Test of the Linearity of Quantum Mechanics in an Atomic System with a Hydrogen Maser

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We extend a nonlinear generalization of quantum mechanics, recently formulated by Weinberg, to systems of composite spin, such as atoms. We have used hydrogen (H) masers to set a limit of 3.7×10^{-20} eV (8.9 μHz) on the magnitude of a nonlinear correction to the quantum mechanics of atomic spins. This result is of comparable magnitude to the limit recently set in a single-valued (nuclear) spin system. In the absence of nonlinear effects, the experiment provides a new and stringent test of H-maser clock performance and the applicability of standard maser theory.

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Linear superposition is a fundamental principle of quantum mechanics (QM). As early as 1939, however, it was noted by Wigner[1] that the linearity of quantum mechanics is merely an assumption, given credence by experimental evidence, and that the possibility of a nonlinear generalization of quantum mechanics should be investigated. Recently, Weinberg[2,3] has developed a generalized theory of (nonrelativistic) nonlinear quantum mechanics (NLQM) for the internal degrees of freedom of single-valued spin particles. Requiring NLQM to satisfy Galilean invariance, Weinberg finds that nonlinear terms are allowed in the generalized Hamiltonian function, and thus in the equation of motion, of particles of spin ≥ 1. The magnitudes of these nonlinear terms are set by parameters, assumed small, such that the generalized theory reduces to linear quantum mechanics in the limit that the parameters go to zero. Many of the physical implications of this theory are analogous to the behavior of a classical anharmonic oscillator, and clearly differ from the tenets of linear quantum mechanics. For example, the resonant frequency of transitions between eigenstates depends on the relative populations of the states. This property has recently been exploited experimentally by Bollinger *et al.*[4] to significantly improve confidence in the linearity of the quantum mechanics of the spin of the ^{9}Be nucleus; further experiments on different nuclear systems are currently being performed.[5,6]

In this Letter we report an extension of NLQM to systems of composite (multivalued) spin, such as atoms or molecules. We find that nonlinear behavior is allowed for the spin-0 or spin-$\frac{1}{2}$ states, unlike with single-valued spin systems. Applied to the ground-state hyperfine structure of the hydrogen atom (H), this analysis predicts a frequency shift in the H-maser clock transition of a unique form, proportional to the population difference between the two masing states. We have performed an experiment that places a limit on this atomic NLQM effect, of comparable absolute magnitude to the limit previously set in a nuclear system.[4] The maser population inversion was controllably and measurably varied using a new technique, changing the H beam flux and monitoring the resultant change in the oscillation linewidth. In the absence of NLQM corrections, this experiment provides a new, high-precision test of the agreement between H-maser performance and simple, semiclassical maser theory.

In the NLQM developed in Refs. 2 and 3, the equation of motion for the state vector ψ of a particle of spin s is taken to be

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

where the subscript refers to the particle's discrete m_s states, and h is a (real) generalized Hamiltonian function satisfying the requirement of first-degree homogeneity: $h(\lambda\psi,\psi^*)=h(\psi,\lambda\psi^*)=\lambda h(\psi,\psi^*)$ for any complex λ. Homogeneity must be retained in a realistic theory as it rules out special normalization of physical states and is essential to a proper treatment of noninteracting separated systems.[3] As is discussed in Ref. 3, previous realistic NLQM theories have failed to require both homogeneity and Galilean invariance. For $h(\psi,\psi^*)$ taken to be bilinear ($h=\psi_k^*H_{kl}\psi_l$), Eq. (1) reduces to the usual linear equation of motion of quantum mechanics. Foregoing the distributive property of the linear superposition principle, however, it is possible for $h(\psi,\psi^*)$ to be nonbilinear and remain homogeneous. The resulting expression for h can be given as a sum over all non-negative powers of ψ and ψ^*:

$$h=\sum_r\frac{h_r}{n^r}=\sum_{kl}\psi_k^*H_{kl}\psi+\frac{1}{n}\sum_{klmn}\psi_k^*\psi_l^*G_{klmn}\psi_m\psi_n+\cdots, \qquad (2)$$

where the first term is the linear QM term, the second term is the lowest-order nonlinear term, and $n = \sum_k |\psi_k|^2$ is the normalization factor. Small nonlinear corrections to quantum mechanics, if they exist, will result in small nonlinearities in the equation of motion. Hence, it will be sufficient to consider only the lowest-order nonlinear term ($h_{\rm NL}$) in the expansion. In the quantum mechanics of atoms, the spins of the atomic constituents are often coupled, resulting in a "particle" (the atom) that can exist in more than one spin state. The significant difference with multivalued spin is that each ψ (and ψ^*) in the expansion for h can now be in any of two or more spin states. As a result[7] physically significant nonlinear effects are possible for states of spin 0 or spin $\frac{1}{2}$, unlike for single-valued spin.

