Manifestation of intermediate spin for Fe$_8$.

S. E. Barnes

Department of Physics, University of Miami, Coral Gables, Florida 33124

(March 24, 2022)

Intermediate spin, which occurs in the theory of anyons, can also be exhibited by mesoscopic magnetic particles. The necessary broken time reversal symmetry is due to a suitably directed magnetic field. As a function of this field a system passes periodically through points which correspond to whole or half-integer spin. Intermediate spin is defined by fields which lie between these points. Since the tunnel splitting in the ground doublet vanishes for half-integer spin, this splitting becomes periodic. The doubly periodic oscillations observed in the magnetic molecular cluster Fe$_8$ represent the first unequivocal observation of this phenomena. Here the whole or half-integer nature of the system, i.e., the parity, is periodic for a field which is along either the easy or hard axis or a suitable combination of the two. A detailed theory is presented.

The search for quantum effects in mesoscopic systems has become quite fashionable. In particular, relatively recently advances in several technologies have encouraged the investigation of the cross-over between the classical and quantum regimes in magnetic molecular clusters. A particularly interesting quantum effect which can occur in such large spin clusters has been emphasized by Loss et al. and von Delft and Henley. For half-integer spin the amplitude for tunneling with a complete reversal of the magnetization of a small ferromagnet is zero due to interference effects arising from the so called topological term in the effective action. This parity effect implies that, while a whole integer spin system can have a tunnel split ground state doublet, the half-integer analogue will not. Garg has shown, for a small easy axis ferromagnet with a field perpendicular to the direction of the magnetization, the same tunneling amplitude oscillates as a function of field.

The author has shown that mesoscopic magnets, in a suitably directed magnetic field, can exhibit intermediate spin. Intermediate spin values occur naturally in the theory of anyons and are usually associated with the SO(2) group algebra which contains the single operator $\hat{S}_z$ with the spectrum $S_z = n - \alpha/2$, where $n$ an integer, and where $\alpha$ is the usual statistical parameter. Evidently, anyons imply broken time reversal symmetry and, in the present context of a real physical magnet, the presence of a magnetic field. The necessary reduction in symmetry from SO(3) (or SU(2)) to SO(2) (U(1)) will occur for large spin and an easy plane magnet. For such a magnet, it has been shown the parity changes smoothly and periodically as a function of the magnetic field. The absence of a tunnel splitting for half-integer points directly implies oscillations in the tunnel splitting, i.e., the oscillations of Garg are a trivial consequence of intermediate spin.

Very recently Wernsdorfer and Sessoli have experimentally observed double oscillations in the tunnel splitting for Fe$_8$ which is a molecular cluster with a net spin $S = 10$. Oscillations which occur with a field along the hard axis are interpreted in terms of intermediate parity (i.e., spin) while those which are induced by a field along the hard axis are discussed in terms of the Garg interference effect. The principle purpose of this contribution is to show that both effects find a natural interpretation and a relatively simple quantitative description in terms of the author’s approach to intermediate spin in magnets. It will be shown, in the context of Fe$_8$, that periodic changes in parity can occur even for an easy ($y$-) axis magnet when the full SO(3) or SU(2) algebra is important. However, it must be that the symmetry in the plane perpendicular ($x-y$) plane is broken, so that it remains the case that there is an easy plane for tunneling between the classically defined minima.

It has been shown by numerical simulation that Fe$_8$ is adequately described by the Hamiltonian:

$$\mathcal{H} = -DS_z^2 + E(S_x^2 - S_y^2) + g\mu_B\hat{S}\cdot\vec{H}$$

where $D > E$ so that the $z$-axis is easy while the $x$-axis is hard. The term involving $S_y^2$ might be eliminated using $S_y^2 = S(S+1) - (S_x^2 + S_z^2)$ so that, to within a constant, $\mathcal{H} = -(D-E)S_z^2 + 2ES_x^2 + \vec{S}\cdot\vec{h}$ where $\vec{h} = g\mu_B\vec{H}$ is the field in energy units.

Formally this Hamiltonian has been considered in some detail in and by Preda and Barnes using the auxiliary particle method. This approach will be used for the detailed calculations.

