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Zero-Point Spin Reduction of an Antiferromagnet

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Zero-point spin reduction in the low-dimensional antiferromagnet is discussed by the spin wave theory taking into account kinematical interaction due to finite magnitude of spin. This method is applied to $KCuF_3$ and the result is in good agreement with the observed one. Furthermore the values of zero-point spin reduction of several other chain-structure antiferromagnets are predicted.

§ 1. Introduction

A large zero-point spin reduction in the antiferromagnet of the low dimensionality has recently been observed. In the layer-structure antiferromagnet K_2MnF_4, the observed values of zero-point spin reduction agree with those of a simple two-dimensional spin-wave theory.[1] It has been found that the zero-point spin reduction in the one-dimensional antiferromagnet $KCuF_3$ is nearly 45%.[2] However, the kinematical interaction due to the restriction on the number of spin deviations possible at any given site cannot always be neglected in the antiferromagnet of the low dimensionality even at absolute zero. Especially the zero-point spin reduction of the ideal spin-wave theory diverges in the one-dimensional Heisenberg antiferromagnet. As the zero-point spin reduction should not be over the spin magnitude S, the divergence means that the spin-wave theory which does not take account of the kinematical interaction cannot be trusted.

The effect of the kinematical interaction for zero-point spin reduction was first estimated by Herbert[3] for the Heisenberg antiferromagnet with each spin $S=1/2$, but he did not give any attention to the low-dimensional antiferromagnet. In the present paper his method is extended to the case of general S, and the effect of the kinematical interaction is taken into account in the calculation of the zero-point spin reduction, and several antiferromagnetic chain-structure compounds are discussed.

§ 2. The boson representation

For simplicity, we consider the Heisenberg ferromagnet in this section. The Hamiltonian of the system can be written as follows:

$$H=-2J\sum_{\langle i,j\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j\,, \tag{2·1}$$

where $\boldsymbol{S}_i$ is the spin operator at the i-th atom, the summation $\sum_{\langle i,j\rangle}$ is taken over all nearest neighbor pairs in the crystal and $J(>0)$ is the exchange integral.

At first, let us consider a spin space spanned by the spin operators concerned with the j-th atom. All eigenstates of the operator S_j^z forms a complete set in this space.

$$\begin{aligned} &S_j^z|n_j\rangle=(S-n)|n_j\rangle, \quad (n=0, 1, 2, \cdots, 2S) \\ &\langle n_j|m_j\rangle=\delta_{nm}\,. \end{aligned} \tag{2·2}$$

These eigenstates can be represented by

$$|n_j\rangle=[(2S)^n n!\, F_n]^{-1/2}(S_j^+)^n|0_j\rangle, \tag{2·3}$$

where we use usual notations $S_j^{\pm}=S_j^x\pm iS_j^y$ and

$$F_n=1\cdot\left(1-\frac{1}{2S}\right)\cdots\left(1-\frac{n-1}{2S}\right). \tag{2·4}$$

Let a_j^* and a_j be the creation and annihilation operators labelled by the site index j, and the following orthonormal boson states are defined:

$$\begin{aligned} &|n_j)=[n!]^{-1/2}(a_j^*)^n|0_j), \\ &(n_j|m_j)=\delta_{nm}\,, \end{aligned} \tag{2·5}$$

where $|0_j)$ stands for the vacuum state.

Since any physical quantity can be expressed in terms of sums of products of the matrix elements $\langle n|S_j^{\alpha}|m\rangle$ $(\alpha=+, -, z)$, we define the boson operator $\hat{S}_j^{\alpha}$ corresponding to the spin operator S_j^{α} as follows:[4]

$$\langle n|S_j^{\alpha}|m\rangle=(n|f^{-1}(n)\hat{S}_j^{\alpha}f(m)|m), \quad (n,m=0, 1, 2, \cdots, 2S) \tag{2·6}$$

where $f(n)$ is an arbitrary function of n provided that $f(n)$ and $f^{-1}(n)$ exist. Various representations of $\hat{S}_j^{\alpha}$ are possible by this arbitrariness. For simplicity, we choose the Holstein-Primakoff transformation which is given by (2·6) under $f(n)=$const for all n:[4]

$$\begin{aligned} &\hat{S}_j^{+}=(2S)^{1/2}(1-a_j^*a_j/2S)^{1/2}a_j\,, \\ &\hat{S}_j^{-}=(2S)^{1/2}a_j^*(1-a_j^*a_j/2S)^{1/2}\,, \\ &\hat{S}_j^{z}=S-a_j^*a_j\,. \end{aligned} \tag{2·7}$$

