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Test of the Linearity of Quantum Mechanics by rf Spectroscopy of the $^9Be^+$ Ground State

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A hyperfine transition in the ground state of $^9Be^+$ was used to test a nonlinear generalization of quantum mechanics recently formulated by Weinberg. We searched for a dependence of the frequency of a coherent superposition of two hyperfine states on the populations of the states. We are able to set a limit of 4×10^{-27} on the fraction of binding energy per nucleon of the $^9Be^+$ nucleus that could be due to nonlinear corrections to quantum mechanics.

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Since the 1920s, quantum mechanics has passed numerous tests as illustrated by the agreement between the predictions of specific quantum-mechanical theories and experimental measurements. For example, the measured energy levels of the hydrogen atom are in excellent agreement with the predictions of quantum-mechanical theory. However, this could be regarded as a test of the accuracy of the Hamiltonian rather than a test of quantum mechanics itself. It should be possible to test the basic framework of quantum mechanics independently of and more precisely than any particular quantum-mechanical theory. Recently, Weinberg[1,2] has formulated a general framework for introducing nonlinear corrections to quantum mechanics which enables such a test. He has suggested that a sensitive way to search for possible nonlinearities is to look for a change in a transition frequency as the wave function of a system changes between initial and final states. Monochromatic radiation used to drive the transition would therefore not stay in resonance throughout the entire transition. If the transition can be driven experimentally in time T, then the maximum frequency shift due to the nonlinearity must be on the order of $1/T$. Because nuclear magnetic resonance transitions in $^9Be^+$ have been observed with T as long as 1 s, Weinberg sets a limit of $\sim 10^{-15}$ eV on the magnitude of any such nonlinear corrections to the energy of the ^9Be nucleus. In this Letter we report an experiment which improves this limit by 5 orders of magnitude.

In the formalism developed in Refs. 1 and 2, the equation which describes the time evolution of the wave function $\psi(t)$ is nonlinear and derivable from a Hamiltonian function $h(\psi,\psi^*)$. For a discrete system, it takes the form

$$i\hbar\frac{d\psi_k}{dt}=\frac{\partial h(\psi,\psi^*)}{\partial\psi_k^*}, \tag{1}$$

where ψ_k is the amplitude of state k. In general, h is not a bilinear function of ψ and ψ^* as in ordinary quantum mechanics, but the property of homogeneity $[h(\lambda\psi,\psi^*)=h(\psi,\lambda\psi^*)=\lambda h(\psi,\psi^*)$ for any complex $\lambda]$ is retained. Homogeneity guarantees that if $\psi(t)$ is a solution of Eq. (1) then $\lambda\psi(t)$ is also a solution representing the same physical state. Homogeneity ensures the proper treatment of physically separated systems and distinguishes this formalism from previous nonlinear generalizations[3,4] and tests[5,6] of quantum mechanics.

Consider a two-level system which in the absence of nonlinear corrections has eigenvalues E_k, $k=1,2$. Because any nonlinear corrections to quantum mechanics are expected to be small, it is reasonable to write the Hamiltonian function as the sum of the bilinear term $h_0(\psi,\psi^*)=\sum_{k=1,2}E_k\psi_k^*\psi_k$ of ordinary quantum mechanics and a term $h_{nl}(\psi,\psi^*)$ that is not bilinear and contains the small nonlinear corrections. A form of h_{nl} appropriate for the work discussed here is $h_{nl}=n\bar{h}(a)$, where $n\equiv|\psi_1|^2+|\psi_2|^2$, $a\equiv|\psi_2|^2/n$, and $\bar{h}$ is a real function.[1,2] The nonlinear time-dependent Schrödinger equation then takes the form

$$i\hbar\frac{d\psi_1}{dt}=\left[E_1+\bar{h}-a\frac{d\bar{h}}{da}\right]\psi_1\equiv\hbar\omega_1(a)\psi_1\,,$$
$$i\hbar\frac{d\psi_2}{dt}=\left[E_2+\bar{h}+(1-a)\frac{d\bar{h}}{da}\right]\psi_2\equiv\hbar\omega_2(a)\psi_2\,,$$

which has solutions

$$\psi_k(t)=c_k e^{-i\omega_k(a)t},\quad k=1,2\,, \tag{2}$$

where a and the c_k's can be parametrized by $c_1=\sin(\theta/2)$ and $c_2=a^{1/2}=\cos(\theta/2)$. The relative phase of the two components of the wave function (specifically, the time dependence of the coherence $\psi_1\psi_2^*$) evolves with a frequency

$$\omega_p\equiv\omega_1(a)-\omega_2(a)=\omega_0-(d\bar{h}/da)/\hbar\,,$$

where $\omega_0=(E_1-E_2)/\hbar$ is the atomic transition frequency in the absence of nonlinearities. A two-level system is mathematically equivalent to a spin-$\frac{1}{2}$ system in an external, uniform magnetic field, where θ is the angle by which the spin is tipped with respect to the magnetic field and ω_p is the precession frequency of the spin about the magnetic field.[7] In the language of the equivalent spin-$\frac{1}{2}$ system, the effect of the nonlinear correction $d\bar{h}/da$ is to create a dependence of the precession frequency ω_p on the tipping angle θ between the spin and the magnetic field.

