac{\hbar\omega}{2k_B T}. \tag{6}$$

Now, consider the same object, in the absence of bulk rotation, but in the presence of a background magnetic field $\vec{B}_0$. The eigenvalues of the energy associated with a single spin-1/2 particle are given by

$$E = -m(\gamma\hbar B_0) = -m\hbar\omega_0 \qquad m = \pm\frac{1}{2}, \tag{7}$$

where $\vec{\omega}_0 = \gamma\vec{B}_0$. Now in a similar scenario that a particle flips its direction, the energy of the particle changes by $\Delta E = \hbar\omega_0$. From here on, the same discussion of equilibrium statistical mechanics as presented above will lead to the same magnetization as Eq. (6) where ω is replaced by ω_0. In the presence of both bulk rotation and external magnetic field, and if the magnetic field and the axis of rotation are parallel [9], these two effects (magnetization due to rotation and magnetization due to magnetic field) just add.

Even though the above argument proves that rotation can create a magnetization equal to the magnetization created by a magnetic field $\vec{B}_{\text{eff}}$, we must emphasize that the magnetic field $\vec{B}_{\text{eff}}$ does not really exist in the rotating sample. The relationship between the rotating frame of reference and the magnetic field has been discussed in great detail by Heims and Jaynes [10] and we refer the interested reader to this article for a thorough discussion.

Equation (3) tells us that the relative average number of particles aligned with the direction of rotation is independent of the type of particle and is a function only of angular frequency and temperature. Therefore, the size of the Barnett magnetization for every ensemble of particles depends only on the magnetic moment of the specific particle. Quantum mechanics gives the following equation for the gyromagnetic ratio

$$\gamma = g\frac{q}{2m}, \tag{8}$$

where m is the mass of the particle, q is its charge, and g is the g factor. Because the mass of the electron is much smaller than the proton mass, its gyromagnetic ratio, and consequently its magnetic moment, is much larger than that of the proton [11]

$$\mu_e = 9.3\times10^{-24}\ \mathrm{J}T^{-1} \qquad \mu_p = 1.4\times10^{-26}\ \mathrm{J}T^{-1}, \tag{9}$$

which makes the *electronic* Barnett effect much larger and much easier to detect than the nuclear Barnett effect. This explains why the electronic Barnett effect has been observed more than a century ago but the nuclear Barnett effect has not been detected until now.

Because this experiment explores the effect of rotation on the nuclear polarization of protons in the sample, a natural way to measure this polarization is with proton nuclear magnetic resonance (NMR) [12,13].

In an NMR experiment, a sample of nuclear spins is magnetized in the presence of an external magnetic field $\vec{B}_0$, which is by convention in the direction of the $\hat{z}$ axis. Then by the application of a short radio frequency (RF) pulse of appropriate duration and frequency, the magnetization tilts away from the $\hat{z}$ axis and begins to precess about the $\hat{z}$ axis. The result is a changing magnetic field at the Larmor frequency $\omega_0 = \gamma B_0$. An RF coil around the object picks up a signal at this frequency. The amplitude of the signal is proportional to the magnetization times the frequency.

If we conduct the NMR experiment with a rotating sample (rotation parallel to the magnetic field axis), the total magnetization of the sample originates from both the magnetic field and the Barnett effect ($\vec{M} = \vec{M}_B + \vec{M}_0$ where $\vec{M}_B$ is the Barnett magnetization and $\vec{M}_0$ is the magnetization from the magnetic field). Fixing the magnetic field and changing the rotational frequency of the sample will result in a change in the Barnett magnetization (and the total magnetization) equal to δ_{M_B}

$$\delta_M = \delta_{M_B} = \frac{\Delta M_B}{M} \approx \frac{\Delta M_B}{M_0} = \frac{\Delta B_{\text{eff}}}{B_0} = \frac{\Delta\omega}{\omega_0} = \frac{\Delta f}{f_0}, \tag{10}$$

where Δf is the change in rotational frequency and f_0 is the NMR frequency. In addition, if rotation creates a real magnetic field inside the sample, the NMR spectrum will be shifted by Δf (we previously mentioned that the theoretical explanation suggests that such a magnetic field does not exist, and we will later show that our experimental results support the theoretical prediction. Even though the observed Barnett frequency shift is zero, one should note that in our experiment the RF coil is fixed in the laboratory frame. If the RF coil rotates with the sample, a frequency shift should be observed [14]). The following sections describe how we investigated the nuclear Barnett effect by measuring these changes (more detailed explanation of the experiment is also available [15]).

