Nonlinear quantum mechanics for systems of composite spin

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A nonlinear generalization of quantum mechanics for internal (spin) degrees of freedom has recently been developed by Weinberg [Phys. Rev. Lett. **62**, 485 (1989); Ann. Phys. (N.Y.) **194**, 336 (1989)]. We extend this theory to systems of composite, or multivalued, spin, such as atoms and molecules. Results for the generalized Hamiltonian function, energy eigenvalues, and state-vector time dependence are found to be similar for single- and multivalued spin systems, with the important difference that states of spin 0 or $\frac{1}{2}$ can exhibit observable nonlinear behavior in a composite spin system. Examples of the application of the composite spin analysis are given for the ground-state hyperfine structure of the hydrogen and deuterium atoms.

INTRODUCTION

Wigner[1] pointed out in a 1939 paper: "It is perhaps the most fundamental principle in quantum mechanics that the system of states forms a linear manifold." In a footnote to that statement, he commented: "the possibility of a future nonlinear character of the quantum mechanics must be admitted of course." Since that time, outstanding agreement has been found between specific quantum-mechanical theories and experimental measurements (e.g., quantum electrodynamics). But such results do not necessarily serve as precision tests of the linearity of quantum mechanics, as neither the theoretical nor the experimental methodology have been designed for that purpose. Recently, Weinberg[2,3] has developed a generalized theory of (nonrelativistic) nonlinear quantum mechanics (NLQM) as a framework for the development and analysis of experimental tests of the linearity of quantum mechanics. In this theory, as in linear quantum mechanics, physical states are represented by rays (state vectors) in a complex vector space, and physical observables by the (real) generating functions for operators acting on these rays. In NLQM, however, these operators are not in general linear: the distributive property of the operators is loosened, while the homogeneity condition and Galilean invariance are retained. Together, the distributive and homogeneity properties constitute the linear superposition principle. Homogeneity must be kept in a realistic theory as it rules out any special normalization of physical states and is essential to a proper treatment of noninteracting separated systems.[3] Weinberg finds that nonlinear terms are allowed in the Hamiltonian, and thus in the equation of motion, of the internal (spin) degrees of freedom of a particle 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 NLQM are analogous to the behavior of a classical anharmonic oscillator: energy eigenvalues can be shifted; the time dependence of state vectors is not in general an exponential function of a single, fixed frequency; and the resonant frequency of transitions between eigenstates depends on the relative populations of the states. This last property has recently been exploited experimentally by Bollinger *et al.*,[4] to put a low upper limit on the size of a nonlinear contribution to the quantum mechanics of the spin of the ^{9}Be nucleus; more experiments on different nuclear systems are currently being performed.[5,6]

In Weinberg's formulation of NLQM he does not address the possibility of nonlinear terms in the Hamiltonian of the translational (spatial) degrees of freedom or deal with the interacting multiple-body problem. Nor does the current form of the theory allow for systems of composite (i.e., multivalued) spin, such as atoms or molecules. In the present paper we consider the last of these problems. The construction of a nonlinear multivalued spin Hamiltonian is found to be a straightforward extension of Weinberg's theory for single-valued spin systems. Most physically significant results, regarding energy eigenvalues, the time dependence of state vectors, etc., are similar in the single- and multivalued spin cases. A notable difference is that nonlinear behavior is allowed for the spin-0 and spin-$\frac{1}{2}$ states of composite spin systems, because of the mixture of state probability among the different spin manifolds. This new result has allowed an experimental upper limit to be placed on the magnitude of nonlinear corrections to the quantum mechanics of the spin of the hydrogen atom.[7] The similarities and differences between the NLQM of single- and multivalued spins are demonstrated in two example systems: the hyperfine structure of the hydrogen atom (H) and the deuterium atom (D).

