ATOMIC PHYSICS TESTS OF NONLINEAR QUANTUM MECHANICS*

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ABSTRACT

Atomic physics experiments which test a nonlinear generalization of quantum mechanics recently formulated by Weinberg are described. The experiments search for a dependence of hyperfine transition frequencies or nuclear spin precession frequencies on the relative populations of the hyperfine or nuclear spin states. The experiments set limits less than 10 μHz on the size of the possible nonlinear contributions to these frequencies. In some cases this can be interpreted as a limit of less than ~ 10^{-26} on the fraction of binding energy per nucleon that could be due to a nonlinear correction to a nuclear Hamiltonian. The possibility that a nonlinear addition to quantum mechanics violates causality is discussed.

INTRODUCTION

Quantum mechanics has survived numerous tests since its inception in the 1920's. Many of these tests can be regarded as tests of a specific quantum-mechanical theory as well as tests of the basic formalism of quantum mechanics. For example, the fact that the measured energy levels of the hydrogen atom are in agreement with the predictions of quantum-mechanical theory is a test of both the formalism of quantum mechanics and the accuracy of the Hamiltonian for the hydrogen atom. However, this only tests quantum mechanics at the level we know the Hamiltonian is accurate. Because of its importance in modern physics, quantum mechanics should be tested independently of and, if possible, more precisely than any particular quantum-mechanical theory. Surprisingly few such tests exist. Some examples are the experiments [1] which have conclusively ruled out hidden variable theories in favor of quantum mechanics. These experiments, however, do not provide a test of quantum mechanics itself to much less than 1%.

The lack of precise, independent tests of quantum mechanics is at least partially due to the absence of generalized versions of quantum mechanics with which to plan or interpret an experimental test. Generalized versions of quantum mechanics which have a logically consistent interpretation have been difficult to construct. Recently, however, Weinberg has developed a formalism for introducing nonlinear corrections to quantum mechanics.[2,3] The size of the nonlinear corrections is determined by a parameter ϵ. In the limit that $\epsilon = 0$, Weinberg's nonlinear generalization reduces to ordinary

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linear quantum mechanics. This paper includes a discussion of atomic physics experiments[4-7] which search for nonlinear corrections permitted by Weinberg's formalism. The experiments completed to date have found null results which put upper bounds on the parameter ϵ. In some cases these bounds are 26 orders of magnitude smaller than the size of the ordinary, linear quantum-mechanical terms.

The following simple discussion gives an idea of how the atomic physics tests work. Consider a two-level system with energy eigenstates $|k\rangle$, k = 1,2 and energies $\hbar\omega_k$, with $\omega_1 > \omega_2$. The time evolution of these eigenstates is given by $e^{-i\omega_k t}|k\rangle$. Now suppose at time t = 0, the state

$$|\psi(0)\rangle = (1-a)^{\frac{1}{2}}|1\rangle + a^{\frac{1}{2}}|2\rangle \qquad (1)$$

is formed from a superposition of the two energy eigenstates. The linearity of quantum mechanics (specifically, the linearity of the Hamiltonian) requires that the time evolution of $|\psi(0)\rangle$ be given by

$$|\psi(t)\rangle = e^{-i\omega_1 t}[(1-a)^{\frac{1}{2}}|1\rangle + a^{\frac{1}{2}}e^{i(\omega_1-\omega_2)t}|2\rangle]. \qquad (2)$$

Except for an overall phase factor, the time evolution of the coherent superposition is determined by the frequency $\omega_p \equiv \omega_1 - \omega_2$, which is independent of the parameter a, the probability of finding the system in the eigenstate $|2\rangle$. This result can be stated graphically by using the mathematical equivalence between a two-level system and a system with a spin-½ particle in a magnetic field.[8] The eigenstates $|1\rangle$ and $|2\rangle$ correspond to spin states antiparallel and parallel to the magnetic field. The state $|\psi(0)\rangle$ corresponds to a state where the spin is tipped by an angle θ with respect to the magnetic field, where $\cos(\theta/2) = a^{\frac{1}{2}}$. The frequency ω_p is then the precession frequency of the spin about the magnetic field (or z axis). In the language of the equivalent spin-½ picture, the linearity of quantum mechanics requires that the precession frequency ω_p be independent of the tipping angle θ of the spin.

A test for a nonlinear addition to quantum mechanics can therefore be made by searching in a coherent superposition of states for a dependence of the frequency ω_p on the state probabilities (the parameter a). After a brief discussion of Weinberg's formalism, we will describe the different experimental tests. They include an ion trap experiment[4] which uses a ground-state hyperfine transition in $^9Be^+$, two different experiments[5,7] which use optical pumping techniques to polarize Ne and Hg nuclei in gas cells, and an experiment[6] which uses a hydrogen maser. The first three experiments may be viewed as tests of the linearity of quantum mechanics in a nuclear system. The hydrogen maser experiment may be viewed as a test of the linearity of quantum mechanics in an atomic system. In addition to the experimental tests, Weinberg's work has stimulated theoretical discussions[9-14] on the possibility of a nonlinear addition to quantum mechanics. In particular, a nonlinear addition to quantum mechanics may violate causality and make it possible, at least in principle, to have arbitrarily fast communications.[11,12,14] We summarize this discussion and give an example of how a nonlinear correction to quantum mechanics in the Be^+ experiment could be used to communicate faster than the speed of light.

WEINBERG'S NONLINEAR QUANTUM MECHANICS

In order to help motivate tests of nonrelativistic quantum mechanics, Weinberg has developed a formalism[2,3] which generalizes quantum mechanics by permitting possible nonlinear terms. In this formalism, observables are represented as operators on a Hilbert space of states just as in ordinary quantum mechanics. The difference with ordinary quantum mechanics is that the operators no longer need be linear. In order to have a sensible physical interpretation, Weinberg requires that any nonlinear additions do not violate Galilean invariance. He shows that there is a possible class of such additions to the Hamiltonian describing the internal degrees of freedom of a particle with spin. This means that the time evolution of the wave function $\psi(t)$ is no longer given by a linear Schrödinger equation. Instead it takes the form

$$i\hbar\frac{d\psi_k}{dt} = \frac{\partial h(\psi,\psi^*)}{\partial\psi_k^*} \qquad (3)$$

where $\psi_k = \langle k|\psi\rangle$ is the amplitude of state $|k\rangle$ ($k = -I, -I+1,.., I-1, I$ for a particle of spin I) and the Hamiltonian function $h(\psi,\psi^*)$ is a homogeneous function of ψ and ψ^* [$h(\lambda\psi,\psi^*) = h(\psi,\lambda\psi^*) = \lambda h(\psi,\psi^*)$ for any complex λ]. Homogeneity guarantees that if $\psi(t)$ is a solution of Eq. (3) then $\lambda\psi(t)$ is also a solution representing the same physical state. This ensures the proper treatment of physically separated and uncorrelated systems.[3] In general, $h(\psi,\psi^*)$ is not bilinear as it would be in ordinary quantum mechanics. A possible form of $h(\psi,\psi^*)$ which Weinberg has investigated is

