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The Spin Wave Theory in Antiferromagnetic Heisenberg Model on Face Centered Cubic Lattice

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The classical ground states of face centered cubic lattice with nearest neighbor antiferromagnetic Heisenberg interactions are shown to have continuous degeneracy. The spin wave theory removes the degeneracy and determines the ground state, in which the energy E_g and spin reduction are calculated. E_g is also obtained by calculating the lowest energy of finite size systems and extrapolating to infinite lattice. Both results are in fairly good agreement with each other. Finally it is concluded that the sublattice long range order exists.

The magnetic properties at low temperatures of quantum mechanical antiferromagnetic Heisenberg spin system on bi-partite lattices have been well studied. On the other hand, those on the non-bipartite lattice such as triangular lattice or face centered cubic (fcc) lattice with nearest neighbor antiferromagnetic interactions have not been solved unambiguously. As is well known the antiferromagnetic Heisenberg model on non-bipartite lattice has necessarily frustrations.

In the classical ground state on triangular lattice, the ordered 120° structure, in which the spins on the three sublattices are set at relative angle of 120° to each other, is stable. The spin wave theory based on the classical ground state shows that the 120° structure is stable, although there exists the spin reduction.[1,2] Miyashita[3] treated the antiferromagnetic Heisenberg and XY models by a variational study of the ground state and found a sublattice long range order for both cases. On the other hand, Anderson[4,5] proposed the resonating valence bond state in which the sublattice long range order does not exist. Recently we[6] have numerically analyzed finite size systems and extrapolated them to the infinite lattice to conclude that the XY model has a sublattice long range order in the ground state while the Heisenberg model does not.

A typical example of three dimensional non-bipartite lattice is the face centered cubic lattice. No researches have been done about the frustration effect to quantum spin systems on fcc. In this paper we apply the spin wave theory to fcc with nearest neighbor antiferromagnetic Heisenberg interactions.

We assume that the second nearest neighbor interactions J_2 are ferromagnetic so that fcc would be reasonably divided into four sublattices, a, b, c and d as shown in Fig. 1. Henceforth J_2 is not taken into account explicitly, so that the Hamiltonian is $\mathcal{H}=2|J|\Sigma_{\langle ij\rangle}\boldsymbol{S}_i\cdot\boldsymbol{S}_j$, where i, j are nearest neighbors.

Let us explain the crystal structure of fcc. Each site has 12 nearest neighbor sites which belong to other 3 sublattices. We denote the interactions between a and b sites by ab bond and so on. The total number of bonds is $6N$, where N is the total number of sites. The numbers of ab, ac, ad, bc, bd and cd bonds are N, respectively. The tetrahedron shown in Fig. 1 is composed of a, b, c and d sites, and has every one of the six kinds of bonds.

First of all, we have to determine the spin orientations on four sublattices in the ground state for the classical Heisenberg model. Let us denote the spins on four sublattices by $\boldsymbol{S}_a$, $\boldsymbol{S}_b$, $\boldsymbol{S}_c$ and $\boldsymbol{S}_d$, respectively. Since the interactions between spins are isotropic, we may take the plane containing $\boldsymbol{S}_a$ and $\boldsymbol{S}_b$ as the xz plane, and take the bisector of the angle between $\boldsymbol{S}_a$ and $\boldsymbol{S}_b$ as the z axis (polar axis). We express the orientations of spins by the polar coordinates as follows,

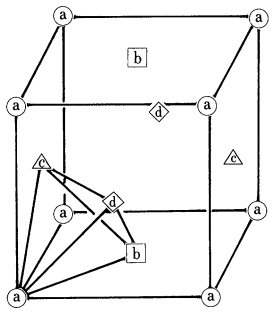

Fig. 1.

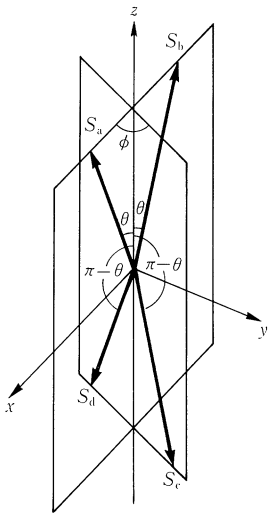

Fig. 2.

Fig. 1. Four sublattices in fcc.

