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**LETTER TO THE EDITOR**

# Intermediate spin and certain small magnets

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**Abstract.** An isolated spin $S$ can have only whole or half-integer values. It is shown that under certain circumstances a system composed of such spins can have low-lying excitations which correspond to a different value of the spin $S_{\rm eff}$ which is intermediate between these standard possibilities. A general demonstration valid in any dimension is given followed by a detailed examination of the simplest, zero-dimensional example, namely a small easy plane ferromagnet. Some experimental implications are worked out.

Reflecting the effect of the so-called *topological term* (see, e.g., Fradkin 1991), Haldane (1985) has conjectured that one-dimensional quantum field theories based on half and whole integer spins differ in a significant fashion, i.e., the latter have a gap in the low-energy spectrum while the former do not. A similar effect, reflecting a topological term, occurs for small zero-dimensional magnets. Loss *et al* (1992) and von Delft and Henley (1992) have shown that the amplitude for tunnelling with a complete reversal of the magnetization of a small ferromagnet is strictly null for half-integer spin in situations where it is finite for a similar system with integer spin. Garg (1993) has shown, for a small easy axis ferromagnet with a field perpendicular to the direction of the magnetization, this same amplitude oscillates as a function of field.

Every elementary quantum mechanics text offers a proof that only half and whole integer spin values are allowed. It is interesting to investigate the simplest situations when this standard result can be called into question *for low-lying excitations*. The point of the exercise is not to challenge the authenticity of the basic theorem, any more than the demonstration of the possible existence of anyons in condensed matter systems is intended to disprove that such a system is ultimately composed of bosons and fermions. The goal is to show that the low-lying excitations reflect the physics of a spin $S_{\rm eff}$ which has a value which is intermediate between a whole and half-integer. It remains the case that the system is constructed from spins with a whole or half-integer value of the spin $S$.

Consider the following Hamiltonian describing such whole or half-integer spins:

$$\mathcal{H}(H) = g\mu_B H \sum_n S_{nz} + K_\parallel \sum_n S_{nz}^2 + \mathcal{H}_1(\boldsymbol{S}_n) \tag{1}$$

where $\mathcal{H}_1$ is a fairly general (scalar) function of the $\boldsymbol{S}_n$ and $n$ is some real space site index. The *key* assumptions are that $S$ and the positive anisotropy energy $K_\parallel$ are sufficiently large that $S^2K_\parallel \gg g\mu_B H$, $\langle\mathcal{H}_1\rangle$, and that $S$ is not *too* small. (The classical limit is *not* assumed, e.g., $S = 2$ or 3 will exhibit the predicted phenomena.) The expectation values are for the low-lying states of interest. This inequality forces the spins to lie close to the $x$–$y$ plane, i.e., the relevant states are constructed from eigenstates of $S_z$ such that $|S_z| \ll S$. When this is the case the matrix elements of $S^\pm$ are $S$, or equivalently $[S^+, S^-] = 0$, to a good approximation. Then to within an additive constant, the magnetic field $H$ can be absorbed into the anisotropy energy by the substitution

$$\hat{S}_z \to \hat{S}_z - a \qquad a = \frac{g\mu_B H}{2K_\parallel} \tag{2}$$

and the new spin operator $\hat{S}_z - a$ obeys the approximate spin commutation rules. This observation suffices to prove that $\mathcal{H}(H)$ is equivalent to $\mathcal{H}(H = 0)$ *if for the latter* the spectrum of allowed values of $S_z$ are

$$n - \frac{g\mu_B H}{2K_\parallel} \tag{3}$$

where $n$ is a whole or half-integer. In general this spectrum does *not* correspond to either whole or half-integer spin.

In fact, in all respects the problem in finite field, $\mathcal{H}(H)$, is equivalent to that in zero field, $\mathcal{H}(H = 0)$, *but* with a value of the total spin $S_{\mathrm{eff}} = S - (g\mu_B H/2K_\parallel)$ which differs from the whole or half-integer value $S$ of the constituting particles by the quantity $g\mu_B H/2K_\parallel$. The resulting effective spin value is neither whole or half-integer unless $g\mu_B H/2K_\parallel$ is either a whole or half-integer. Thus the Hamiltonian $\mathcal{H}(H)$ is equivalent to a problem with intermediate spin which is *periodic* with a field period $H_p = (2K_\parallel/g\mu_B)$. A field of $H_{1/2} = K_\parallel/g\mu_B$ converts the half-integer problem into the whole integer equivalent and vice versa.

