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Intermediate Spin, Schrödinger Cat States, and Nanomagnets

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Quantum tunneling in nanomagnets finds a natural description in terms of intermediate spin. Periodic magnetic effects correspond to a change of flux by the flux quantum \( \Phi_0 \). Schrödinger cat states with different superpositions of the applied magnetic field occur. The molecular magnet \( \text{Fe}_8 \) is discussed and new effects are predicted for \( \text{Mn}_{12} \).

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There is currently considerable interest in quantum effects for nanoscale systems. In this context quantum tunneling in small magnetic clusters such as \( \text{Fe}_8 \) and \( \text{Mn}_{12} \) is of particular interest [1–5]. The purpose of this Letter is to point out that these nanomagnets exhibit a number of fairly unique quantum effects. The problem maps rather accurately to that of a localized anyon characterized by a statistical parameter \( \alpha_s \) and viewed as a spinless charged particle bound to a \( \Phi \) flux tube in two dimensions [6]. Modulo unity, \( \alpha_s = 0 \) \((\alpha_s = 1)\) corresponds to bosons (fermions) and whole (half) integer spin. Other values of \( \Phi \) and \( \alpha_s \) define intermediate spin. The problem is periodic in the \( \Phi \) flux with period \( \Phi_0 \), the flux quantum. For the nanomagnet it is the applied external magnetic field component \( B \) along a certain symmetry direction (the hard transverse direction for \( \text{Fe}_8 \)) which breaks time reversal symmetry and attaches a flux \( \Phi(B) \) to the equivalent particle. The tunnel splitting for all levels is periodic with period \( \Phi_0 \). A flux \( \Phi(B) = \pm \Phi_0/2 \) transmutes a physical whole integer spin \( S \) system with \( \alpha_s = 0 \) into its half integer spin \( S - \frac{1}{2} \) and \( \alpha_s = 1 \) equivalent and following Kramer’s theorem the tunnel splitting of all states is necessarily zero [7] at such points. Theoretically and experimentally [1] for the simplest case corresponding to \( \text{Fe}_8 \), to a good approximation, \( \Delta(B) = \Delta[\cos \pi \Phi/\Phi_0] \) and \( \Delta(B) \) is indeed zero for half-integer points. While a change \( \Delta \Phi = \Phi_0 \) maps the problem back to itself, it also causes a change \( \Delta M = g \mu_B \hbar \) in the magnetic moment, per period, where \( \mu_B \) is the Bohr magneton and \( g \) an appropriate \( g \)-factor. It is possible to change this relationship between \( \Delta \Phi \) and \( \Delta M \) by constructing a Schrödinger cat, defined to be a superposition of states where a macroscopic quantity is controlled by a microscopic degree of freedom. The nanomagnet \( \text{Fe}_8 \) is a realization of such a cat since the eigenstates comprise superpositions of states with opposite flux values and the sign of the flux is controlled by the quantum state of the spin. The immediate experimental consequence is that the tunnel splitting has a period which is less than \( g \mu_B \hbar \).

For the \( \text{Fe}_8 \) symmetry the other components of the field \( B \) play a different role. Special values of the field component \( B_t \) along the easiest axis also cause the system to be transmuted from whole to half-integer spin or the inverse but in such a fashion that quantum tunneling is effectively absent for intermediate values. The (transverse) component \( B_t \) along the remaining orthogonal direction can be incorporated in the mapping to the particle problem as an imaginary field \( B \), and so for such a field alone \( \Delta(B) = \Delta[\cos \pi \Phi/\Phi_0] = \Delta \cosh \pi \Phi/\Phi_0 \). In general the effective \( \Phi \) is complex. For the \( \text{Mn}_{12} \) symmetry, see below, a real physical field leads in an unavoidable fashion to such a complex \( B \) field.

