Intermediate spin approach to the tunneling of nano-magnets.

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The theory for the quantum tunneling of nano-magnets is developed within the intermediate spin framework. Periodic magnetic effects are seen to reflect that associated with a change of flux by a single flux quantum $\Phi_0$. Essential are Schrödinger cat wave functions which involve superpositions of different magnitudes of the applied magnetic field. For systems in which the tunneling paths are not co-planar the theory leads to essentially complex magnetic fields although the expectation values of the fields remain real. In general the ground states correspond to minimal uncertainty squeezed states. The degree of squeezing depends on the anisotropy parameters.

75.45+j, 75.50Tt, 75.60Ej

I. INTRODUCTION

The quantum properties of very small magnets often called nano-magnets have generated a good deal of interest over the past decade. Much of the activity has focused upon the demonstration of quantum mechanical tunneling in its various aspects. Here attention is drawn to another purely quantum phenomenon namely intermediate spin. Intermediate spin comes as part of the baggage of intermediate statistics, i.e., anyons and two dimensions. The intermediate statistics and spin reflect a statistical parameter $\alpha_s$ which is conventionally $\alpha_s = 0$ for bosons and whole integer spin and $\alpha_s = 1$ for fermions and half-integer spin. It does not seem to be generally appreciated that there are a good number of (real and numerical) experimental manifestations of intermediate spin in relatively simple magnet systems in a magnetic field. The principal purpose of this paper is to develop a formalism for quantum tunneling of nano-magnets based upon the theory of intermediate spin.

It will be shown that a magnetic field directed along a suitable symmetry axis can cause, e.g., a physical whole integer nano-magnet to have a low energy level structure which is identical to the equivalent half-integer system, this including a Kramer’s degeneracy. Zero and a certain number of fields lying between the half-integer values correspond to whole integer points. Intermediate fields correspond to intermediate spin.

The suppression of the ground doublet tunnel splitting which necessarily occurs at half-integer points has been previously recognized by Garg. What has been labeled “topological quenching” is in fact just a pseudo-Kramer’s degeneracy of the ground state. The implications of intermediate spin are much stronger since all low lying levels must be degenerate at exactly the same field.

It is a commonplace observation, for the situation illustrated in Fig. (1), that magnetic effects are periodic with period $\Phi_0$, the flux quantum. For the problem explained in the figure caption in particular the tunnel splitting,

$$\Delta = \Delta_0 |\cos 2\pi \Phi / \Phi_0|.$$  

(1)

For the magnetic systems the symmetry which will be considered in most detail here is that appropriate to $\text{Fe}_8$, Fig. (2a). This corresponds to the most general case of 4-fold symmetry.

FIG. 1. A particle of charge $q$ is confined to a unit circle. A tunneling problem might be defined by adding a potential $V$ such that $V(\phi) = V(\phi + \pi)$. If this potential has minima at $\phi = 0$ and $\pi$ and barrier between is higher than the zero point energy then there will be a ground doublet with a splitting $\Delta$. A perpendicular field exerts no force on the particle but does change the splitting as explained in the text.

FIG. 2. The length of the axis indicates the energy gain when the spin $\tilde{S}$ lies along that direction, thus (a) for the symmetry appropriate to $\text{Fe}_8$ when all three axes are inequivalent the vertical $y$-axis is easy while the field is directed along the hardest $x$-direction. For (b) appropriate to $\text{Mn}_{12}$ with equivalent $x$ and $y$ directions, the $z$-direction is again easy. The field is along the hardest direction in the $x - y$-plane. For cubic symmetry, illustrated is the possibility with the field along a hard four fold axis. The perpendicular axes directed to the corners of the cube are all equivalent easy directions (in the absence of a field). Intermediate spin effects also occur if the field is directed along a three fold axis.
quadratic nano-magnet Hamiltonian:

$$\mathcal{H} = -DS_z^2 + E \left[ S_x^2 - S_y^2 \right]$$  \hspace{1cm} (2)

which without loss of generality has two positive anisotropy parameters $E < D$, see the Appendix. A magnetic field is directed along the hardest, i.e., $z$-direction. This is fully equivalent to the particle problem of Fig. (1) with $\Phi/\Phi_0 = g \mu_B H/4 \hbar^2 \sqrt{2E(D + E)}$. The agreement with Eqn. (1) is illustrated by Fig. (3). The deviations from the $|\cos|\phi$ behavior for larger fields is due to the finite value of $S$. It is larger for the value $S = 10$ relevant for Fe$_8$.

Under an adiabatic change of magnetic field by $\Phi_0$ the $S_z$ quantum number $m \rightarrow m + 1$ for the angular momentum quantized along the magnetic field direction. Whatever the mixture of states with different $m$, the period measured in terms of the magnetization along the field direction is automatically $h \mu_B$. However it is a trivial consequence of time reversal invariance that the states with fluxes $\pm \Phi$ are degenerate and so there is the possibility to construct “Schrödinger cat” states which comprise a linear combination of states with the macroscopic field $\vec{H}$ in both its positive and negative directions. In this case the magnetization period need no longer be $h \mu_B$. It will be shown, with this symmetry, such a combination exists in all energy eigenstates and that the period is indeed a fraction of $h \mu_B$.

An even more bizarre phenomenon occur when either the field is not directed exactly along the appropriate symmetry axis and/or if the least action tunneling paths are not co-planar. For these cases the energy eigenstates correspond to suitably weighted “Schrödinger cat” combinations with oppositely directed complex magnetic fields. The complex weighting is such that the expectation value of the physical magnetic field remains real. Thus, again for the Fe$_8$ symmetry, a field $H_z$ which is perpendicular to both the hard and easy directions corresponds to a pure imaginary field $iH_z$ directed along the symmetry axis, i.e., the tunnel splitting is given by Eqn. (1) with $\Phi/\Phi_0 = i g \mu_B H_z/4 \hbar^2 \sqrt{2E(D + E)}$ so that $\cos \rightarrow \cosh$. In the presence of both fields

FIG. 4. To the right the thick line is approximation Eqn. (1) with a complex field $B_0 = B(\cos \theta + i \sin \theta)$. The thinner line corresponds to exact diagonalization. To the left is experiment with a logarithmic scale. The imaginary part of the flux causes the tunnel splitting to increase with field.

$$\Phi/\Phi_0 = g \mu_B (H + iH_z)/4 \hbar^2 \sqrt{2E(D + E)}.$$  \hspace{1cm} (3)

The resulting approximation is compared with the exact result and experiment in Fig. (4).

For the much studied molecular magnet Mn$_{12}$ it is impossible to not have a complex field. With the physical field in the direction indicated in Fig. (2b) the resulting complex field has equal real and imaginary parts in the large $S$ limit, see Fig. (5). There are two equivalent tunneling planes which both make an angle $\theta = \pi/4$ with the applied field and $\pi/2$ with each other. The wave function for the planes is added so that there are two sets of paths, one from each plane, which are equivalent and which interfere. The resulting expression for the tunnel splitting is shown in the figure caption. As the figure illustrates this splitting increases very rapidly with field and by several orders for the $S$ oscillations.

The last important principle is illustrated by applying a field in the hardest direction for a system with cubic symmetry, Fig. (1c). This is a four fold axis and group theory insists that the lowest tunnel split multiplet comprises four states. For whole integer $S$ it turns out that there are two singlets and one doublet, however for a half-integer point this must reduce to two doublets, reflecting the pseudo-Kramer’s theorem. That this is the case for large $S$ is illustrated by the exact results of Fig. (6).

