Coherence in Freely Precessing ^{21}Ne and a Test of Linearity of Quantum Mechanics

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We have observed coherence among the four magnetic sublevels of freely precessing ^{21}Ne for times of 4.5 h. This has allowed direct detection of the quadrupole component of coherence relaxation and a test of linearity of quantum mechanics. The quadrupole component of relaxation is evident in the different relaxation rates for the three lines of the NMR spectrum. Nonlinear corrections to quantum mechanics are found to be less than 1.6×10^{-26} of the binding energy per nucleon of ^{21}Ne.

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The coherence among the magnetic projections of a system with angular momentum provides a fundamental demonstration of the principle of linear superposition that underlies quantum mechanics. For a spin-$\frac{3}{2}$ system such as ^{21}Ne this coherence can be demonstrated for an ensemble of spins by measuring the component of magnetization transverse to a static magnetic field during free precession. Since the magnetization is due to the ensemble average, a coherent superposition of amplitudes, the time dependence of the transverse magnetization shows that the relative phases of the amplitudes for the magnetic projections evolve at different rates. For ^{21}Ne the result is a spectrum of three frequencies with a time dependence such as that shown in Fig. 1 which illustrates the beat pattern and coherence over a time interval of 4.5 h. (It is important to note that, in contrast to spin 1 or greater, the magnetization in a two-state, spin-$\frac{1}{2}$, system has a purely classical description provided by the Bloch equations.) However, any test of a fundamental principle such as linear superposition is best interpreted within some framework that allows for violation of that principle. Such a framework could be provided by a specific theory or by a generalization, such as that provided by Weinberg[1,2] to include nonlinearity as a correction to quantum mechanics.

Weinberg's approach provides for the time dependence of a wave function to be described by an equation of the Hamiltonian form [with $q_k=(\psi_k+\psi_k^*)/\sqrt{2}$ and $p_k=(\psi_k-\psi_k^*)/i\sqrt{2}$]

$$i\hbar\, d\psi_k/dt = dh(\psi,\psi^*)/d\psi_k^*\,, \tag{1}$$

where $h(\psi,\psi_k^*)$ is a real function satisfying the requirement of homogeneity but not necessarily linearity. Equation (1) becomes the Schrödinger equation when

$$h(\psi,\psi^*)=\sum_{i,j}\psi_i^* H_{ij}\psi_j\,. \tag{2}$$

Small nonlinear corrections appear as additional terms on the right-hand side of Eq. (2) of the form $h'(a)$, where a is a function of the amplitudes ψ_j^2. The nonlinearity is therefore manifest in the dependence of the time evolution of ψ_k on the amplitudes ψ_i.

For ^{21}Ne, a spin-$\frac{3}{2}$ system, the nonlinear Hamiltonian function of lowest degree that satisfies rotational invariance consists of a product of two ψ's and two ψ^*'s, though it is possible to construct terms of higher order.[2] The time dependence of each of the four states labeled by magnetic quantum number $m_I=\frac{3}{2}$, $\frac{1}{2}$, $-\frac{1}{2}$, and $-\frac{3}{2}$ will exhibit the oscillatory behavior expected in quantum mechanics with the frequencies dependent on the probability amplitudes ψ_{m_I}. The result of time averaging then provides the frequency shifts due to nonlinear corrections which can be derived from Eq. 4.42 of Ref. 2. They are

$$\begin{aligned}
\hbar\delta\omega_{3/2}&=\epsilon(6|\psi_{-1/2}|^2+9|\psi_{-3/2}|^2)\,,\\
\hbar\delta\omega_{1/2}&=\epsilon(4|\psi_{1/2}|^2+|\psi_{-1/2}|^2+6|\psi_{-3/2}|^2)\,,\\
\hbar\delta\omega_{-1/2}&=\epsilon(6|\psi_{3/2}|^2+|\psi_{1/2}|^2+4|\psi_{-1/2}|^2)\,,\\
\hbar\delta\omega_{-3/2}&=\epsilon(9|\psi_{3/2}|^2+6|\psi_{1/2}|^2)\,,
\end{aligned} \tag{3}$$