Choosing the appropriate Larmor frequency.—In an NMR experiment, the signal-to-noise ratio is proportional to the cube of the magnetic field. (On the one hand, the signal amplitude is proportional to the square of the magnetic field and therefore the signal power is proportional to the fourth power of the magnetic field. On the other hand, thermal noise [16] is proportional to bandwidth and therefore proportional to the magnetic field). As such, a larger magnetic field results in a larger signal-to-noise ratio and is therefore favorable. In this experiment, however, we are looking for a small change in the magnitude of the signal which is inversely proportional to the magnetic field [Eq. (10)]. A larger magnetic field makes this change smaller and therefore harder to detect. Many factors can change the NMR signal but the dominant effect is temperature variation. Changing the temperature changes the polarization of the magnets and results in a change in the Larmor frequency. Because the signal is proportional to the square of the Larmor frequency and inversely proportional to the temperature, the relative change of the signal resulting from the temperature change is equal to

$$\delta_S \equiv \frac{\Delta S}{S} = \frac{2\Delta f}{f} - \frac{\Delta T}{T}, \tag{11}$$

where S is the signal amplitude. In fact, the sensitivity of the NMR pulse (given by Eq. (11) is a function of temperature and varies by time. We will show that, under the conditions of our experiment, this sensitivity is high enough to detect the Barnett effect.

Choosing the appropriate material.—The Bloch equations [17] tell us that the transverse NMR magnetization (and consequently the signal) decays by the spin-spin relaxation time constant, T_2. To be able to collect enough data, we should choose a material with a large T_2. For this purpose, liquids are better candidates. In solids, because of the strong dipolar and quadrupole interaction of the adjacent molecules, T_2 is very short (typically on the order of a few microseconds). In liquids, however, the tumbling motion [18] of the molecules effectively eliminates the dipolar interaction and increases T_2 considerably (on the order of milliseconds to seconds). In addition, to reduce the statistical noise we should be able to repeat the experiment and average over time. We can repeat the experiment with a rate inversely proportional to the spin-lattice relaxation time constant, T_1. Therefore, a smaller T_1 is preferred. For solids this time is normally larger than for liquids. In liquids, it is possible to change the relaxation time constants by adding paramagnetic materials or by changing the viscosity. For example, adding copper sulfate to water will decrease the value of T_1 by a large factor [19].

Choosing the appropriate sample size.—To maximize the effect, we need the fastest possible spinning rate. The drawback is that the faster the sample spins, the smaller the size of the sample must be. A smaller sample produces a smaller signal, decreasing the signal-to-noise ratio.

Choosing the appropriate time parameters.—The longer duration of the experiment results in a higher signal-to-noise ratio. However, a long experiment increases the risk of introducing large systematic errors. In addition to the total duration of the experiment, our simulations and measurements show that in a Carr-Purcell-Meiboom-Gill (CPMG) echo experiment (the technique that we used for this experiment), in the presence of magnetic field inhomogeneity, the signal might become unstable if $f\tau = (k + 1/2)$, where τ is the time between two consecutive echoes and k is an integer (at least for very small values of k). To get a stable signal we set the rotational frequency close to k/τ. This helps us get a more stable signal, which results in fewer repetition of the experiment to eliminate the statistical noise.

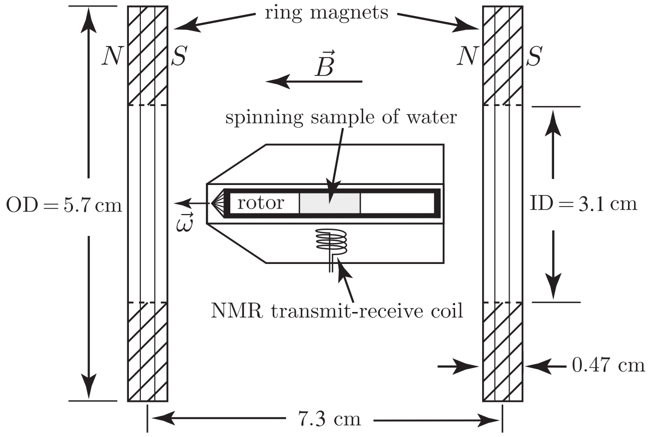

FIG. 1. Diagram of the experiment. Outer diameter (OD); inner diameter (ID).

