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Macroscopic quantum coherence in small antiferromagnetic particles

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Abstract. The Néel vector of a small antiferromagnetic particle can resonate between degenerate directions. The total moment of the particle will generally be non-zero, and this resonance cannot then occur without transverse anisotropy. Previous work is extended to include such anisotropy via a two-spin model representing the two sublattice moments. The WKB exponent in the tunnel splitting is found exactly when the sublattice moments are equal, and by a variational method that holds over the entire parameter space when the moments are unequal.

1. Introduction

Consider a small (~ 50 Å radius) antiferromagnetic (AFM) particle at a temperature well below its anisotropy gap, in zero magnetic field. Let us assume a simple two-sublattice AFM, and denote the sublattices A and B, and the easy direction for the moments by $\pm\hat{z}$. This system has a classical ground state where moments on A point along $\hat{z}$ and on B along $-\hat{z}$, and another state, degenerate with the first, obtained by reversing all the moments. One can now envisage quantum mechanical resonance between these degenerate states, and since this would require the simultaneous tunnelling of $\sim 10^5$ moments, this would be an example of macroscopic quantum coherence (MQC) according to Leggett [1].

The above system was proposed as a candidate for MQC in parallel by Barbara and Chudnovsky [2], and by Krive and Zaslavskii [3] shortly after a similar proposal [4] for ferromagnetic FM particles [5]. The vast majority of macroscopic systems are unsuitable for the observation of MQC simply because the tunnelling frequency (or splitting) is astronomically small, although even for those that pass this test, formidable obstacles remain, arising from the inescapable environmental interactions [1]. It is in the first regard that AFM particles are attractive candidates for MQC. To see this, note that the tunnel splitting Δ for a degenerate double-well system is generally given by

$$\Delta = c\omega_0 e^{-c'U/\hbar\omega_0} \tag{1}$$

where U is the barrier between the wells, ω_0 is the small oscillation frequency in one of the wells, and c and c' are constants of the order of unity. The energy barrier is governed by anisotropy and is similar for both FM and AFM particles. Taking an anisotropy energy of 10^5 erg cm^{-3}, we obtain $U \simeq 10^{-14}$ erg in both cases. The frequency ω_0 is the AFM or FM resonance frequency (ω_{AFM} or ω_{FM}, respectively) and these are very different:

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$\omega_{\mathrm{AFM}} \propto (H_{\mathrm{ex}} H_{\mathrm{a}})^{1/2}$, while $\omega_{\mathrm{FM}} \propto H_{\mathrm{a}}$, where H_{ex} and H_{a} are the exchange and local anisotropy fields, respectively. Since $H_{\mathrm{ex}}/H_{\mathrm{a}}$ can be as large as 10^4, ω_{AFM} can be much larger than ω_{FM}, and is often $\sim 10^{12}$ s^{-1}, yielding $\hbar\omega_{\mathrm{AFM}} \simeq 10^{-15}$ erg. This gives hope that the exponent in (1) will not be too large (less than 30, say) for AFM particles.

Though (1) provides an adequate first estimate, a proper calculation of Δ is somewhat harder. As noted in [2], a finite-sized AFM particle will have unequal numbers of A and B lattice sites, giving a net or uncompensated magnetic moment $\boldsymbol{M}$, which must reverse direction along with the Néel vector $\hat{\boldsymbol{l}}$. Experimental detection of MQC would be most simply done by looking at the reversal of $\boldsymbol{M}$†. Early calculations, however, were done using a Lagrangian for the Néel vector (a non-linear sigma model), effectively assuming equal sublattice moments, and an anisotropy energy with exact cylindrical symmetry. But, with such symmetry, $\boldsymbol{M} \cdot \hat{\boldsymbol{z}}$ is strictly conserved and cannot tunnel at all, so that if one is to connect with real experiments, and theoretically understand particles with equal sublattices, one must at the same time include transverse anisotropy. A model that does this is introduced in section 2. We will see below (section 3) that the WKB exponent in Δ is given by the least value of a certain action, so the problem is effectively one of classical mechanics. With cylindrical symmetry one has enough conserved quantities to solve this mechanical problem completely. When transverse anisotropy is included, this is not so. It is our goal in this paper to study the problem with transverse anisotropy. We will solve the problem exactly for equal sublattice spins (in spite of the lack of integrability) in section 4, and then variationally for unequal spins in section 5. A comparison with previous work (section 6) concludes the paper.