Fig. 2. Polar angles of S_a, S_b, S_c and S_d.

$$S_a(\theta, 0), S_b(\theta, \pi), S_c(\theta_c, \phi_c), S_d(\theta_d, \phi_d). \quad (1)$$

These angles can be determined so as to minimize the interaction energies of classical Heisenberg model. As the calculations are tedious, only the results are written, as follows,

$$S_a(\theta, 0), S_b(\theta, \pi), S_c(\pi-\theta, \phi), S_d(\pi-\theta, \phi+\pi), \quad (2)$$

where θ and ϕ are arbitrary. (see Fig. 2). The energy of the classical ground state is $E_{cl}=-4|J|NS^2$ which is independent of θ and ϕ, so that the ground states have continuous degeneracy. This result could also be obtained by use of the Fourier transform method developed by Yamamoto and Nagamiya.[7] When $\theta=0$, S_a and S_b point to the $+z$ direction, while S_c and S_d to the $-z$ direction. When $\theta=\pi/2$, they are in the xy plane. When $\theta=\cos^{-1}\sqrt{1/3}$, $\phi=\pi/2$, the angles between any two spins are equal to each other. This last spin structure is regarded as the extension of the 120° spin structure in the classical antiferromagnetic Heisenberg model on the triangular lattice to the three-dimensional case.

We have expected the degeneracy of the ground states would be removed when we take account of the quantum property. Thus we consider the quantum antiferromagnetic Heisenberg model by the spin wave theory. For the sake of simplicity, we put $\phi=\pi/2$ in the following calculations.

We express the spin operators S_p^x, S_p^y, S_p^z on the p sublattice (p=a, b, c or d) in terms of $S^{p\xi}$, $S^{p\eta}$, $S^{p\zeta}$, where x, y, z are the coordinate axes of crystal and pξ, pη, pζ are those in each sublattice. The pζ axis is taken to be the orientation of spins on the p sublattice in the classical ground state. The transformation between them are given by

$$\left.\begin{aligned}
&S_a^x=S^{a\xi}\cos\theta+S^{a\zeta}\sin\theta,\ S_a^y=S^{a\eta},\\
&S_a^z=-S^{a\xi}\sin\theta+S^{a\zeta}\cos\theta,\\
&S_b^x=S^{b\xi}\cos\theta-S^{b\zeta}\sin\theta,\ S_b^y=S^{b\eta},\\
&S_b^z=S^{b\xi}\sin\theta+S^{b\zeta}\cos\theta,\\
&S_c^x=S^{c\eta},\ S_c^y=S^{c\xi}\cos\theta+S^{c\zeta}\sin\theta,\\
&S_c^z=S^{c\xi}\sin\theta-S^{c\zeta}\cos\theta,\\
&S_d^x=S^{d\eta},\ S_d^y=S^{d\xi}\cos\theta-S^{d\zeta}\sin\theta,\\
&S_d^z=-S^{d\xi}\sin\theta-S^{d\zeta}\cos\theta.
\end{aligned}\right\} \quad (3)$$

We introduce the annihilation and creation Holstein-Primakoff operators[8] α_p and α_p^* for the spin deviation from pζ axis in each sublattice as follows,

$$\left.\begin{aligned}
&S^{p\xi}=\sqrt{S/2}(\alpha_p+\alpha_p^*),\ S^{p\eta}=i\sqrt{S/2}\,(\alpha_p^*-\alpha_p),\\
&S^{p\zeta}=S-\alpha_p^*\alpha_p.
\end{aligned}\right\} \quad (4)$$

Here and henceforth we neglect terms of more than third order of operators. The scalar product of nearest neighbor spins which are on the different sublattices can be written by

$$\begin{aligned}
&S_a\cdot S_b=S^2\cos 2\theta+S\{-\cos 2\theta(\alpha_a^*\alpha_a+\alpha_b^*\alpha_b)\\
&\qquad+[-\sin^2\theta\alpha_a\alpha_b+\cos^2\theta\alpha_a\alpha_b^*+\text{h.c.}]\},\\
&S_c\cdot S_d=(\text{replacing a}\to\text{c, b}\to\text{d in the above eq.}),\\
&S_a\cdot S_c=-S^2\cos^2\theta+S\{\cos^2\theta(\alpha_a^*\alpha_a+\alpha_c^*\alpha_c)\\
&\qquad+[(-1/2\sin^2\theta-i\cos\theta)\alpha_a\alpha_c\\
&\qquad-1/2\sin^2\theta\alpha_a\alpha_c^*+\text{h.c.}]\},\\
&S_b\cdot S_d=(\text{replacing a}\to\text{b, c}\to\text{d in the above eq.}),\\
&S_a\cdot S_d=-S^2\cos^2\theta+S\{\cos^2\theta(\alpha_a^*\alpha_a+\alpha_d^*\alpha_d)\\
&\qquad+[(1/2\sin^2\theta-i\cos\theta)\alpha_a\alpha_d\\
&\qquad+1/2\sin^2\theta\alpha_a\alpha_d^*+\text{h.c.}]\},\\
&S_b\cdot S_c=(\text{replacing a}\to\text{b, d}\to\text{c in the above eq.}).
\end{aligned} \quad (5)$$