NLQM OF SINGLE-VALUED SPIN PARTICLES

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

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

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:

$$h(\lambda\psi,\psi^*)=h(\psi,\lambda\psi^*)=\lambda h(\psi,\psi^*)\ . \tag{2}$$

Euler's theorem shows that Eq. (2) is equivalent to the expression

$$\sum_k \psi_k\frac{\partial h}{\partial\psi_k}=\sum_k \psi_k^*\frac{\partial h}{\partial\psi_k^*}=h(\psi,\psi^*)\ . \tag{3}$$

In linear quantum mechanics, $h=\sum_{k,l}\psi_k^*H_{kl}\psi_l$, so that Eq. (1) reduces to the familiar equation of motion of ordinary quantum mechanics:

$$i\hbar\frac{d\psi_k}{dt}=\sum_l H_{kl}\psi_l\ . \tag{4}$$

Foregoing the distributive property of the linear superposition principle, 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^*:

$$h=\sum_r\left[\frac{h_r}{n^r}\right]=\left[\sum_{k,l}\psi_k^*H_{kl}\psi\right]+\left[\frac{1}{n}\sum_{k,l,m,n}\psi_k^*\psi_l^*G_{klmn}\psi_m\psi_n\right]+\cdots\ , \tag{5}$$

where the first term in large parentheses of the second equation is the linear QM term, the second is the lowest-order nonlinear term, and $n=\sum_k|\psi_k|^2$ is the normalization factor. The small nonlinear corrections to quantum mechanics, if they exist, will result in small nonlinearities in the equation of motion. Hence it will usually be sufficient to consider only the lowest-order nonlinear term in the expansion. This term will be referred to as $h_{\rm NL}$.

A realistic generalization of quantum mechanics must retain Galilean invariance. NLQM, as currently developed, assigns nonlinearities to the internal (spin) degrees of freedom of a particle, and not to the translational degree of freedom, so the requirement of Galilean invariance reduces to the condition that h be rotationally invariant: $[h,\mathbf{s}]=0$. For present purposes, this is equivalent to the requirement that $[h_{\rm NL},s]=0$. The condition can be satisfied by treating each factor $\psi_m\psi_n$ (similarly $\psi_k^*\psi_l^*$) in $h_{\rm NL}$ as a single "effective" state vector Ψ_{SM_S} for spin $S=2s,2s-1,\ldots,0$. The N nonzero values of Ψ_{SM_S} are those symmetric in the exchange of ψ_m and ψ_n. Thus the G_{klmn} terms in $h_{\rm NL}$ are proportional to the relevant Clebsch-Gordan coefficients $A(SM_S|kl)$, and $h_{\rm NL}$ becomes

$$h_{\rm NL}=\sum_{S,M_S}\left[\frac{\varepsilon_S}{n}\right]\left|\sum_{k,l}A(SM_S|kl)\psi_k\psi_l\right|^2\ . \tag{6}$$

Here the ε_S are unconstrained parameters that characterize the magnitude of the nonlinear terms.

A straightforward analysis shows that $\sum_{S,M_S}(1/n)|\Psi_{SM_S}|^2=\frac{1}{n}\sum_{k,l,m,n}\psi_k^*\psi_l^*\psi_m\psi_n=n$. Therefore, of the N terms in $h_{\rm NL}$, corresponding to the N nonzero values of Ψ_{SM_S}, only $N-1$ are independent. So one of the N terms in $h_{\rm NL}$, or any linear combination of these terms, can be expressed in a form proportional to n (bilinear in ψ and ψ^*). Such a term would result only in an unobservable offset in the energy of all states. Thus a system must have $N\geq 2$ for there to be any observable nonlinear effects from $h_{\rm NL}$. In terms of single-valued spin, this condition becomes $s\geq 1$, and so particles of spin 0 or $\frac{1}{2}$ cannot exhibit nonlinear behavior. For comparison with the composite spin examples given below, the $h_{\rm NL}$ expressions for $s=1$ and $\frac{3}{2}$ (equivalent to those derived by Weinberg[2,3]) are presented here:

$$h_{\rm NL}=\begin{cases}\left[\dfrac{\varepsilon_0-\varepsilon_2}{3n}\right]|2\psi_1\psi_{-1}-\psi_0^2|^2, & s=1 \qquad (7)\\[2ex] \left[\dfrac{\varepsilon_1-\varepsilon_3}{5n}\right](|\sqrt{6}\psi_{3/2}\psi_{-1/2}-\sqrt{2}\psi_{1/2}^2|^2+|3\psi_{3/2}\psi_{-3/2}-\psi_{1/2}\psi_{-3/2}|^2+|\sqrt{6}\psi_{-3/2}\psi_{1/2}-\sqrt{2}\psi_{-1/2}^2|^2), & s=\tfrac{3}{2}\ . \qquad (8)\end{cases}$$