Readers who do not wish to see the details of our solution should read section 2 where the model and basic parameters are introduced, skip to (6) which relates Δ to the least action, and skip again to our final result, contained in (25) and (26), and the ensuing discussion of this result.

2. The two-spin model

The above situation is summed up in figure 1. We note that a calculation including transverse anisotropy that holds close to the X–Y or easy plane limit has previously been done by Loss and co-workers [9]. They start from an indirect model, consisting of an AFM spin chain, and a single large spin representing the excess moment, to which the spins in the chain are coupled with alternating sign. We study instead a more obvious model where we treat the total moments on each sublattice as spins of large, fixed, unequal magnitudes [10]. Denoting their directions by unit vectors $\hat{\boldsymbol{n}}_{\mathrm{a}}$ and $\hat{\boldsymbol{n}}_{\mathrm{b}}$, we have the Hamiltonian

$$\mathcal{H}(\hat{\boldsymbol{n}}_{\mathrm{a}}, \hat{\boldsymbol{n}}_{\mathrm{b}}) = J\hat{\boldsymbol{n}}_{\mathrm{a}} \cdot \hat{\boldsymbol{n}}_{\mathrm{b}} - \sum_{\alpha=\mathrm{a,b}} (K_{1\alpha}\hat{n}_{\alpha z}^2 + K_{2\alpha}\hat{n}_{\alpha x}^2). \tag{2}$$

Here $J \gg K_{i\alpha} > 0$ is the exchange energy, and the $K_{i\alpha}$ values are anisotropy energies. We take $K_{i\alpha} = N_\alpha k_i$, where N_α ($\alpha = \mathrm{a}, \mathrm{b}$) are the numbers of sites in the two sublattices, and $k_1 > k_2 > 0$. It is convenient to define

$$N = N_{\mathrm{a}} + N_{\mathrm{b}} \qquad p = (N_{\mathrm{a}} - N_{\mathrm{b}})/N. \tag{3}$$

† Precisely such a detection of MQC has been claimed in particles of ferritin [6], although one of us has argued that this claim is implausible [7, 8].

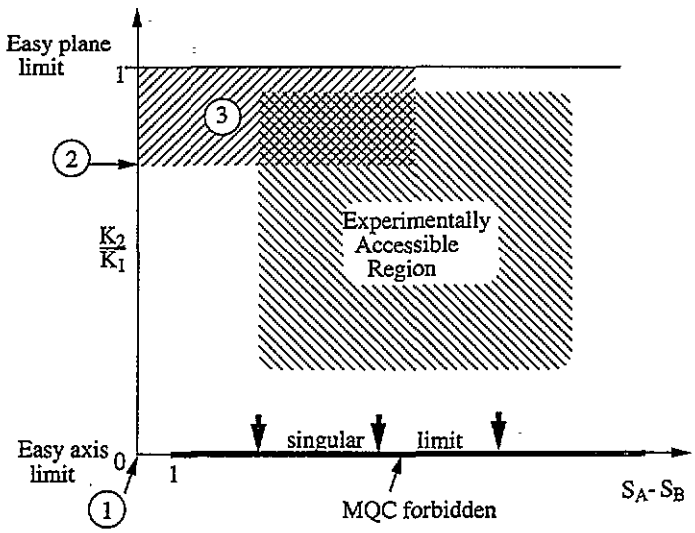

Figure 1. The parameter space of the MQC problem in small AFM particles. The encircled numbers denote the following: 1, the equal-sublattice, easy axis solution of [2] and [3]; 2, our exact equal-sublattice result (10) for any K_2/K_1; 3, the result of [9] close to the X–Y limit. Our variational result (25) and (26) holds over the entire parameter space.

Table 1. The correspondence between this work and [9]. E_e, E_m, and E_h denote the energy of the particle when all moments lie along the easy, medium, or hard axes, respectively.

Axes/energy	This work	[9]
easy/medium/hard	$z/x/y$	$x/y/z$
$E_h - E_e$	Nk_1	k_z
$E_m - E_e$	$N(k_1 - k_2)$	k_y

p is thus the fractional excess spin. It is also convenient to write $K_{1,2} = k_{1,2}N/2$ and $J = \tilde{j}N/2$.