Here we have neglected the linear terms of operators, because they will be cancelled each other after we sum up all interactions in the crystal which means the classical spin orientations are in equilibrium. Although we have four sublattices, we do not distinguish them when we make the Fourier transformations of α_r and α_r^*, (r expresses a position of every site), which will turn out to be correct below. Therefore we have

$$\alpha_r=\frac{1}{\sqrt{N}}\sum_k \alpha_k\, e^{ik\cdot r},$$

$$\alpha_r^*=\frac{1}{\sqrt{N}}\sum_k \alpha_k\, e^{-ik\cdot r}, \tag{6}$$

where the summation of the wave vector $\boldsymbol{k}$ is taken over the first Brillouin zone of fcc. Let us denote the Hamiltonian which is composed of all interactions between sites on the a sublattice and their nearest neighbors by $\mathcal{H}_a$, and similarly by $\mathcal{H}_b$, $\mathcal{H}_c$ and $\mathcal{H}_d$. They become the same formula if expressed in terms of α_k and α_k^* using (6). This verifies the use of (6) is allowed. This result is due to (3), because other several choices of the transformation of coordinates are not successful by such a simple procedure. Adding $\mathcal{H}_a$, $\mathcal{H}_b$, $\mathcal{H}_c$ and $\mathcal{H}_d$ and dividing by 8, as each pair is counted twice and the number of sites of each sublattice is $N/4$, we get

$$\tilde{H}\equiv\frac{\mathcal{H}}{2|J|S}$$
$$=\sum_k\left(P_k\alpha_k^*\alpha_k-\frac{1}{2}Q_k\alpha_k\alpha_{-k}\right.$$
$$\left.-\frac{1}{2}Q_k^*\alpha_{-k}^*\alpha_k^*\right), \tag{7}$$

where we have dropped the energy of classical ground state $E_{cl}=-4|J|S^2N$, and

$$P_k\equiv 1+\cos^2\theta C_xC_y$$
$$+1/2\,\sin^2\theta(C_zC_x-C_yC_z), \tag{8}$$

$$Q_k\equiv\sin^2\theta(C_xC_y+1/2C_yC_z$$
$$-1/2C_zC_x)+i\cos\theta(C_yC_z+C_zC_x),$$

$$C_i\equiv\cos(k_i/2)\ (i=x,y,z). \tag{9}$$

In order to diagonalize (7), we use the following transformations,

$$\alpha_k=\frac{1}{\sqrt{2}}\{\cosh\Theta_k(\xi_k+\xi_{-k})$$
$$+\sinh\Theta_k(\xi_k^*-\xi_{-k}^*)\}e^{-i\Phi_k},\ (k>0)$$

$$\alpha_{-k}=\frac{1}{\sqrt{2}}\{\cosh\Theta_k(\xi_k-\xi_{-k})$$
$$+\sinh\Theta_k(\xi_k^*+\xi_{-k}^*)\}e^{-i\Phi_k},\quad (k>0)$$

$$\alpha_0=(\cosh\Theta_0\xi_0+\sinh\Theta_0\xi_0^*)e^{-i\Phi_0}, \tag{10}$$

where

$$[\xi_k,\xi_{k'}^*]=\delta_{k,k'},\ [\xi_k,\xi_{k'}]=[\xi_k^*,\xi_{k'}^*]=0. \tag{11}$$

Substituting (10) into (7), we obtain

$$\tilde{H}=\sum_k\left\{-\frac{1}{2}P_k+\sqrt{P_k^2-|Q_k|^2}\right.$$
$$\left.+\sqrt{P_k^2-|Q_k|^2}\xi_k^*\xi_k\right\}, \tag{12}$$

provided that

$$\tanh 2\Theta_k=\frac{|Q_k|}{P_k},\quad \tan 2\Phi_k=\frac{\mathrm{Im}\,Q_k}{\mathrm{Re}\,Q_k}. \tag{13}$$

The energy $\hbar\omega_k=8|J|S\sqrt{P_k^2-|Q_k|^2}$ of a spin wave with wave vector $\boldsymbol{k}$, is given from (8),

$$\hbar\omega_k=8|J|S\{1+2\cos^2\theta C_xC_y$$
$$+\sin^2\theta C_z(C_x-C_y)$$
$$+(\cos^2\theta-\sin^2\theta)C_x^2C_y^2$$
$$+\sin^2\theta C_xC_yC_z(C_x-C_y)$$
$$-\cos^2\theta C_z^2(C_x+C_y)^2\}. \tag{14}$$

As expected from the original classical configurations (put $\phi=\pi/2$ in Fig. 2), at a glance, it seems to be strange that (14) does not have the symmetry between x and y. However, if we shift the origin in the $\boldsymbol{k}$ space to $(\pi,-\pi,-\pi)$, $\hbar\omega_k$ has clearly the symmetry between x and y. Furthermore it has the symmetry among x, y and z when $\cos\theta=1/\sqrt{3}$ as it should.