$$h(\psi,\psi^*) = \sum_r \frac{h_r}{n^r}$$

$$= \sum_{k\ell} \psi_k^* H_{k\ell}\psi_\ell + \frac{1}{n}\sum_{k\ell mn} \psi_k^*\psi_\ell^* G_{k\ell mn}\psi_m\psi_n + \ldots \qquad (4)$$

where the first term is the linear term which describes ordinary quantum mechanics, the second term is the lowest order nonlinear addition, and $n = \sum_k \psi_k{}^*\psi_k$. Galilean invariance puts restrictions on the coefficients $H_{k\ell}$, $G_{k\ell mn}$, ... If we ignore translational degrees of freedom, Galilean invariance reduces to rotational invariance. The Hamiltonian function must transform as a scalar under rotations. The only scalars that can be formed from the two components of a spin-½ wave function are functions of the norm n. Therefore, in the absence of external fields, the potential nonlinear contributions of Eq. (4) to the Hamiltonian function of a spin-½ particle are functions of the norm and have no effect other than to set the zero of energy for the spin. Weinberg shows that in the presence of external fields (where the $G_{k\ell mn}$ transform according to the rotation properties of a tensor), a spin-½ particle can have a first-order quadrupole interaction. Particles with spin $I \geq 1$ can have a nonlinear contribution to the Hamiltonian function even in the absence of external fields. The lowest order nonlinear addition $h_{n\ell} (\equiv h_1/n)$ for a spin $I = 3/2$ can be written as[3]

$$h_{n\ell} = \frac{\epsilon}{n} \{ |\sqrt{6}\psi_{3/2}\psi_{-\frac{1}{2}} - \sqrt{2}\psi_{\frac{1}{2}}\psi_{\frac{1}{2}}|^2 + |3\psi_{3/2}\psi_{-3/2} - \psi_{\frac{1}{2}}\psi_{-\frac{1}{2}}|^2 + |\sqrt{6}\psi_{-3/2}\psi_{\frac{1}{2}} - \sqrt{2}\psi_{-\frac{1}{2}}\psi_{-\frac{1}{2}}|^2 \}. \quad (5)$$

For a nucleus with $I = 3/2$, Eq. (5) gives a possible nonlinear addition to the nuclear Hamiltonian. The parameter ϵ gives the strength of the nonlinear term. Weinberg does not make a prediction for a value of ϵ; he only constructs a theory in which $\epsilon \neq 0$ is allowed.

As a simple example, consider the two-level system consisting of the $m_I = -3/2$ and $-\frac{1}{2}$ states of a spin $I = 3/2$ nucleus. Let $|1\rangle \equiv |m_I = -3/2\rangle$ and $|2\rangle \equiv |m_I = -\frac{1}{2}\rangle$ and $h_0(\psi,\psi^*) = \hbar\omega_1\psi_1\psi_1^* + \hbar\omega_2\psi_2\psi_2^*$ denote the bilinear Hamiltonian function for the system. (Suppose the degeneracy of the levels is split by a magnetic field and that no population exists in the $+\frac{1}{2}$ and $+3/2$ sublevels.) The Hamiltonian function for the lowest order nonlinear contribution given by Eq. (5) reduces to

$$h_{n\ell} = n2\epsilon a^2, \quad (6)$$

where $a = |\psi_2|^2/n$ is the probability of finding the system in $|2\rangle$ and is identical to the parameter a in Eqs. (1) and (2). From Eq. (3) the time evolution of the superposition state $|\psi(0)\rangle$ from Eq. (1) is

$$|\psi(t)\rangle = e^{-i\omega_1(a)t} \left[(1-a)^{\frac{1}{2}}|1\rangle + a^{\frac{1}{2}} e^{i(\omega_1(a)-\omega_2(a))t}|2\rangle\right], \quad (7)$$

where $\omega_1(a) = \omega_1 - 2(\epsilon/\hbar)a^2$ and $\omega_2(a) = \omega_2 - 2(\epsilon/\hbar)a^2 + 4(\epsilon/\hbar)a$. The coherence frequency $\omega_p \equiv \omega_1(a) - \omega_2(a)$ now depends on the state probabilities according to

$$\begin{aligned} \omega_p &= \omega_0 - 4(\epsilon/\hbar)a \\ &= \omega_0 - 4(\epsilon/\hbar)\cos^2(\theta/2), \end{aligned} \quad (8)$$

where $\omega_0 \equiv \omega_1 - \omega_2$ is the transition frequency in the absence of nonlinearities. In the language of the equivalent spin-½ system, the precession frequency ω_p of the spin now depends on the tipping angle θ between the spin and the magnetic field. The equation of motion for a two-level system with the nonlinear addition given by Eq. (6) is formally identical to that of Jaynes' neoclassical theory of the interaction of the electromagnetic field with atoms.[15,16] However, the origin of the nonlinearity is quite different. In Jaynes' theory the nonlinearity is due to a postulated radiation reaction of the atomic dipole on itself.

The frequency ω_p is the "instantaneous" transition frequency of the two-level system. That is, if an external field of frequency ω is used to induce transitions between the two states, this transition rate is maximized for $\omega = \omega_p$. Suppose all the population is initially in state $|1\rangle$. Application of an external field with frequency $\omega = \omega_0 = \omega_p(\pi)$ starts driving population from state $|1\rangle$ to state $|2\rangle$. If the Rabi frequency Ω due to the applied field is much smaller than the nonlinear correction ϵ $[\Omega \ll (\epsilon/\hbar)]$, then the population can never be completely inverted. This is because the transition frequency given by ω_p is chirped as the state of the system evolves. Consequently, unless the frequency of the applied field is similarly chirped, a transfer of all the population to state $|2\rangle$ never occurs. Weinberg used this fact, along with the experimental observation that nuclear magnetic resonance transitions in $^9Be^+$ have been driven in times as long as 1 s to set a limit of $\sim 10^{-15}$ eV on the magnitude of a nonlinear correction to the energy of the ^9Be nucleus.[2]

Very few proposals for nonlinear terms in quantum mechanics have been made and experimentally tested. One proposal that has been experimentally tested is the addition of a logarithmic term $-b\psi(\vec{x})\ell n|\psi(\vec{x})|^2$ to the one-particle Schrödinger equation discussed by Bialynicki-Birula and Mycielski.[17] This nonlinear addition can be derived from a Hamiltonian function, but the Hamiltonian function is not homogeneous. Physically, it predicts that the time evolution of a wave function depends on its normalization, in contrast to Weinberg's theory. Nevertheless it gives a correct treatment of physically separated systems. Shimony[18] suggested an experimental test based on neutron interferometry for this nonlinear term. Experiments[19,20] have put a bound of 3×10^{-15} eV on the magnitude of the parameter b.