Since the Hamiltonian (1), for one dimension, lies in the class of models believed to be covered by Haldane's conjecture, it is implied that as a function of applied field parallel to the hard axis there will be periodic half-integer 'phases', without a gap, with whole-integer gapped 'phases' between. Also since there are no phase transitions as a function of such an applied field, the tunnelling amplitude for a complete reversal of spin in zero dimensions must be periodic with a zero amplitude occurring at points on the field axis. In addition, the present demonstration is valid for arbitrary dimensions and implies an applied field periodicity in the properties of a whole class of Hamiltonians.

These predicted oscillations *have* been observed by Taft *et al* (1994), in the quasi-zero-dimensional antiferromagnet $Fe_{10}$, although the interpretation offered by those authors is different.

In the following, this general result will be investigated for the simplest example, namely a small easy axis ferromagnet. Such a zero-dimensional ferromagnet can be modelled by a single large spin subject to the external and anisotropy fields, i.e., the Hamiltonian $\mathcal{H} = g\mu_B \boldsymbol{S} \cdot \boldsymbol{H} + K_\parallel {S_z}^2 + K_\perp {S_x}^2$ where without loss of generality it is assumed that $|K_\parallel| > |K_\perp|$ (see, e.g., Chudnovsky 1995).

Experimentally it is important to observe that the energy parameters $K_\parallel$, $K_\perp$ all scale as $S^{-1}$. The equivalent physical quantities are $g\mu_B H_\parallel = K_\parallel/S$, and $g\mu_B H_\perp = K_\perp/S$. Thus a field of $H = H_\parallel/S$ transforms the integer easy axis ferromagnet into the half-integer equivalent and vice versa.

The detailed formulation is in terms of auxiliary particles (see Barnes *et al* 1997). A basis $|S_z\rangle \equiv |n\rangle$ is chosen, and an auxiliary particle, a fermion $f_n$, is associated with each state via the mapping $|n\rangle \to f_n^\dagger|\rangle$ where $|\rangle$ is a non-physical vacuum without any auxiliary particles. Define a bi-quadratic version of an operator $\hat{O}$ via: $\hat{O} \to \sum_{n,n'} f_n^\dagger \langle n|\hat{O}|n'\rangle f_{n'}$. The constraint $Q = \sum_n \hat{n}_n = \sum_n f_n^\dagger f_n = 1$ holds. It has been shown (see, e.g., Barnes 1981) that such schemes preserve all operator multiplication rules including commutation.

With $h = g\mu_B H$, the replacement rule is applied to $\mathcal{H}$ to yield:

$$\mathcal{H} = \sum_n \left(K_\parallel n^2 - nh + \tfrac{1}{4}K_\perp[(M_n^{n+1})^2 + (M_n^{n-1})^2]\right) f_n^\dagger f_n + \tfrac{1}{4}K_\perp \sum_n M_n^{n+1} M_{n+1}^{n+2} (f_{n+2}^\dagger f_n + \text{H.C.}) \tag{4}$$

where the $M_n^{n+1} = [S(S+1) - n(n+1)]^{1/2}$ are the matrix elements of $S^\pm$. This represents *two* tight binding models of spinless fermions $f_n^\dagger$. The constraint $Q = 1$ implies this is a single-particle problem.