where ϵ defines the size of the nonlinear correction. We assume that $|\psi_{-3/2}|^2+|\psi_{-1/2}|^2+|\psi_{1/2}|^2+|\psi_{3/2}|^2=1$ and rewrite Eq. (3) in terms of the orthogonal tensor multipole moments of the polarization and alignment. The terms dependent on the vector polarization

$$\langle K_z\rangle=\tfrac{3}{2}|\psi_{3/2}|^2+\tfrac{1}{2}|\psi_{1/2}|^2-\tfrac{1}{2}|\psi_{-1/2}|^2-\tfrac{3}{2}|\psi_{-3/2}|^2$$

and the tensor polarization or alignment

$$T_2=|\psi_{3/2}|^2-|\psi_{1/2}|^2-|\psi_{-1/2}|^2+|\psi_{-3/2}|^2$$

are

$$\begin{aligned}
\tfrac{5}{42}\hbar\delta\omega_1+\tfrac{8}{42}\hbar\delta\omega_2+\tfrac{5}{42}\hbar\delta\omega_3&=-\epsilon\langle K_z\rangle\,,\\
\hbar\delta\omega_1-\hbar\delta\omega_3&=\epsilon(2+T_2)\,,
\end{aligned} \tag{4}$$

where $\delta\omega_1$, $\delta\omega_2$, and $\delta\omega_3$ are the corrections to the coherence frequencies $\omega_{1,2,3}$ defined in Ref. 3. In particular, $\omega_1-\omega_3=\omega_Q$, the quadrupole component of the frequency splittings. Our test of linearity of quantum mechanics is the search for the linear dependence of ω_Q on the tensor polarization T_2 given by Eq. (4). The dipole component of the frequency splittings is prone to spin-exchange shifts and changes of the net magnetic field due to changing magnetization within the nonspherical cell and instability of the applied field.

The apparatus used for this measurement has been developed for precision frequency spectroscopy of freely