Considering these limitations, we carefully selected the different parameters of the experiment. We used neodymium ring magnets to create a magnetic field equal to $B_0 =$ 208 Gauss, which is equivalent to a Larmor frequency of around $f_0 = 885$ kHz for hydrogen atoms. (We will later discuss that we fixed the rotational frequency and reversed the orientation of magnetic field to measure the Barnett effect. The exact value of B_0 is not important as long as its absolute value stays the same when we reverse the magnetic field.) To spin the sample, we used a commercial spinner turbine (typically used in solid state NMR [20], spinning at the magic angle [21,22] to quench the dipolar and quadrupole interactions and make a narrower line width) built by Revolution NMR, [23] with a spinning rate up to $f_{\max} =$ 15 kHz. In our experiment, the axis of rotation was *parallel* to the magnetic field. The sample holder had an internal diameter of 2 mm, and the length of the sample was 8 mm (please note that the effective sizes of the sample that created the signal were smaller than these numbers). The sample was water, doped with 2.5 m*M* copper sulfate to reduce the spin-lattice relaxation time (the estimated value for the spin-lattice relaxation time is 400–500 ms [19]). We used the CPMG echo add technique [24,25], in which a train of spin echoes in a CPMG sequence are added together to maximize the signal-to-noise ratio, and looked at the Fourier component of the signal at the Larmor frequency as an indicator of the magnetization. Each scan consisted of 300 echoes (1.1 ms apart), and the acquisition time for each echo was set to 1.024 ms (when the sample rotates, each train of echoes starts from a random position of the sample). The duration of the RF pulse was 8.5 μs, the repetition time of the experiment was 1500 ms, and the number of scans for a single experiment was 600. Figure 1 is a diagram of the experiment.

Suppose that the magnetic field changes over time with a standard error σ_{B_0} (due to the temperature change σ_T), which is equivalent to $\sigma_{f_0} = \gamma\sigma_{B_0}$ for the standard error of the NMR frequency. As mentioned earlier, we are measuring the

Fourier component of the signal at the resonance frequency. Two different errors occur due to the change in the signal frequency. The first error is due to the change in the amplitude of the signal. Equation (11) tells us that the relative change of the signal amplitude is approximately

$$\delta_S = \frac{2\sigma_{f_0}}{f_0} - \frac{\sigma_T}{T}. \tag{12}$$

The second source of error is that changing the frequency of the signal changes the value of its Fourier components. To estimate this change, suppose that we have a very low frequency signal in the form of $S = A_0 \exp(i\Omega t)$, where $\Omega t \ll 1$ for all t. The dominant component of the Fourier transform is the dc component (other components are approximately zero). A simple calculation shows that the amplitude of the dc component of the signal is almost equal to $A_0[1 - (\Omega T_{\mathrm{acq}})^2/24]$, where T_{acq} is the acquisition time. Therefore, we can conclude that the relative change in the amplitude of the Fourier component of the signal at the Larmor frequency is approximately

$$\delta_A = -\frac{(\sigma_\Omega T_{\mathrm{acq}})^2}{24} = -\frac{(2\pi\sigma_{f_0} T_{\mathrm{acq}})^2}{24}. \tag{13}$$

Under the conditions of our experiment, $\delta_A \gg \delta_S$, and therefore the error in measurement of the magnetization is equal to

$$\delta_M \approx \delta_A. \tag{14}$$

A simple calculation shows that if $\sigma_{f_0} < 55$ Hz then $\delta_M < 0.5\%$ which is accurate enough for our purpose. Therefore, we needed to ensure that σ_{B_0} is not much larger 13 mG. In addition, we needed to ensure that there was no shift in the magnetic field larger than 13 mG after reversing the direction of the magnetic field (this sets the upper limit for the accepted systematic error).