High-precision measurements employing H hyperfine transitions are possible with the use of an H maser.[8] Both the hydrogen nucleus and electron have a spin of $\frac{1}{2}$; so neither can exhibit nonlinear behavior, according to the NLQM of single-valued spin particles.[3] Combining a proton and electron to form H, however, results in a composite atomic spin of 1 or 0: Thus the hydrogen *atom* may have NLQM effects. Applying homogeneity and Galilean-invariance conditions to Eq. (2), one finds for the H ground-state hyperfine structure,

$$h_{\rm NL} = \frac{\epsilon_1'}{n} |2\psi_d\psi_b - \psi_c^2|^2 + \frac{\epsilon_0'}{n} |\psi_a|^4 , \tag{3}$$

where the mixed nonlinearity parameters ϵ_1' and ϵ_0' include a possible dependence on the constituent atom's spin quantum numbers.[7] The four (F,m_F) hyperfine states are labeled a,b,c,d in order of increasing energy. An application of the NLQM equation of motion [Eq. (1)], to solve for the nonlinear contribution to the resonant frequency of the $c \to a$ (clock) transition $\omega_{ca}^{\rm NL}$, yields

$$\hbar\omega_{ca}^{\rm NL} = \left[\frac{\epsilon_1' + \epsilon_0'}{n}\right] (|\psi_c|^2 - |\psi_a|^2) . \tag{4}$$

Thus, the resonant frequency gains a unique dependence on the eigenstate (c-a) population difference. This discussion assumes that the c and a states are pure (F,m_F) states.

We searched for a dependence of the frequency of the H clock transition on the ensemble-averaged population difference between the c and a states ($\equiv \langle \rho_{cc} - \rho_{aa} \rangle_{\rm ens}$), with the use of an H maser built at the Smithsonian Astrophysical Observatory (SAO). In an H maser,[8] a beam of atoms in the upper (c) state enters a Teflon lined quartz storage bulb located in an evacuated microwave cavity ($Q_{\rm cavity} \approx 40\,000$) resonant at the hyperfine transition frequency ($\nu_0 \approx 1.42$ GHz), and is stimulated to radiate to the lower (a) state by the electromagnetic field in the cavity. An H maser's output frequency is shifted from the unperturbed H hyperfine frequency by the atom's interactions with the Teflon wall coating, with an imposed static magnetic field, and with each other via collisions. The output frequency can be controllably shifted, or pulled, by an appropriate mistuning of the cavity resonance frequency:

$$\Delta\nu_{\rm pull} = \frac{Q_{\rm cavity}}{Q_l} \Delta\nu_{\rm cavity} . \tag{5}$$

Here, Q_l is the line Q of the maser signal, $\Delta\nu_{\rm cavity}$ is the detuning of the cavity from resonance, and $\Delta\nu_{\rm pull}$ is the resultant pulling of the output frequency. We employed a new technique of controllably and measurably varying $\langle \rho_{cc} - \rho_{aa} \rangle_{\rm ens}$ by changing the input H flux and monitoring the change in Q_l. The measurements were performed with SAO maser P24, a high-stability state-of-the-art H maser; SAO maser P13 was used as the reference oscillator. The resultant beat-signal periods (≈ 1 sec in this work) were averaged by counters and the average beat period, over 100 periods, was recorded along with the time at the beginning and end of each measurement.

The H beam flux determines the steady-state H density in the maser storage bulb; this affects the rate of H-H collisions in the maser, and thereby the oscillation linewidth. Equivalently, one finds that Q_l varies inversely with the H flux. An application of standard maser theory shows that $\langle \rho_{cc} - \rho_{aa} \rangle_{\rm ens}$ and Q_l are related by[9]

$$\langle \rho_{cc} - \rho_{aa} \rangle_{\rm ens} = \alpha \frac{1/Q_l}{1/Q_l - 1/Q_l^0} , \tag{6}$$

where Q_l^0 is the value of Q_l in the limit of zero H flux, and α is a system constant.[9] For P24, $\alpha = 0.025$ with an uncertainty $\lesssim 10\%$. In addition, H-H collisions cause a shift in the maser output frequency of the form[9] $\Delta\nu_{\rm coll} = \gamma/Q_l$ to first order in $1/Q_l$, where γ is a system constant related to α. A comparison of the expressions for $\Delta\nu_{\rm coll}$ and $\Delta\nu_{\rm pull}$ [Eq. (5)] shows that the maser output frequency can be made insensitive to H-flux, and thus $1/Q_l$, variations in the absence of NLQM effects by an appropriate tuning of the cavity frequency.[9] In practice, the experiment reported here is also a high-precision test of this widely used method of "flux-insensitive tuning."

