RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 37, NUMBER 4 1 FEBRUARY 1988

Ground-state staggered magnetization of two-dimensional quantum Heisenberg antiferromagnets

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(Received 2 November 1987)

The ground-state staggered magnetization of spin-S quantum Heisenberg antiferromagnets may be estimated in spin-wave theory (effectively a $1/S$ expansion) or by perturbation in $J_\perp = J_x = J_y$ away from the Ising ($J_\perp = 0$, $J_z > 0$) limit. The latter series in $J_\perp^2$ is poorly convergent for the square lattice and a naive summation of the available terms yields overestimates of the staggered magnetization. A new way of analyzing the perturbation series that should remove the slow convergence is proposed. The resulting ground-state staggered magnetization per spin for spin-$\frac{1}{2}$ is 0.313, while spin-wave theory yields 0.303, in units where the full, classical staggered magnetization is 0.500.

The ground-state staggered magnetization per spin, $m^\dagger$, in spin-S quantum antiferromagnets is reduced from its saturated or classical value, $m^\dagger_{\rm cl} = S$, by zero-point quantum spin fluctuations. This "spin reduction," Δ, defined by

$$m^\dagger = S - \Delta , \tag{1}$$

has been calculated in spin-wave theory,[1-3] in perturbation theory,[4-6] on small finite-size lattices,[7] and has been measured experimentally.[8,9] The experimental measurements,[8,9] made in the quasi-two-dimensional systems K_2NiF_4, K_2MnF_4, and Rb_2MnF_4, found results in good agreement with spin-wave theory. Previous analysis of the perturbation-theory results and finite-size lattices significantly underestimated the spin reduction for the square-lattice case appropriate for modeling these systems. In this paper I reanalyze the perturbation series, taking into account the expected behavior of this two-dimensional system, and obtain a spin reduction very close to that found in spin-wave theory. For spin-$\frac{1}{2}$ I obtain $\Delta \cong 0.187$, while spin-wave theory, which is, in principle, valid only in the limit of large S, yields $\Delta \cong 0.197$. The spin-wave theory appears to only slightly overestimate the spin reduction in this most unfavorable (smallest-S) case; production and analysis of a longer perturbation series will presumably determine how accurate the present estimate of $\Delta \cong 0.187$ is. It is also argued below that the available finite-lattice results[7] cannot yield a useful estimate of $m^\dagger$. This reexamination of this old problem is motivated by the current interest in the quasi-two-dimensional spin-$\frac{1}{2}$ antiferromagnet La_2CuO_4.[10]

Let us focus on the square-lattice antiferromagnet with exchange anisotropy, $J_x = J_y = J_\perp = J \le J_z$. Only nearest-neighbor exchange is considered, so the Hamiltonian is

$$H = \sum_{\langle ij \rangle} [S_i^z S_j^z + J(S_i^x S_j^x + S_i^y S_j^y)] , \tag{2}$$

where the sum runs over nearest-neighbor pairs and $\mathbf{S}_i$ are quantum spins. (Note that J_z has been set to unity, so $J = 1$ is the isotropic Heisenberg case.) For $J = 0$ we have an Ising system; its ground state is simply $S^z = +S$ on one sublattice and $S^z = -S$ on the other sublattice. The ground-state and spin reduction can be calculated order by order in perturbation theory[4-6] away from this Ising limit for $J \le 1$. For all $S \le \frac{5}{2}$ this has been done to order J^6 by Parrinello and Arai.[6] For $S = \frac{1}{2}$ the series for the spin reduction[6] is

$$\Delta = \tfrac{1}{9} J^2 + \tfrac{4}{225} J^4 + \tfrac{17}{1800} J^6 + \cdots . \tag{3}$$

(I have confirmed the first two terms of this series.) This series, truncated at order J^6, is shown in Fig. 1 as the *lower* curve. For the isotropic Heisenberg antiferromagnet, $J = 1$, this series gives the estimate $\Delta \cong 0.138$, well under the $\Delta \cong 0.197$ of spin-wave theory. One might think this is because S is so small and spin-wave theory is valid only for large S. However, the disagreement between the estimates of Δ from perturbation theory to order J^6 and