EXPERIMENTAL TESTS

Weinberg's generalization of quantum mechanics has motivated recent experimental tests which improve upon the limit set by Weinberg by more than 5 orders of magnitude. The first experiment to be described here was done at the National Institute of Standards and Technology and used ion storage techniques to measure the frequency of a hyperfine transition in $^9Be^+$ to high precision.[4] Trapped ions provide very clean systems for testing calculations of the dynamics of quantum transitions. The ions can be observed for long periods, relatively free from perturbations and relaxations. Their levels can be manipulated easily with rf and optical fields. This has led to their use in observing quantum effects such as quantum jumps, photon antibunching, sub-Poissonian statistics,[21] and the quantum Zeno effect[22]. In addition, trapped ions have been used to test the isotropy of space[23] and search for a spin-dependent fifth force.[24] For a test of nonlinear quantum mechanics, we searched for a state population dependence (a θ dependence) of the (m_I, m_J) = (-1/2, +1/2) → (-3/2, +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, called the clock transition, depends only quadratically on magnetic field fluctuations. With $|1\rangle \equiv |-3/2, +1/2\rangle$ and $|2\rangle \equiv |-1/2, +1/2\rangle$, the lowest order nonbilinear addition to the Hamiltonian function of the free $^9Be^+$ nucleus for the two states is given by Eq. (6). Equation (8) then gives the dependence of the precession frequency ω_p on the tipping angle θ, which describes the relative amplitude admixture of the two states. A search for a nonlinear addition to quantum mechanics was made by measuring ω_p at two different tipping angles.

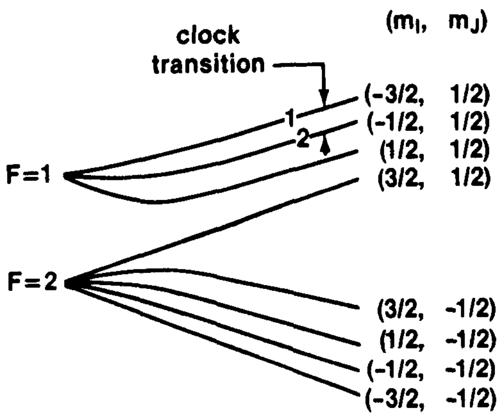


Fig. 1. Hyperfine energy levels (not drawn to scale) of the $^9Be^+$ 2s $^2S_{1/2}$ ground state as a function of magnetic field. At B = 0.8194 T the 303-MHz clock transition is independent of magnetic field to first order. (From Ref. 4)

Between 5000 and 10 000 $^9Be^+$ ions and 50 000 to 150 000 $^{26}Mg^+$ ions were simultaneously stored in a cylindrical Penning trap[25] with B ≃ 0.8194 T under conditions of high vacuum ($\lesssim 10^{-8}$ Pa). To minimize second-order Doppler shifts of the clock transition, the $^9Be^+$ ions were cooled to less than 250 mK by the following method. The $^{26}Mg^+$ ions were directly laser cooled[26] and the $^9Be^+$ ions were then sympathetically cooled[27] by their Coulomb interaction with the cold Mg^+ ions. Narrow-band 313-nm radiation was used to optically pump and detect the $^9Be^+$ ions. With the 313-nm source tuned to the 2s $^2S_{1/2}$(3/2, 1/2) to 2p $^2P_{3/2}$(3/2, 3/2) transition, 94% of the $^9Be^+$ ions were optically pumped into the 2s $^2S_{1/2}$(3/2, 1/2) ground state.[28,29] The 313-nm source was then turned off to avoid optical pumping and ac Stark shifts.

The clock transition was detected by the following method. After the 313-nm source was turned off, the ions in the (3/2, 1/2) state were transferred to the (1/2, 1/2) state and then to the (-1/2, 1/2) state by two successive rf π pulses. The clock transition was then driven by Ramsey's method of separated oscillatory fields[30] 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 (-1/2, 1/2) state to the (-3/2, 1/2) state. Those ions remaining in the (-1/2, 1/2) state were then transferred back to the (3/2, 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 (-3/2, 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 (-3/2, 1/2) state to the (3/2, 1/2) state was an order 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 in a coherent superposition of states |1> and |2> of the clock transition given by Eq. (7) for a particular value of $a = \cos^2(\theta/2)$. 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 $T \gg \tau_\theta$, $T \gg \tau_{\pi/2}$, the Ramsey line shape (specifically, the number of ions remaining in the state |2> as a function of the rf frequency ω in the Ramsey excitation) is

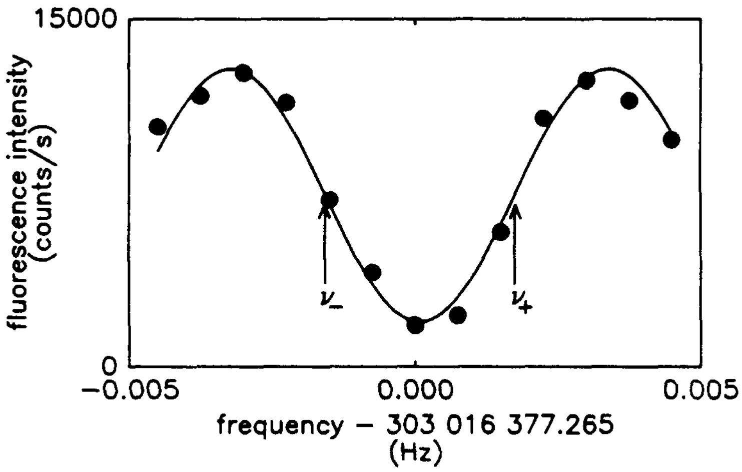


Fig. 2. Ramsey signal of the $^9Be^+$ clock transition with T = 150 s and $\theta = \pi/2$. The data are the result of one sweep (that is, one measurement per point). The sweep width is 9 mHz. The dots are experimental and the curve is a least-squares fit. The signal-to-noise ratio is limited by the frequency stability of the reference oscillator. The full-width-at-half-minimum frequencies are indicated by ν_+ and ν_-. (From Ref. 4)

proportional to

$$1 - \sin\theta \cos\{[\omega - \omega_p(\theta)]T\}, \qquad (9)$$

where $\omega_p(\theta)$ is given by Eq. (8). 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 $\theta = \pi/2$.