The molecular magnet \( \text{Fe}_8 \), reflecting a spin model,

\[
\mathcal{H} = -(D - E)S_z^2 + 2E S_x^2 - ES_z - hS_z - h_xS_x - h_yS_y ,
\]

is a good illustration of intermediate spin. The positive anisotropy constants have \( E < D \) and, e.g., \( h = g \mu_B B \) is a field component in energy units. In the absence of fields, classical equilibrium has \( S_x = \pm S \). Tunneling occurs along two equivalent paths perpendicular to the \( z \) axis. A \( z \)-axis applied field \( B = h/g \mu_B \) determines \( \Phi/\Phi_0 = g \mu_B B/2h\sqrt{2E(D + E)} \) and a number of formalisms, including the present one, then yield \( \Delta(B) = \Delta_0[\cos \pi \Phi/\Phi_0] \) which agrees well with experiment and direct diagonalization [1] for both the ground and excited states. [Rigorously [8] the period, for Eq. (1) is \( 2h\sqrt{2E(D + E)}/g \mu_B \), independent of which levels are involved, the parameters, and even if the system is in the tunneling regime. Experimental deviations [1] reflect additional, e.g., \( C(S_x^4 + S_y^4) \), terms.]

For \( \text{Mn}_{12} \) an appropriate large spin model is [2]:

\[
\mathcal{H} = -DS_z^2 + a(S_x^4 + S_y^4) + hS_z ,
\]

with \( a \) and \( D \) positive. The magnetic field lies along one of the four equivalent hardest axes, and controls the intermediate spin. For \( h = 0 \), the \( four \) tunneling paths are close to one of two planes. Each plane makes an angle of 45° with the field and each of the hard axes. These planes taken separately would be equivalent to that in the \( \text{Fe}_8 \) problem but because of the 45° field angle the flux \( \Phi \) has equal real and imaginary parts. Adding the amplitude for each plane gives the final result [9] illustrated, along with those of exact diagonalization, in Fig. 1. There are periodic, zero splitting, half-integer points but now the maximum splitting increases by almost an order of magnitude per period.
A similar behavior follows when the field is directed along a hard threefold or fourfold axis of a cubic material [9].

The remainder of this Letter develops the mapping to the intermediate spin problem for the model of Eq. (1). More details and a longer discussion of other models is to be found elsewhere [9].

The simplest model for a localized anyon and thereby the intermediate spin problem [6] corresponds to a spinless particle bound to a unite circle in two dimensions. The generator of rotations $S_z$ is the single operator for the $U(1)$ Abelian group. A rotation $R(\phi) = e^{iS_z \phi}$ involves $\phi$ the angle to the $x$ axis. The momentum conjugate to $\phi$ is

$$p_\phi = S_z,$$

i.e., $[\phi, p_\phi] = i\hbar$. The eigenstates of $p_\phi$ and $S_z$ are $\psi_m(\phi) = \frac{1}{\sqrt{(2\pi)n}} e^{im\phi} \text{ but } m = n + \frac{1}{2}, n \text{ an integer, reflecting the continuous spectrum appropriate for this group [6].}$ This defines the statistical parameter $\alpha_s$. In this singular gauge $\psi_m(\phi)$ is multivalued. An equivalent single valued function is defined by the analytic continuation of some branch from the physical interval $[0, 2\pi]$. This continued wave function is the solution of a Schrödinger’s equation $[e - (p_\phi^2/2m)]\psi_m(\phi) = V(\phi)\psi_m(\phi)$ in which the potential $V(\phi)$ has period $2\pi$, i.e., reflects an extended periodic solid. Floquet’s (Bloch’s) theorem then implies $\psi_m(\phi) = e^{ik\phi}\psi_m(\phi)$ where $u_k(\phi)$ is periodic. The reciprocal lattice vector $k = 1$ and the energy $\epsilon(k)$ has period $K$. This periodicity implies $u_k(\phi) = \sum_{n \text{-integer}} A_n e^{in\phi}$ and so $e^{ik\phi}u_k(\phi) = \sum_{n \text{-integer}} A_n e^{i(n+k)}$ which leads to the identification: $k = \alpha_s/2$. The strategy is then to write Schrödinger’s equation in this singular gauge whence the periodic effects are all contained in $e^{i\alpha_s/2}$.