Classically for magnet such as Fe$_8$ the equilibrium

FIG. 3. Properties are periodic in the field $h = H/g \mu_B$ for $S = 35$. The deviations from $\Delta = \Delta_0 \cos 2\pi \Phi/\Phi_0$ become smaller as $S$ increases.
magnetization lies along its easy axis and is maximal. The existence of quantum fluctuations is reflected in the fact that the expectation value of this easy axis magnetization is $\hbar S$ and not $\hbar \sqrt{S(S+1)}$. In the absence of a hard direction in the plane perpendicular to the easy axis, the quantum fluctuations measured by $\langle S_x^2 \rangle$ and $\langle S_y^2 \rangle$ are equal and in fact minimal. In the terminology used for fluctuations in quantum optics, these are minimal un-squeezed states. Adding an anisotropy energy $E$, which creates a hard axis in this plane as is appropriate to Fe$_8$, squeezes the quantum fluctuations so that $\langle S_x^2 \rangle \neq \langle S_y^2 \rangle$ but leaves minimal states. The present approach thereby lends itself to a natural definition of squeezed spin states \cite{9}.

Apart from the simple periodicity \cite{6} of Eqn. (1) the intermediate spin effects described above have yet to be experimentally verified.

II. INTERMEDIATE SPIN

Returning to the basic charged particle problem of Fig. (1), the relevant group is SO(2) which is equivalent to U(1), i.e., is abelian with a single operator $S_z$, the generator of rotations $e^{iS_z\phi}$, where the $z$-axis is perpendicular to the circle and where $\phi$ is the angle to the $x$-axis. For a unit circle $S_z = p_\phi$ the momentum conjugate to $\phi$, i.e., $[\phi, p_\phi] = i\hbar$. The eigenstates of $S_z$ and $p_\phi$ are $\psi_m(\phi) = \frac{1}{\sqrt{2\pi}}e^{im\phi}$ but for particles on a circle there is no reason it insist on $m$ being an integer reflecting the fact that the spectrum of $S_z$ for SO(2) is continuous. In general for this singular gauge $\psi_m(\phi)$ is multi-valued. A convenient single valued function can be defined by taking some convention for $\psi_m(\phi)$ in the interval $\{0, 2\pi\}$ and then analytically continuing this function to the interval $\{-\infty, +\infty\}$. For any physical potential $V(\phi) = V(\phi + 2\pi n)$; $n = 0, \pm 1, \pm 2, \ldots$, and implies that only the basis states $\psi_{m+n}(\phi)$: $n = 0, \pm 1, \pm 2, \ldots$, mix.

It follows for a given statistical parameter $\alpha_s$ the energy spectrum is discrete. This can be stated differently: Since $V(\phi)$ is periodic the solutions obey Floquet’s (Bloch’s) theorem, i.e., $\psi_k(\phi) = e^{ik\phi}u_k(\phi)$ where $u_k(\phi)$ is periodic. The reciprocal lattice vector $K = 2\pi/(a = 2\pi) = 1$ and the energy $\epsilon(k) = \epsilon(k + nK)$; $n = 0, \pm 1, \pm 2, \ldots$, i.e., is periodic. An examination of the Fourier components immediately implies $k = \alpha_s/2$. Given that $V(\phi)$ has a point with reflection symmetry the $\psi_{\pm k}(\phi)$ are degenerate and the most general solution for a given energy is $Au_k(\phi) + Bu_{-k}(\phi)$ for which the spectrum of $m = n \pm \alpha_k/2$, i.e., there are twice the number of observable $S_z$ values except importantly for the whole and half-integer points when $\alpha_s = 0$ or 1. This more general method of extrapolating between whole and half-integer spin which will be found applicable here.

In the non-singular gauge the basis set $\psi_n(\phi)$ has $n = \pm 1, \pm 2, \ldots$ and the charged particle encircles a magnetic flux $\Phi = \alpha_s\Phi_0/2$ where $\Phi_0$ is the appropriate flux quantum. For the same $V(\phi)$ there exists an eigenstate $u_k(\phi)$ with the same energy as in the singular gauge, i.e., the energy spectrum are identical and two problems can mapped to each other and both reflect intermediate spin. In the non-singular gauge the combination $Au_k(\phi) + Bu_{-k}(\phi)$ comprises a linear combination of states with the flux $\Phi$ in both its positive and negative senses. Evidently the expectation value of the flux $\langle \Phi \rangle$ is in general smaller than $\alpha_s\Phi_0/2$. This possibility of a linear combination of different flux states does not seem to have been envisaged in connection with intermediate statistics although it would arise if, e.g., quantum coherence in SQUIDS could be observed. It is realized in the systems, e.g., Fe$_8$, discussed here.

III. SPIN MODELS

It has become usual to re-write Eqn. (2) with the $x$ and $z$-axes interchanged. Thus with fields the general quadratic nano-magnet Hamiltonian becomes,

$$H = -(D - E)S_z^2 + 2ES_x^2 - ES^2 - hS_z - h_xS_x - h_yS_y,$$

see the Appendix. Unless stated otherwise it will be assumed that the physical spin value $S$ is whole integer. The value $S$ is that which would be assigned by the usual arguments, e.g., for a set of ferromagnetically ordered spins this is simply the sum of the spin values. The field $h$ is parallel to the hardest and transverse to the easiest axis. It is equivalent to that introduced in the non-singular gauge. The other transverse $h_x$ and the longitudinal $h_y$ fields simply modify the potential $V$.

Since the length of $S$ is fixed, classically the energy,

$$E(\theta, \phi) = h^2S^2\sin^2\theta \left[-(D - E)\cos^2\phi + 2ES^2\phi\right] - hS\cos\theta - h_xh_S\sin\theta\cos\phi - h_yh_S\sin\theta\sin\phi - Eh^2S^2,$$

(4)
Quantum mechanically Eqn. (4) is the energy expectation value for the coherent state $|\theta, \phi\rangle$ obtained by rotating $|S_z = S\rangle$ through $\theta, \phi$. It is these coherent states which appear in the usual functional integral formulation (1). However the $|\theta, \phi\rangle$ are very much over-complete. In fact the states $|\phi\rangle \equiv \{\theta = \frac{\pi}{2}, \phi = (2S + 1)^{1/2}\sum_{m=-S}^{S} e^{-im\phi} |m\rangle$, where $|m\rangle \equiv |S_z = m\rangle$, remain over-complete. If $|\psi\rangle = \sum_{m=-S}^{S} a(m) |m\rangle$ then $\psi(\phi) \equiv \langle \psi | \psi \rangle = \sum_{m=-S}^{S} e^{im\phi} a(m)$, i.e., is a Fourier transform of the m-space wave function $a(m)$. The function $\psi(\phi)$ is defined on the interval $(-\infty, +\infty)$ and the inverse transform $\psi(m) \equiv \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\phi \psi(\phi) = \sum_{m'} \delta(m - m') a(m')$ so that $|\psi\rangle = \int_{-\infty}^{+\infty} dm \psi(m) |m\rangle$, i.e., $\psi(m)$ is a definition of the m-space wave function appropriate when $m$ is considered as a continuous variable. Clearly $\hat{S}_z \psi(\phi) = \int_{-\infty}^{+\infty} dme^{im\phi} hm\psi(m) = p_\phi \psi(\phi)$ where $p_\phi = -i\hbar \frac{d}{d\phi}$ is the same as that defined earlier. Also evident is that multiplication by $e^{i\phi}$ amounts to a displacement of $m$ by unity and so, e.g., $\hat{S}^+ = e^{i\phi} [\hbar^2S(S + 1) - p_\phi (p_\phi + 1)]^{1/2}$.