We searched for a θ dependence of the precession frequency of the $(m_I,m_J)=(-\frac{1}{2},+\frac{1}{2})\to(-\frac{3}{2},+\frac{1}{2})$ hyperfine transition at ~303 MHz in the ground state of $^9Be^+$ (see Fig. 1). At a magnetic field B of 0.8194 T, this transition, referred to as the clock transition, depends only quadratically on magnetic field fluctuations. With $\psi_1\equiv\psi(-\frac{3}{2},+\frac{1}{2})$ and $\psi_2\equiv(-\frac{1}{2},+\frac{1}{2})$ the simplest nonbilinear addition to the Hamiltonian function of the free $^9Be^+$ nucleus for the two states is[1,2]

$$\bar{h}(a)=2\epsilon a^2\,, \tag{3}$$

where ϵ is a measure of the strength of the nonlinear correction. This gives rise to a dependence of ω_p on θ of

$$\omega_p=\omega_0-4(\epsilon/\hbar)\cos^2(\theta/2)\,. \tag{4}$$

This discussion assumes that the $^9Be^+$ nuclear spin is decoupled from the valence electron spin and therefore the $(-\frac{1}{2},\frac{1}{2})$ and $(-\frac{3}{2},\frac{1}{2})$ states are pure (m_I,m_J) states. At a magnetic field of 0.8194 T these states have a 0.02 to 0.03 amplitude admixture of $m_J=-\frac{1}{2}$ states. This creates small corrections to Eqs. (3) and (4) which we neglect.

Between 5000 and 10000 $^9Be^+$ ions and 50000 to 150000 $^{26}Mg^+$ ions were simultaneously stored in a cylindrical Penning trap[8] with $B\simeq0.8194$ T under conditions of high vacuum ($\lesssim10^{-8}$ Pa). To minimize second-order Doppler shifts of the clock transition, the $^9Be^+$ ions were cooled to less than 250 mK. The $^{26}Mg^+$ ions were directly laser cooled and compressed by a narrow-band (~1 MHz) radiation source at 280 nm.[9] The $^9Be^+$ ions were then sympathetically cooled[10] by their Coulomb interaction with the cold Mg^+ ions. A narrow-band 313-nm radiation source was used to optically pump and detect the $^9Be^+$ ions.[11,12] With the 313-nm source tuned to the $2s\,^2S_{1/2}(m_I=\frac{3}{2},m_J=\frac{1}{2})$ to $2p\,^2P_{3/2}(\frac{3}{2},\frac{3}{2})$ transition, 94% of the $^9Be^+$ ions were optically pumped into the $2s\,^2S_{1/2}(\frac{3}{2},\frac{1}{2})$ ground state.[11,12] The 313-nm source was then turned off to avoid optical pumping and ac Stark shifts. The sympathetic cooling of the $^9Be^+$ ions by the Mg^+ ions provided a steady cooling source independent of the 313-nm radiation and therefore permitted the use of long transition times.

The clock transition was detected by the following method. After the 313-nm source was turned off, the ions in the $(\frac{3}{2},\frac{1}{2})$ state were transferred to the $(\frac{1}{2},\frac{1}{2})$ state and then to the $(-\frac{1}{2},\frac{1}{2})$ state by two successive rf π pulses. Each pulse was 0.2 s long and resonant with the appropriate transition frequency (around 321 and 311 MHz, respectively). The clock transition was then driven by Ramsey's method of separated oscillatory fields[13] with rf pulses of about 1-s duration and a free-precession time on the order of 100 s. This transferred some of the ions from the $(-\frac{1}{2},\frac{1}{2})$ state to the $(-\frac{3}{2},\frac{1}{2})$ state. Those ions remaining in the $(-\frac{1}{2},\frac{1}{2})$ state were then transferred back to the $(\frac{3}{2},\frac{1}{2})$ state by reversing the order of the two rf π pulses. The 313-nm source was then turned back on, and the population of ions in the $(-\frac{3}{2},\frac{1}{2})$ state was registered as a decrease in the $^9Be^+$ fluorescence, relative to the steady-state fluorescence, during the first second that the 313-nm source was on. [The optical repumping time of the ions from the $(-\frac{3}{2},\frac{1}{2})$ state to the $(\frac{3}{2},\frac{1}{2})$ state was an or-