The correspondence between our notation and that of [9] is given in table 1 to assist in comparing our results with theirs. By equating *differences* in the energies of various spin orientations, we eliminate any possible confusion due to the choice of zero of energy, and we see that the coefficients k_y and k_z of [9] correspond to $N(k_1 - k_2)$ and Nk_1, respectively, in our notation.

The model (2) is often used to understand bulk AFM resonance and (with suitable gradient energies added) spin waves [11]. It is useful to relate its parameters to ω_{AFM} and the transverse susceptibility $\chi_\perp$. Due to the K_2 term, we should distinguish two AFM resonance modes, and two $\chi_\perp$ values. Because $\tilde{j} \gg k_1, k_2$, however, the difference between the $\chi_\perp$ values is negligible, but the same is not necessarily true of the resonance frequencies. Dropping terms of order $k_1/\tilde{j}$, $k_2/\tilde{j}$, we have

$$\omega_I^2 = 4(k_1 - k_2)\tilde{j}/\hbar^2 s^2 \qquad \omega_{II}^2 = 4k_1\tilde{j}/\hbar^2 s^2 \qquad \chi_\perp = N(\hbar\gamma s)^2/v_0\tilde{j} \quad (4)$$

where v_0 is the particle volume, s is the spin on one site, and γ is the spectroscopic splitting ratio.

3. The formalism for calculating tunnel splitting

The quantal dynamics of the spins can be specified via an action to be used in a Feynman path integral. The Euclidean version of this action is given by [12]

$$S[\hat{n}_\alpha(\tau)] = i\hbar s \sum_{\alpha=a,b} N_\alpha \int \phi_\alpha \dot{z}_\alpha \, d\tau + \int \mathcal{H}(\hat{n}_\alpha(\tau)) \, d\tau. \tag{5}$$

We have written $z_\alpha = \cos\theta_\alpha$, where θ_α and ϕ_α are the polar angles of $\hat{n}_\alpha$. Also, $\dot{z}_\alpha = dz_\alpha/d\tau$. The tunnel splitting is readily formulated using (5) and instanton methods [13]. The central quantity in Δ is the WKB exponent ($c'U_0/\hbar\omega_0$ in (1)) and this is given by the least value of $S/\hbar$ subject to the boundary conditions $\hat{n}_a(\pm\infty) = -\hat{n}_b(\pm\infty) = \mp\hat{z}$. The problem is thus reduced to finding the classical trajectory (the instanton) and the associated action.

We briefly note that the first term in (5) gives rise to interference effects that have been studied in several papers recently [9, 14, 15]. What happens is that there are symmetry related instantons with the same value for the real part of the action, but whose imaginary parts differ by $2i\pi Ns$ due to the first term in (5). For our problem, this phenomenon is entirely equivalent to Kramers' theorem, which states that all energy levels are degenerate if the total spin Ns is a half integer, and which therefore implies that $\Delta = 0$ strictly. If Ns is an integer, the interference is constructive, and Δ is twice the single instanton answer. Since Ns is a half integer if and only if the excess spin Nps is, the final answer for Δ can be written as

$$\Delta = \begin{cases} 2c\omega_{\mathrm{AFM}} e^{-S_{\mathrm{cl}}/\hbar} & Nps = \text{integer} \\ 0 & Nps = \text{half integer.} \end{cases} \tag{6}$$

Here S_{cl} is the least action, and c is a single-instanton determinantal prefactor [16] that we shall not find as it affects the answer only weakly.

The equations of motion for the instanton, i.e., the Euler–Lagrange conditions, are easily found to be

$$\begin{aligned} iN_a\hbar s\dot{z}_a &= J[(1-z_a^2)(1-z_b^2)]^{1/2}\sin(\phi_a-\phi_b) - K_{2a}(1-z_a^2)\sin 2\phi_a \\ iN_a\hbar s\dot{\phi}_a &= J[z_b - z_a[(1-z_b^2)/(1-z_a^2)]^{1/2}\cos(\phi_a-\phi_b)] - 2z_a(K_{1a} - K_{2a}\cos^2\phi_a) \end{aligned} \tag{7}$$

and an identical pair obtained by interchanging the subscripts a and b everywhere†. It is easy to see that the energy $\mathcal{H}(\hat{n}_a, \hat{n}_b)$ is conserved, but that the total spin along the z axis, $N_a z_a + N_b z_b$, is *not* conserved due to the presence of the K_2 terms.