IV. TOPOLOGICAL CONSIDERATIONS

In terms of the unit sphere, the system of coordinates $\phi, p_\phi$ might be considered as a particular projection in which the axis of quantization plays the role of the North-South axis as shown in Fig. (7). In these terms, the angle $\phi$ is evidently the longitude while $p_\phi$ is the projection on the $z$-axis and is therefore the sine of the latitude. That these are canonical coordinates and momentum emphasizes the evident fact that the unit sphere is the phase rather than simply the coordinate space.

In order to calculate the Berry phase (10) a unit vector $\vec{R}$ is directed along the easy, i.e., $x$-axis for Eqn. (3) (the $z$-axis for Eqn. (1)). It is particularly easy to determine this phase for an adiabatic process in which the system is rotated about the $z$-axis (the $x$-axis for Eqn. (1)). For the singular gauge, if the wave function comprises a single Floquet (Bloch) wave $e^{ik\theta}u_k(\phi)$ this is trivially

$$\Theta_B = 2\pi k = 2\pi \frac{\alpha}{2}$$

which is the obvious generalization of the result $\Omega S$ where $S_{\Omega}$ here $2\pi$, is the solid angle subtended by the path of $\vec{R}$. However for the combination $A\psi_k(\phi) + B\psi_{-k}(\phi)$ the result is $\tan\Theta_B = [(A - B)/(A + B)] \tan 2\pi \frac{\alpha}{2}$ which agrees with $\Omega 2\pi$ only at integer and half-integer points.

V. PSEUDO KRAMER’S OPERATORS

If it is possible to transmute a whole into a half-integer spin then at the half-integer points there should exist an operator $K$, the equivalent of the time reversal operator for $h = 0$. This must commute with $\hat{H}$ and if $|E_n\rangle$ is an eigenstate then $K|E_n\rangle$ should be orthogonal (or zero).

For the current model Preda and Barnes (11) have constructed such operators.

The simplest case is with $h_{\ell} = h_{\ell} = 0$ and $h = \hbar \sqrt{2E(D + E)}$. The operator $K_1 = Tr$ where $T$ is the conventional time reversal operator and where

$$r = \sqrt{D + ES_x} + i\sqrt{2ES_y}.$$ 

It is easy to show that with this field the negative semi-definite

$$\hat{H} = -(Tr)^2$$

so that trivially $[\hat{H}, K_1] = 0$ independent of $S$ and the parameters $D$ and $E$. As will be shown in Sec. VII, with $h_{\ell} = h_{\ell} = 0$ there is an eigenstate of the form $|E_n\rangle = \sum a(2p)|2p\rangle$, where $S_z|m\rangle = \hbar m|m\rangle$, i.e., only states with even values of $m$ are present. Since only has matrix elements between even and odd such states it follows that $K_1|E_n\rangle$ is orthogonal to $|E_n\rangle$ and hence for this value of the field all states are double degenerate or $K_1|E_n\rangle = 0$. Clearly this latter state has energy $E_n = 0$. It is evident that when $E = 0$ there is only one such state and by a continuity argument this must be the case for all $E$. It follows that there are $S$ degenerate pairs when $h = \hbar \sqrt{2E(D + E)}$ fully mimicking a half-integer spin except for the highest energy singlet.

It is therefore a remarkable fact that this degeneracy for fields which are exact multiples of $\hbar \sqrt{2E(D + E)}$ exists for all but the highest energy states for arbitrary values of integer $S$ and independent of the existence of a tunneling barrier, etc. For example it is possible to explicitly construct the $S = 1$ matrices and show that
for \( h = h \sqrt{2E(D+E)} \) the ground state is degenerate independent of the parameters \( E \) and \( D \).

It is to be concluded that this degeneracy corresponds to a hidden symmetry of this class of Hamiltonian, akin to time reversal symmetry for the half-integer equivalent in zero field. This pseudo-time-reversal symmetry then implies a pseudo-Kramer’s degeneracy.

VI. QUANTUM FLUCTUATIONS - SQUEEZED STATES

If \( S \) are not too small then \( \vec{S} \) remains always close to the classical energy minima at \( \theta = \pi/2 \) and \( \phi = 0 \) or \( \pi \), and to an excellent approximation near the first of these classical ground state Eqs. (6), after dropping a constant \( E^0 = -h^2S^2(D - E) \) and with \( \hbar = \hbar = 0 \), simplifies to,

\[
\mathcal{H} = (D + E)p_\phi^2 + \hbar p_\phi + h^2(D - E)S^2\phi^2. \tag{6}
\]

The field can be absorbed into the kinetic energy so

\[
\mathcal{H} = (D + E)\langle p_\phi + \delta \rangle^2 + h^2(D - E)S^2\phi^2 - (D + E)\delta^2, \tag{7}
\]

where \( \delta = \hbar/2(D + E) \). Recall, \( \delta \) corresponds to displacement of \( \delta \) in \( m \)-space, however it is not equal to \( \alpha_s/2 \). In \( \phi \) space it adds a phase factor \( e^{i\delta\phi} \) to the wave function. Even for finite \( \hbar \), this is just a harmonic oscillator with a ground state energy

\[
\epsilon_n = E^0 - \frac{h^2}{4(D + E)} + (n + \frac{1}{2})\omega_n, \tag{8}
\]

where \( \omega_n = S h\sqrt{D^2 - E^2} \) and where only the last term is of quantum origin. The wave function

\[
a(\phi) = \left( \frac{S}{\pi} \right)^{1/2} \left( \frac{D - E}{D + E} \right)^{1/4} e^{i\delta\phi} e^{-S(D-E)/2} \tag{9}
\]

Directly, the magnetic moment,

\[
\langle S_z \rangle = \langle p_\phi \rangle = \hbar\delta = \hbar\frac{h}{2(D + E)}. \tag{10}
\]

The same result follows from differentiating \( \epsilon_n(\hbar) \).

This ground state wave function of a harmonic oscillator necessarily corresponds to the minimum of the uncertainty relationship \( \Delta \phi \Delta p_\phi \geq \hbar/2 \). This permits a natural definition of squeezed spin states. The transverse spin fluctuations are a minimum, e.g., for \( E = 0 \), \( \sigma_x = \sigma_y = \hbar(S/2)^{1/2} \) (e.g., \( \sigma_x^2 \equiv \langle S_x^2 \rangle - \langle S_x \rangle^2 \)). The \( \vec{S} \) can be pictured as having fluctuating transverse components \( \sim \hbar(S/2)^{1/2} \) such that the total magnitude square is correctly \( h^2[S^2 + 2 \times (S/2)] = h^2S(S + 1) \). Clearly the two quadrature components \( \sigma_x \) and \( \sigma_y \) have equal uncertainties and this minimum uncertainty state is unsqueezed. For finite \( E \),

\[
\sigma_x^2 = \left( \frac{h^2S}{2} \right) \sqrt{\frac{D + E}{D + E}}; \quad \sigma_y^2 = \left( \frac{h^2S}{2} \right) \sqrt{\frac{D + E}{D + E}}.
\]

Maximum squeezing occurs as \( E \to D \).