Ramsey-signal measurements were taken near both of the full-width-at-half-minimum frequencies $\omega_+ \equiv 2\pi\nu_+$ and $\omega_- \equiv 2\pi\nu_-$, 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[28] the average frequency of a synthesized rf source 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 $\theta = \theta_B$. Runs were taken with free-precession periods of T = 30, 60, and 100 s. A weighted average of the synthesizer frequency differences for $\theta = \theta_A$ and $\theta = \theta_B$ from about 110 h of data gave a possible dependence of the precession frequency on θ of $[\omega_p(\theta_B) - \omega_p(\theta_A)]/2\pi = 3.8 \pm 8.3$ μHz. From Eq. (8) a value for the parameter ϵ of $\epsilon/(2\pi\hbar) = 1.8 \pm 4.0$ μHz is obtained. The uncertainty is expressed as a 1 standard deviation.

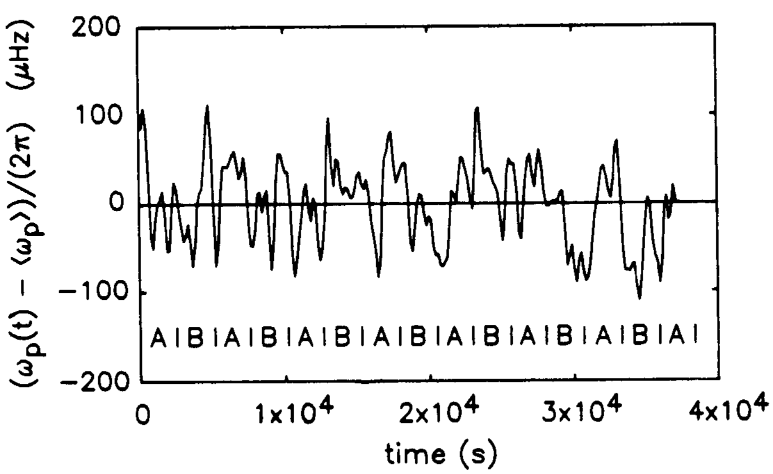


Fig. 3. $^9Be^+$ precession frequency $\omega_p(t)$ as a function of time for a single run with T = 100 s. The periods A(B) during which the initial Rabi pulse created a mixed state with angle $\theta_A = 1.02$ rad ($\theta_B = 2.12$ rad) are indicated. (From Ref. 4)

This 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. Our experimental result is limited by statistical fluctuations due to the frequency instability of the reference oscillator used with the synthesizer (typically a commercial Cs atomic clock). The largest known systematic errors of our measurement of $\omega_p(\theta)$ are an apparent, surprisingly large pressure shift[31] of ~ 30 μHz and a second-order Doppler shift of ~ 3 μHz due to the $\vec{E}\times\vec{B}$ rotation of the ion cloud in the trap.[28] We believe both shifts are constant to better than 1 μHz over the time required to make a frequency difference measurement (about 40 min).

Experiments[5,7] with ^{21}Ne and ^{201}Hg nuclei have produced comparable results to the $^9Be^+$ experiment described above. All three experiments use spin-3/2 nuclei to test the linearity of quantum mechanics. In the ^{21}Ne and ^{201}Hg experiments, optical pumping techniques are used to polarize the nuclear spins of an atomic vapor along a weak magnetic field. Let $\hbar\omega_k$, k = +3/2, +1/2, -1/2, -3/2 denote the energies of the nuclear spin levels. In a coordinate system with the z axis along the polarization axis, the density matrix ρ describing the spin-3/2 state populations is diagonal and can be expressed[32] in terms of irreducible spherical tensors T_{L0} according to $\rho \equiv \sum_{L=0}^{3} \rho_{L0}T_{L0}$. The multipole polarizations

$\rho_{L0} \equiv Tr(\rho T_{L0})$, L = 0,1,2,3 (the monopole, dipole, quadrupole, and octupole atomic polarizations) are given by

$$\begin{aligned}
\rho_{00} &= \tfrac{1}{2}\{<|\psi_{3/2}|^2> + <|\psi_{\frac{1}{2}}|^2> + <|\psi_{-\frac{1}{2}}|^2> + <|\psi_{-3/2}|^2>\},\\
\rho_{10} &= \frac{1}{2\sqrt{5}}\{3<|\psi_{3/2}|^2> + <|\psi_{\frac{1}{2}}|^2> - <|\psi_{-\frac{1}{2}}|^2> -3<|\psi_{-3/2}|^2>\},\\
\rho_{20} &= \tfrac{1}{2}\{<|\psi_{3/2}|^2> - <|\psi_{\frac{1}{2}}|^2> - <|\psi_{-\frac{1}{2}}|^2> + <|\psi_{-3/2}|^2>\},\\
\rho_{30} &= \frac{1}{2\sqrt{5}}\{<|\psi_{3/2}|^2> - 3<|\psi_{\frac{1}{2}}|^2> + 3<|\psi_{-\frac{1}{2}}|^2> - <|\psi_{-3/2}|^2>\},
\end{aligned} \tag{10}$$

where < > denotes the ensemble average over the spin-3/2 state populations. A coherent superposition of all of the nuclear spin levels is created by tipping the polarized nuclear spins with respect to the magnetic field by an angle θ. The transverse polarizations of the nuclear spins then precess about the magnetic field at frequencies determined by the Zeeman frequency splittings. Measurement of these free precession frequencies as a function of the state populations provides a test of the linearity of quantum mechanics. The Zeeman frequencies ω_i, i = 1,2,3 are[33]

$$\begin{aligned}
\omega_1 &\equiv \omega_{3/2} - \omega_{\frac{1}{2}} = 2\pi(D + Q + \frac{13}{12}\,O),\\
\omega_2 &\equiv \omega_{\frac{1}{2}} - \omega_{-\frac{1}{2}} = 2\pi(D + \frac{1}{12}\,O),
\end{aligned} \tag{11}$$

$$\omega_3 \equiv \omega_{-\frac{1}{2}} - \omega_{-3/2} = 2\pi(D - Q + \frac{13}{12} O),$$

where D, Q, and O are the dipole, quadrupole, and octupole contributions to the splitting of the Zeeman levels. If the Zeeman frequencies are resolved (for example, if Q^{-1} is less than the relaxation time of the coherent superposition), Weinberg's nonlinear corrections to these frequencies from Eq. (5) can be expressed in terms of $\rho_{L0}(\theta)$, the multipole polarizations along the precession axis, where $\rho_{L0}(\theta) = \rho_{L0} P_L(\cos\theta)$ and $P_L(\cos\theta)$ is the L^{th}-order Legendre polynomial, according to[7]