Because the energy is conserved, there can be no real trajectory connecting our initial and final states. To decide which of the components of the trajectory should be made complex, it is very useful to think in terms of a discrete WKB approach [17]. Imagine rewriting the Hamiltonian (2) by formally replacing $\hat{n}_a$ and $\hat{n}_b$ by scaled spin operators, S_a/S_a and S_b/S_b. If we expand the wavefunctions in the $|m_a, m_b\rangle$ basis, where $S_a^z|m_a, m_b\rangle = m_a|m_a, m_b\rangle$ etc, then Schrödinger's equation becomes a recursion relation for the expansion coefficients. We now view this recursion relation as a tight-binding model for an electron moving on a two-dimensional lattice with slowly varying site dependent hopping matrix elements and on site energies. The discrete WKB approach is equivalent to solving this problem using semiclassical electron dynamics, with z_a and z_b as the position coordinates and ϕ_a and ϕ_b as the crystal momentum components. The corresponding semiclassical equations are precisely (7). While this insight does not make the actual calculation any easier, it does suggest that, as for a continuum problem, we take the positions z_a and z_b to be real and to lie in the interval $[-1, 1]$, and allow the momenta, i.e., ϕ_a and ϕ_b, to become complex.

† These equations are also profitably viewed in Cartesian components.

4. Equal sublattices—the exact solution

We now present the exact solution when $N_a = N_b$. Our problem is not among the class of integrable models of pairs of interacting spins found by Magyari *et al* [18], so one cannot solve the equations of motion for arbitrary initial conditions. There is no bar, however, to finding a special solution, which is all we need. Indeed, one can see from (7) that if either of the following sets of conditions holds at a given instant of time, it holds at all others:

$$\begin{aligned} &\text{(I)} \qquad z_b = -z_a \qquad \phi_b = \pi - \phi_a \\ &\text{(II)} \qquad z_b = -z_a \qquad \phi_b = -\phi_a. \end{aligned} \tag{8}$$

Since these are met by our boundary conditions, the problem is effectively turned into an integrable single-spin problem. Energy conservation shows that ϕ_a is constant in time for both types of solution. For type (I), we have

$$z_a = -\tanh \omega_I \tau \qquad \cos^2 \phi_a = (\tilde{j} + k_1)/(\tilde{j} + k_2) \tag{9}$$

$$S_I = 2N\hbar s \log \left[\sqrt{\frac{\tilde{j} + k_1}{\tilde{j} + k_2}} + \sqrt{\frac{k_1 - k_2}{\tilde{j} + k_2}} \right]. \tag{10}$$

The type (II) solution is obtained by replacing k_1 and k_2 by $k_1 - k_2$ and $-k_2$ everywhere, and writing $\sin^2 \phi_a$ instead of $\cos^2 \phi_a$ in (9). In particular, the action is

$$S_{II} = 2N\hbar s \log \left[\sqrt{\frac{\tilde{j} + k_1 - k_2}{\tilde{j} - k_2}} + \sqrt{\frac{k_1}{\tilde{j} - k_2}} \right]. \tag{11}$$

These solutions are degenerate when $k_2 = 0$, and go over into those found in [10]. For $k_2 \neq 0$, $S_I < S_{II}$, so the dominant contribution to the WKB exponent for Δ is given by S_I. In the physically relevant limit when $\tilde{j} \gg k_1 > k_2$, we can write $S_I \approx 4N\hbar s(k_1 - k_2)/\omega_I$, which is of the generic form (1), or $S_I \propto (\chi_\perp (k_1 - k_2))^{1/2}$ as in [2]. Another form that will prove useful later is

$$S_I = 2I\omega_I \tag{12}$$

where

$$I = (N\hbar s)^2/4J \tag{13}$$

is the effective moment of inertia of the Néel vector. Note that $I = \chi_\perp v_0/\gamma^2$.