Maser P24's maximum H flux was roughly a factor of 7 above the threshold for oscillation. Over this range $\langle \rho_{cc} - \rho_{aa} \rangle_{\rm ens}$ varied from about 0.45 just above threshold to 0.10 at the highest H fluxes. P24's dynamic range in $\langle \rho_{cc} - \rho_{aa} \rangle_{\rm ens} \approx 0.35$ is exceptional for an H maser. Referring to Eq. (6), most of the change in $\langle \rho_{cc} - \rho_{aa} \rangle_{\rm ens}$, and thus the majority of any NLQM frequency shift, comes when Q_l is close to Q_l^0; that is, very near the oscillation threshold. This sensitive dependence of frequency on H flux, near the oscillation threshold, is unique among previously considered and observed H-maser frequency-shift mechanisms.[10] A recent, quantum-mechanical treatment[11] of H-H collisions in H masers has predicted a dependence of the oscillation frequency on $\langle \rho_{cc} + \rho \rangle_{\rm ens}$, although the magnitude of the effect is calculated to be negligible at room temperature.

Nonetheless, such a dependence is functionally distinct from the NLQM shift of Eq. (4). Thus a possible NLQM effect could be cleanly distinguished in an analysis of the maser frequency as a function of $1/Q_l$.

For each of the eight data taking runs, a preliminary flux-insensitive tuning of the cavity was followed by measurements of the beat frequency and $1/Q_l$ at five or six H-flux levels. Frequency measurements were taken in blocks of 20 of the 100-beat-period averages discussed above. The mean value of the block beat frequency $\bar{\nu}_b$ was calculated along with an uncertainty $\sigma(\bar{\nu}_b)$, taken as the estimated error of the mean. Typically, two to five consecutive $\bar{\nu}_b$ measurements were made at each H-flux level. The precision of the $\bar{\nu}_b$ measurements was limited by the frequency stability of the masers. Their characteristic fractional frequency stability[12] is $\sigma_y(\tau) \lesssim 10^{-13} \times \tau^{-1/2}$ for averaging time τ in seconds, over the time scale of $\bar{\nu}_b$ measurements. Measurements of $1/Q_l$ were made by varying the cavity frequency and observing the resultant pulling of the maser output frequency. The cavity frequency was tuned to three or four values over a range $\sim$4 kHz, allowing $1/Q_l$ to be ascertained with a certainty of 1%–2%. The value of $1/Q_l^0$ was determined by a fit of the maser signal power to $1/Q_l$, extrapolated to zero H flux, with an uncertainty of 1%–2%.

After measurements were made for the five or six chosen H-flux (i.e., $1/Q_l$) levels, a second round of data were taken for some of these $1/Q_l$ values. This allowed a characterization of maser frequency drift and served as a check for spurious frequency shifts. H masers built at SAO are known to exhibit upward drifts in their cavity frequencies because of the slow settling of cavity materials.[13] Typically, this will shift the maser output frequency at a rate $\lesssim 10$ μHz/d. The cavity settling has a time constant of several months to years, and so the frequency drift is quite linear on the time scale of the hours to days of the data runs reported here. A small number of spurious frequency shifts unrelated to H-flux or cavity frequency changes did occur during or between the data runs. These were usually correlated with environmental disturbances (thermal, magnetic, etc.) of the masers or the maser storage room. Because of these systematic shifts, the data were segregated such that no measurements from across shift boundaries were combined into a single run.

The raw data for each run (see Fig. 1 for an example) were first fitted for frequency drift. Least-squares fits of $\bar{\nu}_b$ as a function of time were performed on groups of data points of *constant* $1/Q_l$. The only statistically significant effect found was a linear drift in time. Together, the drift rates for different $1/Q_l$ were best fitted to a constant. We found that including a term linear in $1/Q_l$, to model the differential pulling effect of the various H-flux rates [see Eq. (5)], did not improve the fits, in general. This differential effect was expected to be $\sim$1 μHz/d, and so was below the typical level of uncertainty in a run's averaged drift rate ($\approx$2 μHz/d). The raw data were then corrected for the frequency drift, with the $\sigma(\bar{\nu}_b)$ values adjusted to account for the uncertainty in the drift. In the fits described above (and those below) the χ^2 per degree of freedom (χ_ν^2) was usually < 1. For fits with $\chi_\nu^2 > 1$ the associated parameter variances were corrected by multiplying by χ_ν^2. Residuals for $\bar{\nu}_b$ were fitted by

$$(\bar{\nu}_b)_{\text{res}} = A + \frac{B}{Q_l} + \left(\frac{\epsilon}{2\pi\hbar}\right)\alpha\frac{1/Q_l}{1/Q_l - 1/Q_l^0}\,, \quad (7)$$

where the fit parameter A accounts for $1/Q_l$-independent frequency shifts between the two masers, as well as the frequency synthesizer offset, the second term accounts for residual-cavity mistuning [see Eq. (5)], and the third term represents the possible NLQM effect ($\epsilon \equiv \epsilon_1' + \epsilon_0'$). The results from this "residual" method of data analysis, for $\epsilon/2\pi\hbar$ and the associated (1-standard-deviation) uncertainty, are shown in Fig. 2 for each of the eight runs. The uncertainties include the contribution from the uncertainty in $1/Q_l$ and $1/Q_l^0$. Calculating the weighted average of these $\epsilon/2\pi\hbar$ values, one finds

$$\epsilon/2\pi\hbar = 1.5(7.4)\ \mu\text{Hz}, \quad \chi_\nu^2 = 0.56\,. \quad (8)$$