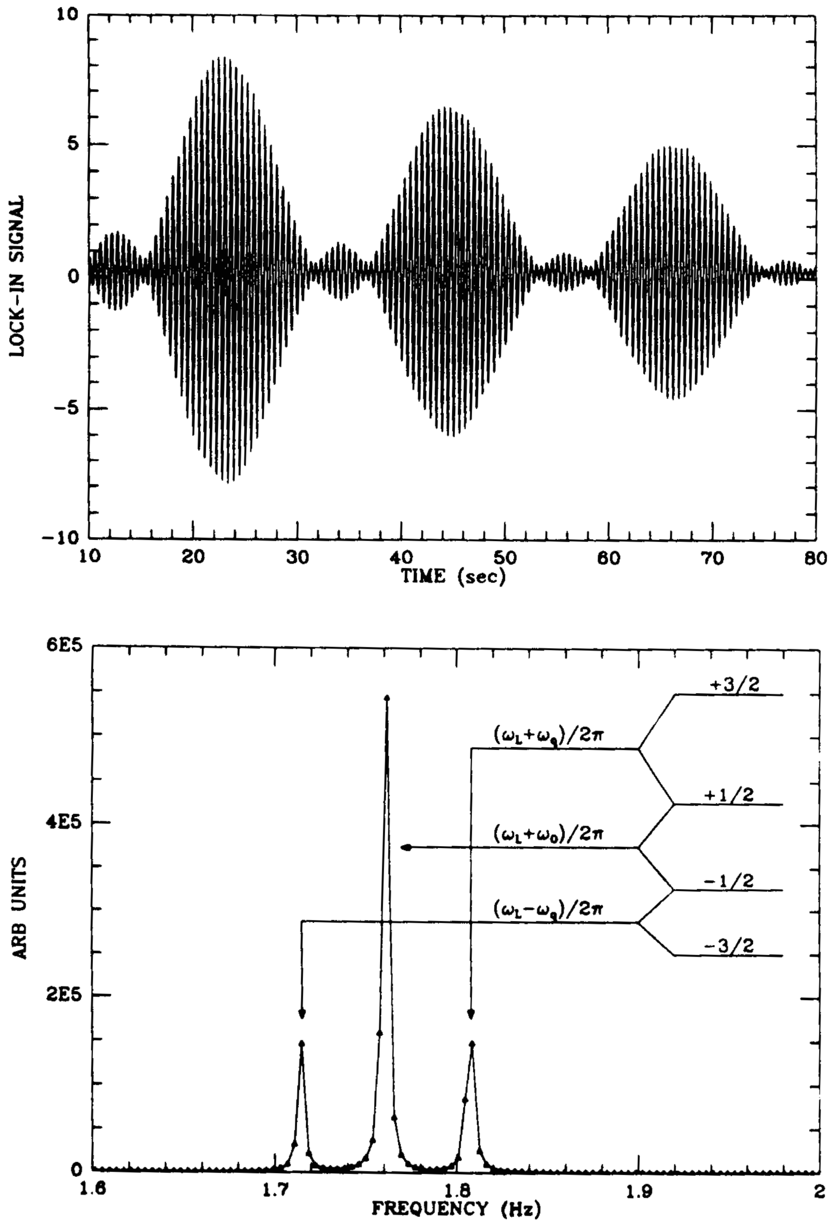
$$\begin{aligned}
\delta\omega_1 &= \frac{\epsilon}{\hbar}\left[-\frac{9}{\sqrt{5}}\rho_{10}(\theta,t) + \rho_{20}(\theta,t) + \frac{12}{\sqrt{5}}\rho_{30}(\theta,t)\right] \\
\delta\omega_2 &= \frac{\epsilon}{\hbar}\left[-\frac{15}{\sqrt{5}}\rho_{10}(\theta,t) - \frac{15}{\sqrt{5}}\rho_{30}(\theta,t)\right] \\
\delta\omega_3 &= \frac{\epsilon}{\hbar}\left[-\frac{9}{\sqrt{5}}\rho_{10}(\theta,t) - \rho_{20}(\theta,t) + \frac{12}{\sqrt{5}}\rho_{30}(\theta,t)\right].
\end{aligned} \tag{12}$$

The monopole component of the polarization has been omitted because it is unimportant to the discussion of these experiments. The variable t explicitly denotes the dependence of the multipole polarizations on time due to relaxation.

The precession frequencies in these experiments are relatively low (< 1 kHz) compared to the ~ 300 MHz frequency in the $^9Be^+$ experiment. In addition, the number of atoms used in these experiments is very large (10^{13} - 10^{19} atoms) compared with the number of ions (~ 5000) used in the $^9Be^+$ experiments. As a result, the second order Doppler shifts in these experiments are negligible and the signal-to-noise ratio is typically high enough to measure the precession frequencies to much better than 1 μHz. Time-dependent, systematic frequency shifts, however, typically limit the precision of the precession frequency measurements. In addition to the test of quantum mechanics discussed here, experiments like these have been used to test the isotropy of space[32-35] and to search for T-violating interactions.[36]

In the ^{201}Hg experiment done at the University of Washington,[7] circularly polarized light from a ^{198}Hg discharge lamp is used to optically pump about 10^{13} ^{201}Hg atoms contained in a quartz cell and polarize the ^{201}Hg nuclei along a few milligauss (1 mG = 10^{-7} T) magnetic field collinear with the light. A polarization of 45% of the value for a maximally polarized four-level system (all atoms in the m_I = +3/2 or -3/2) was obtained. After optical pumping, the polarized nuclear spins are adiabatically rotated by an angle ϕ by slowing rotating the magnetic field. The magnetic field is then suddenly changed to a direction perpendicular to the incident light and at an angle $\theta = 90° - \phi$ with respect to the polarized spins. By changing the angle θ the spin polarization moments given in Eq. (12) could be varied. The 2 Hz precession of the spins about the magnetic

(a)



(b)

Fig. 4. (a) Typical ^{201}Hg free-precession signal for $\theta = 90°$. The signal-to-noise is photon shot-noise limited (roughly 500 to 1 here). (b) A fast Fourier transform power spectrum of the data in (a). Also shown are the relevant ^{201}Hg Zeeman levels and the parametrization of the three coherence frequencies used in Ref. 7. (From Ref. 7)

field is measured from a modulation of the light transmitted through the cell for about 240 s which is roughly 2.5 coherence lifetimes. The three precession frequencies are resolvable due to a Q = 50 mHz quadrupole interaction between the ^{201}Hg spins and the cell wall. A typical free precession signal and its fast Fourier transform are shown in Figs. 4a and 4b. In this experiment, the difference in the center frequency and the average of the outer two frequencies, $\omega_2 - (\omega_1 + \omega_3)/2$, was fitted for a possible dependence on the dipole and octupole polarization given by Eq. (12). This combination of frequencies is sensitive only to the octupole contribution [O in Eq. (11)] of the Zeeman splitting. This provides good isolation from possible systematic shifts; the only known octupole shift is due to a misalignment of the cell axis with the magnetic field and was measured to be small [$O \simeq 2.5 \pm 0.5$ μHz]. Furthermore, this octupole shift is independent of atomic polarization as well as precession angle θ. The result of several thousand 240-s runs taken at different precession angles θ between 15° and 45° is $\epsilon/(2\pi\hbar) = 1.1 \pm 2.7$ μHz. Thus the fraction of the binding energy per ^{201}Hg nucleon which could be due to nonlinear quantum mechanics is less than 2×10^{-27} (3.8 μHz).