However, the representations (2·7) are restricted to the boson-state where occupation number of the boson is smaller than $2S+1$. In order to remove this restriction, a projection operator P_j is introduced:

$$P_j=\theta(2S-a_j^*a_j), \tag{2·8}$$

where $\theta(x)$ is the unit step function,

$$\begin{aligned} \theta(x)&=1\,, \quad x\geq 0\,, \\ &=0\,, \quad x<0\,. \end{aligned} \tag{2·9}$$

It should be noted that the projection operator P_j commutes with $\hat{S}_j^{\alpha}$ $(\alpha=+,-,z)$. Using this projection operator, the boson representation of any physical quantity $A(S_j^+, S_j^-, S_j^z)$ which is the function of S_j^{α} can be expressed as follows:

$$\begin{aligned} A &= A(P_j\hat{S}_j^+P_j, P_j\hat{S}_j^-P_j, P_j\hat{S}_j^zP_j) \\ &= P_jA(\hat{S}_j^+, \hat{S}_j^-, \hat{S}_j^z). \end{aligned} \tag{2·10}$$

At this stage, the extension of this representation to full spin space which is the tensor product of each spin space spanned by S_j^{α} may be done, and the thermal average of the operator A is obtained from

$$\langle A\rangle = \sum_n (n|P\hat{\rho}\hat{A}|n). \tag{2·11}$$

Here, $\{|n)\}$ is any complete set of full boson states, and

$$\begin{aligned} &P=\prod_j P_j, \\ &\hat{\rho}=\exp\left(-\frac{1}{k_BT}\hat{H}\right)/\sum_n\left(n|P\exp\left(-\frac{1}{k_BT}\hat{H}\right)|n\right), \end{aligned} \tag{2·12}$$

where k_B is Boltzmann's constant and T the temperature.

§ 3. Zero-point spin reduction in antiferromagnet

The Hamiltonian of the antiferromagnet we wish to consider in this section is written by

$$H=2J\sum_{\langle i,j\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j+2J'\sum_{\langle l,m\rangle}\boldsymbol{S}_l\cdot\boldsymbol{S}_m, \tag{3·1}$$

where the summation $\sum_{\langle l,j\rangle}$ is taken over all nearest neighbor (n.n.) pairs along the z-axist and $\sum_{\langle l,m\rangle}$ over all n.n. pairs perpendicular to the z-axis, $J(>0)$ and $J'(>0)$ are the intra-chain, and inter-chain exchange integrals, respectively.

Two sublattice model is introduced as usual. The Holstein-Primakoff representations are the following in this case:

$$\begin{aligned} \hat{S}_i^+ &= (2S)^{1/2}(1-a_i^*a_i/2S)^{1/2}a_i, \\ \hat{S}_i^- &= (2S)^{1/2}a_i^*(1-a_i^*a_i/2S)^{1/2}, \\ \hat{S}_i^z &= S-a_i^*a_i, \end{aligned} \tag{3·2}$$

for a spin on the A sublattice,

$$\begin{aligned} \hat{S}_j^+ &= (2S)^{1/2}b_j^*(1-b_j^*b_j/2S)^{1/2}, \\ \hat{S}_j^- &= (2S)^{1/2}(1-b_j^*b_j/2S)^{1/2}b_j, \\ \hat{S}_j^z &= -S+b_j^*b_j, \end{aligned} \tag{3·3}$$

for a spin on the B sublattice. Here two different sets of boson operators $\{a, a^*\}$ and $\{b, b^*\}$ have been introduced.

The method of the preceding section can be applied to the calculation of the sublattice magnetization $\langle S_i^z \rangle$. We define the Fourier transformations,

$$a_k = \sqrt{\frac{2}{N}} \sum_i e^{ikR_i} a_i , \qquad b_k = \sqrt{\frac{2}{N}} \sum_j e^{-ikR_j} b_j , \tag{3·4}$$

where N is the total number of atoms, and use the canonical transformations,

$$a_k = \alpha_k \cosh \theta_k - \beta_k^* \sinh \theta_k , \qquad b_k = -\alpha_k^* \sinh \theta_k + \beta_k \cosh \theta_k . \tag{3·5}$$