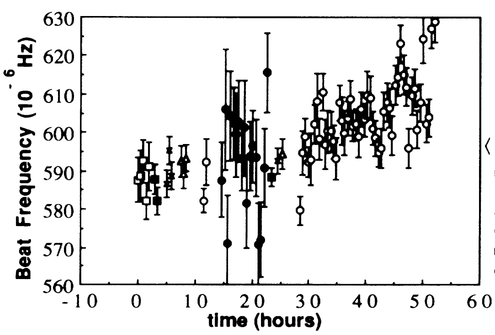

FIG. 1. The raw beat-frequency data from a single run (No. 6). Six different H-flux levels were used in this run. An overall linear frequency drift $\sim$6 μHz/d is evident in the data. The cavity pulling is small (< 10 μHz) due to flux-insensitive tuning of the cavity prior to the run.

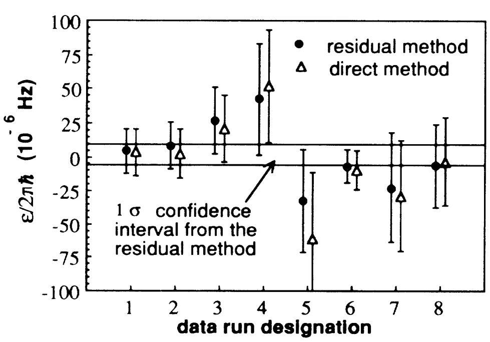

FIG. 2. The results for the characteristic NLQM parameter ϵ from each of the eight runs, as given by both the "residual" and "direct" methods of data analysis.

The error is a 1-standard-deviation uncertainty, and includes the contribution from the uncertainty in the system parameter α. As a check, a second method of analysis was employed, fitting the raw data of each run directly for $1/Q_l$ and time t, by χ_ν^2 minimization, with Eq. (7) with the addition of a term Dt. Here, D is like the averaged frequency drift rate discussed above. The results for $\epsilon/2\pi\hbar$ from this "direct" method of analysis, as shown in Fig. 2, are quite similar to those obtained via the residual method. A weighted average over the eight runs gives $\epsilon/2\pi\hbar = -0.4$ (8.1) μHz, with $\chi_\nu^2 = 0.68$.

Equation (8) sets an upper limit of $|\epsilon| < 3.7 \times 10^{-20}$ eV (8.9 μHz) on the nonlinear contribution to the quantum mechanics of the internal (spin) degrees of freedom of the H atom. This is comparable in magnitude to the limit placed recently on NLQM effects in a nuclear system.[4] If it is appropriate to compare limits on nonlinearities with the characteristic energy scale to change the spin of the particle under consideration,[2,3] then the test with the nuclear system sets a fractional limit on NLQM $< 4 \times 10^{-27}$, whereas the work presented here places a much less stringent limit ($< 3 \times 10^{-21}$ compared to the 13.6-eV electronic binding energy; $< 6 \times 10^{-15}$ compared to the hyperfine interaction). However, it is not clear which form of normalization of limits on NLQM effects is correct.[14] Our experimental results were limited by statistics due to the limited availability of maser P24, and due to the frequency instability of the masers. In general, including the possible NLQM correction in the analysis of $\bar{\nu}_b$ did not improve the quality of the fits, as judged by the resultant χ_ν^2 values. Similarly, including terms of different powers of $1/Q_l$ did not improve the fits. Therefore, an analysis of each of the data runs was performed to determine the deviation of $\bar{\nu}_b$ from low- to high-H-flux levels, in the absence of nonlinear corrections and corrected for residual-cavity mistuning. For both the residual and direct methods, a weighted average over the eight runs sets an upper limit of $|(\bar{\nu}_b)_{\text{high flux}} - (\bar{\nu}_b)_{\text{low flux}}| < 4.0$ μHz, exceptional confirmation of the applicability of standard maser theory. To our knowledge, this is the most stringent test to date of the method of flux-insensitive tuning, widely used in H-maser clocks.

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[14]For example, if nonlinear behavior arises from new physics, such as an internal coupling of the system's (linear) states, then the magnitude of the effect could depend on the nature of this coupling and the system in question.