5. Unequal sublattices—a variational solution

We now obtain a variational estimate for the action for all values of k_2/k_1 assuming only that $p \ll 1$, but non-zero. The conditions (8) no longer hold at all times, but it is apparent from (2) that the trajectories should be such that $\hat{n}_a \cdot \hat{n}_b \approx -1$ at all times, i.e., $z_b(\tau) \approx -z_a(\tau)$, and $\phi_b(\tau) - \phi_a(\tau) \approx \pi$. With this in mind we take as a variational trajectory

$$z_a(\tau) = -z_b(\tau) = \cos \theta_0(\tau) = -\tanh \omega\tau. \tag{14}$$

The frequency ω, and the optimal trajectories for $\phi_a(\tau)$ and $\phi_b(\tau)$, will be found variationally, given this form for z_a and z_b. This trajectory has the property that $\dot{z}_a = -\omega \sin^2\theta_0(\tau)$. If we recall that for an instanton calculation of the tunnel splitting, we should subtract the energy of the classical ground state from $\mathcal{H}(\hat{n}_\alpha(\tau))$ in (5), we see that each term in the action integral (5) is proportional to $\sin^2\theta_0(\tau)$. We can thus write

$$S = \int_{-\infty}^{\infty} \sin^2\theta_0(\tau) F(\phi_a(\tau), \phi_b(\tau), \omega)\, d\tau \tag{15}$$

where F is a function unspecified at present, but one plainly independent of $\dot{\phi}_{a,b}(\tau)$. If we minimize S with respect to $\phi_{a,b}(\tau)$, we see immediately that the optimal trajectories are

$$\phi_a(\tau), \phi_b(\tau) = \text{constants}. \tag{16}$$

Noting that $\sin\theta_0 = \operatorname{sech}\omega\tau$, the τ integration in (15) can now be done immediately, and we obtain

$$S = (2/\omega) F(\phi_a, \phi_b, \omega). \tag{17}$$

The problem is now reduced to minimizing an ordinary function. Let us parametrize ϕ_a and ϕ_b as follows:

$$\phi_a = i(\alpha + \beta) \qquad \phi_b = \pi + i(\alpha - \beta). \tag{18}$$

We expect (as will be shortly confirmed) that $\phi_b - \phi_a \approx \pi$, i.e., that $\beta \ll 1$. It is therefore enough to expand F in powers of β up to terms of the second order. It is straightforward to show that this gives†

$$F = 2K_1 - 2K_2 \cosh^2\alpha + N\hbar\omega s(p\alpha + \beta) - 2J\beta^2 - 2K_2 \sinh 2\alpha(p\beta + \beta^2). \tag{19}$$

Although further analysis can be done without backing into the answer, it is simplest to anticipate future results and assume that

$$\begin{aligned} N\hbar\omega s &\gg 2K_2 p \sinh 2\alpha \\ J &\gg K_2 \sinh 2\alpha \end{aligned} \tag{20}$$

and show that these assumptions hold self-consistently. If this is done, the terms containing $K_2 \sinh 2\alpha$ in (19) can be neglected, and the extremization of F with respect to β is trivial‡. The optimal values§ of β and F are given by

$$\beta^* = N\hbar\omega s/4J \tag{21}$$

$$F(\alpha, \beta^*, \omega) = 2K_1 + \tfrac{1}{2}I\omega^2 + N\hbar\omega sp\alpha - 2K_2 \cosh^2\alpha. \tag{22}$$

† We omit a constant term that gives a pure imaginary contribution to S, as it leads to the interference effects that are fully accounted for in (6).

‡ It is startling at first that $\partial^2 S/\partial\beta^2$ (and also $\partial^2 S/\partial\alpha^2$) is negative, i.e., that our extremum is a *maximum* as a function of α and β. Recall, though, that the instanton method is essentially a steepest-decent approximation for a Laplace integral, albeit a path integral, and here we are merely finding the critical point in the exponent. It still remains to expand this exponent and perform the remaining Gaussian integral along the steepest-decent path—this is what gives the prefactor c in (6). The variation in (18) lies along the path of steepest *ascent*, on which the exponent grows without bound. To find c we must integrate over *real* variations of ϕ_a and ϕ_b around the answer given by (21) and (23), and we would indeed come to grief if we forgot this point!

§ We shall denote the optimal values of α and β by α^*, β^*.

Setting $\partial F/\partial\alpha = 0$, we obtain

$$2K_2 \sinh 2\alpha^* = N\hbar\omega sp. \tag{23}$$

The first of the conditions in (20) is thus equivalent to $p^2 \ll 1$, which is a basic condition for the validity of the model, and the second can be written as $J \gg N\hbar\omega sp/2$. Given that $p \ll 1$, this condition holds automatically if $\beta^* \ll 1$, i.e., if $4J \gg N\hbar\omega s$.