In the ^{21}Ne experiment done at Harvard University,[5] ^{21}Ne and ^{3}He (about 2×10^{19} atoms of each) are contained in a sealed glass cell and polarized along a static 0.3 mT magnetic field by spin exchange with optically pumped Rb. The ^{21}Ne spins are then tipped by $\theta = 20°$ with respect to the static magnetic field axis by a pulse of oscillating magnetic field at the precession frequency. The free precession of the ^{21}Ne spins is then measured over a period of 4.5 h by monitoring the transverse magnetization with the voltage induced in a pickup coil. The ^{21}Ne spins remain coherent over the entire measurement. The ~ 995 Hz ^{21}Ne precession frequency is mixed with the ^{3}He precession frequency (~ 9600 Hz) divided by 9.649 to produce a ~ 1/60 Hz carrier frequency which is relatively free of the effects of magnetic field fluctuations. Figure 5 shows the ^{21}Ne precession data for one 4.5-h run. The three precession frequencies are resolvable due to the interaction of the electric quadrupole moment of the ^{21}Ne nucleus with the glass cell wall. The quadrupole contribution to the Zeeman splitting is Q ~ 240 μHz. Because the dipole component of the frequency splitting is sensitive to spin-exchange shifts, originally the quadrupole component of the Zeeman splitting was used to test the linearity of quantum mechanics.[5] Specifically the difference frequency $\omega_1 - \omega_3$ was extracted from the precession frequency data and fit for a possible dependence on the quadrupole polarization. The quadrupole polarization would decay during a run due to relaxation. The result of five 4.5 h runs is $\epsilon/(2\pi\hbar) = 12 \pm 19$ μHz. The upper limit on $|\epsilon/(2\pi\hbar)|$ of 31 μHz is 1.6×10^{-26} of the binding energy per nucleon of the ^{21}Ne nucleus. This measurement is limited at least in part by the small quadrupole polarization ($\rho_{20} \leq 0.025$). The quadrupole polarization is small since it arises only from quadrupole relaxation in the presence of spin exchange, where spin exchange is a dipole interaction that produces dominantly dipole polarization. Recently, these ^{21}Ne data

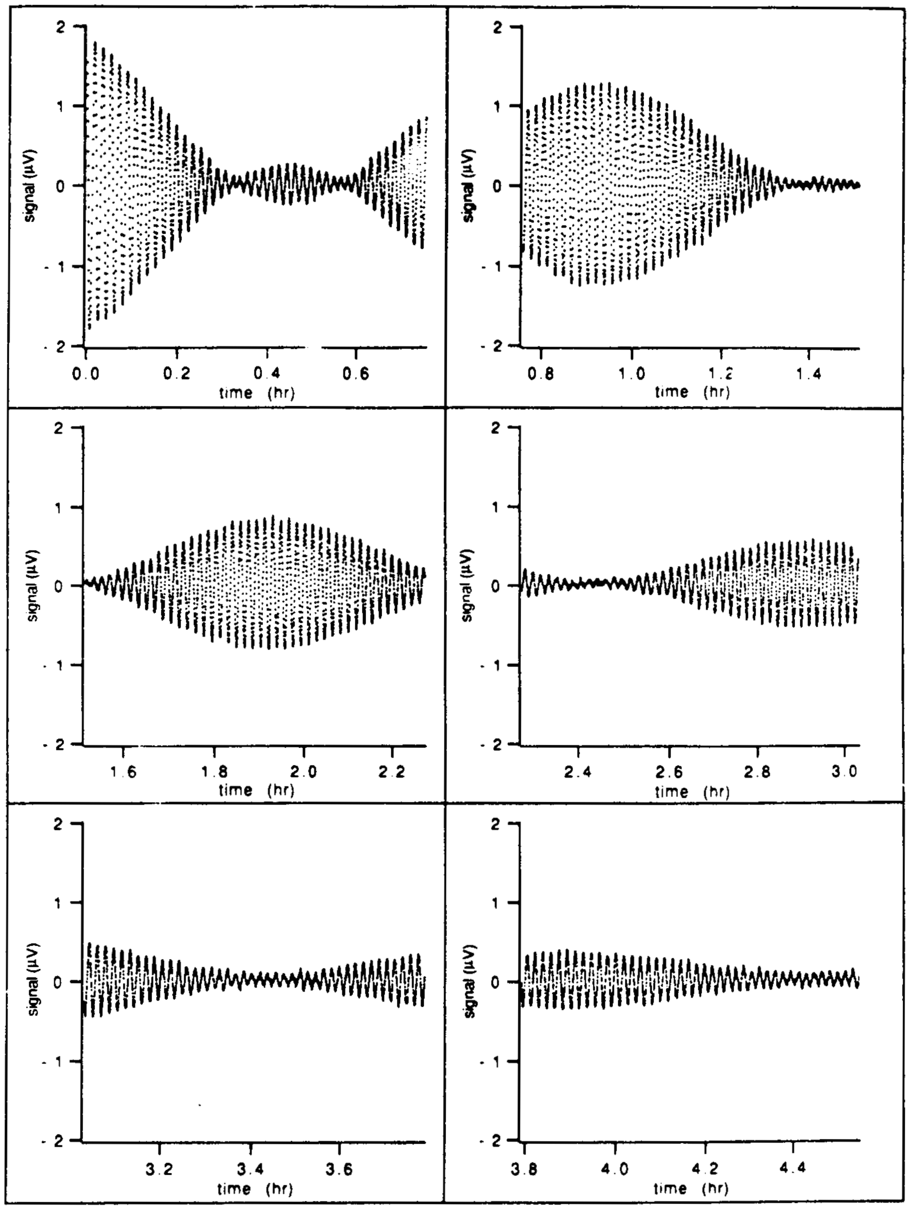


Fig. 5. Data for coherent free precession of ^{21}Ne over 4.5 h. Each panel represents a 45-min measurement of the beat frequency between the ^{21}Ne signals and a reference frequency derived from freely precessing ^{3}He. ^{3}He is not coherent from panel to panel; ^{21}Ne is. (From Ref. 5)

have been reanalyzed[37] by searching for a dipole and octopole polarization-dependent shift in the frequency component 0, similar to the technique developed by the University of Washington group in their ^{201}Hg experiment. The new limit on a possible nonlinear correction to the ^{21}Ne nuclear Hamiltonian of $|\epsilon|/(2\pi\hbar) < 1.5$ μHz is about a factor of 20 improvement over the previous limit.