The 'two-chain' structure reflects the fact that the 'hopping' term in (4) couples 'sites' with indices which differ by two, resulting in distinct even and odd site chains. The present formulation and the resulting chains are a convenient method to reflect the underlying structure of the characteristic determinant. As noted by a number of authors, this structure implies immediately a spin-parity effect found by Loss *et al* (1992) and von Delft and Henley (1992). For integer spin, the two chains comprise the sites

$$n = -S, -(S-2), -(S-3), \ldots, (S-3), (S-2), S$$

and

$$n = -(S-1), -(S-3), -(S-5), \ldots, (S-5), (S-3), (S-1)$$

which for $h = 0$ are both symmetric relative to $n = 0$. On the other hand for half-integer spin the chains are

$$n = -S, -(S-2), -(S-3), \ldots, (S-5), (S-3), (S-1)$$

and

$$n = -(S-1), -(S-3), -(S-5), \ldots, (S-3), (S-2), S$$

which for $h = 0$ are equivalent to each other through the map $n \to -n$ but which lack the symmetry about $n = 0$. Because of the equivalence of the two chains there must always be a double (Kramers') degenerate ground state, without a tunnel splitting, for half-integer spin and $h = 0$, while, because of symmetry about $n = 0$, tunnel split pairs for integer spin can exist.

To see how a whole integer spin is converted into a half-integer equivalent, consider first the *fixed point* defined by $K_\perp \to 0$. All off-diagonal matrix elements are zero, the diagonal energies are $K_\parallel n^2 - hn$, and for an *integer spin easy plane ferromagnet*, i.e., with $K_\parallel > 0$, and in zero field ($h = 0$), the ground state lies on the site $n = 0$, and therefore on the even site chain. The first excited states, at an energy $K_\parallel$, are located on the sites $n = \pm 1$ and correspond to the odd site chain. For a similar system but of *half-integer spin* the two chains are fully equivalent and there are two grounds states. One chain contains the ground state at $n = +1/2$ and the other at $n = -1/2$ and all excited states, corresponding to the sites $n = \pm 3/2$, $n = \pm 5/2$, etc, also occur in pairs. Now consider the situation when $h = K_\parallel$ and first whole integer spin. The pairs of states $n = 0, +1$, $n = -1, +2$, etc, are degenerate and might be mapped onto the states $\pm 1/2$, $\pm 3/2$, etc, of the half-integer system in *zero* field. Similarly, for half-integer spin and the same field, the energy of the site $n = +1/2$ is $K_\parallel$ lower than that with $n = -1/2$ and this latter is now degenerate with $n = +3/2$, so the non-degenerate ground state is on the chain which contains the site $n = +1/2$ with the first excited states on the other chain which contains the pair $n = -1/2, +3/2$, thus mimicking the whole-integer spin chain in zero field. In fact for whole integer spin there is a level crossing whenever

$$h = (2n+1)K_\parallel \qquad n = 0, 1, 2, 3 \ldots \tag{5}$$

and at such points all the ground state and low-lying excited states are identical to those of the half-integer chain in zero field, and, evidently, these same fields transform a half-integer chain into its integer twin. Other fields correspond to intermediate spin values.

These crossings have period $2K_\parallel$, and with this period the splitting between the ground state and the first excited state undergoes triangular oscillations with amplitude $\sim K_\parallel$ and, with each level crossing, the ground state passes from one chain to the other. Since physical qualities are analytic in $K_\perp$, this fixed point will govern the behaviour for all values of this parameter. This is verified by both numerical calculations and by the analytic results described below.

The vicinity of the fixed point with

$$S^2 K_\perp < K_\parallel \tag{6}$$

defines the *small particle limit*. In the large spin limit all small $n$ states undergo a constant shift of $S^2 K_\perp/2$, otherwise the corrections to the ground state $|n = 0\rangle$ are perturbative, however the excited states split. Ignoring the constant shift, these are $(1/\sqrt{2})[|1\rangle \pm |-1\rangle]$ with energies $K_\parallel \pm S^2 K_\perp/4$. The $h = 0$ ground to lowest exicted state splitting $K_\parallel - S^2|K_\perp|/4$ determines the amplitude of the near triangular oscillations. This splitting has nothing to do with tunnelling. However the ground state is clearly 'coherent', e.g., $\langle \boldsymbol{S}\rangle = 0$, i.e., the order parameter is not localized in a particular direction in the $x$–$y$ plane.