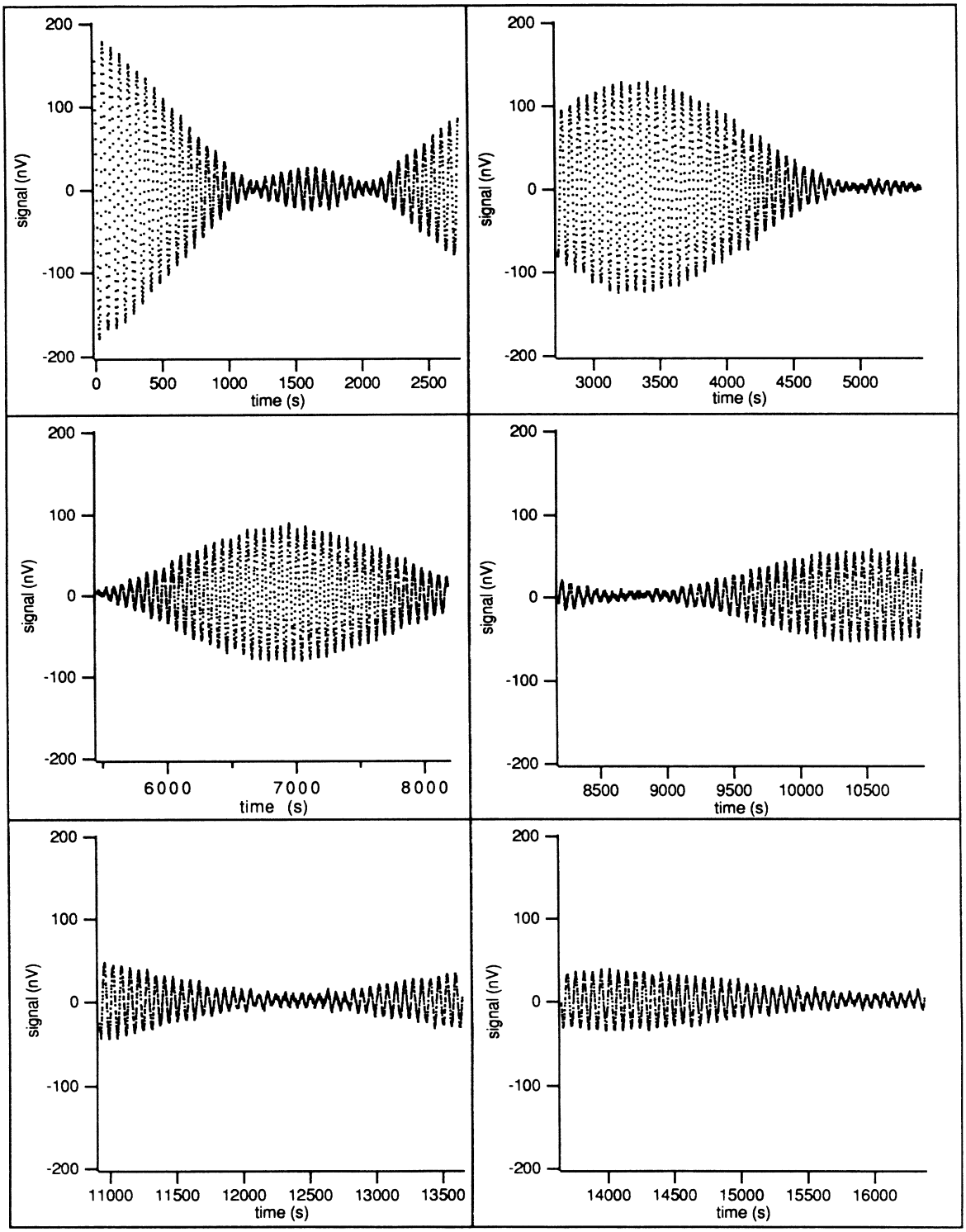

FIG. 1. Data for coherent free precession of ^{21}Ne over 4.5 h. Each panel represents a 45-min measurement of the beat frequency between the ^{21}Ne signals and a reference frequency derived from freely precessing ^{3}He which is divided by 9.469. ^{3}He is not coherent from panel to panel; ^{21}Ne is.

precessing polarized noble-gas nuclei and is described in detail elsewhere.[3,4] A brief description is provided here. The technique employs mixtures of noble-gas species, in this case ^{21}Ne and ^{3}He, in order to compare the free-precession frequencies and monitor systematic effects such as magnetic-field fluctuations. Because of the requirement of rotational

invariance, ^{3}He is not sensitive to the nonlinear corrections we seek.[1,2] ^{21}Ne and ^{3}He (about 2×10^{19} atoms of each) are contained in a sealed glass cell of 1 cm^3 volume and are polarized along a static 3-G magnetic field by spin exchange with a laser-optically-pumped Rb vapor.[5,6] The optical pumping light is supplied by a GaAlAs-diode-laser array.[7] Initially the polarization is built up over a period of several hours with the cell heated to about 150°C to maintain a sufficiently high Rb vapor. The cell is then cooled to eliminate the Rb vapor which would otherwise cause relaxation and frequency shifts due to spin-exchange collisions. A measurement is started by tipping the ^{21}Ne spins with respect to the static-magnetic-field axis and monitoring the transverse magnetization with the voltage induced in a pickup coil. The tipping angle is 20°, provided by a pulse of a resonant oscillating magnetic field. The free precession is monitored over a period of 4.5 h by extracting the beat frequency between the ^{21}Ne signals and a reference frequency derived from the freely precessing ^{3}He and divided down in frequency by 9.649, approximately the ratio of the precession frequencies. This greatly mitigates the effects of fluctuations of the static magnetic field which are about 5×10^{-6} G peak to peak. The ^{3}He free-precession coherence time is limited to about 2000 s by magnetic-field-inhomogeneity coupling to the large magnetic moment, and we take separate runs of 2700 s—each with a new pulse of transverse ^{3}He magnetization provided by a 45° resonant pulse. The ^{21}Ne spins remain coherent over the entire 5-h measurement.