In the nonsingular gauge the basis set $\psi_m(\phi)$ has $n = \pm 1, \pm 2, \ldots$ and in Schrödinger’s equation $[\varepsilon + h p_\phi - (p_\phi^2/2m)]u_k(\phi) = V(\phi)u_k(\phi)$ the energy $\varepsilon = \epsilon + (\hbar^2k^2/2m)$ contains a $k^2$ shift. The term $h p_\phi$ with $h = \hbar/k$ reflects a magnetic flux $(\Phi/\Phi_0) = k = \alpha_s/2$. Since $\epsilon(k) = \epsilon(-k)$, the Schrödinger cat, $u_k(\phi) + Bu_{-k}(\phi)$ is an eigenstate which superimposes solutions with oppositely directed applied fields for which the expectation value $\langle \Phi \rangle$ is smaller than $\alpha_s\Phi_0/2$.

For the spin model Eq. (1), it is necessary to carefully formulate the problem in terms of the basis set $[S_z = m]$. If $|\psi(m)\rangle = \sum_{m = -S}^{S} a(m)|m\rangle$ [defining $\psi(m)$ then Schrödinger’s equation with $\hbar = \hbar_1 = 0$ is

$$[\varepsilon - (2\hbar^2m^2 - \hbar\hbar m)]a(m) = \frac{1}{2}\hbar^2(D - E)[M_{m+1}^2M_{m+2}a(m + 2) + M_{m-1}^2M_{m-2}a(m - 2) + [M_{m+1}^2 + M_{m-1}^2]a(m)],$$

where $M_s = [S(S + 1) - st]^{1/2}$. The structure of this equation is worth noting. It comprises two distinct tight binding models. The distribution $\psi(m)$ has a well-defined Fourier transform $\hat{\psi}(\phi) = \pi^{-1/2} \int dm e^{-im\phi} \psi(m) = \pi^{-1/2} \sum \xi e^{-im\phi} a_m$. When acting on $\psi(\phi)$, $S_z = -\hbar^2\frac{d}{d\phi}$ which is just the explicit definition for the momentum $p_\phi$. With, e.g., $S_+ = e^{i\phi}[\hbar^2S(S + 1) - p_\phi(p_\phi + 1)]^{1/2}$ an arbitrary spin Hamiltonian can be expressed in terms of $p_\phi$ and $\phi$. Special care is needed when $h = E = 0$ tunneling is absent. The wave function for the relevant eigenstate $[S_z = S]$ is $a^0(m) = \frac{1}{\sqrt{2(S-m)(S+m+1)}}^{1/2}$. With Stirling’s formula $a^0(m) \approx e^{-m^2/2S}$, however, this approximation is inadequate since it lifts the degeneracies. To avoid this pitfall $f(m)$ is defined via $a(m) = a^0(m)f(m)$. With this, Schrödinger’s equation reduces exactly to

$$[\varepsilon - 2\hbar^2p_\phi^2 + h p_\phi]f(\phi) = V(\phi, p_\phi)f(\phi),$$

with

$$V(\phi, p_\phi) = \frac{1}{2}(D - E) \times [2\sin^2\phi[\hbar^2S(S + 1) + p_\phi^2] - i\hbar(2S - 1)\sin2\phi p_\phi],$$

where $\varepsilon = (e - E^0); E^0 = -\hbar^2S^2(D + E)$. This is of the intermediate spin form but with a momentum dependent potential $V(\phi, p_\phi)$. This is quite consistent with earlier Schrödinger equation treatments [10,11] which reduce the problem to the motion of a particle in a periodic potential. In this nonsingular gauge $\psi(\phi) = e^{ik\phi}u_k(\phi)$, and the whole or half-integer nature of the physical spin only appears implicitly as an appropriate value of $k$, i.e., with $k = (\alpha_s/2) - 0 [k = (\alpha_s/2) = 1/2]$ the values of $m$ which appear in $\psi(\phi)$ are whole (half) integer.