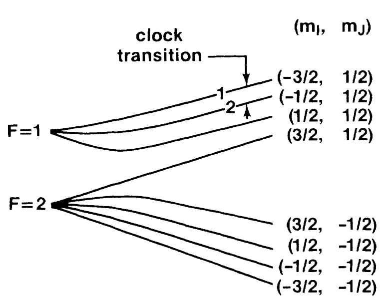

FIG. 1. Hyperfine energy levels (not drawn to scale) of the $^9Be^+$ $2s\,^2S_{1/2}$ ground state as a function of magnetic field. At $B=0.8194$ T the 303-MHz clock transition is independent of magnetic field to first order.

der of magnitude longer than this.]

For the test of nonlinearities, Ramsey's method with unequal rf pulses was used to drive the clock transition and measure ω_p for different values of θ. First an rf θ pulse of duration τ_θ was applied. This prepared the ions into a coherent superposition of the $(-\frac{1}{2},\frac{1}{2})$ and $(-\frac{3}{2},\frac{1}{2})$ states given by Eq. (2) for a particular value of θ. The value of θ was determined from $\theta=(\tau_\theta,\tau_\pi)\pi$, where τ_π was the length of time to drive a π pulse at the same rf power. After the rf θ pulse, the ions freely precessed for a time T. This was followed by an rf $\pi/2$ pulse coherent with the first pulse which completed the Ramsey excitation. In the limit that $T\gg\tau_\theta,\tau_{\pi/2}$, the Ramsey line shape [specifically, the number of ions remaining in the $(-\frac{1}{2},\frac{1}{2})$ state as a function of the rf frequency ω in the Ramsey excitation] is proportional to

$$1-\sin\theta\cos\{[\omega-\omega_p(\theta)]T\},$$

where $\omega_p(\theta)$ is given by Eq. (4). The center frequency of the Ramsey line shape is the precession frequency $\omega_p(\theta)$. Figure 2 shows a Ramsey signal obtained with $T=150$ s and $\theta=\pi/2$.

The Ramsey signal was used to steer the frequency of a synthesized rf source.[11] Ramsey-signal measurements were taken near both of the full-width-at-half-maximum frequencies $\omega_+\equiv2\pi\nu_+$ and $\omega_-\equiv2\pi\nu_-$, where ν_+ and ν_- are indicated in Fig. 2. The difference in the measured signal strengths on either side of the line center was used to electronically steer the average frequency of the synthesizer to $\omega_p(\theta)$. Eight pairs of measurements were taken with an angle $\theta_A=1.02$ rad followed by eight pairs of measurements with an angle $\theta_B=2.12$ rad. This pattern was repeated for the length of an entire run as indicated in Fig. 3. The average frequency of the synthesizer for $\theta=\theta_A$ was then subtracted from the average frequency of the synthesizer for $\theta=\theta_B$. Run lengths varied between 4 and 10 h. The uncertainty was due to the frequency instability of the reference oscillator used with the synthesizer. Most runs were taken with a commercial cesium beam clock [fractional frequency stability[14] $\sigma_y(\tau)\sim6\times10^{-12}\tau^{-1/2}$ for measurement time τ in seconds] as the reference oscillator. For a limited time we had access to a passive hydrogen maser[15] [$\sigma_y(\tau)\sim(2\text{-}3)\times10^{-12}\tau^{-1/2}$] and a few runs were taken with the passive hydrogen maser as the reference oscillator.

Runs were taken with free-precession periods of $T=30$, 60, and 100 s and rf pulse lengths of $\tau_A=0.65\tau_{\pi/2}$, $\tau_B=1.35\tau_{\pi/2}$ with $\tau_{\pi/2}=0.5$, 1, and 2 s. A weighted average of the synthesizer frequency differences for $\theta=\theta_A$ and $\theta=\theta_B$ from 25 runs is 2.7(6.0) μHz. The uncertainty (in parentheses) is the external error calculated from the scatter of the 25 measurements from the weighted average and is in good agreement with the internal error of 5.7 μHz calculated from the uncertainties of each of the 25 runs. The time constant of the servo would have decreased the apparent size of a real frequency difference by 28%. This results in a possible dependence of the precession frequency on θ of $[\omega_p(\theta_B)-\omega_p(\theta_A)]/2\pi=3.8(8.3)$ μHz and from Eq. (4) a value for the parameter ϵ of

$$\epsilon/2\pi\hbar=1.8(4.0)\ \mu\text{Hz}. \tag{5}$$

The error is a 1 standard deviation uncertainty. A few runs were also taken with $\theta=\pi/2$. The frequency $\omega_p(\pi/2)/2\pi$ was compared with the frequencies $\omega_p(\theta_A)/2\pi$ and $\omega_p(\theta_B)/2\pi$ for runs taken within a few days of each other. The standard deviation of the frequencies from their average was 6 μHz, consistent with the 7-μHz uncertainty of the frequencies. The $\theta=\pi/2$ runs do not improve the limit of Eq. (5) on a possible correction to ω_p linear in a, but in general can be used to help place limits