The ψ subscripts refer to the m_s states. Note that only *differences* of the ε_S nonlinearity parameters appear in the $h_{\rm NL}$. Physically significant results such as changes in energy eigenvalues and frequency shifts are found to depend on these "mixed" ε parameters. Experimental tests[4-6] of the linearity of quantum mechanics, inspired by Weinberg's analysis, attempt to measure or put upper limits on the magnitude of the ε parameters. Also, only specific ε_S values appear in each $h_{\rm NL}$, those that correspond to the values of the effective state vector Ψ_{SM_S} that are symmetric in the exchange of the ψ_{m_s}.

EXTENSION OF NLQM TO SYSTEMS OF COMPOSITE SPIN

Quantum mechanics often involves the coupling of spins, resulting in a "particle" (e.g., an atom) that has multiple spin states. Extending NLQM to such compos-

ite spin systems, a significant difference is that each ψ (and ψ^*) in the expansion for h can now be in any of two or more spin states. That is, there is an additional degree of freedom in the construction of the effective state vector Ψ_{JM_J}. Thus for composite spin systems one has $\psi_m\psi_n=\psi_{j_1m_{j_1}}\psi_{j_2m_{j_2}}\rightarrow\Psi_{JM_J}$, where the spin quantum numbers j_1 and j_2 are no longer fixed. Homogeneity constrains each ψ to "pair up" with a ψ^* in the same term in the expansion for h, such that the ψ and ψ^* assume the same j quantum number (but not necessarily the same m_j). Extending the sum over states in $h_{\rm NL}$ to include this additional degree of freedom gives

$$h_{\rm NL}=\sum_{j_1,j_2}\sum_{JM_J}\left[\frac{\varepsilon_J^{j_1j_2}}{n}\right]\left|\sum_{m_{j_1},m_{j_2}}A(JM_Jj_1j_2|m_{j_1}m_{j_2})\times\psi_{j_1m_{j_1}}\psi_{j_2m_{j_2}}\right|^2. \quad (9)$$

Here the normalization factor is $n=\sum_{j,m_j}|\psi_{jm_j}|^2$, so that the different j_1j_2 manifolds in $h_{\rm NL}$ are coupled, as long as there is a nonzero probability for the atom to be in states within those manifolds. Note that the characteristic nonlinearity parameters $\varepsilon_J^{j_1j_2}$, are generalized to include the possibility that they depend on the constituent spin quantum numbers, as well as the effective spin value of the nonlinear term.

The j_1 and j_2 sums run over all the system's spin states. At each particular value of j_1 and j_2, the effective spin J is constrained to the values $j_1+j_2, j_1+j_2-1, \ldots, |j_1-j_2|$. For $j_1=j_2$ one can consider $\psi_{j_1m_{j_1}}\psi_{j_2m_{j_2}}$ to be a single effective state vector Ψ_{JM_J} with spin J and $2J+1$ M_J states. As int he NLQM of single-valued spins, only the M_J states that are symmetric in the exchange of $\psi_{j_1m_{j_1}}$ and $\psi_{j_2m_{j_2}}$ will be nonzero. Thus the $j_1=j_2$ terms in $h_{\rm NL}$ follow directly from the single-valued spin analysis. For $j_1\neq j_2$ the analysis is not so simple, since the $\psi_{j_1m_{j_1}}$ and $\psi_{j_2m_{j_2}}$ state vectors are not identical fields; therefore, antisymmetric combinations of $\psi_{j_1m_{j_1}}$ and $\psi_{j_2m_{j_2}}$ are allowed. These $j_1\neq j_2$ "cross terms" can have effective spin J values not present in any of the $j_1=j_2$ terms, thereby enriching the system's behavior beyond that for single-valued spin systems. The $j_1\neq j_2$ cross terms also mix the $\varepsilon_J^{j_1j_2}$ parameters, for most observables, and make the construction of a rotationally invariant $h_{\rm NL}$ (via Clebsch-Gordan coefficients) more involved.

