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Paired excitations in the antiferromagnetic Heisenberg model with unaxial anisotropy

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Abstract. The antiferromagnetic Heisenberg model with uniaxial anisotropy, arbitrary spin and dimension is solved approximately for $D > 0$. The ground-state wavefunction, its energy, and zero-spin excitations are obtained. The validity range of the analytic approach is determined by comparing it with numerical computation and perturbative results in the large-D limit. It is shown that the paired, non-magnetic excitation method presented here yields more accurate results than linear spin-wave theory.

1. Discussion

Studies on this class of compounds, of which TMMC, $K_3Fe(CN)_6$ [1, 2] are prototypical examples, show that they can be described by the antiferromagnetic Heisenberg Hamiltonian with uniaxial anisotropy:

$$\mathcal{H} = J\frac{\alpha}{2}\sum_{r,\delta}\left[S^+(r)S^-(r+\delta) + S^+(r+\delta)S^-(r)\right] + J\sum_{r,\delta}S^z(r)S^z(r+\delta) - D\sum_{r}S^z(r)S^z(r) \tag{1}$$

where $S(r)$ is the spin at the site r, the spin size S depends on the material (for TMMC $S = \frac{5}{2}$), δ is a vector connecting a point in one sublattice to any of its neighbouring points in the other sublattice, J is the exchange coefficient, α is the anisotropy parameter, and D measures the uniaxial anisotropy.

In one dimension, for $S = \frac{1}{2}$, this Hamiltonian was completely solved by the Bethe anzats [3–5], but for larger spin or higher dimension no exact analytical solution is known. Numerical solutions become more difficult as well, and for the 2D-$\frac{1}{2}$ Heisenberg antiferromagnetic lattice of 96×96 seems to be the state-of-the-art [6]. The study of the spin dynamics in real lattices thus resorts to approximate approaches. Among them, spin-wave theory has been shown to describe accurately the magnetic excitations of a number of antiferromagnetic crystals. However, the minimun error for the ground-state energy of spin-wave theory is obtained for $\alpha = 1$ (3 and 2% error in one and two dimensions, respectively), and rapidly increases as α departs from unity (see figures 5 and 6 later).

Recently, an approximate solution for this Hamiltonian in the $D = 0$ case was proposed [7–9]. This solution is valid for a wide range of values of the anisotropy

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parameter α, arbitrary dimension and spin. The method allows one to formulate the problem in terms of new elementary paired, non-magnetic excitations and yields analytical expressions for the ground state, spectrum and, mean values with respect to the stationary states. The comparison of these analytical results with numerical computations in one and two dimensions shows excellent agreement (figures 5 and 6 later).

In this work we extend the method to include uniaxial anisotropy. A positive D is expected to favour the orientation of the antiferromagnetic order in the z direction, thus diminishing the spin fluctuations associated with the transversal (x, y) term of the Heisenberg Hamiltonian; the former increases the region of the parameter space in which the method suitable. The calculations are done for arbitrary spin and dimension and, are compared with numerical results obtained first in a small spin cluster and second, by perturbation theory calculations.

We define the zero-spin excitation operators as follows:

$$\phi_\delta^\dagger(k) = \frac{1}{\sqrt{2S^2N}} \sum_r e^{ik\cdot r} S^+(r+\delta) S^-(r) + Q\delta_{k,0} \tag{2}$$

with

$$Q = \left(\frac{N}{2}\right)^{1/2} \frac{\alpha JS}{(2zS-1)J + 2(2S-1)D} \tag{3}$$

where r runs over the sublattice with spin up, k is a vector in the reduced Brillouin zone of one sublattice given by the antiferromagnetic order, z is the number of near neighbours and N is the number of spin S atoms in the lattice.

The operators defined by relation (2) obey a complicated algebra. One has, for example,

$$[\phi_\delta(k), \phi_{\delta'}^\dagger(k')] = \frac{1}{2S^2N} \sum_r e^{i(k'-k)\cdot r} \{S^+(r+\delta)S^-(r+\delta')S^z(r) - e^{ik'\cdot(\delta-\delta')} S^+(r)S^-(r+\delta-\delta')S^z(r+\delta)\} \tag{4}$$

$$\begin{aligned}[\mathcal{H}, \phi_\delta^\dagger(k)] = \frac{1}{\sqrt{2S^2N}} \sum_{r,\delta'} e^{ik\cdot r} \{ & J\alpha S^+(r+\delta)S^-(r+\delta')S^z(r') \\ & - J\alpha S^+(r+\delta-\delta')S^-(r)S^z(r+\delta) \\ & + 2\delta_{\delta\delta'} DS^+(r+\delta)(S^z(r+\delta) - S^z(r) - 1) \\ & + JS^+(r+\delta)S^-(r)((S^z(r+\delta-\delta') - S^z(r+\delta) - \delta_{\delta\delta'})\}. \end{aligned} \tag{5}$$