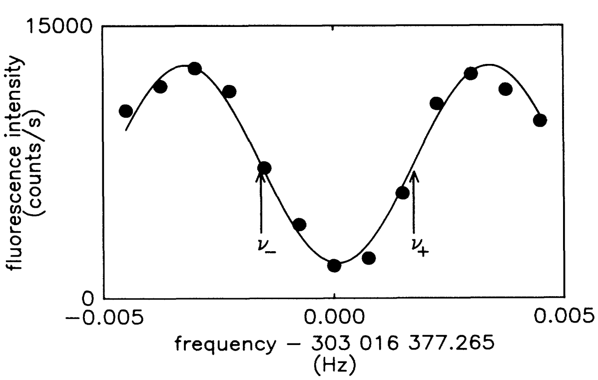

FIG. 2. Ramsey signal of the clock transition with $T=150$ s and $\theta=\pi/2$. The data are the result of one sweep (that is, one measurement per point). The sweep width is 9 mHz and the frequency interval between points is 0.750 mHz. The dots are experimental and the curve is a least-squares fit. The signal-to-noise ratio is limited by the frequency stability of the reference oscillator. The full-width-at-half-maximum frequencies are indicated by ν_+ and ν_-.

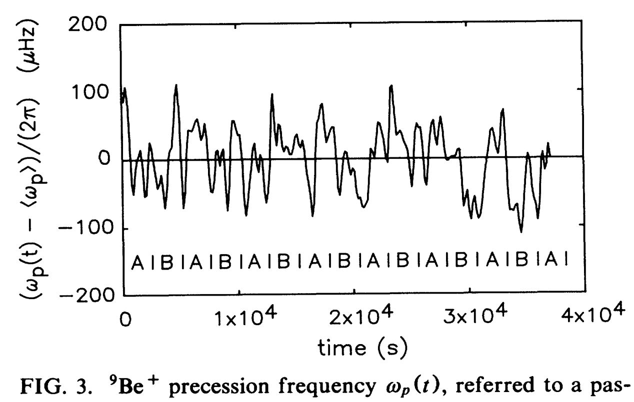

FIG. 3. $^9Be^+$ precession frequency $\omega_p(t)$, referred to a passive hydrogen maser, as a function of time for a single run with $T=100$ s. The periods A (B) during which the initial Rabi pulse created a mixed state with angle $\theta_A=1.02$ rad ($\theta_B=2.12$ rad) are indicated.

on a more complicated form for $\bar{h}(a)$.

Equation (5) sets an upper limit of $|\epsilon| < 2.4\times 10^{-20}$ eV (5.8 μHz) for a nonlinear contribution to the $^9Be^+$ nuclear Hamiltonian. This is less than 4 parts in 10^{27} of the binding energy per nucleon of the $^9Be^+$ nucleus and improves the limit set in Ref. 1 by roughly 5 orders of magnitude. The limit on $|\epsilon|$ is also 5 orders of magnitude smaller than experimental limits placed by neutron interferometry[5,6] on $|b|$, where b is the coefficient of a logarithmic addition $-b\psi(\mathbf{x})\ln|\psi(\mathbf{x})|^2$ to the one-particle Schrödinger equation. However, this nonlinearity does not satisfy the property of homogeneity and therefore these experiments[5,6] test for a nonlinearity which does not satisfy the requirements of the framework developed by Weinberg. Our experimental result is limited by statistics due to the frequency instability of the reference oscillator. The largest known systematic error of our measurement of $\omega_p(\theta)$ is the second-order Doppler (time dilation) frequency shift due to the temperature and $\mathbf{E}\times\mathbf{B}$ rotation of the ions in the trap.[11] Its size is less than 3 μHz (1×10^{-14}). We believe it can be held constant to significantly better than 10% over the time required to make a frequency difference measurement. With a better reference oscillator or a second $^9Be^+$ clock, it should therefore be possible to improve our limit on $|\epsilon|$ by more than an order of magnitude. Improvements on this measurement may also be possible using nuclear magnetic resonance techniques on neutral atoms.[16,17]

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