Transactions of the Materials Research Society of Japan 24 [2] 201-204 (1999)

Calculation for the Magnetic Properties in Triangulated Kagomé Lattice -The Approach by Spin-Wave Theory and Monte Carlo Simulation -

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The dispersion of linear Holstein-Primakoff spin-waves for Heisenberg spin state in triangulated Kagomé lattice is obtained by Bogoliubov transformation. This Heisenberg model with nine-sublattice system has been proposed by Mekata for the compound $Cu_9Cl_2(cpa)_6 \cdot xH_2O$ on the basis of experimental results. Because obtained modes have both characters of triangular and Kagomé antiferromagnetic lattices, remarkable features can be found in the behavior of magnons: as well as "the flat mode" with zero-energy excitation corresponding to that in Kagomé lattice, two "dispersionless modes" with finite energy appear. Furthermore, we made Monte Carlo simulation for this systems in order to discuss spin dynamics. The specific heat and the susceptibility is discussed on the basis of the simulation. In particular, we obtain characteristic features of spin dynamics are from analyzing averaged distributions of angles between spins.

KEYWORDS: spin-waves, Heisenberg spin state, triangulated Kagomé lattice, $Cu_9Cl_2(cpa)_6 \cdot xH_2O$, flat modes, Monte Carlo simulation

§1. Introduction

By the theoretical and experimental work on spin systems in triangular and Kagomé lattices, the properties of the non-collinear Néel state for antiferromagnetic systems has been made clear, in last several years. In addition to this, the triangulated Kagomé lattice joins with this kind of remarkable magnetic systems.[1,2] Recently, Mekata and his coworkers have investigated [3–5] magnetic properties of $Cu_9Cl_2(cpa)_6 \cdot xH_2O$. This magnetic system for spin = 1/2 of Cu^{2+} ions is expected to have essential quantum effects originating from the frustration of exchange interactions. In fact, they have pointed out that both the ferromagnetic and antiferromagnetic properties are observed in experiments. As a result, the obtained magnetization has the tendency of saturating at $0.27\mu_B$ for each Cu^{2+} ion.

1.1 Compounds

The structure of this compound is illustrated in Fig.1. It should be noted that this structure is made by pasting of small triangles to each triangle in Kagomé lattice. As mentioned above, the exchange interactions (J_{AF}) between sites in pasted small triangles A, B, C, and D, E, F are assigned [4,5] to be antiferromagnetic, while the bonds(J_F) between spins of original Kagomé sites P, Q, R and sites on pasted small triangles are expected to be ferromagnetic. Further, the strength of the latter is expected to be relatively small in comparison with that of the former.

§2. Present Model

2.1 Hamiltonian

The Hamiltonian is, therefore, expressed as

$$\begin{aligned}\mathcal{H} = \sum_n [\ & J_{AF}(\boldsymbol{S}_{An}\cdot\boldsymbol{S}_{Bn}+\boldsymbol{S}_{Bn}\cdot\boldsymbol{S}_{Cn}+\boldsymbol{S}_{Cn}\cdot\boldsymbol{S}_{An}) \\ & +J_{AF}(\boldsymbol{S}_{Dn}\cdot\boldsymbol{S}_{En}+\boldsymbol{S}_{En}\cdot\boldsymbol{S}_{Fn}+\boldsymbol{S}_{Fn}\cdot\boldsymbol{S}_{Dn}) \\ & -|J_F|(\boldsymbol{S}_{Pn}\cdot\boldsymbol{S}_{An}+\boldsymbol{S}_{Pn}\cdot\boldsymbol{S}_{Bn} \\ & \quad +\boldsymbol{S}_{Pn}\cdot\boldsymbol{S}_{Dn}+\boldsymbol{S}_{Pn}\cdot\boldsymbol{S}_{En}) \\ & -|J_F|(\boldsymbol{S}_{Qn}\cdot\boldsymbol{S}_{Bn}+\boldsymbol{S}_{Qn}\cdot\boldsymbol{S}_{Cn} \\ & \quad +\boldsymbol{S}_{Qn}\cdot\boldsymbol{S}_{En}+\boldsymbol{S}_{Qn}\cdot\boldsymbol{S}_{Fn}) \\ & -|J_F|(\boldsymbol{S}_{Rn}\cdot\boldsymbol{S}_{Cn}+\boldsymbol{S}_{Rn}\cdot\boldsymbol{S}_{An} \\ & \quad +\boldsymbol{S}_{Rn}\cdot\boldsymbol{S}_{Fn}+\boldsymbol{S}_{Rn}\cdot\boldsymbol{S}_{Dn})\ ]\ ,\end{aligned} \tag{1}$$

where $J_{AF} > 0$.