However, if one substitutes the right hand sides of equations (4) and (5) by their projections over the Néel state $|\mathcal{N}\rangle$ which assigns spin up to sublattice r and down to the other, they reduce to

$$[\phi_\delta(k), \phi_{\delta'}(k')] = 0 \qquad [\phi_\delta(k), \phi_{\delta'}^\dagger(k')] = \delta_{k,k'}\delta_{\delta,\delta'} \tag{6}$$

$$[\mathcal{H}, \phi_\delta^\dagger(k)] = [(2zS-1)J + 2(2S-1)D]\phi_\delta^\dagger(k) \equiv \varepsilon\phi_\delta^\dagger(k). \tag{7}$$

It is quite clear that the commutation rules (4) can be replaced by the much simpler ones given by (6) if either α is small or D is large enough, as both conditions assure that the system is close to a Néel order. Reference [9] shows that the procedure used here remains valid in a non-trivial region of parameter space; a precise criterion for the associated accuracy is also given there.

Equation (6) shows that in the limit where antiferromagnetic order prevails (small α or large D) the ϕ operators represent a set of independent spin zero excitations which obey Bose statistics. Using these relations the Hamiltonian can be written as

$$\mathcal{H} = \varepsilon \sum_{k,\delta} \phi_\delta^\dagger(k)\phi_\delta(k) + E_g \tag{8}$$

where E_g is the ground-state energy and is given by the following expression

$$E_g = -\frac{N}{2} JzS^2 - NDS^2 - \frac{NJzS^2\alpha^2}{2[(2zS-1)J + 2(2S-1)D]} \tag{9}$$

As expected, for spin one-half there is no contribution from the uniaxial term to the energy except for the trivial constant factor NDS^2.

The ϕ operators have the property

$$\phi_\delta(k)|\mathcal{N}\rangle = Q\delta_{k,0}|\mathcal{N}\rangle\,, \tag{10}$$

where Q is a constant. From this property one readily obtains that the ket

$$|g\rangle = \exp\left[-Q\sum_\delta (\phi_\delta^\dagger(0) - \phi_\delta(0))\right]|\mathcal{N}\rangle \tag{11}$$

satisfies

$$\phi_\delta(k)|g\rangle = 0 \qquad \forall k, \delta \tag{12}$$

and is thus the ground state of the asymptotic version equation (9) of $\mathcal{H}$.

In order to compare our results for the ground-state energies equation (9) with those obtained from an entirely different method, we solved a square arrangement of four $S = 1$ spins. As was shown in [9] the theory presented here works better in the large spin or high dimensionality limit. Then the configuration, which was solved numerically, corresponds to the more stringent condition in order to check the theory. The finite size of the cluster limits the accuracy of the comparison. We use the fact that for spin one in the isotropic case ($\alpha = 1$ and $D = 0$) numerical results with negligible error are available [10, 11] to determine an upper bound for the error in the small cluster calculations (7%). Our calculations are asymptotically exact in the Ising limit.

Figures 1 and 2 shows how expression (9) for the ground-state energy (full curve) compares with the numerically obtained data for several choices of the parameters. The error expected in the numerical data relative to the case of large N is represented as a shadowed area. As expected, large values of D favour the antiferromagnetic ordering of the spins and makes (9) applicable in a wider range of α. What is surprising is the magnitude of this effect, in fact, a change of D from 0.0 to 0.4

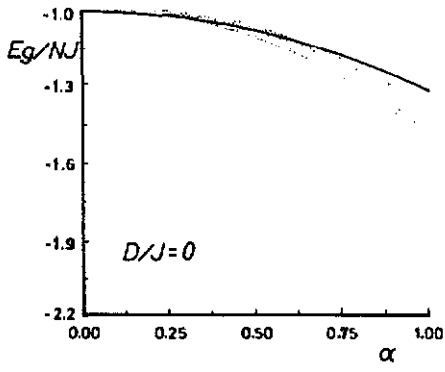

Figure 1. Ground-state energy E_g as function of α (full curve) for $D = 0$. The error expected in the numerical data relative to the case of large N is represented by the shaded area.

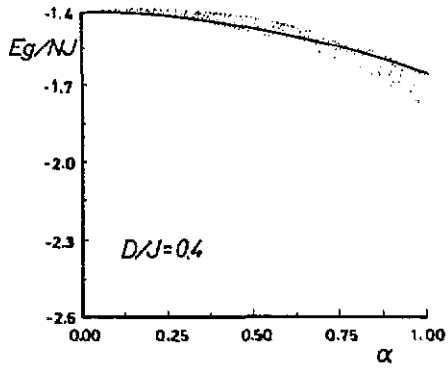

Figure 2. Same as figure 1, but for $D = 0.4$.

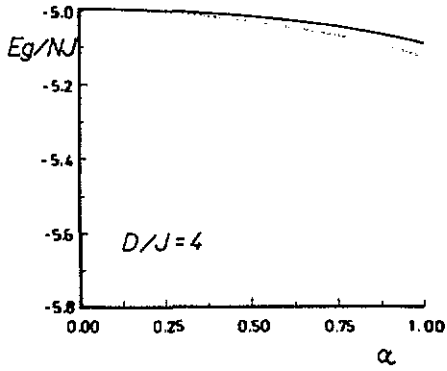

Figure 3. Ground-state energy E_g as function of α (full curve) for $D/J = 4$. The segmented line correspond to perturbative calculations.