The physically significant results of the extension of NLQM to systems of composite spin are found to be similar to that of the single-valued spin case, if somewhat more complex. A new result is that observable nonlinear effects are possible for the spin-0 or spin-$\frac{1}{2}$ states in a composite spin system, unlike for single-valued spin-0 or spin-$\frac{1}{2}$ particles. Examples of the application of NLQM to atomic systems of multivalued spin are given in the following two sections.

GROUND-STATE HYPERFINE STRUCTURE OF ATOMIC HYDROGEN

The H nucleus has a spin of $\frac{1}{2}$, and so it cannot exhibit nonlinear behavior, according to the NLQM of single-valued spin particles. The single H electron also has a spin of $\frac{1}{2}$, resulting in a composite atomic spin of 1 or 0: thus the H *atom* can have NLQM effects. The four hyperfine states (three composite spin $F=1$ levels and one $F=0$ level) are shown in Fig. 1 as a function of static magnetic field, as given by linear quantum mechanics in the "low-field" limit (where F and m_F are good quantum numbers). The states are labeled a,b,c,d in order of increasing energy. The derivation of the lowest-order nonbilinear term ($h_{\rm NL}$) in the generalized Hamiltonian function for the H ground-state hyperfine structure follows directly from the formalism developed above. Homogeneity and rotational invariance conditions imply that there are four distinct, nondegenerate terms in $h_{\rm NL}$. These four terms are labeled by the quantum numbers f_1,f_2, and the composite spin F: $f_1=f_2=1$, $F=2$ or 0; $f_1=1$, $f_2=0$, $F=1$; and $f_1=f_2=0$, $F=0$. But the sum of the four terms in $h_{\rm NL}$ is proportional to the norm n; this just adds a physically insignificant constant to the energy of all states. Thus there are only three physically significant terms in $h_{\rm NL}$, which can be simplified to the following:

$$h_{\rm NL}=\frac{\varepsilon_1'}{n}\left|2\psi_d\psi_b-\psi_c^2\right|^2+\frac{\varepsilon_0'}{n}\left|\psi_a\right|^4, \quad (10)$$

where $\varepsilon_1'\equiv\frac{1}{3}(\varepsilon_0^{11}-\varepsilon_2^{11})$ and $\varepsilon_0'\equiv\varepsilon_0^{00}-2\varepsilon_1^{10}+\varepsilon_2^{11}$. As will be shown below, the physically significant implications of this nonlinear term in the Hamiltonian depend on the "mixed" nonlinearity parameters ε_1' and ε_0'. The ε_1' term in Eq. (10) contains only $F=1$ state vectors and is analogous to the single-valued spin-1 $h_{\rm NL}$ [Eq. (7)]. The ε_0' term is like a single-valued spin-0 $h_{\rm NL}$ (here, however, $|\psi_a|^2\neq n$ in general). The ε_1' and ε_0' terms have the form of $f_1=f_2=1$ and 0 terms, respectively. The $f_1\neq f_2$ cross term has been absorbed into the ε_0' term (contribut-

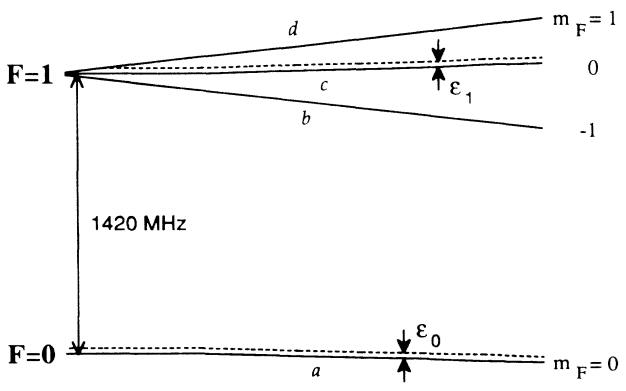


FIG. 1. Atomic hydrogen ground-state hyperfine energy levels as a function of magnetic field, for fields <200 G. Energy-level shifts due to possible nonlinear effects are shown by the dashed lines.

ing the $-2\varepsilon_1^{10}$ to ε_0'); it also adds an unimportant bilinear term to the a state energy. Comparing Eq. (10) to Eq. (7), one sees that the generalization from a single-valued $s=1$ spin to the $F=1$ or 0 multivalued spin case amounts to the addition of a term dependent on the a state ($F=0$, $m_F=0$) population, and a generalization of the ε parameters.