The shift of the energy ΔE of the ground state due to the quantum effect is the 1st and 2nd terms in (12). Hereafter we treat the case of $S=1/2$. It was calculated as the function of θ by the computer. The energy E_g of the ground state including E_{cl} is shown in Fig. 3. As is seen, the degeneracies of the ground states are removed and the most stable state is obtained when $\theta=0$, and $E_g(\theta=0)=-1.488N|J|$.

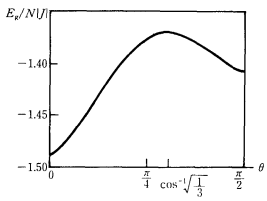

Fig. 3.

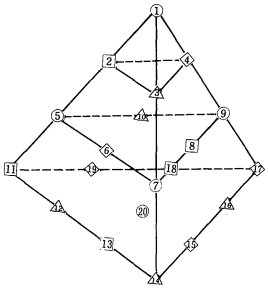

Fig. 4.

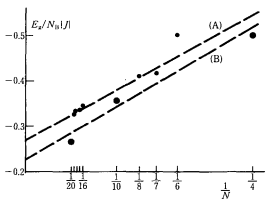

Fig. 5.

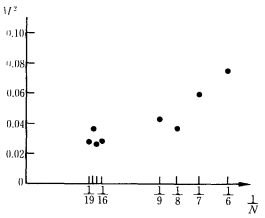

Fig. 6.

Fig. 5. $E_g/N_B|J|$ versus $1/N$ for each N. Large dots are data for $N=4$, 10 and 20.

Fig. 6. M^2 versus $1/N$ for each N.

Fig. 3. $E_g/N|J|$ as functions of θ.

Fig. 4. Finite number of spins on the planes perpendicular to [111] direction in fcc.

The spin reduction in the ground state ($\theta=0$) is given by

$$\Delta S=\frac{1}{N}\sum_r \langle \alpha_r^* \alpha_r \rangle = \frac{1}{N}\sum_k \frac{1}{2}\left\{\frac{P_k}{\sqrt{P_k^2-|Q_k|^2}}-1\right\}=0.33. \quad (15)$$

It is very large in spite of three dimensional lattice. This large spin reduction is probably caused by the frustration effect.

Aside from the spin wave theory, we have numerically computed the lowest energy of the finite number of spins ($N=4$, 6, 7, 8, 10, 16, 17, 18, 19, 20) on fcc lattice sites to obtain E_g of infinite lattice. (see ref. 6). Figure 4 shows the sites on the planes perpendicular to [111] direction in fcc, where ○, □, △ and ◇ indicate four sublattices. When $N=4$, 10 and 20, the form is a regular tetrahedron, but for other N's, the form is truncated, although we choose sites so as to have symmetry as much as possible. For example, when $N=6$ the sites are 2, 3, 4, 6, 8 and 10. In this way the computed values $E_g/N_B|J|$ for each N are plotted against $1/N$ in Fig. 5, where N_B is the number of bonds for N sites, which tends to $6N$ when $N\to\infty$. By the method of least squares, we draw the dashed line (A) using all available data and (B) using those for $N=4$, 10 and 20. The extrapolated value of E_g in the case of $N\to\infty$ are $-1.6N|J|$ for (A) and $-1.4N|J|$ for (B). These values are in good agreement with that obtained by the spin wave theory.

Finally we check whether sublattice long range order exists in the correct ground state $|g\rangle$ or not. Although the spin wave theory predicts the sublattice long range order, the exact state may fail to have one as in the triangular lattice. (see ref. 6). We calculate the sublattice magnetization for the finite number of spins,

$$M^2=\langle g|(\sum_{i\in a} S_i^z+\sum_{i\in b} S_i^z -\sum_{i\in c} S_i^z-\sum_{i\in d} S_i^z)^2|g\rangle/N^2. \quad (16)$$

The numerical values are plotted for each $1/N$ in Fig. 6.

By the spin wave theory we have

$$M^2=\left(\frac{1}{2}-\Delta S\right)^2\simeq 0.03. \quad (17)$$

From (17) and Fig. 6, we can conclude that the sublattice long range order exists in the ground state.

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