In the singular gauge the field $h$ is eliminated in favor of a change $\Delta k = (\Delta \alpha_s/2)$ in the statistical parameter: The passage to this gauge is facilitated by defining $g(\phi)$ such that $f(\phi) = [1 \pm (\Delta \alpha_0/2\hbar S)]g(\phi)$. To leading order in $S$ and using the fact that $V(\phi, p_\phi)$ localizes the wave
function near $\phi = 0$ and/or $\pm \pi$, the solution of,

$$\left[ \mathcal{E} - 2E(p_\phi \pm \hbar \Delta \alpha_1/2)^2 + \hbar^2 E \Delta \alpha_1^2/2 \right] g(\phi) = V\left( \phi, p_\phi \pm \hbar \Delta \alpha_1/2 \right) g(\phi),$$

is a solution to Eq. (4) if $h = \pm 2\hbar(D+E)\Delta \alpha_1/2$. In the singular gauge Eq. (6) becomes $(\mathcal{E} - 2E p_\phi^2)u_{z,k}(\phi) = V(\phi, p_\phi)u_{z,k}(\phi)$ with $u_{z,k}(\phi) = e^{-ik\phi}g(\phi)$ and $\mathcal{E} = \mathcal{E}_0 + \hbar^2 E \Delta \alpha_1^2/2$. The solution gives $\mathcal{E} = \epsilon(k) = \epsilon(-k)$ where $\epsilon(k)$ includes both the energy of the harmonic motion within a well and that associated with tunneling between wells.

The field induced energy shift $\hbar^2 E \Delta \alpha_1^2/2$ is determined by solving Eq. (4) in the harmonic approximation, say, near $\phi = 0$. This gives for the energy $E_n = E_0 - \frac{h^2}{4(D+E)} + (n + \frac{1}{2})\hbar \omega_n$ where $\omega_n = \hbar \sqrt{D^2 - E^2}$ and $n$ is the quantum number for the $n$th harmonic oscillator level. Equating $\hbar^2 E \Delta \alpha_1^2/2$ and $\hbar^2/4(D+E)$ gives $\Delta k = \frac{\Delta \alpha_1}{2} = \hbar/2 \hbar \sqrt{2E(D + E)}$. This is the shift in the $m$ from whole or half-integer values. But, as a consequence the Zeeman term in Eq. (4) is $\pm 2\hbar(D+E)\Delta \alpha_1^2/2$ which is in larger magnitude than the $h \alpha p$, which actually appears in Eq. (4). The fact that there are two solutions with the same energy but with different signs for this Zeeman term permits the approximative solution of Eq. (4), in terms of those of Eq. (6), by the construction of a Schrödinger cat state. It is observed that while, for $h \neq 0$ and a given energy, these are independent solutions they are by no means orthogonal. In fact, their expansions in terms of the $h = 0$ solution differ only by small terms involving odd derivatives and hence higher (odd) excited states. A perturbation theory can easily be developed for a cat wave function of the form

$$A(1 - \frac{\Delta \alpha_1 p_\phi}{2\hbar S})u_k(\phi) + B(1 + \frac{\Delta \alpha_1 p_\phi}{2\hbar S})u_{-k}(\phi)$$

by Taylor expanding the Zeeman term in terms of this wave function plus odd derivatives of the $h = 0$ solution. In this way it is easy to obtain $A = 1 + \sqrt{2E(D+E)}$ and $B = 1 - \sqrt{2E/(D+E)}$ and to show that convergence requires $2E \rho < S^2(D - E)$ where $p$ is the number of whole flux quanta for a given field $h$.

In the tunneling regime, the tight binding approximation is valid and band energy $\epsilon_k = \epsilon(k) - (n + \frac{1}{2})\hbar \omega_0$ is determined by the very small tunneling matrix elements between wells. Before describing this, the other fields, $h_{\ell}$ and $h_1$, need to be included. With $h_{\ell} > 0$ finite, but still with $h_1 = 0$, the effect is that $V(\phi, p_\phi) \Rightarrow V(\phi, p_\phi) + \hbar \hbar_{\ell} \cos \phi$ which removes the symmetry between $\phi = 0$ and $\pm \pi$. The harmonic levels near $\phi = \pm \pi$ have quantum numbers designated by $n$ and are higher than those near $\phi = 0$ with labels $n'$. Ignoring tunneling these have energies, $E_n = E_0 - \frac{h^2}{4(D+E)} + \hbar \hbar_{\ell} + (n + \frac{1}{2})\hbar \omega_0$, and, $E_{n'} = E_0 - \frac{h^2}{4(D+E)} - \hbar \hbar_{\ell} + (n' + \frac{1}{2})\hbar \omega_0$. There are now two possibilities (i) $E_n \neq E_{n'}$ and there is a very narrow band formed about both these ener-