First consider a longitudinal field $h_{\parallel}$, i.e., one along the $z$-axis, and to be specific physical whole integer spin value $S$. The Hamiltonian only connects states $|S_z = S - n >$; $n$ an integer, for which $\Delta n$ is a multiple of two. It follows that when the levels $S_z = -S$ and $S_z = S - n$ cross there can only be a tunnel splitting if $n$ is even. The crossings at fields $h = sD$, with $s$ even correspond to whole integer physics while those with $s$ odd behave as if the spin was half-integer, i.e., a longitudinal applied field causes a periodic change of parity.

At a more formal level, following ref. it is observed that the longitudinal field $h_{\parallel}$ can be eliminated by re-defining the spin operator to be
\[ \dot{\hat{S}}_z \rightarrow \dot{\hat{S}}_z - \frac{\alpha}{2}, \quad \alpha = \frac{\hbar t}{D}. \]  

which clearly has the intermediate spin spectrum \( S_z = m - \frac{\alpha}{2} \), \( m \) an integer. This transformation does not change \( [\dot{\hat{S}}_z, S^\pm] = \pm S^\pm \) but is not consistent with \( [S^+, S^-] = 2S_\hat{z} \). The argument that the problem maps to itself is more subtle than in ref. 2.

The periodic behavior as a function of a transverse field can be understood in terms of the fixed point \( E = D \). Here, to within a constant, \( \mathcal{H} \) reduces to \( +2ES_x^2 - S_z^2h_t \) where \( h_t \) is the transverse field in energy units. This defines another intermediate spin, or level crossing problem. For an integer \( S \), the \( h_t = 0 \) ground state is non-degenerate, while it is doubly degenerate corresponding to a half-integer point when \( h_t = 2E \). The “splitting” is periodic with period \( \Delta h_t = 4E \). This fixed point behavior explains qualitatively the tunnel splitting oscillations as a function of \( h_t \), however a calculation of the general result for the period, the tunnel splitting, and the combined effects of longitudinal and transverse fields requires a detailed formalism and some tedious calculations.

In outline the calculation goes as follows. The axis of quantization is put, \( x \leftrightarrow z \), along the direction of the transverse field direction. A basis \(|S_z > = | n > \) is chosen, and an auxiliary particle, a fermion \( f_n \), is associated with each state via the mapping \( | n > \rightarrow f_n^\dagger | > \) where \(| > \) is a non-physical vacuum without any auxiliary particles. Defined is a bi-quadratic version of an operator \( \hat{O} \) via: \( \hat{O} \rightarrow \sum_{n,n'} f_n^\dagger f_{n'} \). The constraint \( Q = \sum_n \hat{n}_n = \sum_n f_n^\dagger f_n = 1 \). This procedure gives

\[
\mathcal{H} = \sum_n \left( (2En^2 - h_t n) \right. \\
- \frac{1}{4}D \left[ (M_n^{n+1})^2 + (M_n^{n-1})^2 \right] f_n^\dagger f_n \\
- \frac{1}{4}D \sum_n M_n^{n+1}M_n^{n+2}(f_n^\dagger f_{n+2} + f_{n+2}^\dagger f_n) \\
+ \frac{1}{2}h_t \sum_n M_n^{n+1}f_n^\dagger f_n + f_n^\dagger f_n + f_{n+1}^\dagger f_{n+1},
\]

where the \( M_n^{n+1} = [S(S+1) - n(n+1)]^{1/2} \) are the matrix elements of \( S^z \). Without a longitudinal magnetic field \( h_t \), this is two isolated tight binding chains of spinless fermions \( f_n^\dagger \). A longitudinal field \( h_t \) provides a coupling between these two chains. The constraint \( Q = 1 \) implies this is a single particle problem. Notice that the “site” indices are either whole of half-integer following the parity of the spin.

Schrödinger’s equation

\[
(\epsilon - (2En^2 - h_t n))a_n = \\
+ \frac{1}{4}D \left[ M_n^{n+1}M_n^{n+2}a_{n+2} + M_n^{n+1}M_n^{n-1}a_{n-2} \\
+ [(M_n^{n+1})^2 + (M_n^{n-1})^2]a_n \right] \\
- \frac{1}{2}h_t (M_n^{n+1}a_{n+1} + M_n^{n-1}a_{n-1}),
\]

involves finite differences, where the wave-function \( \Psi = \sum_n a_n f_n^\dagger \). The difference in the site energies is \( \sim 2E \) near the center \( n \sim 0 \) of the chains while the “hopping term” is \( \sim S^2D \). For the problem of interest \( D \) is positive and the ground state wave function does not change sign.