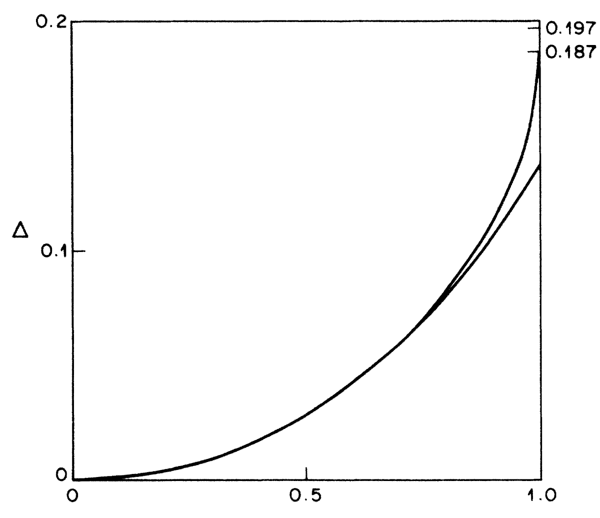

FIG. 1. The spin deviation $\Delta = \frac{1}{2} - m^\dagger$ for the square-lattice spin-$\frac{1}{2}$ antiferromagnet (2) with nearest-neighbor exchanges $J_z = 1$ and $J_x = J_y = J$. The lower curve is the perturbation series in J^2 to order J^6 as given by Eq. (3). The upper curve is the transformed series, Eq. (6), taking into account the expected square-root cusp at $J = 1$. The latter series appears well converged and gives us the estimate $\Delta(J = 1) \cong 0.187$. The spin-wave-theory estimate is $\Delta(J = 1) \cong 0.197$.

spin-wave theory actually *increases* with increasing S. The reason is that Δ as a function of J^2 has a square-root cusp for $J^2 \to 1$, so the series is poorly convergent. Added Ising exchange anisotropy is a *singular* perturbation on the isotropic, $J=1$, Heisenberg antiferromagnet. The spin-wave theory tells us that Δ is actually an analytic function of $(1-J^2)^{1/2}$ for J^2 near unity.[3] This is because Δ is an integral over zero-point spin-wave amplitudes of the form

$$\Delta \propto \int \frac{d^2k}{[1-J^2+k^2+O(k^4,J^2k^2)]^{1/2}}$$
$$\propto 1-A(1-J^2)^{1/2}+O(1-J^2)\,, \qquad (4)$$

where A is a number of order unity.[3] For finite S the precise zero-point amplitudes will differ a little from low-order spin-wave theory, but there is every reason to expect the functional form of $\Delta(J)$, namely the square-root cusp for $J^2 \to 1$ and analyticity in $(1-J^2)^{1/2}$, to apply for all S. This should be correct as long as there is long-range antiferromagnetic order at $J=1$, so the higher-order spin fluctuations merely renormalize the moments, stiffness, etc. by *finite* amounts and the *phenomenology* of spin-wave theory remains correct.

This suggests that the singularity in $\Delta(J)$ at $J=1$ can simply be removed by the variable change

$$1-\delta=(1-J^2)^{1/2}\,,$$
$$J^2=2\delta-\delta^2\,. \qquad (5)$$

Making this change, we obtain the series

$$\Delta=\tfrac{2}{9}\delta-\tfrac{1}{25}\delta^2+\tfrac{1}{225}\delta^3+\cdots \qquad (6)$$

for $S=\frac{1}{2}$. This series, truncated at order δ^3, is shown in Fig. 1 as the upper curve. This series appears more convergent than that in J^2 [Eq. (3)], and yields the estimate $\Delta \cong 0.1867$ for the isotropic case $J=\delta=1$. The Padé approximants to this series of the form $\Delta \cong (a\delta+b\delta^2)/(1+c\delta)$ and $\Delta \cong d\delta/(1+e\delta+f\delta^2)$ yield the estimates $\Delta \cong 0.1862$ and 0.1864, respectively, and show no poles in the vicinity of the interval of interest, $0 \le \delta \le 1$. That all three estimates based on (6) give essentially the same answer [unlike Padé estimates based on (3)] suggests that the difference between this result and spin-wave theory, $\Delta \cong 0.197$, is real. Generation and analysis of a longer series would check this. This difference from spin-wave theory, as expected, decreases for larger S; the series for $S \ge 1$ agree with spin-wave theory within the accuracy suggested by their Padé approximants.