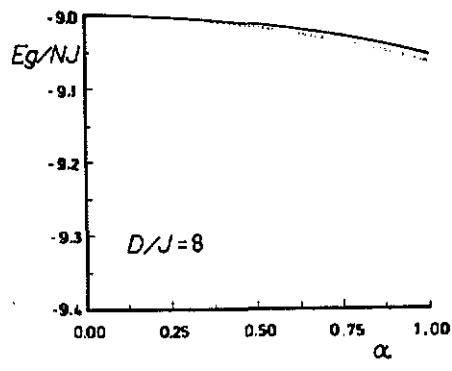

Figure 4. Same as figure 3, but for $D/J = 8$.

enlarges the range of validity of the approximation from α by a factor of two. The same situation occurs in [12] where the antiferromagnetic Heisenberg Hamiltonian with coupling to nearest ($J_1 > 0$) and second nearest neighbours ($J_2 < 0$) was solved, the change of J_2/J_1 from 0 to -0.2 enlarges the range of α by a factor of two.

Another verification of the theory presented in this work can be obtained in the large D limit. Let us think in a one dimensional, $S = 1$ chain of spins described by the Hamiltonian in equation (1). As $D \to \infty$ only the $S^z = 1$ and -1 components will survive, the energy of the $S^z = 0$ state being much higher. Therefore, an effective $S = 1/2$ model can be obtained by perturbation theory in $1/D$. This has been done

by Sólyom and Ziman [13]. Their result is the following:

$$\mathcal{H}_{\text{eff}} = -N\frac{\alpha^2 J^2}{4D} - ND + \left(4J + \frac{\alpha^2 J^2}{D}\right)\sum_l \tilde{S}_l^z \tilde{S}_{l+1}^z$$
$$- \frac{\alpha^2 J^2}{2D}\sum_l \left(\tilde{S}_l^+ \tilde{S}_{l+1}^- - \tilde{S}_{l+1}^+ \tilde{S}_l^-\right) \tag{13}$$

where $\tilde{S}^z$, $\tilde{S}^+$ and $\tilde{S}^-$ are effective spin one-half operators which obey

$$\tilde{S}^z|\pm 1\rangle = \pm\tfrac{1}{2}|\pm 1\rangle \qquad \tilde{S}^{\pm}|\pm 1\rangle = |\pm 1\rangle\,. \tag{14}$$

The spectrum of the Hamiltonian presented in equation 13 coincides with that of the antiferromagnetic anisotropic Heisenberg problem solved exactly by Orbach (due to the fact that $\mathcal{H}(J_{xy}, J_z)$ is related to $\mathcal{H}(-J_{xy}, J_z)$ by a unitary transformation). Then using Orbach calculations for spin one-half we can test our theory for the $S = 1$ case in the limit $D \gg J$. The results are shown in figures 3 and 4. It is seen that the difference between our results (full curve) and perturbative ones are less than 0.1% even for $\alpha = 1$ (or 1% if we do not consider the constant term $-D$). We remark that the theory presented here works for arbitrary dimension (d) and spin (S) (the results are better as d and S becomes larger) giving closed expressions for the ground-state and excitation energies.

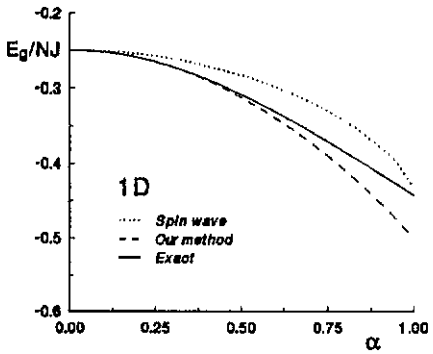

Figure 5. Ground-state energy as a function of the anistropy parameter α for the $S = \frac{1}{2}$ one-dimensional case.

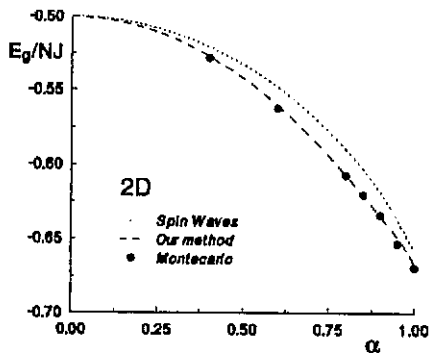

Figure 6. Ground-state energy as a function of the anistropy parameter α for the $S = \frac{1}{2}$ two-dimensional case.

Finally it is interesting to compare the results of the present method with linear spin-wave theory. This is done for spin $\frac{1}{2}$ in one and two dimensions, where exact analytic [4,5] or numerical calculations [14] are available in the literature. The results are shown in figures 5 and 6. It is seen that our 'paired non-magnetic excitation theory' yields more accurate results than linear spin-wave theory, particularly in the two-dimensional case.

Acknowledgments

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