VII. FORMULATION WITH THE \(|S_z\rangle\) BASIS

The zero point energy associated with the transverse fluctuations is \( \omega_0/2 \) and correctly the low lying excited states have energies \( (n + (1/2))\hbar\omega \sim h^2SD \) above the ground state. However from Eqn. (4) the energy barrier is also \( \sim S^2\hbar^2D \) and this would apparently correspond to a problem with a tunneling amplitude of order \( e^{-S/2} \) which is not inconsistent with the Fourier transform of Eqn. (4). However the unsqueezed states when \( E = 0 \) are in fact eigenstates of \( S_z \), i.e., in this case tunneling is absent.

It is necessary to carefully formulate the problem in terms of the basis set \(|m\rangle\), i.e., in \( m \)-space. If \( |\psi\rangle = \sum_{m=-S}^{+S} a(m)\langle m| \), Schrödinger’s equation in this basis can be written as \( (\hbar = \hbar = 0) \):

\[
(\epsilon - 2\hbar^2m^2 - hh\omega) a(m)
= \frac{1}{2} \hbar^2(D - E) \left[ M_{m+1}^2 + M_{m-1}^2 \right] a(m + 2)
+ M_m^2 a(m - 2)
+ [M_m^2 + M_{m+1}^2 + M_{m-1}^2] a(m)], \tag{11}
\]

where \( M_m^2 = [S(S+1) - s^2]^{1/2} \). The structure of this equation is worth noting. It has the form of two tight binding models, one for even \( m \) values and the other for odd \( m \). The two chains are not connected as illustrated.
in Fig. (8). This is not a periodic solid since (i) the diagonal energy increases steadily going away from $m = 0$ (ii) the chains have ends at $m = \pm S$. This is rather a pair of discrete harmonic oscillator problems for large $S$ when (ii) is not important since the wave function is well localized near $m = 0$.

Making a continuum approximation, e.g., $a(m \pm 2) \approx a(m) \pm 2(d^2a/dm^2)$, valid if $S^2(D - E) \gg 2E$, and assuming large spin $S \gg 1$, this reduces to

$$
2EJ^2m^2 - \epsilon) a(m) + 4h^2S^2(D - E)\frac{d^2a(m)}{dm^2} = 0.
$$

This describes a simple harmonic oscillator with wave function $a(m) = e^{-m^2Q/2S}$ where, $Q = \sqrt{(D + E)/(D - E)}$ characterizes the degree of squeezing. This should be recognized as the Fourier transform of Eqn. (4). There is evidently no tunneling barrier and no tunnel ground doublet splitting! However there is a degeneracy since this same equation results for both the odd and even site chains. The splitting of these two states disappeared upon making the continuum approximation. As will become evident below, the discrete problem described by Eqn. (11) has asymptotic corrections which differ as the $m$ arguments are even or odd. It is an interesting observation that the discrete harmonic oscillator and tunneling problems are equivalent.

The wave function for the eigenstate $|S_x = S\rangle$ is $a^0(m) = \frac{1}{\sqrt{[S-m+1]!S+m)!}}$ and $a^0(m)$ has to be a solution to Eqn. (11) with $h = E = 0$, this corresponding to the absence of tunneling. Using Stirling’s approximation this reduces to $a(m) \propto e^{-m^2Q/2S}$ with $E = 0$, however the deviations from this approximation are very important. The absence of tunneling when $h = E = 0$ is put in evidence by a transformation: $a(m) = a^0(m)f(m)$. After some algebra Schrödinger’s equation for $f(m)$ reduces to:

$$(\epsilon - (2EJ^2m^2 - hhm + E^0)) f(m) = \frac{1}{2}h^2(D - E)$$

$$\times \left[(S-m+1)!S+m)!\right)(f(m+2) + f(m-2) - 2f(m))$$

$$- n(2S-1)\left[(f(m+2) - f(m-2))\right].
$$

When $h = E = 0$ this evidently admits the solutions $f(m) = 1$ and $f(m) = e^{imn}$ corresponding to the degenerate states $|S_x = \pm S\rangle$.

**VIII. INTERMEDIATE SPIN FORMULATION**

With again the definition $f(\phi) = \sum_{m} e^{im\phi} f(m)$ this yields the exact Schrödinger’s equation:

$$\left(\mathcal{E} - 2Ep_{\phi}^2\right)f(\phi) = V(\phi, p_{\phi})f(\phi)
$$

with

$$V(\phi, p_{\phi}) = \frac{1}{4}(D - E) \times$$

$$\left[2\sin^2(\phi^{2}(S-1) + p_{\phi}^2) - i\hbar(2S-1)\sin 2\phi p_{\phi}\right],
$$

where $\mathcal{E} = (E - E^0)$. This is of the intermediate spin form but with a momentum dependent potential, $V(\phi, p_{\phi})$.

The effect of the transverse field $h$ is twofold (i) near the classical ground state the spin sees a longitudinal field $S(D - E)$ along with the transverse $h$ and this tilts the axis of quantization towards the $z$-direction. (ii) The field $h$ modifies the tunneling amplitude.

In accounting for (i) the term $2Ep_{\phi}^2$ is not relevant and so what is required is a solution of,

$$\mathcal{E} + h_{\phi})f(\phi) = V(\phi, p_{\phi})f(\phi).
$$

Needed is a small rotation about $y$-axis. The new wave function is defined by $f(\phi) = (1 - (a_{\alpha}p_{\alpha}/2hS))g(\phi)$ where $h = h(D - E)\alpha_{\phi}/2$, this admixes, e.g., a little of the first excited into ground state. For large $S$, $g(\phi)$ obeys

$$\mathcal{E}g(\phi) = V(\phi, p_{\phi} + h_{\phi})g(\phi),
$$

i.e., the field has been replaced by the displacement $\frac{\alpha_{\phi}}{2}$. In order to account for (ii), a similar displacement is included in the full equation. If $g(\phi)$ is the solution of

$$\left(\mathcal{E} - 2E(p_{\phi} + h_{\phi})^2 + \hbar^2E\frac{\alpha_{\phi}^2}{2}\right)g(\phi)$$

$$= V(\phi, p_{\phi} + h_{\phi})g(\phi),
$$

then $f(\phi) = (1 - (a_{\alpha}p_{\alpha}/2hS))g(\phi)$ is the solution to

$$\left(\mathcal{E} - 2Ep_{\phi}^2 + 2h(D + E)\frac{\alpha_{\phi}^2}{2}\right)f(\phi)$$

$$= V(\phi, p_{\phi})f(\phi).
$$

Following the discussion of intermediate spin, the shift determines the wave vector $k = \alpha_{\phi}/2$. In turn the statistical parameter must be chosen to give ground state energy determined without tunneling, i.e., Eqn. (8). Required is that $\hbar^2E\frac{\alpha_{\phi}^2}{2} = \frac{\hbar^2}{4(D + E)}$, so $k = \frac{\alpha_{\phi}}{2} = \hbar/2\sqrt{2E(D + E)}$. This fixes the Zeeman term in Eqn. (9): $2h(D + E)\frac{\alpha_{\phi}^2}{2} \approx \sqrt{(D + E)/2Eh_{\phi}}$. Since, in general, this is larger than $h_{\phi}$ the solution in the non-singular gauge $A(1 - (a_{\alpha}p_{\alpha}/2hS))u_{\phi}(k) + B(1 + (a_{\alpha}p_{\alpha}/2hS))u_{-\phi}(k)$ involves a linear combination of both bare fields $\pm \sqrt{(D + E)/2Eh}$ such that the expectation value is equal the physical applied field. This requires $A = 1 + \sqrt{2E/(D + E)}$ and $B = 1 - \sqrt{2E/(D + E)}$. The total energy for level $n$ and wave vector $k$ is,

$$E_{\phi}(k) = E_{\phi}^0 - \frac{\hbar^2}{4(D + E)} + \left(n + \frac{1}{2}\right)\hbar\omega_0 + \epsilon_{\phi}(k).
$$