The energy differences have a very small dependence on $k$ and hence $h$, see below. (ii) When $E_n = E_{n'}$ there is resonant tunneling. There are again two cases (iia) when both $n$ and $n'$ are odd or even and (iib) when one of $n$ and $n'$ is odd and the other even, see Fig. 2. The tunneling matrix elements are defined to be $\Delta_{n,n'}$ for the barrier between $\phi = 0$ and $\pi$, and $\Delta_{n',n}$ for that between $\phi = -\pi$ and $0$ (or $\pi$ and $2\pi$). If now $h = 0$, Eq. (6) admits real solutions $g(\phi)$, and these matrix elements can be made real. From the symmetry of the potential it must be that $\Delta_{n,n'} = \mp \Delta_{n',n}$. The relative sign is reflected in the ground state wave functions as illustrated in Fig. 2. For case (iia) this wave function does not change sign between $\phi = -\pi$ and $\pi$ reflecting matrix elements such that $\Delta_{n,n'} = \Delta_{n',n}$. For (iib), the wave function changes sign as $\phi$ advances by $2\pi$ implying that $\Delta_{n,n'} = -\Delta_{n',n}$ and that the ground state wave vector $k_0 = 1/2$ is finite. The alternation in sign of the $\Delta_{n,n'}$ can be eliminated by adopting a convention in which the phase advances by $\pi k$ from one well to the next within the cell, whence for (iia): $\Delta_{n,n'} = \Delta_{n',n} = i\Delta$. With the same convention, for finite $h$, $\Delta_{n,n'} = \Delta_{n',n} = e^{i\pi k/2} \Delta$ for case (iia), while $\Delta_{n,n'} = \Delta_{n',n} = e^{i\pi k + (1/2)} \Delta$ for case (iib), i.e., there is an additional statistical shift $(\Delta_{n,n}/2) = k_0 - 1/2$ beyond the $\Delta k = h/2 \hbar \sqrt{2E(D + E)}$ associated with $h$.

At this point it is possible to introduce the second transverse field $h_{\ell}$ by slight of hand. It is observed that if $E$ changes sign then the two transverse axes change role and thus $h$ becomes the equivalent of $h_{\ell}$. If $\mathcal{H}$ is considered to be a function of a complex parameter $E$ the solution might be analytically continued from positive to negative values. In particular, $k = h/2 \hbar \sqrt{2E(D + E)}$ becomes pure imaginary, i.e., $k = ik_{k_0} = \hbar |k_0|/2 \hbar \sqrt{2E(D - E)}$ and it follows, e.g., for (iia) $\Delta_{n,n'} = e^{i\pi (k + i k_{k_0})} \Delta$ with $\Delta_{n',n} = e^{-i\pi (k + i k_{k_0})} \Delta$. This is reflected by a two level model:

$$\mathcal{H} = \frac{(E_n - E_{n'})}{2} \sigma_z + \Delta \cdot \sigma^+ + \text{H.c.},$$

where the center of gravity energy $(E_n + E_{n'})/2$ has been dropped and where the $\sigma$ are the Pauli matrices.

FIG. 2. When, e.g., $n = 0$ and $n' = 2$, both wave functions are even and there is no sign change. If $n = 0$ and $n' = 1$ the wave function must change sign as the $\phi$ argument advances by $2\pi$. This produces a $\pi$ phase change in addition to that due to the field $h_{\ell}$ and transmutes, e.g., a whole into a half-integer spin.
The solution of Eq. (8) gives eigenvalues
\[ \epsilon_k = \pm \frac{1}{2}\sqrt{(E_n - E_0)^2 + 2\Delta^2 \cos \pi(k + ik_i)^2}, \]
for at resonance \( \epsilon_k = \pm (\Delta/2)\cos \pi(k + ik_i), \) with \( k = h/\sqrt{2h(E(D + E)} \) and \( k_i = h_1/2h\sqrt{2E}(D - E) \).