It remains to minimize S with respect to ω. Using (17), $\partial S/\partial\omega = 0$ is seen to be equivalent to $F - \omega\partial F/\partial\omega = 0$. This gives

$$2K_1 - 2K_2 \cosh^2\alpha^* = \tfrac{1}{2}I\omega^2. \tag{24}$$

Using (23), this can be transformed into a quadratic equation for ω^2, with the solution

$$\tfrac{1}{2}I\omega^2 = 2K_1 - K_2 + Jp^2 - [K_2^2 + 2(2K_1 - K_2)Jp^2 + J^2p^4]^{1/2}. \tag{25}$$

Note that $\omega = \omega_{\mathrm{I}}$ when $p = 0$. (Also, $\omega = 0$ when $K_2 = K_1$.) The other sign of the root can be seen to give ω_{II}, which leads to a larger action. Finally, using (17) and (22)–(24), we obtain our variational estimate for the least action:

$$S_{\mathrm{cl}} = 2I\omega + (N\hbar sp)\sinh^{-1}(N\hbar\omega sp/2K_2). \tag{26}$$

We recall that $I = (N\hbar s)^2/4J$.

We must still show that $4J \gg N\hbar\omega s$ for self-consistency. It is not difficult to show that the right-hand side of (25) does not exceed $2(K_1 - K_2)$, i.e., that $\omega \leqslant \omega_{\mathrm{I}}$, no matter what the value of p. Thus the condition $4J \gg N\hbar\omega s$ amounts to $(\tilde{j}/k_1)^{1/2} \gg 1$, which is a basic precondition for the model to be sensible at all.

Equations (25) and (26) are the main result of our paper, and provide an estimate for the WKB exponent for all parameter values. Let us now examine this result in several limits.

(i) $p = 0$. In this case, $\omega = \omega_{\mathrm{I}}$ and the action reduces to the exact answer (12) (modulo irrelevant corrections of relative order $(k_1/j)^{1/2}$).

(ii) *The X–Y limit,* $k_1 - k_2 \to 0$. In this case

$$\omega^2 \approx \omega_{\mathrm{I}}^2(k_2/k_2')(1 + [(k_1 - k_2)/k_2]\tilde{j}^2p^4/k_2'^2 + \cdots) \tag{27}$$

where we have defined

$$k_2' = k_2 + \tilde{j}p^2. \tag{28}$$

Note that the expansion (27) is valid if $(k_1 - k_2) \ll \tilde{j}p^2/(k_2')^2$. The same condition with $(k_2')^2$ replaced by $k_2'k_2$ allows the inverse hyperbolic sine in (26) to be expanded in a Taylor series, and the final result for the action can be written as

$$S_{\mathrm{cl}} = 2I\omega_{\mathrm{I}}\left(1 + \frac{\tilde{j}p^2}{k_2}\right)^{1/2}\left\{1 + \left(\frac{k_1 - k_2}{k_2}\right)\left(\frac{\tilde{j}^2p^4}{k_2'^2}\right)\left(\frac{k_2 - 2\tilde{j}p^2}{6k_2}\right) + \cdots\right\}. \tag{29}$$

Equation (29) is a modest extension of the answer found by Loss and co-workers [9]. Their analysis also holds close to the X–Y limit, and they obtain (see their (10))

$$S_{\mathrm{cl}} = 2I\omega_{\mathrm{I}}(1 + \tilde{j}p^2/2k_1) \tag{30}$$

which follows from (29) if we make a second expansion in powers of p, neglect terms of order p^4, and replace k_2 by k_1 in the square root. Even if the correction terms in brackets are neglected, we believe that (29) is a better way of writing the answer. Firstly, it does not explicitly require the quantity $\tilde{j}p^2/k_2$, which is a ratio of two small parameters, to itself be small. Secondly, by writing the p^2 correction with a k_2 instead of k_1 in the denominator, the correction terms in $k_1 - k_2$ appear at higher order in p: p^4 as opposed to p^2.