Solving for the eigenvalues and eigenvectors of the H $h_{\rm NL}$, one finds that the energies of the d and b states are unchanged by the nonlinearity, whereas $E_c \rightarrow E_c+\varepsilon_1'$ and $E_a \rightarrow E_a+\varepsilon_0'$ (see Fig. 1). This result is for nonlinearities small relative to the static magnetic field.[3] The energy eigenvalues do not satisfy the equal-level-spacing rule of ordinary quantum mechanics, as this rule is a consequence of both rotational invariance *and* linearity. Applying the NLQM equation of motion [Eq. (1)] to a Hamiltonian function that includes the hyperfine interaction, the Zeeman interaction, and the $h_{\rm NL}$ of Eq. (10), the general time behavior of the four states, in the low-field limit, is found to be

$$
\begin{aligned}
\psi_{d,b}(t)&=\frac{1}{\sqrt{2}}e^{-i\omega_A t}e^{\pm i\omega_B t}\{\alpha_0(\pm c_x+ic_y)e^{-i\omega_C t}+[1+(1-\alpha_0^2)^{1/2}](\pm c_x^*+ic_y^*)e^{i\omega_C t}\}\ ,\\
\psi_c(t)&=e^{-i\omega_A t}\{\alpha_0 c_z e^{-i\omega_C t}+[1+(1-\alpha_0^2)^{1/2}]c_z^* e^{i\omega_C t}\}\ ,\\
\psi_a(t)&=|\psi_a(0)|e^{-i\omega_A t}e^{-i\omega_D t}\ ,
\end{aligned}
\tag{11}
$$

where

$$
\omega_A=\omega_{F=1}+\varepsilon_1'(2-\alpha_0^2)-\varepsilon_0'\frac{|\psi_a(0)|^4}{n^2}\ ,
$$

$\omega_B=\mu B$, μ is the magnetic dipole moment in d,b states, B is the static magnetic field, $\omega_C=2\varepsilon_1'(1-\alpha_0^2)^{1/2}$,

$$
\omega_D=\omega_{F=0}-\omega_{F=1}-2\varepsilon_1'+2\varepsilon_0'\frac{|\psi_a(0)|^4}{n^2}\ ,
$$

$\alpha_0=|2\psi_d\psi_b-\psi_c^2|/n$ at $t=0$, and c is a three-vector fixed by the relative state populations. Note that the characteristic frequencies of the state vectors are no longer constants, as they are in linear quantum mechanics, but depend on the state mixture. Thus the energy difference between two states is not simply proportional to the resonant frequency at which the transition occurs: $E_2-E_1\neq\hbar(\omega_2-\omega_1)$. Rather, the resonant frequency must be averaged over the possible state distributions: $E_2-E_1=\hbar\langle\omega_2-\omega_1\rangle_{\text{state dist}}$. This result is similar to that found in the single-valued spin case, but here the spin-0 state can also have these effects, as long as there is a finite probability for the system to be in both the spin-1 and the spin-0 states. Thus the $c\rightarrow a$ hyperfine transition, commonly used in hydrogen maser clocks,[8] can exhibit nonlinear behavior.

Applying the time averaging method outlined by Weinberg[3] to Eq. (10), allowing the complex, time-dependent terms in $h_{\rm NL}$ to be averaged out, and using the NLQM equation of motion to solve for the nonlinear contribution to the resonant frequency of the $c\rightarrow a$ transition ($\omega_{ca}^{\rm NL}$), one finds

$$
\hbar\omega_{ca}^{\rm NL}=(\varepsilon_1'-\varepsilon_0')+\left[\frac{\varepsilon_1'+\varepsilon_0'}{n}\right](|\psi_c|^2-|\psi_a|^2)\ . \tag{12}
$$