The first two terms are purely classical in origin while the third corresponds to the harmonic motion about the
bottom of the well. It is the last term which reflects the tunneling between wells.

The problem therefore reduces to the determination of $\epsilon_n(k)$ and when the tunneling approximation is well justified this reflects the tight binding approximation. Actually for the case consider so far, with only a transverse field $h$, the problem has a higher symmetry, see Fig. (9). The potential $V(\phi, p_\phi)$ has a $\phi$ period of $a = \pi$ rather than $2\pi$ so the reciprocal space unit vector $K = 2$. Since $K$ corresponds to the smallest relevant displacement in $m$-space, it follows that a given solution only involves $m$ values which differ by 2, as already noted in Sec. VII. The tunneling energy,

$$\epsilon_n(k) = \frac{\Delta_0}{2} \cos \pi k$$

(21)

where $\Delta_0/2$ is the yet undetermined (diagonal) matrix element for tunneling for level $n$. The evaluation of this will be addressed in Sec. XIII. There are two solutions which are acceptable for $h = 0$, namely $k = \frac{\pi}{2} = 0$ corresponding to even $m$ values and $k = \frac{\pi}{2} = 1$ and odd $m$. Clearly a shift by one brings the set of integers with $m$-values back to itself. These two $k$ values correspond to $\epsilon(k) = \pm \frac{\Delta_0}{2}$ and so the zero field tunnel splitting is reflected by the parameter $\Delta_0$. For finite transverse fields $h$ the corresponding $k = \frac{\pi}{2} = (h/2\sqrt{2E(D + E)})$ and $k = (1 + \frac{\pi}{2}) = 1 + (h/2\sqrt{2E(D + E)})$ and so the tunnel splitting is,

$$\Delta = \Delta_0 \sin \frac{\pi h}{2\sqrt{2E(D + E)}}$$

(22)

equivalent to a result first obtained by Garg and attributed to topological quenching.

IX. HALF-INTEGER SPIN

Nowhere in the development has use been made of the assumption that $S$ is an whole integer. The only difference when $S$ is a half-integer is that the values of $m$ are also half-integer and thus the solutions correspond to $k = \frac{1}{2} + \frac{\phi}{2} = \frac{1}{2} + (h/2\sqrt{2E(D + E)})$ and $k = -\frac{1}{2} + \frac{\phi}{2} = -\frac{1}{2} + (h/2\sqrt{2E(D + E)})$ which yields a splitting,

$$\Delta = \Delta_0 \sin \frac{\pi h}{2\sqrt{2E(D + E)}}$$

(23)
i.e., the cosine is replaced by a sine.

X. CONTINUATION OF SOLUTION

The solution described above is based on a small $h$ approximation it is valid in the very large $S$ limit. That the matrix elements of $h_p = ph_1$; $h_1 = 2h\sqrt{2E(D + E)}$ to excited states be small requires $2E_p < S^2(D - E)$. For the first non-trivial whole integer point when $h = h_1$ this is equivalent to the requirement that the continuum approximation be valid, or that the shifts $\pm \frac{\pi}{2}$, or $\pm k$, for a field $h = h_p$ are smaller than the width of Gaussian wave function $e^{-m^2/2S}$. The wave function

$$A(1 - (\alpha_s p_\phi/2hS))u_k(\phi) + B(1 + (\alpha_s p_\phi/2hS))u_{-k}(\phi)$$

is therefore a valid approximation for $h = h_1$ for not too small $S$. However it will fail for modest values of $2E$ and large fields $h$. The theory can be extended to cover such larger fields by a process similar to analytic continuation. The current approximation can always be developed to construct the essentially exact solution $a^1(m)$ for $h = h_1$. The dimensionless moment $m_1 \approx h/2h(D + E)$ for $h_1$ can be determined. Re-defined are the states $|\phi\rangle = \sum_m e^{i(m-m_1)\phi}|m\rangle$ so that $S_2 = -i\hbar(d/d\phi) + \hbar m_1$. This translates back to the origin the wave function $a^1(m)$. Using the original $a^0(n)$ an equation for $f(n)$ is developed. Important is that in this equation there can be no term linear $n$ reflecting an effective field, since this would imply a finite displacement $\frac{\pi}{2} = k$ and hence a finite first derivative of the tunneling energy. This contradicts the assumption that the initial $h = h_1$ reflects a whole integer point and hence a maximum in the tunnel splitting. The rather obvious point being made is that the whole (and half-) integer points can be represented without a mixture of different effective fields while in between these points this is not possible. Continuing the development for points near to $h_p$, the new wave vector $k = \frac{\pi}{2} = \delta h/2\sqrt{2E(D + E)}$ where $\delta h = h - h_p$. (The period cannot change since otherwise the current solution would predict a whole integer point which is not exactly at zero field.) Always the shifts are smaller than the Gaussian width and the new $\frac{\pi}{2}$ differs from the old value by an integer and is hence equivalent. The fact that there is more than a single fashion by which to construct the wave function reflects the fact that each of the sets $|\phi\rangle$ independent of $m_1$ are (over-) complete. Important is the observation that away from the whole and half-integer points the wave function reflects an essential
mixture of fields. The description with a minimum of difference has fields \( h_p \pm \sqrt{(D + E)/2E} \delta h; \delta h = h - h_p \), and where \( h_p \) corresponds to the closest whole or half-integer point.

**XI. EFFECT OF OTHER FIELD COMPONENTS**

Consider first the effect of \( h_\ell \) alone. This adds a potential,

\[
V(\phi, p_\ell) \to V(\phi, p_\ell) + h\ell \cos \phi
\]

and removes the symmetry between \( \phi = 0 \) and \( \pi \). The period of the potential is now the full \( \phi = 2\pi \), with \( K = 1 \), but with two wells per unit cell. The harmonic levels near \( \phi = \pi \) have quantum numbers designated by \( n \) and are higher than those near \( \phi = 0 \) and which will be labeled \( n' \). Ignoring tunneling these have energies,

\[
E_n = E_0 - \frac{\hbar^2}{4(D + E)} + n\hbar\ell + \left(n + \frac{1}{2}\right)\hbar\omega_0,
\]

and,

\[
E_{n'} = E_0 - \frac{\hbar^2}{4(D + E)} - n'\hbar\ell + \left(n' + \frac{1}{2}\right)\hbar\omega_0.
\]

For a given \( h \) there is only one solution with \( k = \frac{a}{2} = (\hbar/2\hbar\sqrt{2E(D + E)}) \), the solution displaced by unity being the same state. There are now two possibilities (i) \( E_n \neq E_{n'} \) and there is a very narrow band formed about both these energies. The energy differences have a very small dependence on \( k \) and hence \( h \), see below. (ii) When \( E_n \approx E_{n'} \) 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.