For the simplest case with \( h_1 = h_0 = 0 \) (i.e., \( n = n' \)) and for whole integer \( S \) for which \( k = (\Delta \alpha_s/2) = h/2h\sqrt{2E(D + E)} \) the doublet splitting is
\[ \Delta = \Delta_n,n' \cos \frac{\pi h}{2h\sqrt{2E(D + E)}}, \tag{9} \]
i.e., of the form mentioned in the introductory remarks. For half integer \( S \), \( k = (1/2) + (\Delta \alpha_s/2) = (1/2) + \frac{h}{2h\sqrt{2E(D + E)} \) and the cosine is replaced by a sine. An equivalent result for the ground state tunneling was obtained some time ago by Garg [12] and attributed to “topological quenching.”

For a finite \( h_1 \) and case (ii), at resonance \( E_n = E_0 \), there is the extra shift \( \Delta k_h = 1/2 \) which interchanges the role of whole and half integer \( S \) as is observed [1].

Returning to case (i), i.e., away from resonance, Eq. (8) gives
\[ \epsilon_k = E_n + (\Delta_{n,n'}^2/2)[\cos \pi(k + ik_i)^2/(E_n - E_0)] \]
for the level near \( E_n \) and where the center of gravity has been included. (Implicitly the level \( n' \) is the closest in energy. More generally there is a sum over \( n' \).) Since for typical values, away from resonance, \( \Delta_{n,n'} \ll (E_n - E_0) \) the tunneling energy correction is extremely small.

The outstanding problem remains the calculation of \( \Delta_{n,n'} \). The presence of \( p_\phi \) in the potential complicates the matter and depending upon the ratio \( E/D \), there are three different regimes. For each regime the appropriate \( k \) and \( h = 0 \) problem reduces to one found in the literature [8,10,11,13] as described elsewhere [9].

There are a number of predictions which are subject to experimental verification. The existence of a Schrödinger’s cat is the most striking claim. In the absence of intrinsic length scale, the projection of the cat by a local field measurement is not possible. However, this cat state is reflected by a reduction of the period as measured by the magnetic moment along the field direction. The dimensionless moment \( m = M_i/g \mu_B h \) which should have period one is reduced to \( \Delta m = \sqrt{2E(D + E)} \leq 1 \). Here \( Fe_B \) is a suitable system. For the very heavily studied Mn12, the applied magnetic field is necessarily complex in the equivalent intermediate spin mapping. This is reflected by a tunnel splitting, Fig. 1, which increases very strongly with field but retains zeros at half-integer points. These zeros have never been observed.

There are also implications for concentrated magnets. Consider a \( \widetilde{H} \) which has, e.g., Eq. (1) as the site diagonal part and \( J S_i \cdot S_j \) interactions. If \( J < D \) and \( S \) is not too small, a finite \( h \) will lead to intermediate spin values. The phase diagram will be periodic in \( h \) and again a period \( \Delta m = \sqrt{2E(D + E)} \), e.g., for one dimensions where whole integer points are reflected by a gap [14] and a magnetization plateau. These do not occur at simple multiples of \( g \mu_B h \) as is often speculated to be the case.

[7] An interpretation of such points in terms of Berry phase interference, for the ground state, was given by D. Loss, D. P. DiVincenzo, and G. Grinstein, Phys. Rev. Lett. 69, 3232 (1992); J. von Delft and C. L. Henley, Phys. Rev. Lett. 69, 3236 (1992); generalized in Ref. [12].
[14] When \( D = E = 0 \), and integer spin, this corresponds to a “Haldane” gap [F. D. M. Haldane, J. Appl. Phys. 57, 3359 (1985)]. For an easy plane anisotropy, i.e., when \( E \approx D \) and \( D \gg J \) the ground state is a singlet at each site and there is trivially a gap. It is known that this “large-\( D \)” phase is separated from the “Haldane” phase by a critical value \( D_c = 2J \) for which the gap is zero, see, e.g., M. Oshikawa et al., Phys. Rev. Lett. 78, 1984 (1997); K. Totsuka, Phys. Rev. B 57, 3454 (1998); T. Tsuneto and T. Murao, Physica (Amsterdam) 51, 3454 (1998); A. Garg, Europhys. Lett. 22, 205 (1993).