Thus a dependence of the transition frequency on the $c-a$ population difference is created; the constant term in Eq. (12) would have no observable effect. Recently, an upper limit on the magnitude of $\omega_{ca}^{\rm NL}$ has been set using H masers.[7] Similarly, for the other allowed (one-photon) transitions within the H hyperfine structure, the nonlinear corrections to the resonant frequencies are

$$
\begin{aligned}
\hbar\omega_{dc}^{\rm NL}&=6\varepsilon_1'+\frac{2\varepsilon_1'}{n}(|\psi_b|^2-|\psi_c|^2)\ ,\\
\hbar\omega_{da}^{\rm NL}&=(2\varepsilon_1'+\varepsilon_0')+\left[\frac{2\varepsilon_1'-\varepsilon_0'}{n}\right](|\psi_b|^2-|\psi_a|^2)\ ,\\
\hbar\omega_{cb}^{\rm NL}&=6\varepsilon_1'+\frac{2\varepsilon_1'}{n}(|\psi_d|^2-|\psi_c|^2)\ ,\\
\hbar\omega_{ba}^{\rm NL}&=(2\varepsilon_1'+\varepsilon_0')+\left[\frac{2\varepsilon_1'-\varepsilon_0'}{n}\right](|\psi_d|^2-|\psi_a|^2)\ .
\end{aligned}
\tag{13}
$$

The nonlinear contributions to the resonant frequencies depend on the state populations and on the mixed nonlinearity parameters ε_1' and ε_0'. Note the interesting result that the transitions involving the d (b) state depend on the b (d) state population, but not on the d (b) state population.

GROUND-STATE HYPERFINE STRUCTURE OF ATOMIC DEUTERIUM

The spin-one deuteron can have observable nonlinear behavior, following the NLQM of single-valued spin particles. The deuterium atom (D) is a system of composite spin $F=\frac{3}{2}$ or $\frac{1}{2}$, and so it can exhibit nonlinear effects according to the multivalued spin analysis. Thus nonlinearities are allowed for both the single-valued spin nucleus and the composite spin atom, each leading to energy-level and resonant frequency shifts. These nuclear and atomic nonlinear effects are not mutually exclusive, and their relative and absolute magnitudes are undetermined as they depend on the various ε parameters. Deuterium has six

hyperfine states, four $F=\frac{3}{2}$ levels and two $F=\frac{1}{2}$ levels, that are labeled $\alpha,\beta,\gamma,\delta,\epsilon,\zeta$ in order of increasing energy. An energy-level diagram as a function of static magnetic field is given in Fig. 2, in the low-field limit where F and m_F are good quantum numbers.

Homogeneity and rotational invariance conditions yield five distinct, nondegenerate terms in the D $h_{\rm NL}$. Each term is labeled by the spin quantum numbers f_1 and f_2, and the composite spin quantum number F: $f_1=f_2=\frac{3}{2}$, $F=3$ or 1; $f_1=\frac{3}{2}$, $f_2=\frac{1}{2}$, $F=2$ or 1; and $f_1=f_2=\frac{1}{2}$, $F=1$. As with the NLQM analysis for H, the sum of all terms in $h_{\rm NL}$, with equal prefactors, is proportional to the norm n. Since a term proportional to n has no physically significant consequences, only four of the terms in $h_{\rm NL}$ need to be retained. Dropping the most algebraically involved term (the $f_1=f_2=\frac{3}{2}$, $F=3$ term), $h_{\rm NL}$ reduces to the following:

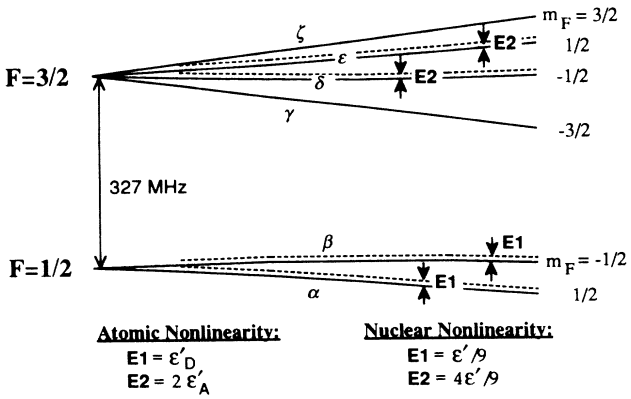


FIG. 2. Atomic deuterium ground-state hyperfine energy levels as a function of magnetic field, for fields < 100 G. Energy-level shifts due to possible atomic *and* nuclear spin nonlinear effects are shown by the dashed lines.