Generally Schrödinger's equation

$$(\epsilon - (K_\parallel n^2 - nh))a_n = \tfrac{1}{4}K_\perp\big[M_n^{n+1}M_{n+1}^{n+2}a_{n+2} + M_n^{n-1}M_{n-1}^{n-2}a_{n-2} \\ -[(M_n^{n+1})^2 + (M_n^{n-1})^2]a_n\big] \tag{7}$$

involves finite differences, where the wavefunction $\Psi = \sum_n (-)^{n/2} a_n f_n^\dagger|\rangle$. When the inequality $S^2 K_\perp < K_\parallel$ is reversed the particle is *large*, and assuming $K_\perp < 0$, $a_n$ varies slowly as a function of the discrete $n$. However the wavefunction *remains well localized near* $n \sim 0$, so the matrix elements $M_n^{n+1}$ can be replaced by $S$ and for low-lying states the wavefunction does not 'see' the chain ends.

That the low-lying states are insensitive to the boundary conditions is the physical reason why intermediate spin values are possible and even physically necessary. In the standard proof where the spin must take whole or half-integer spin values it is precisely the insistence that, in the present language, the chains must terminate which limits the spin to these values. Since the low-lying excitations do not 'know' the chains terminate it is possible to find a parameter, here the applied magnetic field, which determines an arbitrary effective spin value for these excitations. More precisely, in (7) the magnetic field can be absorbed, e.g., for even spin, into shifts of $d = h/2K_\parallel$ and $h^2/4K_\parallel$ in the origin and the energy, respectively. In fact to within unimportant energy shifts, it is possible to define a single problem which accommodates *all values of the field and both whole and half-integer spin* by defining the origin to be at the centre of the harmonic potential and then displacing the chains by $d = h/2K_\parallel$ for whole integer, and $d = -1/2 + h/2K_\parallel$ for half-integer spin. It is immediately seen that the only difference between whole and half-integer spin is a *discrete* shift of $1/2$ and that a fully equivalent *continuous* shift is induced by the external field for small fields. Whole and half-integer spins with a value $\sim S$ correspond to whole or half-integer $d$ while other values of $d$ correspond to intermediate spin.

In order to compare this with previous calculations, and for an eventual comparison with experiment, it is interesting to obtain explicit expressions for the tunnel splitting for this large particle case. To this end a wavefunction, continuous in $n$, is defined by $a(n+d) = a_n$.

After a Fourier transform $f(y) = \frac{1}{\sqrt{2\pi}} \int dn\, e^{iny} a(n)$, equation (7) becomes

$$\left(E + K_\parallel \frac{\partial^2}{\partial y^2}\right) f(y) = -\frac{S^2 K_\perp}{2} [\cos 2y - 1] f(y) \tag{8}$$

which is Mathieu's equation (see Blanch 1964), and where $E = \epsilon - (h^2/4K_\parallel)$. The potential is periodic with period $\Delta y = \pi$ and the solutions form bands. (In the relevant limit, this agrees with Zaslavskii (1990) who uses a different method to reduce the easy plane problem to that of a particle in a periodic potential. He also considers the field dependence of the solution but for a different, less interesting direction of the field.) For a given energy, a solution might be characterized by a wavevector $k$ and Floquet's (Bloch's) theorem implies that solutions are of the form $f_k(y) = e^{iky} u_k(y)$ where $u_k(y) = u_k(p+\pi)$. That the solution is finite on the even site chain implies $k = d - 2m$ where $m$ is a unique integer which reduces $k$ to the first Brillouin zone $\{0, 2\}$. Similarly for the odd site chain $k = d + 1 - 2m$.

Around $y = 0$, $f(y) = (1/\sqrt{\beta\sqrt{\pi}}) e^{-y^2/2\beta^2}$ with $\beta^2 = S\sqrt{K_\parallel/K_\perp}$, and the nominal ground state energy is $(h^2/4K_\parallel) + (\omega_0/2)$; $\omega_0 = 2S\sqrt{K_\parallel K_\perp}$. The band energies are: $\epsilon_k = (h^2/4K_\parallel) + (\omega_0/2) + (w/2)\cos \pi k$ where the width (Blanch 1964) $w = 8\sqrt{2/\pi}\,\omega_0 S_f^{1/2} e^{-S_f}$ and where the action $S_f = 2S\sqrt{(K_\perp/K_\parallel)}$. For whole integer spin, the result for the 'tunnel splitting', i.e., the difference in energy between the ground and first excited states is

$$\delta E = 8\sqrt{\frac{2}{\pi}}\,\omega_0 S_f^{1/2} e^{-S_f} \left|\cos\left(\pi \frac{SH}{2H_\parallel}\right)\right| \qquad S_f = 2S\sqrt{(H_\perp/H_\parallel)}. \tag{9}$$