Here, $\tanh 2\theta_k = [\cos k_z + \xi(\cos k_x + \cos k_y)]/(1+2\xi)$, and $\xi = J'/J$. If the terms higher than bilinear are neglected, the boson representation of the Hamiltonian (3·1) can be written by the above transformations as

$$H = E_0 + \sum_k \varepsilon_k (\alpha_k^* \alpha_k + \beta_k^* \beta_k), \tag{3·6}$$

where

$$E_0 = -2JS^2N - 4J'S^2N + \sum_k \{\varepsilon_k - 4JS - 8SJ'\},$$

$$\varepsilon_k = 2S\sqrt{(2J+4J')^2 - (2J\cos k_z + 2J'\cos k_x + 2J'\cos k_y)^2} . \tag{3·7}$$

The expectation value of a spin on the A sublattice at absolute zero can be calculated by

$$\langle S_i^z \rangle_{T=0} = S - \frac{(g|Pa_i^* a_i|g)}{(g|P|g)} , \tag{3·8}$$

and the zero-point spin reduction is

$$\Delta S = (g|Pa_i^* a_i|g)/(g|P|g), \tag{3·9}$$

where $|g)$ is the ground state of the full boson Hamiltonian.

Now, the true ground state $|g)$ is approximated by the ground state $|0)$ of the free spin-wave Hamiltonian (3·6). This is defined by

$$\alpha_k|0) = \beta_k|0) = 0 . \tag{3·10}$$

Futhermore the following approximation is introduced:

$$\Delta S = \frac{(0|Pa_i^* a_i|0)}{(0|P|0)} \simeq \frac{(0|P_i a_i^* a_i|0)}{(0|P_i|0)} . \tag{3·11}$$

The operator P_i and $P_i a_i^* a_i$ can be represented as follows:[3]

$$P_i = \sum_{l=0}^{\infty} B_l (a_i^*)^l (a_i)^l , \qquad P_i a_i^* a_i = \sum_{l=0}^{\infty} C_l (a_i^*)^l (a_i)^l , \tag{3·12}$$

where $B_0 = 1$, $B_1 = B_2 = \cdots = B_{2S} = 0$,

$$B_n = (-1)^{n-2S} \frac{(n-1)(n-2)\cdots(2S+2)(2S+1)}{n!(n-2S-1)!} , \quad (n \geq 2S+1)$$

and $C_1 = 1$, $C_2 = C_3 = \cdots = C_{2S} = 0$,

$$C_n=(-1)^{n-2S}\frac{(n-2)(n-3)\cdots(2S+1)(2S)}{(n-1)!(n-2S-1)!}. \quad (n\geq 2S+1)$$

From (3·4) and (3·5), we get

$$(0|(a_i^*)^n(a_i)^n|0)=n!\,v^n \tag{3·13}$$

with

$$v=\frac{2}{N}\sum_k \sinh^2\theta_k . \tag{3·14}$$

Hence, we have

$$(0|P_i|0)=\sum_{n=1}^{\infty} B_n n! v^n , \tag{3·15}$$

$$(0|P_i a_i^* a_i|0)=\sum_{n=1}^{\infty} C_n n! v^n . \tag{3·16}$$

The zero-point spin reduction is

$$\Delta S=v-w \tag{3·17}$$

with

$$w=\frac{(2S+1)v^{2S+1}}{(1+v)^{2S+1}-v^{2S+1}} . \tag{3·18}$$

Here, v defined in (3·14) is the zero-point spin reduction obtained by the simple spin-wave theory, while w is the correction by the kinematical interaction. It is interesting that (3·17) and (3·18) agree with that obtained by the Green function method.[5]

The numerical values of ΔS are shown in Table I for the case of pure two dimensional ($J=0$, $J'\neq 0$) and three dimensional ($J=J'\neq 0$) antiferromagnets. The correction w is very small except the case of two-dimensional lattice with $S=1/2$. Consequently the reason why the observed values of ΔS agree with those of a simple spin-wave theory in $KMnF_3$, $RbMnF_3$, K_2MnF_4, Rb_2MnF_4 ($S=5/2$)[1] can be understood.

On the other hand, w is very important for the case $J'/J\equiv\xi\ll 1$ (one-dimen-

Table I. Zero-point spin reduction of antiferromagnetic lattices.