(iii) *The Ising limit, $k_2 \to 0$.* In this case, one does not obtain formulas that are significantly simpler than (25) and (26) in general. If we assume, however, that $k_1 \gg \tilde{j}p^2 \gg k_2^2/k_1$, then ω can be expanded in powers of $|p|$, and the argument of the inverse hyperbolic sine is large compared to unity. We obtain

$$\omega \approx 2(K_1/I)^{1/2} - N\hbar s|p|/2I \tag{31}$$

$$S_{\mathrm{cl}} \approx 4(K_1 I)^{1/2} + N\hbar sp \ln\left(4p\sqrt{K_1 J}/eK_2\right). \tag{32}$$

There are two noteworthy points about this result. First, it correctly shows that the action diverges and the tunnel splitting vanishes in the limit $K_2 \to 0$. In fact, the $\ln K_2$ dependence in the action, is exactly as surmised by us [19] on the basis of the discrete WKB approach [10, 17]. In this view, the motion takes place on a square net (the $\{m_a, m_b\}$ space) with a rectangular boundary. The sides of the rectangle differ in length by $2Nsp$. When $K_2 = 0$, motion on the net is confined to straight lines $m_a + m_b$ =constant and takes place via the J term. The K_2 term gives rise to jumps between these lines, and connects every second line. To move from the corner $m_a = S_a$, $m_b = -S_b$ to the corner $m_a = -S_a$, $m_b = S_b$ requires at least Nsp jumps†. Perturbation theory then suggests that the tunnelling rate will be multiplied by a factor K_2^{Nsp} (the energy denominator is rather hard to find), and this expectation is confirmed in (32). Second, (29) and (32) show that there is no simple formula in the $p \to 0$ limit that is uniformly valid for all values of K_2/K_1.

In figures 2 and 3 we plot $S_{\mathrm{cl}}/2I\omega_{\mathrm{I}}$, which gives the $p \neq 0$ multiplicative correction to the action. This is plotted versus k_2/k_1 in figure 2, and versus p in figure 3 for several values of k_2/k_1. Note that $S_{\mathrm{cl}}/2I\omega_{\mathrm{I}}$ is a function only of the combinations k_2/k_1 and $\tilde{j}p^2/k_1$.

6. Conclusion

We conclude in this section with some remarks about our work in comparison to [9], its strengths and limitations, and avenues for further work.

Firstly, let us compare our treatment with [9]. The reader will no doubt have noted that the approximation $z_a = -z_b$ is on the same footing as $\phi_b - \phi_a \simeq \pi$, and that one could also try to minimize the action expanded to quadratic order in $(z_a + z_b)$. One can start directly from (5), in fact, and integrate out $z_a + z_b$ and the variable we call β, provided one makes the self-consistently verifiable assumption that the frequency scale ω on which the paths vary satisfies $\omega \ll \tilde{j}/s$. This leads to an action (or Lagrangian) for the Néel vector, $\hat{l}$, that is much the same as (8) of [9]

$$L_{\text{Néel}} = \tfrac{1}{2}(\dot{\theta}^2 + \sin^2\theta\dot{\phi}^2) - \mathrm{i}Nsp\cos\theta\dot{\phi} + E(\theta, \phi) \tag{33}$$

† Because the K_2 term only connects every other line, it is clearly impossible to do this unless $2Nsp$ is an even integer, i.e., unless Nsp is integral. This is a simple proof of the Kramers result, (6), as it pertains to our problem.

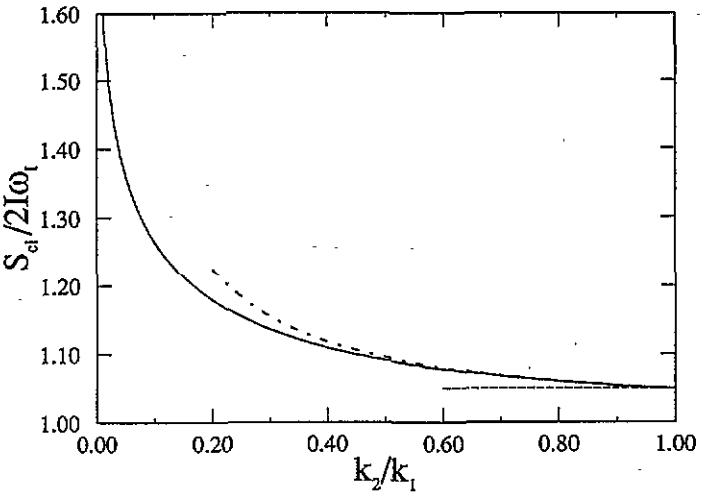

Figure 2. The multiplicative correction to the action against the transverse anisotropy, k_2/k_1 (solid line). We have taken $\tilde{j}p^2/k_1 = 0.1$. Also shown for comparison is the result of Loss and co-workers [9] (dashed line), and our (29), without the correction term in curly brackets (dot–dashed line).