The ground state is located near the center of the chains and for most purposes it will suffice to replace the \( M_n^{n+1} \) by a constant matrix element \( S \) whence the problem reduced to that of a (discrete) harmonic oscillator. It might be expected that continuum approximation is valid since \( S^2D \gg 2E \). If, \( h_t = 0 \), and if the passage is made from the discrete to a continuum problem the two chains are identical and there is no tunnel splitting. In this rather unusual representation of a tunneling problem it is the exponentially small difference in energy between the ground state on the even site chain (the absolute ground state) and that on the odd site chain which leads to the “tunnel” splitting.

However, in the absence of fields and if \( E = 0 \) this equation must reproduce the eigenenergies \( -Dm^2 \) where \( m = -S - (S - 1) \ldots S - 1, S \) is an integer quantum number. In particular the ground state is exactly doubly degenerate with \( m = \pm S \) and again with one such state on each chain. In order to obtain correctly this limit use is made of the exact result for \( E = 0 \), namely

\[
a_{\pm S} = \frac{1}{2S}e^{\mp i\pi S} \left( \frac{2S}{(S-n)(S+n)!} \right)^{1/2},
\]

where the two solutions correspond to \( S_z = \pm S \). For large spin \( S \) this approximates well to

\[
a_{\pm S} = \frac{1}{2S}e^{\mp i\pi S} e^{-n^2/2S},
\]

i.e., ignoring the factor \( e^{\mp i\pi S} \) the wave-function is that of a simple harmonic oscillator fairly well localized relative to the limits at \( n = \pm S \). It can be made to lie on one or the other chains by taking the sum or difference of these solutions. The system does not “see” that \( n \) is limited. Adding the term \( 2En^2 \) causes the wave function to become slightly more localized. Since this additional potential is harmonic the wave function remains harmonic.

Consider the problem with \( h_t = 0 \) but with both \( E \) and \( h_t \) finite. The wave function is taken to have the form

\[
a_n = f(n)a_{\pm S}^n.
\]

Substituting this into Schrödinger’s equation gives at a sufficient level of approximation:

\[
\epsilon f(n) = (2En^2 - h_t n)f(n) \\
+ \frac{1}{4}S^2D[f(n + 2) + f(n - 2) - 2f(n)].
\]

At this point the intermediate spin transformation is made, i.e., the field \( h_t \) is absorbed into the term \( 2En^2 \) by the shift \( n \rightarrow n - (h_t/4E) \). There is an apparent displacement of \( \delta = (h_t/4E) \). The small change
in energy is unimportant and will be dropped. However, the real displacement \( d \) is different. With \( h_t = 0 \) to a good approximation, \( f(n) \sim e^{-[(2E/D)1/2]n^2/2S} \) while for finite \( h_t \), \( f(n) \sim e^{-[(2E/D)1/2][n-(h_t/4E)]^2/2S} \) so that \( a_n \sim e^{-n^2/2S} \) which is approximately \( e^{[n-(h_t/2(2ED)1/2)]^2/2S} \), i.e., the real displacement of the net solution is

\[
d = h_t/2(2ED)^{1/2},
\]

which corresponds to a statistical parameter \( \alpha = h_t/(2ED)^{1/2} \). The point to be made is that to a good approximation the solution for finite \( h_t \) is the same as for \( h_t = 0 \) except for a displacement of the site location from integer (or half-integer values for half-integer \( S \)) by \( d \).

A Fourier transform, is made in order to examine the approach to the continuum limit and to generate a tunnel-barrier problem: \( f(y) = \frac{1}{\sqrt{2\pi}} \int dn e^{imn}a(n) \), whence Schrödinger’s equation \( (h_t = h_t = 0) \) is:

\[
(2E\frac{d^2}{dy^2})f(y) = -\frac{S^2D}{2}[\cos 2y - 1]f(y),
\]

which is Mathieu’s equation [3]. The potential is periodic with period \( \Delta y = \pi \) and the solutions form bands. For a given energy, a solution might be characterized by a wave-vector \( k \) and Floquet’s (Bloch’s) theorem [3] implies that solutions of the form \( f_k(y) = e^{iky}u_k(y) \) where \( u_k(y) = u_k(p + \pi) \).