In addition to these two experiments, a similar experiment[38] is planned which uses radioactive spin-3/2 ^{37}Ar nuclei ($t_{\frac{1}{2}}$ = 35 days) polarized by spin exchange with optically pumped Rb. The polarization of the ^{37}Ar is detected by observing the asymmetry in the emission of the internal bremsstrahlung accompanying the electron capture decay of ^{37}Ar to ^{37}Cl.

Hydrogen masers at the Smithsonian Astrophysical Observatory have also been used to search for a possible nonlinear correction to quantum mechanics.[6] The $(F,m_F) = (1,0) \rightarrow (0,0)$ clock transition in a hydrogen maser (see Fig. 6) is a transition between states of two coupled spin-½ particles. As noted earlier, a spin-½ particle cannot have a nonlinear correction to the Hamiltonian describing its internal degrees of freedom. However, the interaction between two spin-½ particles can. Walsworth and Silvera have determined the lowest order allowed nonlinear Hamiltonian function for the H ground-state hyperfine structure.[39] The resulting nonlinear contribution to the clock transition frequency ω_0 of the maser is

$$\delta\omega_0 = \left(\frac{\epsilon_1{}'+\epsilon_0{}'}{\hbar n} \right) (|\psi_1|^2 - |\psi_0|^2), \tag{13}$$

where ψ_1 and ψ_0 are the amplitudes for the atom to be in the F = 1, m_F = 0 and F = 0, m_F = 0 states respectively, the nonlinearity parameters $\epsilon_1{}'$ and $\epsilon_0{}'$ include a possible dependence on the atom's state, and n is the norm. A test for a nonlinear correction to

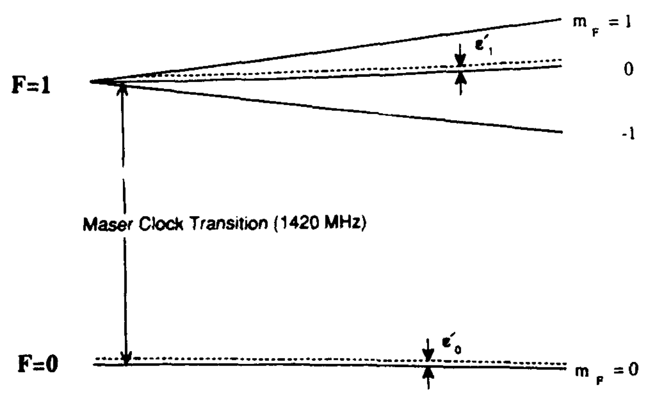


Fig. 6. Atomic hydrogen ground state hyperfine energy levels as a function of magnetic field, for fields < 0.02 T. Energy level shifts due to possible nonlinear effects are shown. (From Ref. 6)

quantum mechanics was made by searching for a dependence of the maser frequency on the relative populations of the clock states as shown in Fig. 7. The relative population of the two states was varied by changing the input H flux to the maser, and it was measured from the dependence of Q_l, the line Q of the maser signal, on the populations.[40] Increasing the H flux decreases both Q_l and the population difference between the states. The result of eight ~50 h runs searching for a dependence of the maser frequency on the state populations was $(\epsilon_1' + \epsilon_0')/(2\pi\hbar) = 1.5 \pm 7.4$ μHz. This limit is comparable in magnitude to the limits placed on nonlinear effects in a nuclear system described in three previous experiments. It sets an upper limit of 3×10^{-21} on the fraction of the 13.6 eV electronic binding energy that could be due to a nonlinear contribution to the atomic Hamiltonian of the hydrogen atom.

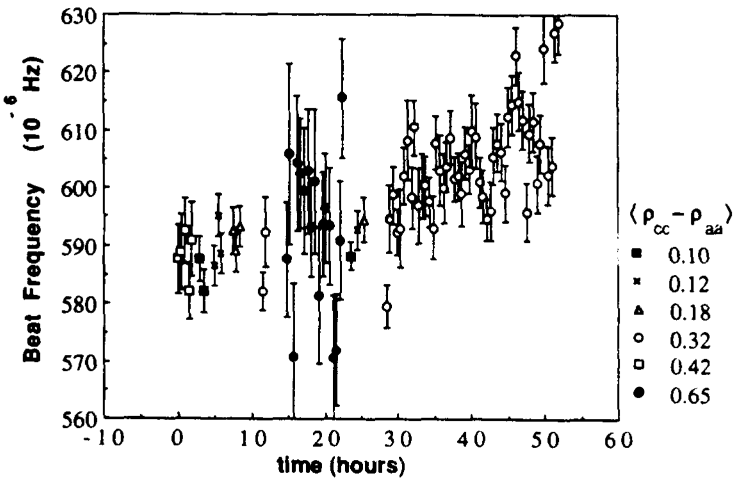


Fig. 7. The raw beat frequency data from a single run (#6). Six different H flux levels were used in this run. An overall linear frequency drift of ~ 6 μHz/day is evident in the data, due to the settling of the materials in the resonant cavity. (From Ref. 6)

Table I summarizes the experimental results. In the absence of a model which gives a specific prediction for ϵ, it is important to have experimental tests in several different systems. Limits have now been set at ~ 10 μHz or less in four different systems. This is 5 orders of magnitude smaller than experimental limits placed by neutron interferometry[19,20] on the size of a logarithmic addition to the one-particle Schrödinger equation. Three of the experiments can be interpreted as setting a fractional limit of better than 10^{-26} on a nonlinear addition to a nuclear Hamiltonian. The hydrogen maser experiment sets a fractional limit of 3×10^{-21} on a possible nonlinear contribution to an atomic Hamiltonian. All of the experiments are first attempts at measuring a nonlinear component of quantum mechanics and have the potential for an order of magnitude improvement in sensitivity.

Experiment	Limit on $\lvert\epsilon\rvert/(2\pi\hbar)$	Interpretation	Reference
^{9}Be	5.8 μHz	nuclear	4
^{201}Hg	3.8 μHz	nuclear	7
^{21}Ne	31 μHz 1.5 μHz	nuclear	5 37
H	8.9 μHz	atomic	6

TABLE I. Summary of experimental tests for a nonlinear addition to quantum mechanics permitted by Weinberg's formalism. The interpretation column gives the type of Hamiltonian which the experiment is interpreted as testing. The limits are one standard deviation.