As illustrated in Fig. (10) there are two tunneling matrix elements one \( \Delta_{n,n'} \) for the barrier between \( \phi = 0 \) and \( \pi \) and \( \Delta_{n,n'} \) for that between \( \phi = -\pi \) and 0 (or \( \pi \) and \( 2\pi \)). With first \( h = (h_\ell = 0) \), Schrödinger’s equation admits real solutions, \( \psi(m) \) or \( \psi(\phi) \), and so these matrix elements can be made real. For case (iia) when the symmetry of the wave functions is the same the tunneling matrix elements \( \Delta_{n,n'} = \Delta_{n',n} \equiv \Delta_0 \). For finite \( h \), for the Floquet (Bloch) wave function, the phase advances by \( 2\pi k \) from one cell to the next while to agree with the smaller unit cell appropriate when \( h_\ell = 0 \) the phase should advance by \( \pi k \) from one well to the next within the cell. This leads to \( \Delta_{n,n'} = e^{i\pi k} \Delta_0 \) and \( \Delta_{n',n} = e^{-i\pi k} \Delta_0 \).

At this point it is possible to introduce the second transverse field \( h_1 \) by slight of hand. It is observed that if \( E \) changes sign in the original version of \( \mathcal{H} \), Eqn. (2), then the two transverse axes change role and thus \( h \) becomes the equivalent of \( h_1 \). 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 \( k = (h/2h\sqrt{2E(D + E)}) \) becomes pure imaginary, i.e., \( k \to ik = i(h/2h\sqrt{2E(D - E)}) \) and it follows that \( \Delta_{n,n'} = e^{i\pi(k+ik)} \Delta_0 \) with \( \Delta_{n',n} = e^{-i\pi(k+ik)} \Delta_0 \). This information can be represented by a two level model:

\[
\mathcal{H} = (E_n - E_{n'})S_z + \frac{\Delta_0}{2} \left[ e^{i\pi(k+ik)} S^+ + e^{-i\pi(k+ik)} S^- \right],
\]

where the center of gravity energy \( \frac{1}{2}(E_n + E_{n'}) \) has been dropped. Degenerate perturbation theory then yields,

\[
\epsilon_k = \pm \frac{1}{2}\sqrt{(E_n - E_{n'})^2 + 4\Delta_0^2} |\cos \pi(k + ik_\ell)|^2
\]

or at resonance \( \epsilon_k = \pm \frac{\Delta_0}{2} |\cos \pi(k + ik_\ell)| \), with \( k = \frac{\hbar}{2h\sqrt{2E(D + E)}} \) and \( k_\ell = \frac{\hbar}{2h\sqrt{2E(D - E)}} \). The \( \hbar \ell = h_\ell = 0 \) result is recovered when \( n = n' \). That this is a good approximation is illustrated by comparison with exact results in Fig. (4). When for (iib) one wave function changes sign while the other does not it is necessarily the case, for \( h = h_\ell = 0 \), that \( \Delta_{n,n'} = -\Delta_{n',n} = \Delta_0 \) whence,

\[
\epsilon_k = \pm \frac{1}{2}\sqrt{(E_n - E_{n'})^2 + 4\Delta_0^2} |\sin \pi(k + ik_\ell)|^2
\]

and \( \epsilon_k = \pm \frac{\Delta_0}{2} |\sin \pi(k + ik_\ell)| \) if resonance and, Eqn. (23), this is equivalent an original half-integer \( S \). For case (i), Eqns. (27,28) serve to illustrate that when \( |E_n - E_{n'}| > 2\Delta_0 \sin \pi k \), e.g., the upper sign of Eqn. (27) gives \( \epsilon_k \approx (E_n - E_{n'}) + 2\Delta_0^2 \sin^2 \pi k / (E_n - E_{n'}) \) which contains a \( k \)-dependent correction which is smaller than those for cases (ii) by a factor of \( \Delta_0/(E_n - E_{n'}) \). This factor is usually extremely small for the region between crossings.

---

**FIG. 10.** The effective potential \( h_\ell = 0, h_\ell \neq 0 \) has two inequivalent minima per unit cell. While the wells near \( \pm \pi \) are physically equivalent, in the singular gauge the wave function is multi-valued and so the phase when \( \phi \) increases by \( 2\pi \) corresponds to the Berry phase \( 2\pi k \) and is relevant. When the field \( h = 0 \) the wave functions can be made real. Illustrated are the two different resonant cases. Above is the case when \( n = 0 \) and \( n' = 2 \), i.e., when both wave functions are even and there is no change in sign. In contrast, below \( n = 0 \) and \( n' = 1 \), i.e., one wave function is odd and so the wave must change sign as its \( \phi \) argument advances by \( 2\pi \).
XII. COMBINED INTERMEDIATE SPIN EFFECTS

Both $h$ and $h_\ell$ cause the effective spin $S - \frac{2}{3}$ to alternate between effective whole and half-integer spin and the two fields can be combined so that, e.g., a field $h = h\sqrt{2E(D + E)}$ transmutes a whole into a half-integer spin. Adding $h_\ell = h\sqrt{D^2 - E^2}$ brings say the $n = 0$ level in the higher well into resonance with the $n = 1$ of the lower well and the system again behaves as whole integer with a maximal tunnel splitting. However the fashion in which the two fields work is different. The transverse field $h$ leads to a smoothly changing (Berry) phase shift of $\delta = \pi(h\hbar/\sqrt{2E(D + E)})$ in the boundary condition $\psi(\phi + 2\pi) = e^{i\delta}\psi(\phi)$ so that values of $h$ which are not multiples of $h\sqrt{2E(D + E)}$ correspond to an intermediate spin value. In contrast the phase shift caused by $h_\ell$ results from the changes in sign of the wave function for the odd $n$ levels, i.e., when this field brings two levels into resonance $\delta = \pi(n - n')$ which causes the effective spin value to alternate between whole and half-integer without a meaningful intermediate case.