Spinning a liquid changes its temperature, causing a change in spin-lattice and spin-spin relaxation times of the sample. In addition, rotation can change the bandwidth of the signal. All of these effects might change the Fourier component of the signal at the Larmor frequency. Therefore, comparing the signals with vs without spinning is not informative. To control all the systematic errors, we compared the signals when the sample was spinning with a constant rate, along vs opposite to the direction of the magnetic field. For this purpose, we rotated the sample with a constant frequency and measured the signal. Then, we reversed the position of the magnets and measured the signal again. We had to make sure that reversing the magnets did not introduce any systematic error into the experiment. Such an error could be added by changing the absolute value of the field at the sample position or by changing the gradient of the field. The change in the absolute value of the magnetic field could be controlled by measuring the magnetic field.

TABLE I. Relative change in magnetization of the sample as a function of rotation frequency f.

f (kHz)	Number of repetitions	δ_{M_B} (theory)	δ_{M_B} (experiment)	Frequency shift
0.0	8	0%	$(-0.08 \pm 0.12)\%$	No
4.5	8	1%	$(1.18 \pm 0.34)\%$	No
9.0	8	2%	$(1.83 \pm 0.28)\%$	No
13.5	4	3%	$(3.21 \pm 0.30)\%$	No

In addition, the CPMG echo experiment and fast rotation of the sample can largely eliminate the effect of magnetic field inhomogeneity. However, in order to completely control the systematic error (we set the maximum systematic error at 0.5% for this experiment) due to the change in the magnetic field direction, we conducted the experiment at zero rotational frequency. Based on the Barnett theory, in this experiment the relative change of the signal after changing the direction of the magnetic field [Eq. (10)] will be $\delta_{M_B} = 2f/f_0$. We repeated the experiment for four different spin rates equivalent to $\delta_{M_B} = 0\%$, 1%, 2%, and 3% (as previously discussed, 0 kHz was conducted to control the error of reversing the field). We repeated each experiment and took the average to decrease the statistical error. We also measured the frequency shift by changing f, and we were not able to detect any shift within the range of experimental error (the sensitivity of frequency measurement is at least equal to $1/T_{\mathrm{acq}} \approx 1$ kHz.)

Table I and Fig. 2 are the first experimental proof of the nuclear Barnett effect, showing that spinning an object can create a nuclear magnetization proportional to the spin rate. The resulting magnetization is the same as the induced magnetization obtained by putting the object in an equivalent magnetic field $\vec{B}_{\mathrm{eff}} = \vec{\omega}/\gamma$. However, this equivalent field does not actually exist in the sample. There is no

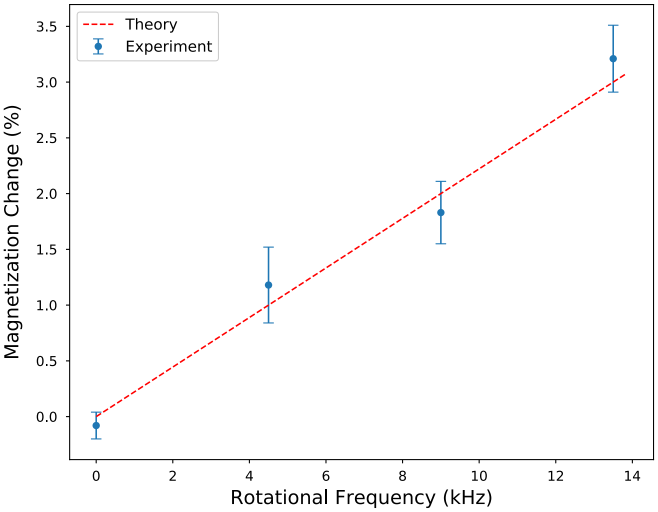

FIG. 2. Signal change vs rotation.

observable shift in the NMR spectrum up to the highest spin rates measured (13.5 kHz).

This work proves the existence of the nuclear Barnett effect in liquids, more than a century after the discovery of the electronic Barnett effect. Even though there was no assumption about the phase of the sample, complementary experiments would need to be done for solids. The difficulty would be short spin-spin and long spin-lattice relaxation times, which would require much longer experiments, leading to the possibility of large systematic errors.

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