	Square	NaCl-type
v (free spin-wave theory)	0.197	0.078
ΔS (present method)	0.141 ($S=1/2$)	0.067 ($S=1/2$)
	0.184 ($S=1$)	0.077 ($S=1$)
	0.194 ($S=3/2$)	0.078 ($S=3/2$)
	0.197 ($S=2$)	0.078 ($S=2$)
	0.197 ($S=5/2$)	0.078 ($S=5/2$)

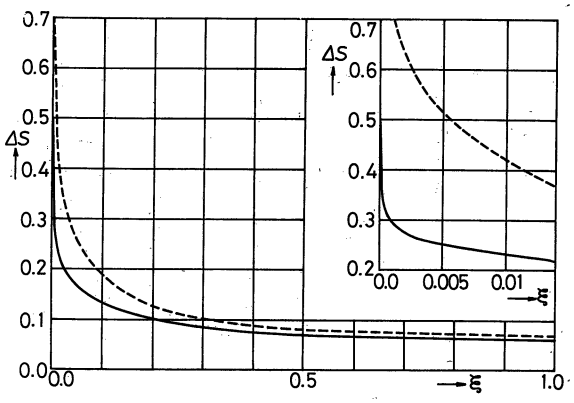

Fig. 1. Zero-point spin reduction of the antiferromagnet with $S=1/2$. The solid line ΔS shows that obtained by the present method and the dashed line v by the free spin-wave theory. The solid line and the dashed line tend to $\Delta S=0.5$ and $+\infty$, respectively, as $\xi\to 0$. The enlarged figure is shown at the right upper corner.

Table II. Zero-point spin reduction of chain-structure antiferromagnets.

	$\xi=J'/J$	v	ΔS
$KCuF_3$	2.7×10^{-2}	0.32	0.20 (42%)
CPC	$0.8\times10^{-3}\sim 5.8\times10^{-3}$	$0.75\sim 0.5$	0.30 (60%) $\sim$ 0.25 (50%)
CTS	$2.1\times10^{-3}\sim 14.4\times10^{-3}$	$0.63\sim 0.35$	0.28 (56%) $\sim$ 0.21 (42%)

sional like). If ξ is less than 6×10^{-3}, v is even bigger than $S=0.5$, and v goes to infinity as $\xi\to 0$. On the other hand, ΔS is never over S, and it tends to S as $\xi\to 0$ thanks to the presence of w. The values of ΔS for $S=1/2$ are plotted against ξ in Fig. 1. However, w is so small for $S\geq 1$ and $\xi\gtrsim 10^{-2}$, that the effect of kinematical interaction can be clearly seen only for the case $S=1/2$ and $\xi\lesssim 0.1$.

We apply now the present method to several chain-structure antiferromagnets with $S=1/2$, such as $KCuF_3$, $CuCl_2\cdot 2NC_5H_5$ (CPC), and $Cu(NH_3)_4SO_4\cdot H_2O$(CTS). The values of ξ are estimated from the observed Curie temperature using Green function method under the following assumptions.[2),6)]

(1) The structure of lattice can be approximated by a tetragonal one.

(2) The exchange integrals are isotropic.

The results are given in Table II. The experimental results for $KCuF_3$ are $\xi=0.027$ at $T=39.8$ K,[7)] and the spin reduction $=0.23$ (45%).[2)] We can see that ΔS is in better agreement than v with the experimental value. Furthermore, as explained already, v is not reliable in this region of ξ, becouse v gets over the spin magnitude 0.5 when $\xi<0.006$. Unfortunately there are no experimental data of the spin reduction available for CTS and CPC, but we give the theoretical values for them in Table II.

§ 4. Discussion

The zero-point spin reduction of the sublattice magnetization in the Heisenberg antiferromagnet was calculated taking into account the kinematical interaction. It

was found that the kinematical interaction acts seriously on this reduction in the case of chain-structure antiferromagnet with $S=1/2$. Two important approximations were introduced in our calculation. The first one is that the true ground state of the spin system is approximated by the magnon ground state. Since the full boson Hamiltonian has source terms which are products of creation operators of magnon only such as $\alpha^*\alpha^*\beta^*\beta^*$, the magnon ground state is not an eigenstate of the spin system. Especially, these source terms would be important in low-dimensional lattice. The corrections due to these source terms is one of the problems which have to be solved in future. The second is that the product of projection operators have been decoupled so that (3·11) is obtained. The meaning of this approximation is not clear. However it should be stressed that our result is in agreement with that by the Green function method with the random phase approximation.

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