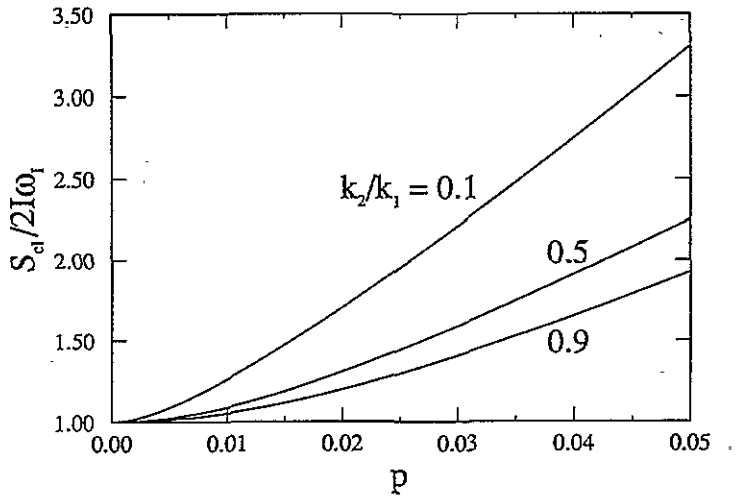

Figure 3. The multiplicative correction to the action against the fractional excess spin p, for three different values of the transverse anisotropy, k_2/k_1. We have taken $\tilde{j}/k_1 = 1000$.

where $E(\theta, \phi)$ is the anisotropy energy for $\hat{n}_b = -\hat{n}_a = \hat{l}$. ($\theta$, ϕ are the polar coordinates of $\hat{l}$.)

There is thus a close relation between our model and that of [9], and which one one prefers may be a matter of taste. In addition to be more direct, however, our model offers several other advantages. (i) It is obvious how to include external magnetic fields. Such fields lead to additional interference effects [10], and while we believe that such effects are strongly suppressed by dissipation [8, 20], should experiments ever reach the stage where

they can be seen, our model would surely be the correct way to analyse them. (ii) If one wishes to numerically solve the least-action trajectory, (33) offers no obvious advantage over (5) as the Euler–Lagrange equations form a fourth-order differential equation system in both cases. (iii) Equation (5) is the natural starting point for a calculation of the prefactor c [16], as the measure for the path integral over $\hat{n}_a$ and $\hat{n}_b$ is known. This is not so for (33), although it could probably be found. (iv) Equation (5) generalizes easily to ferrimagnetic problems.

Secondly, we believe that our variational approximation is rather good over the entire parameter space, as the form (14) accounts for all essential features of the trajectory. The one possible exception is the $k_2 \to 0$ limit. One possible approach to a proper treatment of this limit is as follows. As $\tau \to \pm\infty$, the K_2 terms in (7) are negligible. It is easy to solve (7) when $K_2 = 0$ even if $p \neq 0$ because S^z_{tot} is then conserved. The instanton must match on to these solutions with $S^z_{tot} = Nps$ as $\tau \to -\infty$, and with $S^z_{tot} = -Nps$ as $\tau \to \infty$. There must exist an internal boundary layer connecting these solutions near $\tau = 0$, and where the K_2 terms are essential. We have not succeeded in finding the structure of this boundary layer analytically. A numerical solution is under way and will be reported elsewhere.

Thirdly, we note that while our work settles an important question of principle (namely, how tunnelling occurs at all for unequal sublattices), care should be exercised in extracting numerical values of exchange and anisotropy constants ($\tilde{j}$, k_1, etc) from an experimental measurement of Δ alone. Δ is a very sensitive function of these parameters. By the same token, the WKB experiment is very *insensitive* to the value of Δ, and this means that there is a large range of values that one can assign to the attempt frequency ω_{AFM} in (6). This in turn means that the exchange and anisotropy parameter values that one deduces in this way can have large and highly correlated errors. Further, our treatment leaves out surface and shape effects, which are certainly going to affect the numerical value of Δ significantly. Even more importantly, we have ignored dissipation. To be convinced that one is seeing MQC in an experiment, a study of the systematics of Δ with temperature and RF fields is likely to be far more useful [8].

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