XIII. THE TUNNEL SPLITTING.

There are three regimes for which different approximate methods must be used in order to calculate the tunnel splitting. (i) If $E \ll D$, and for quantization along the easy axis, the maximal tunnel splitting $\Delta_0$ is calculated treating $E$ as a perturbation, see below. (ii) If $S^2(D - E) < 2E$, i.e., very close to the point $E = D$ the energy levels can be calculated using the hard axis for quantization and $(D - E)S_x^2$ is a weak perturbation. To a very good approximation the energies are $2Dh^2m^2 - (h^2/16E)$ where $m = n + (h/4hE)$ and $n$ is an integer [3]. Thus, e.g., the energy between the ground state and first excited state for $h = 0$ is $2D$, i.e., very large and nothing to do with tunneling. As a function of field the splitting is a sawtooth. Otherwise (iii) for $E \approx D$ the problem corresponds to a dominant hard axis anisotropy with again $(D - E)S_x^2$ small. This is the case considered in considerable detail by Preda and Barnes [2]. Following a method described by S. Coleman [13] it is possible to calculate the tunneling matrix elements near to any crossing $m, m'$. The result is [12],

$$\Delta_0 = t_{m,m'} = h\omega_0 \sqrt{2\pi} (\frac{2S'K}{\hbar})^{n+1} \exp(-2\sqrt{2(\frac{2S'K}{\hbar})}) .$$  \hspace{1cm} (29)

For the remaining case (i) the energy levels are $E_m = -Dh^2m^2; m = \pm S, \pm (S - 1), \ldots \pm 1, 0$. There are two cases (a) when the two levels in resonance $n$ and $n'$ both have the same parity and (b) when the parity is different. The whole integer case (a) is simplest since $\Delta_0$ can be determined for $h = 0$. The matrix element of $2ES_x^2$ between the states $m$ and $m + 2$ is

$$p_{m,m+2} = \frac{E}{2} (\frac{m}{m+1}) M_{m+1}^m M_{m+2}^m ,$$  \hspace{1cm} (30)

recalling that $M_{m+1}^m = [S(S + 1) - m(m + 1)]^{1/2}$. Thus the expression for the tunnel splitting is,

$$\Delta_0 = \left(\frac{\hbar^2 E}{2}\right)^{n+1} M_{m+1}^m M_{m+2}^m \frac{1}{E_{m+1} - E_n}$$

$$\times M_{m+3}^{m+1} M_{m+4}^{m+2} \frac{1}{E_{m+4} - E_{m+3}} \ldots$$

$$\times M_{m+1}^{m+1} M_{m+2}^m \frac{1}{E_{m+2} - E_m}$$

$$\times \frac{1}{E_{m+1} - E_n} M_{m+2}^{m+3} M_{m+3}^{m+4} \ldots$$

$$\times M_{m+2}^{m+4} M_{m+3}^{m+5} \frac{1}{E_{m+3} - E_{m+2}}$$

$$= C(n,n') h \left(\frac{E}{2(D + E)}\right)^{n+1} .$$  \hspace{1cm} (33)

so that,

$$\Delta_0 = C(n,n') \frac{\hbar^2}{\pi} \sqrt{2E(D + E)} \left(\frac{E}{2(D + E)}\right)^{n+1}$$

$$= E C(n,n') \left(\frac{E}{2(D + E)}\right)^{n+1} ,$$  \hspace{1cm} (34)

which has the same behavior as Eqn. (31) for transitions between the same parity. In both cases,

$$\Delta_0 \sim 2h^2 E e^{-|2n-n'|S_0};\quad S_0 = \ln\left(\frac{E}{2(D + E)}\right) .$$  \hspace{1cm} (35)
i.e., $2E$ is the attempt frequency and the action agrees with that which is obtained for the ground state to ground state tunneling using the functional integral method \[\text{(1)}\]. There is little interest in reducing the expressions Eqsns. \[\text{(3)}\] and \[\text{(4)}\] since precise values for splitting are easily obtained numerically. It is an interesting result that the attempt frequency is $2\hbar^2 E$ and not perhaps the characteristic energy $2\hbar^3 \sqrt{2E(D+E)}$ or $\hbar^2 \sqrt{D^2 - E^2}$.

**XIV. OTHER SYMMETRIES**

The issue that intermediate spin effects are manifested even when there is no evident easy plane has been raised in the Introduction. Consider cubic symmetry, the appropriate Hamiltonian,

$$
\mathcal{H} = a \left[ S_x^2 + S_y^2 + S_z^2 \right] + \hbar S_z
$$

has eight classical equilibrium directions, Fig. (1c) which with a positive do not coincide with the axes. Quantum mechanically, in the absence of a field the system resonates between each of these equilibrium directions. However in the language of Sec. VII, there are only four chains and for large spin only four closely spaced levels. Symmetry dictates that in zero field the chain with sites ..., $-3, 1, 5, ...$ has levels which are degenerate with the similar chain with $n \rightarrow -n$. To a good approximation for large $S$ Schrödinger’s equation is:

$$
\begin{align*}
[\epsilon - (a\hbar^4 m^4 + \frac{2a\hbar^4}{3}(S(S + 1) - m^2))]a(m) &= \frac{a\hbar^4}{4} \sum (S(S + 1) - (m \pm 2)^2) a(m \pm 4)
\end{align*}
$$

where $a(m)$ is the wave function. An analytic approach to the solution of this equation is facilitated by a modest field $H$ and large $S$. This suppresses the tunneling between the state with positive and negative projections on the $z$-direction. Now tunneling occurs between the four classical equilibrium directions with a negative $S_z$ component. It is the four least action paths between these which map to a circle. Making the continuum approximation near the minimum in the the diagonal energy with a negative such projection defines a discrete simple harmonic oscillator with a potential $\sim 2a\hbar^4 S^2 (m - m_0)^2$ where $m_0$ is the equilibrium dimensionless magnetization. In Fourier space this becomes the kinetic energy and the potential $\sim a\hbar^4 S^4 \cos 4\phi$ so that there are four equivalent minima. Apart from this reduction of the period by a factor of two, the problem is then fully equivalent to that considered in Sec. VIII and the tunnel splitting quasi-periodic as illustrated in Fig. (6). As the field increases the minimum energy directions bend towards the $z$-axis and this reduces the potential barrier between the equivalent wells and increases the kinetic energy, hence the steady increase in the tunnel splitting. The separation between the half-integer points when the splitting is zero is seen to be $\sim 4\hbar^4 S^4 a$ but varies since the coefficient of the $m^2$ kinetic energy term also increases with field. In fact there are two level crossings at small fields which occur before the large field approximation is valid and which are not visible in this figure. At least for larger fields these half-integer points really do correspond to a Kramer’s degenerate half-integer spectrum for the low lying quartet as is explained in the Introduction and the Fig. (6) caption.

The symmetry appropriate to $M_{12}$ has an easy $z$-axis with equivalent $x$ and $y$ directions so that the Hamiltonian \[\text{(5)}\],

$$
\mathcal{H} = -DS_z^2 + a \left[ S_x^2 + S_y^2 \right] + \hbar S_z,
$$

where, with a positive, the $x$-directed transverse field is along a hard direction in the $x - y$-plane. Classically in zero field there are equivalent equilibrium directions which have the magnetization parallel or anti-parallel to the $z$-direction. A finite $h$ does not alter the equivalence of these two directions, the magnetization simply gains a component opposite to the field direction. However the field does have an effect on the tunneling paths. In zero field there are four least action paths which pass via one of the easy directions in the $x - y$-plane. With a field in this particular direction and a positive there remain only two equivalent paths which pass via an easy direction for which the magnetization has a component in the direction away from the field. It is these two least action paths which map to the intermediate spin circle. Here the paths are not co-planar and the field $h$ will enter in the potential but without altering the symmetry.

It is possible to analytically account for all four tunneling paths for large $S$ and not too large a $h$ by imagining that there are two crossed circles. A given circle comprises two easy directions in the $x - y$-plane and $z$-axis, i.e., the two planes containing the circle make an angle of $45^\circ$ to the applied field. The wave function for each circle is calculated and the two are added to give the final result. Because the field is at $45^\circ$ to each circle the wave vector has equal real and imaginary parts and the net result is that displayed in the caption to Fig. (5).