$$h_{\rm NL}=\frac{\varepsilon'_A}{n}(|\sqrt{6}\psi_\zeta\psi_\delta-\sqrt{2}\psi_\epsilon^2|^2+|3\psi_\zeta\psi_\gamma-\psi_\epsilon\psi_\gamma|^2+|\sqrt{6}\psi_\gamma\psi_\epsilon-\sqrt{2}\psi_\delta^2|^2)$$
$$+\frac{\varepsilon'_B}{n}(2|\psi_\zeta|^2|\psi_\alpha|^2+\tfrac{1}{2}|\psi_\zeta\psi_\beta+\sqrt{3}\psi_\epsilon\psi_\alpha|^2+|\psi_\epsilon\psi_\beta+\psi_\delta\psi_\alpha|^2+\tfrac{1}{2}|\psi_\gamma\psi_\alpha+\sqrt{3}\psi_\delta\psi_\beta|^2+2|\psi_\gamma|^2|\psi_\beta|^2)$$
$$+\frac{\varepsilon'_C}{n}(\tfrac{1}{2}|\sqrt{3}\psi_\zeta\psi_\beta-\psi_\epsilon\psi_\alpha|^2+|\psi_\epsilon\psi_\beta-\psi_\delta\psi_\alpha|^2+\tfrac{1}{2}|\sqrt{3}\psi_\gamma\psi_\alpha-\psi_\delta\psi_\beta|^2)+\frac{\varepsilon'_D}{n}(|\psi_\alpha|^2+|\psi_\beta|^2)^2\ , \tag{14}$$

where $\varepsilon'_A\equiv\frac{1}{5}(\varepsilon_1^{3/2,3/2}-\varepsilon_3^{3/2,3/2})$, $\varepsilon'_B\equiv\varepsilon_2^{3/2,1/2}-\varepsilon_3^{3/2,3/2}$, $\varepsilon'_C\equiv\varepsilon_1^{3/2,1/2}-\varepsilon_3^{3/2,3/2}$, and $\varepsilon'_D\equiv\varepsilon_1^{1/2,1/2}-\varepsilon_3^{3/2,3/2}$. The ε'_A term contains only $F=\frac{3}{2}$ state vectors $(\psi_\zeta,\psi_\epsilon,\psi_\delta,\psi_\gamma)$, and is analogous to a single-valued spin-$\frac{3}{2}$ $h_{\rm NL}$ [see Eq. (8)]. The ε'_D term contains the $F=\frac{1}{2}$ state vectors (ψ_α,ψ_β); it is like the $h_{\rm NL}$ for a single-valued spin-$\frac{1}{2}$ particle (here, however, $|\psi_\alpha|^2+|\psi_\beta|^2\neq n$ in general). The ε'_A and ε'_D terms have $f_1=f_2$, and thus follow directly from the single-valued spin analysis. Equation (14) also has cross terms (the ε'_B and ε'_C terms). Unlike the cross term in the above H example, the ε'_B and ε'_C terms are irreducible, because of the higher dimensionality of the spin states ($\frac{3}{2}$ and $\frac{1}{2}$ as opposed to 1 and 0). The cross terms lead to a complication of the mixed nonlinearity parameters associated with observable frequency shift effects, and to phenomena where the population of states uninvolved in a particular transition can nonetheless affect the resonant frequency. A consequence of the composite spin nature of D is that observable nonlinear behavior is allowed for the spin-$\frac{1}{2}$ state, provided there is a finite probability to be in the spin-$\frac{3}{2}$ state.