The data for ^{21}Ne free precession from one such measurement are shown in Fig. 1. The six panels represent successive 2700-s runs each initiated by a pulse producing transverse ^{3}He magnetization. The carrier frequency of about $\frac{1}{60}$ Hz is the difference of the actual ^{21}Ne precession frequency (about 995 Hz) and the ^{3}He precession frequency (about 9600 Hz) divided by 9.649. The existence of the three coherence frequencies of ^{21}Ne is most apparent in the beat pattern during the time interval from 0 to 2700 s. These frequencies are resolvable due to the interaction of the electric quadrupole moment of the ^{21}Ne nucleus with the cell's wall. This shift would average to zero in a perfectly spherical cell, but our cell has a small asymmetry mostly due to the sidearm through which the cell was filled with the gases. The observation of such coherent wall interactions was first reported by Volk, Mark, and Grover[8] for ^{83}Kr and Kwon, Mark, and Volk[9] for ^{131}Xe. Subsequent work by Wu and co-workers[10,11] predicts that the quadrupole wall interaction leads to relaxation rates for the $\frac{3}{2},\frac{1}{2}$ and $-\frac{1}{2},-\frac{3}{2}$ coherences relative to the $\frac{1}{2},-\frac{1}{2}$ coherence in the ratio 3:2. The washing out of the sharp beat pattern shown in Fig. 1 is due to the difference of these relaxation rates. To our knowledge, this is the first clear demonstration of relaxation of coherence due to quadrupole interactions.

The frequencies and relaxation rates are extracted from the time-domain signals $[S(t)]$ for each 2700-s run by fitting with the model

$$\begin{aligned} S(t) = {} & A_0 + A_1\cos[(\omega_D' + \omega_Q)t + \phi]e^{-R\Gamma t} \\ & + A_2\cos(\omega_D' t + \phi)e^{-\Gamma t} \\ & + A_3\cos[(\omega_D' - \omega_Q)t + \phi]e^{-R\Gamma t}. \end{aligned} \tag{5}$$

Here A_0 accounts for electronics offsets and A_1, A_2, and A_3 are the amplitudes of the three coherences. ω_D' is the carrier frequency of $\approx\frac{1}{60}$ Hz. The octupole contribution to the frequency splittings is negligible and is set equal to zero as indicated in Eq. (5). ϕ is the initial phase difference between the ^{21}Ne magnetization and the divided-down ^{3}He frequency. The ratio of the relaxation rates of the coherences R was extracted from separate analysis of the data. The extracted value is $R=1.59\pm0.03$. A value of $R=1.5$ has been predicted by Wu, Schaeffer, Cates, and Happer for pure quadrupole relaxation.[11] Our result shows that the relaxation is dominated by quadrupole couplings.

The data for the test of linearity of quantum mechanics consist of five separate measurements of ^{21}Ne coherence each divided into six or seven 2700-s intervals. We fixed $R=1.59$ in order to extract ω_Q and the average relaxation rate Γ (from the start of the measurement of ^{21}Ne coherence) for each 2700-s interval. For each ^{21}Ne coherence measurement the initial alignment $T_2(t=0)$ can be determined from the measured vector polarization and ratios of the amplitudes, e.g., $A_1/A_2=4|\psi_{-1/2}|/3|\psi_{3/2}|$. The tensor polarization T_2 has a decay rate 2Γ and can therefore be determined at the beginning of each 2700-s interval.[11] The calibration that relates the vector polarization to A_1, A_2, and A_3 could not be directly determined, and only a lower limit is established from our data. This lower limit of 12%–15% vector polarization at the beginning of each measurement satisfies the requirement that $T_2>0$ as indicated by solution of the rate equations for ^{21}Ne polarization under our conditions.[5] The values of ω_Q for each run were corrected for temperature variations of the cell wall by using the relaxation rates as a measure of the wall sticking time in the manner described in Ref. 3. For each of the five measurements of ^{21}Ne coherence, the data for ω_Q were fitted by a model with a constant plus a term linear in Γ. Another temperature-dependent effect which is not corrected by this analysis is that of geometric changes of the apparatus. ω_Q is proportional to $P_2(\cos\beta)$, where β is the angle of the cell's axis of cylindrical symmetry with respect to the static magnetic field, and temperature changes could change β. The errors assigned to each measurement are based on the residual fluctuations after the temperature corrections are made and vary from 0.5 to 1.5 μHz, which is as much as 3 times larger than the statistical error based on the signal-to-noise ratio for each measurement.