Since the \( k \)-space is reciprocal to the \( y \)-lattice it is roughly equivalent to the real \( x \)-space and since this latter is discrete, making an inverse Fourier transform selects a particular wave vector \( k \). Also a shift in \( k \)-space is fully equivalent to a shift in real space. Because of the intermediate spin shift \( d \), the sites are precisely shifted from the whole or half integers. Specifically for a whole integer spin, the solution which is finite on the even site chain implies \( k = d \). Similarly for the odd site chain \( k = d - 1 \). The only difference for a half-integer spin is an addition shift of 1/2. Without an intermediate spin (i.e., statistical shift) this makes the two \( k \)-values equivalent at \( k = \pm 1/2 \) and directly implies a zero splitting in the absence of fields.

In the \( y \)-space, around \( y = 0 \), \( f(y) = (1/\sqrt{3\sqrt{\pi}}e^{-y^2/3S^2})^n/2 \) with \( \beta^2 = S\sqrt{2E/D} \), and the nominal ground state energy is \( (\omega_0/2) \); \( \omega_0 = 2S\sqrt{2ED} \). The band energies are: \( \epsilon_k = (\omega_0/2) + (w/2)\cos \pi k \) where the width \( w = 8\sqrt{2/\pi}\omega_0S^{1/2}e^{-S_f} \) and where the action \( S_f = 2S\sqrt{(D/2E)} \). The result for the “tunnel splitting”, i.e., the difference in energy between the ground and first excited states is

\[
\delta E = 4\sqrt{\frac{\pi}{\omega_0}S^{1/2}e^{-S_f}} |\cos(\pi d)|;
\]

\[
S_f = 2S\sqrt{(D/2E)}.
\]

This result for the tunnel splitting is very similar to that for an easy plane magnet [4]. The most significant difference is that the period for the oscillation, with a field along the hard axis is \( 2(2ED)^{1/2} \) rather than, in the present notation, \( 4E \). For \( E \) small this result for the period agrees with the result of Garg \( 2(2E(D + E))^{1/2} \) quoted by Wernsdorfer and Sessoli [3].

The last step is to account for the longitudinal field \( h_t \). There is a level crossing for the values \( h_t = sD; s \) an integer. Since they reflect the essential physics only results for these crossing points will be presented. Consider a whole integer physical spin \( S \), following the earlier discussion there are two types of crossing corresponding to the value of \( s \). For \( s \) even, including \( s = 0 \), the system behaves as if it is whole integer while for \( s \) odd it has the characteristics associated with a half-integer total spin. The proof goes as follows: The zero order wave functions for the states which cross are, e.g., \( a^{-S}_n \) and \( a^{S}_n \) which are both exact solutions. From Eqn. (15): \( a^{-S}_n = (-1)^n a^S_n \) where \( a^S_n \) is even in \( n \), while \( a^{-S}_n \) is even or odd as \( s \) is even or odd. Given there is tunneling, the appropriate wave functions are of the form, for large enough \( S \):

\[
a_{\pm,n} = f(n) \left[ (-1)^n a^S_n \pm a^{-S}_n \right].
\]

If this function is substituted in Eqn. (15) the terms involving \( h_t \) cancel to \( O(S) \), so that the resulting difference equation and solution for \( f(n) \) are unchanged. However, the function \( a_{\pm,n} \) is quite different depending on the even or odd character of \( s \). With \( f(n) = 1 \), because of the term \( (-1)^n \), \( a_{\pm,n} \) jumps wildly between sites but defines a smooth function on either the even, \( a_{\pm,n}^\pm \), or odd, \( a_{\mp,n}^\pm \), sites. When \( s \) is odd it is almost the case that the function makes \( a_{\pm,n}^\pm = -a_{\mp,n}^\pm \). In fact, if the smooth function which passes through the lattice points is displaced by \( d = 2n = 1/2 \), then these relationships are exact. As discussed above, it is just such a displacement with converts the whole into the half integer problem and demonstrates that the odd crossings are equivalent to a physical half-integer spin \( S + s/2 \). When \( s \) even the functions \( a_{\pm,n}^\pm \) and \( a_{\pm,n}^\mp \) are different, however (to within a sign) \( a_{\pm,n}^\pm = a_{\mp,n}^\mp \), so that difference between the two solutions \( a_{\pm,n}^\pm \) is a displacement by one lattice site or \( d = 1 \) which is just the generalization of the same result for \( s = 0 \) and which results in a the maximal tunnel splitting with \( h_t = 0 \). The tunnel splitting reflects the decrease in effective spin in the sense that it increases strongly with \( s \).

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