Analyzing Eq. (14) for nonlinear shifts of the six (F,m_F) energy levels, one finds that $E_{\zeta,\gamma}$ are unchanged; $E_{\epsilon,\delta}\to E_{\epsilon,\delta}+2\varepsilon'_A$; and $E_{\beta,\alpha}\to E_{\beta,\alpha}+\varepsilon'_D$ (see Fig. 2). The general time behavior of the D state vectors, for the atom in a weak, uniform magnetic field, follows from the NLQM equation of motion, as the problem is integrable. There are six "unknowns" in this system (the state vector amplitudes), and seven conserved quantities (all commuting with each other): h, including $h_{\rm NL}$ and the hyperfine and Zeeman interactions (in the weak field limit); the norms in the $F=\frac{3}{2}$ and $\frac{1}{2}$ manifolds, $n_{3/2}$ and $n_{1/2}$; the z components of angular momenta in these manifolds, $j_z|_{3/2}$ and $j_z|_{1/2}$; and $\mathbf{j}\cdot\mathbf{j}|_{3/2}$ and $\mathbf{j}\cdot\mathbf{j}|_{1/2}$. Nonlinear corrections to the ten allowed (one-photon) D hyperfine transition frequencies can be calculated employing Weinberg's averaging technique.[3] The frequency shift expressions are lengthy, including terms for each of the six (F,m_F) states, in general. As an example, the expression for the ϵ-β transition[9] is given:

$$\omega_{\epsilon\beta}^{\rm NL}=\frac{1}{n}[-\tfrac{1}{2}(\varepsilon'_B+3\varepsilon'_C)|\psi_\zeta|^2+(4\varepsilon'_A-\varepsilon'_B-\varepsilon'_C)|\psi_\epsilon|^2$$
$$+(\varepsilon'_A-\tfrac{3}{2}\varepsilon'_B-\tfrac{1}{2}\varepsilon'_C)|\psi_\delta|^2+2(3\varepsilon'_A-\varepsilon'_B)|\psi_\gamma|^2$$
$$+(\varepsilon'_B+\varepsilon'_C-2\varepsilon'_D)|\psi_\beta|^2$$
$$+(\tfrac{3}{2}\varepsilon'_B+\tfrac{1}{2}\varepsilon'_C-2\varepsilon'_D)|\psi_\alpha|^2]\ . \tag{15}$$

Thus the possible nonlinearity of the D Hamiltonian results in a dependence of hyperfine transition frequencies on the relative state populations, including states not involved in the transition in question. An analysis of the allowed nonlinearity of the spin-1 deuteron in terms of the atomic deuterium states gives similar expressions for the nonlinear corrections to the D transition frequencies (with the mixed ε' parameters now proportional to one another). Unlike the H case, therefore, NLQM analyses

of the composite spin atom and the single-valued spin nucleus do not predict qualitatively different observable behavior. For a system where the nonlinearity can be assigned to either the nucleus or the atom, differences arise between the single- and multivalued spin analyses only in the details (e.g., the ε parameters). Such a distinction would be interesting and useful to study only if precision tests of quantum mechanics began to show evidence of nonlinear behavior.

DISCUSSION

Weinberg has shown that nonlinear terms in the generalized Hamiltonian function of a particle of single-valued spin can be made consistent with the conditions of homogeneity and Galilean invariance. The present paper has extended this treatment to systems of composite spin. Among the observable consequences of such nonlinearity is the dependence of the resonant frequency of transitions between eigenstates on the populations of certain of the system's states. Important questions can be raised about this nonlinear generalization of quantum mechanics. For example, the theory puts no constraints on the magnitude of the nonlinear effects. In addition, the compatibility of NLQM with the second law of thermodynamics[10] and special relativity[11] has recently been questioned.

Weinberg has suggested a scaling of limits on the ε parameters relative to the characteristic energy to change the spin of the particle under consideration.[2,3] This form of comparison may be correct if nonlinearities are inherent in quantum mechanics, independent of the system under study. Then linear and nonlinear effects would scale similarly with the characteristic energy of the system (and single-valued spin nuclear systems would typically exhibit nonlinear behavior of a greater absolute magnitude than composite spin atomic systems). But if the nonlinearity results from new physics, such as an internal coupling of the system's (linear) states, then the magnitude of the effect may depend on the nature of this coupling and the system in question. Also, it should be noted that observable consequences of NLQM can only exist if the ε parameters vary with the effective particle spin (and/or with the constituent particle spins in the composite spin case). The mixed ε' parameters are associated with physically significant results; however, they go to zero if the underlying ε parameters have no spin dependence. This is equivalent to all nonlinear terms in the generalized Hamiltonian function being proportional to the norm n.

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