**XV. EXPERIMENTAL VERIFICATION OF FIELD MIXING**

Certainly the most striking claim is that the wave function $A(1 - (\alpha\phi/2\hbar)h)u_k(\phi) + B(1 + (\alpha\phi/2\hbar)h)u_{-k}(\phi)$ for an intermediate spin system is a mixture of a state with the local applied field of magnitude $h_p = \sqrt{(D + E)/2E\delta h}; \delta h = h - h_p$, i.e., fields both larger and smaller than the distant applied field $h$. (With an equivalent claim for $h_t$ which involves $\sqrt{(D - E)/2Eh_t}$)
For a particle on a ring this would imply a distortion in the near field with a range of order of the ring diameter. Unfortunately the spin has no natural dimension and the ring diameter must be scaled to zero. Experimentally the existence of this superposition is easy to establish. In the absence of the superposition the wave function would have a simple Floquet form: \( e^{i k \phi} u_k(\phi) \), where \( k = \frac{m}{m} \) is determined, modulo unity, from the experimental tunnel splitting: \( \Delta_0 \cos \pi k \). Theory gives \( k = \frac{m}{m} = \delta h/2\sqrt{D/(D+E)} \) which agrees with experiment for Fe8. Following the Fourier transform to \( m \)-space the factor \( e^{i k \phi} \) corresponds to a displacement of \( k \) and would lead to a dimensionless magnetic moment,

\[
m_{\text{non-mix}} = k = \frac{m}{m} = \frac{\delta h}{2h\sqrt{2E(D+E)}},
\]

again modulo unity. In the limit of large \( S \) the theory accounting for the superposition leads to

\[
m_{\text{mix}} \approx \frac{h}{2h(D+E)}
\]

which is smaller except when \( E = D \). This has yet to be verified experimentally.

A more subtle way of stating the same principle goes as follows: The transverse field \( h \) couples to the physical \( S_z \), i.e., this operator in the non-singular gauge, and which has a spectrum with a period \( h \) or in units of magnetic moment \( g \mu_B h \) where \( g \) is the g-factor. Thus if \( M_z \) is the measured moment in the direction of the transverse field, \( m = M_z/g \mu_B h \), the dimensionless magnetization, has a natural period of unity, a statement equivalent to the observation that physical effects are periodic in the flux quantum \( \Phi_0 \). This would be the case if the state did not consist precisely of a superposition of field directions. Because of the admixture, the measurable period is reduced to \( \Delta m \equiv m_1 = \sqrt{2E/(D+E)} \leq 1 \) as illustrated with exact results in Fig. (11). It should be noted that the value of \( m_1 \) given above is not very accurate almost uniquely because dimensionless magnetization \( m > h/2h(D+E) \) for any reasonable \( S \) because of admixture into excited states. For example with \( E/D = 0.35 \) and \( S = 10 \) it was found that \( m = 1.0353718(h/2h(D+E)) \) was indistinguishable from the numerical data for \( m \) in the range \( 0 < h < h_1 \) and gave with five figure precision the numerical \( m_1 \). (Notice \( m_1 h = 4E \) while \( (h/m_1) = 2(D+E) \).

It is also possible to argue for the local field \( \sqrt{(D+E)^2/2h} \) in terms of the two level model. For an integer spin system for the ground state with \( h = h_1 = 0 \) this is \( \mathcal{H} = 2Sh_zS_z + \Delta_0 S_z \) where, as above, \( \Delta_0 \) is the tunnel splitting. For half-integer spin, following Kramer’s theorem this splitting is absent. However since the transverse field breaks time reversal symmetry it should create a splitting. The only satisfactory fashion of introducing this transverse Zeeman effect without introducing a new parameter is to write \( \mathcal{H} = 2Sh_zS_z + \Delta m S_z \) where \( m \) is the dimensionless magnetic moment in the \( x \)-direction. Since \( m = h/2h(D+E) \) this gives a splitting \( 2(\Delta_0/h(D+E))h \). However from Eqn. (23) this is correct only if \( h \) is replaced by the local applied field \( \pi\sqrt{(D+E)^2/2h} \) to give a splitting \( 2\Delta_0(2h/2h(D+E)) \). The field \( h_t \) similarly generates a term \( \pi\Delta(h_t/2h(D-E))S_y \). Clearly this model it is subject to experimental verification and nicely illustrates the effects of squeezing via the different factors involved with the different components of the transverse field.

\[
\text{FIG. 11. The dimensionless moment } m(h). \text{ The field is divided by half the period so that all steps coincide and } S = 5. \text{ The moment period is exactly unity for } E = D \text{ but otherwise is less and implies a Schrödinger’s cat with a superposition of fields.}
\]

\[
\text{APPENDIX}
\]

Given that \( h^2D \) is the dominant anisotropy energy it would seem the most logical to use this easy axis for the quantization, however much of the physics of the problem corresponds to the fact that term proportional to \( E \) creates an easy axis. It is therefore useful to re-write \( \mathcal{H} \) quantized along the hard axis. To this end, first use
\[ S_y^2 = \hbar^2 S(S+1) - S_x^2 - S_z^2 \] 
whence,

\[ \mathcal{H} = -(D - E)S_z^2 + 2ES_x^2 - E\hbar^2 S(S+1), \]

then interchanging the \(x\) and \(z\) axes results in

\[ \mathcal{H} = 2ES_z^2 - hS_z - (D - E)S_x^2 - hS_x - hS_y, \]

where the constant has been omitted and the fields reintroduced.

The case when \(D < E\) can be transformed to \(D' > E'\) where \(D' = 3E - D\) and \(E' = D + E\). To show this, first use again \(S_z^2 = \hbar^2 S(S+1) - S_x^2 - S_y^2\) to write:

\[ \mathcal{H} = -DS(S+1) + (D + E)S_x^2 - (E - D)S_y^2. \]

With the interchange of the \(z\) and \(y\)-axes and to within a constant:

\[ \mathcal{H} = -(E - D)S_z^2 + (D + E)S_x^2. \]

Comparing this with \(\mathcal{H} = -(D - E)S_z^2 + 2ES_x^2\) it is seen that,

\[ D + E \rightarrow 2E, \quad E - D \rightarrow D - E \]

reduces the second to the first version. These manipulations leave the term \(hS_z\) unchanged. Hence it must be, in particular, that the result for the period \(\hbar_1 = 2\hbar\sqrt{2E(D + E)}\) be invariant under the transformation

\[ E \rightarrow \frac{1}{2}(D + E) \quad D \rightarrow \frac{1}{2}(3E - D). \]

Then if \(E > D\) it follows that \(D' = 3E - D > 2E\) while \(E' = D + E < 2E\) so that for the transformed problem \(D' > E'\), i.e., only that case when \(D > E\) need be considered.

Another useful version of \(\mathcal{H}\) is for \(h_t = h_e = 0\),

\[ \mathcal{H} = -(D + E)S_z^2 - 2ES_y^2 - hS_z. \]

---

[1] For a review, see the many articles in *Quantum Tunneling of Magnetization*, edited by L. Gunther and B. Barbara (Kluwer, Dordrecht, 1995); and E. M. Chudnovsky and J. Tejada, *Macroscopic Quantum Tunneling of the Magnetic Moment* (Cambridge University Press, 1997).

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