The temperature-corrected data are shown in Fig. 2,

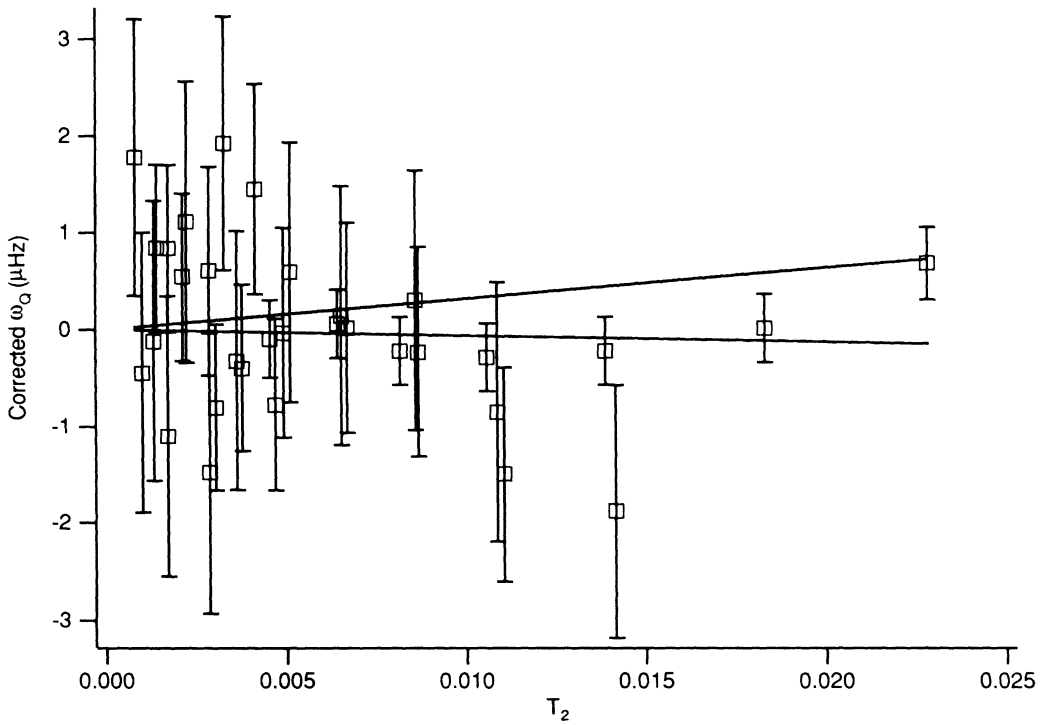

FIG. 2. Data used to set a limit on ϵ defined by Eq. (4). T_2 is the tensor polarization defined in the text. The solid lines indicate the 1σ limits of ϵ.

plotted against the assigned values of T_2. These residuals were fitted by a model linear in T_2 in order to extract ϵ given by Eq. (4). We find that $\epsilon = 12 \pm 19$ μHz. The two extreme 1σ values for ϵ are indicated by the solid lines in Fig. 2. Since the assigned values of T_2 are lower limits, we set an upper limit on ϵ of 31 μHz or 1.3×10^{-19} eV. This should be compared to the binding energy per nucleon of ^{21}Ne, about 8 MeV, which is a measure of the relative size of the nonlinear correction to ordinary quantum mechanics. Our limit on $|\epsilon|$ is therefore less than 1.6×10^{-26} of the binding energy. The tensor polarization T_2 is quite small since it arises only due to quadrupole relaxation in the presence of spin exchange, a dipole interaction that produces dominantly vector polarization. Significantly greater sensitivity to ϵ would result from producing much larger alignment, perhaps with the application of a pulsed quadrupole electric field to initiate the coherence measurement.

Two recent experiments have set limits of about 10 μHz on a quantity analogous to ϵ. These are in the rf spectroscopy of $^9Be^+$ ions[12] and in the hydrogen-maser transition.[13] The $^9Be^+$ experiment provides a limit on nonlinear corrections to the nuclear binding energy 4 times more stringent than our result, though in a different nucleus and with a different observable. The hydrogen-maser experiment provides a limit on the nonlinear corrections to atomic binding energy which is about 6 orders of magnitude less than the nuclear binding energy.

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