The spin-wave theory has been carried to the first nonlinear order and no effect on the spin reduction at $J=1$ is found.[3] Naively, one expects the next-order (uncalculated) terms to be of order $1/(S^2z^3)$, where z is the coordination number of the lattice.[3] For the case we are considering, $S=\frac{1}{2}$, $z=4$, this is not small, $1/(S^2z^3)=0.0625$, while the deviation found above suggests the terms missed are actually of order 0.01. Thus the $1/(S^2z^3)$ term in the $1/S$ expansion of Δ may have a rather small (or vanishing) coefficient at $J=1$.

Another, comparable estimate of the error in spin-wave theory is obtained by considering the wave function at lowest order in $1/S$. It has 0.197 zero-point spin-wave quanta present per spin. These spin waves are bosons, so more than one can sit on each spin; the number of excitations on each spin is Poisson distributed. For large S this is fine because S^z can be reduced by many quanta. However, for $S=\frac{1}{2}$, only one spin excitation is possible per spin and the configurations with more than one are nonexistent. Of the total spin reduction, 0.197, in spin-wave theory, 18% of it, or 0.035, arises from such configurations that are forbidden for $S=\frac{1}{2}$. If we simply replace each such forbidden configuration by the allowed configuration with all the excess spin excitations removed, the spin deviation obtained is $\Delta \cong -0.179$, somewhat under the series estimates. For large S, the number of spins in forbidden configurations vanishes as $(0.197)^{2S+1}/[(2S+1)!]$, so this effect is entirely missed by a $1/S$ expansion.

Oitmaa and Betts[7] found the exact ground states of the isotropic ($J=1$) spin-$\frac{1}{2}$ model on finite square lattices with periodic boundary conditions and up to $N=16$ spins. They measured the mean-square staggered magnetization along the z direction and extrapolated it linearly versus $1/N$ to obtain the estimate[7] $m_z^\dagger=0.243\pm0.006$. Their ground states are all rotationally invariant singlets, so their estimate of the full root-mean-square staggered magnetization per spin is thus $\sqrt{3}$ times this, or $m^\dagger=0.42\pm0.01$. This estimate is far in excess of the $m^\dagger \cong 0.313$ obtained above. The reason is that extrapolation versus $1/N$ is inappropriate here.

The spin-wave theory indicates that the spin-spin correlations decay as

$$|\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle|-(m^\dagger)^2 \sim \frac{1}{r_{ij}} \qquad (7)$$

for large spin separation, r_{ij}. This would imply that the mean-square staggered magnetization per spin of finite-size lattices should converge as

$$(m_N^\dagger)^2-(m^\dagger)^2 \sim N^{-1/2} \qquad (8)$$

for large N. Thus we should expect

$$(m_N^\dagger)^2=(m^\dagger)^2+\alpha N^{-1/2}+\beta N^{-1}+\cdots\,. \qquad (9)$$

The present series estimate is $(m^\dagger)^2 \cong 0.10$, while the largest lattice looked at by Oitmaa and Betts[7] has $(m_{16}^\dagger)^2 \cong 0.28$. This indicates that the sum of the correction term in (9) of order $N^{-1/2}$ and higher are still larger than $(m^\dagger)^2$ itself for $N=16$. A reliable extrapolation for $N \to \infty$ can only be done if there is reason to believe that the higher-order corrections are small, namely if $\beta N^{-1} \ll \alpha N^{-1/2}$. In the absence of knowing the coefficients α and β, the sensible criterion for making a useful extrapolation is $(m_N^\dagger)^2-(m^\dagger)^2 \ll (m^\dagger)^2$. This has clearly not been attained, and would probably require lattices of size $N \gtrsim 100$. Thus the results of Oitmaa and Betts[7] for $N \leq 16$ cannot be used to give a useful estimate of the $N \rightarrow \infty$ staggered magnetization.

Note added in proof. Extrapolation of the series expansion in Ref. 6 for the ground-state energy per bond of (2) yields $E_0 = -0.334 \pm 0.001$ for the isotropic case $J = 1$. This is somewhat below Oitmaa and Betts' estimate of -0.328 ± 0.003;[7] their overestimate occurred because they assumed the finite-size correction is of order N^{-1}, while spin-wave theory tells us it is only of order $N^{-3/2}$. We[11] have a variational wave function that gives a strict bound of $E_0 \leq -0.331$.

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