THEORETICAL DISCUSSIONS

In addition to motivating experimental tests, Weinberg's formalism for introducing nonlinear corrections to quantum mechanics has stimulated theoretical discussions.[9-14] These discussions center on whether there is any theoretical reason why quantum mechanics has to be linear. While the discussions do not rule out the possibility of a nonlinear component to quantum mechanics, they show the difficulty in constructing such a theory.

A. Peres has reported[9] that nonlinear variants of Schrödinger's equation violate the second law of thermodynamics. Weinberg's response[10] was that this may be true if entropy is defined as in ordinary quantum mechanics, but that is not an appropriate definition of entropy in a generalized version of quantum mechanics. An interesting question is how to generalize the concept of entropy in a generalized version of quantum mechanics.

Recently, A. Valentini has shown[13] that small nonlinearities of the Schrödinger equation lead to a violation of the wave-particle complementarity in quantum mechanics. Specifically, if the Schrödinger equation was nonlinear, a double slit experiment could be done where both an interference pattern and the slit the particle went through could be measured. Valentini points out that this does not necessarily rule out nonlinear theories, but that the wave-particle aspect of the physical interpretation of such theories must differ from that of standard quantum mechanics.

An interesting connection between nonlinear time evolution in quantum mechanics and causality has been discussed by

N. Gisin.[11,12] Gisin shows, by the example of an Einstein-Podolsky-Rosen (EPR) experiment consisting of pairs of correlated spin-½ particles traveling in opposite directions, that a nonlinear coupling between a spin-½ particle and an electric quadrupole field (permitted in Weinberg's formalism) would enable arbitrarily fast communications.[11] Gisin argues that the same conclusion follows for any nonlinear time evolution.[12] Gisin points out that there may be ways out of this dilemma by changing the usual interpretation of the EPR experiments. In addition, Weinberg's formalism generalizes nonrelativistic quantum mechanics only. As a nonrelativistic theory it may not necessarily give a correct description of causality. However, as Gisin points out, it does appear to create a problem where before there was none.

Following Gisin's example, we can show how a nonlinear contribution to the $^9Be^+$ clock experiment could result in communication faster than the speed of light. As before, let $|1\rangle$ and $|2\rangle$ denote the two states of the $^9Be^+$ clock transition. Let $|+\frac{1}{2}\rangle_z$ and $|-\frac{1}{2}\rangle_z$ denote the two states of a spin-½ particle, say an electron, along the same quantization axis (the z axis) as the $^9Be^+$ system. Suppose a source of correlated states,

$$\frac{1}{\sqrt{2}}(|1\rangle\ |-\tfrac{1}{2}\rangle_z\ -\ |2\rangle\ |+\tfrac{1}{2}\rangle_z), \qquad (14)$$

is placed between two regions A and B in space. Assume the source continuously emits the correlated states with the electrons moving toward region A along the y axis and the $^9Be^+$ clock (the $^9Be^+$ ion in an 0.8194 T magnetic field) moving in the opposite direction toward region B. (If desired the $^9Be^+$ clock can be at rest and only the electron moves.) In region A an observer measures the spin of the electron in a Stern-Gerlach apparatus. In region B a different observer drives the clock transition and then measures the population of state $|2\rangle$. Specifically, a weak rf pulse (duration $\tau \gg (2\pi\hbar)/\epsilon$, Rabi frequency $\Omega \ll \epsilon/\hbar$) of frequency $\omega = \omega_p(\theta = \pi) = \omega_0$ is applied in region B. Due to the nonlinear contribution to the clock transition frequency, this applied field will start to drive a transition from a pure $|1\rangle$ state to state $|2\rangle$, but not from $|2\rangle$ to $|1\rangle$. After the applied rf pulse, the population of state $|2\rangle$ is measured as described in the previous section.

Suppose the observer in region A orients the Stern-Gerlach apparatus so that the z component of the electron spin is measured. This prepares the $^9Be^+$ clock in either the $|1\rangle$ state or the $|2\rangle$ state. The rf pulse takes the $|1\rangle$ state and puts it into a mixture of $|1\rangle$ and $|2\rangle$ but leaves the $|2\rangle$ state unchanged. Observer B will then measure on the average that state $|2\rangle$ has a population greater than ½. Now suppose the observer in region A rotates the Stern-Gerlach apparatus and measures the x component of the electron spin. The correlated state given by Eq. (14) can be written in terms of $|+\frac{1}{2}\rangle_x$ and $|-\frac{1}{2}\rangle_x$, the electron spin states along the x axis, as

$$\frac{1}{2}(|1\rangle - |2\rangle)|-\tfrac{1}{2}\rangle_x - \frac{1}{2}(|1\rangle + |2\rangle)|+\tfrac{1}{2}\rangle_x . \qquad (15)$$

According to the standard interpretation of the EPR experiments, when observer A measures the x component of the electron spin the $^9Be^+$ clock will be prepared in either the $(|1\rangle - |2\rangle)/\sqrt{2}$ state or the $(|1\rangle + |2\rangle)/\sqrt{2}$ state. Both of these states are described by $\theta = \pi/2$ and will not be affected by the rf pulse with frequency $\omega = \omega_p(\pi) \neq \omega_p(\pi/2)$. In this case, observer B will measure that state $|2\rangle$ has a population equal to ½. When observer A rotates the Stern-Gerlach apparatus, observer B instantly sees a change in the state $|2\rangle$ population. It will take observer B a time greater than $(2\pi\hbar)/\epsilon$ to measure this change, but since regions A and B are arbitrarily far apart, it appears that communication at arbitrarily fast speeds is possible.

Recently, Polchinski[14] has investigated whether nonlinear quantum mechanics necessarily violates causality by determining the constraints imposed on observables by the requirement that communication does not occur in EPR experiments. He shows that this leads to a different treatment of separated systems than that originally proposed by Weinberg, but that it is possible for quantum mechanics to be nonlinear without violating causality. However, he shows that this necessarily leads to another type of unusual communication: the communication between different branches of the wavefunction.[41] Polchinski points out that, in effect, this means the wavefunction is never reduced and that this may in turn lead to a dilution of any nonlinear effects.

Clearly a number of theoretical questions about nonlinear theories of quantum mechanics remain to be fully resolved. These questions at present may not prove that quantum mechanics must be linear, but they do point out some problems with the physical interpretation of a nonlinear theory. Experimental work to date has shown that a nonlinear component to quantum mechanics must be very small. In some cases, the nonlinear terms must be 26 orders of magnitude smaller than the linear terms. The theoretical discussion and the experimental tests given here, involve nonlinear generalizations of the Hamiltonian. A nonlinear version of quantum mechanics could possibly take a different form. For example, Weinberg's formalism allows the possibility that operators other than the Hamiltonian could be nonlinear.

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