The order of magnitude and simple cosine dependence of this result have been confirmed by numerical experiments. (While Garg (1993) has previously shown that $\delta E$ oscillates he gives no explicit expression for the tunnel splitting.) The smooth change in effective spin is reflected by the factor $|\cos(\pi(SH/2H_\parallel))|$. For half-integer spin equation (9) again applies *but* with the cosine replaced by a sine. These two versions of (9) should be compared with a similar, $h = 0$, expression obtained by Loss *et al* (1992) and by von Delft and Henley (1992). Their result contains instead a factor $|\cos(\pi S)|$. Here $(SH/2H_\parallel)$ is identified as the shift from whole or half-integer spin and (9) is seen to be a simple generalization of the earlier result to the case of intermediate spin.

As an aside, it might be noted that Coleman (1985) has discussed how the Schrödinger equation (8) is the simplest model which exhibits a so-called $\theta$-vacuum. In his language, the vacua localized near $y = n\pi$ are connected by instantons. The resulting vacuum is denoted $|\theta\rangle$ and has an energy of the form $E(\theta) = (\hbar\omega_0/2) + 2\hbar K \cos\theta\, e^{-S_0/\hbar}$. The present problem has *two* such vacua with $\theta = \pi k = \pi(h/2K_\parallel)$ and $\theta = \pi[1 + (h/2K_\parallel)]$ for the even and odd site chains, respectively.

From the experimental point of view it is important to inspect the stability of the present periodic-in-field solution with respect to a misalignment of the field. A magnetic field lying in the $x$–$y$ plane couples the two chains. Still, for a larger particle and $K_\perp < 0$, the wavefunction, $a_n$, for $h = 0$ is extended, does not alternate in sign between sites, and is well localized near $n = 0$. The operators $S^\pm$, to a good approximation, cause translations by one lattice site, i.e., they convert the ground state on one chain into that on the other. The matrix elements of $h_x S_x = (h_x/2)[S^+ + S^-]$ are approximately $h_x S/2$ while those of $h_y S_y$ are negligible. Assuming that $h$ is smaller than the splitting, $\sim \omega_0$, between the lowest energy doublet and other excited states, the splitting of the doublet is:

$$\delta E_h = \sqrt{(\delta E)^2 + (Sh_x)^2}.$$

Evidently the field scale is $\sim \delta E/g\mu_B S$ and implies that a very careful alignment is required in order to observe oscillations. If $K_\perp > 0$, $a_n$ alternates in sign between sites and the role of the $x$ and $y$ axes are interchanged.

Together, the above results illustrate the phenomenon of intermediate spin. In all cases there is no difference between integer and half-integer spin except for a $\pi/2$ phase shift while the applied field can induce an arbitrary shift. With a period $H_p = 2H_\parallel/S$ the physics changes smoothly from that appropriate to one parity to the other and back again. In this way, e.g., with a field of $H = H_\parallel/2S$ a system composed of consituent spins $S = 2$ will exibit at low energies the properties of $S_{\rm eff} = 2\frac{1}{4}$. However this might also be thought of as $S_{\rm eff} = 1\frac{3}{4}$ since, to within a change in the sign of definition of the $y$-axis, the two possibilities give the same spectrum for $S_z$, *and* the requirement that the spin be not *too* small implies that $|a| = 1/4$ be much smaller than $S$ (or $S_{\rm eff}$). This latter approximation permits replacing the matrix elements of $S^\pm$ by $S$ or *either* of the two values of $S_{\rm eff}$. Clearly this latter approximation is better satisfied for say $S = 10$ than for $S = 2$ but it remains the case that the smaller spin value will exhibit limited quasi-periodic oscillations.

Finally it perhaps needs emphasizing that an *easy axis* ferromagnet (see Chudnovsky 1995) is quite different from its *easy plane* equivalent. In particular there is no oscillatory field dependence.

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