2.2 Heisenberg Model

Here, we adopt the nine-sublattice structure, which has been proposed on the basis of experimental work.[4,5] This is shown in Fig.1, where arrows mean magnetic moments of spin = 1/2 of Cu^{2+} ions. In detail, the spin structure on pasted small triangles is so-called 120° Néel configuration: the spin for sites of original Kagomé lattice has the direction which makes the angle of 60° to any spin of the pasted small triangle. The classical energy for a unit cell is expressed as $E_{cl} = -3S^2 J_{AF}(1+2|J_F/J_{AF}|)$.

The unit cell for this nine-sublattice state is the rhombus whose sides are $\boldsymbol{u}_1$ and $\boldsymbol{u}_2$ with lengths $4a$ as shown in Fig.1. Here, a is the

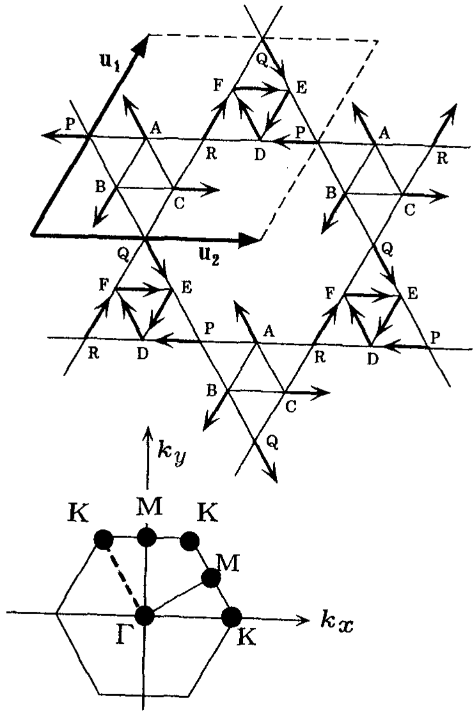

Fig.1 The structure of Cu^{2+} ions in $Cu_9Cl_2(cpa)_6 \cdot xH_2O$. The spin configuration for Heisenberg model is also shown by arrows at Cu^{2+} sites. The primitive unit cell is the rhombus whose sides are u_1 and u_2. The first Brillouin zone is also illustrated, in which points of Γ , K and M are shown.

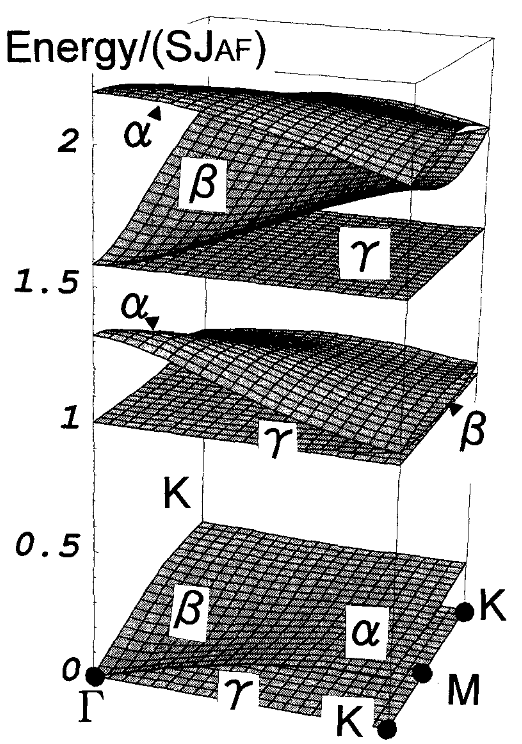

Fig.2 Calculated dispersion for $|J_F/J_{AF}| = 0.5$. The dispersion for $\Gamma - K - M - K - M - K - \Gamma$ is illustrated. The α and β modes of lower-group are not degenerate as well as those of other groups.

length of sides for the pasted triangle.

§3. Spin-Waves

In the present paper, we calculate linear Holstein-Primakoff[6] spin-waves for this nine-sublattice system, on the basis of the canonical transformation called Bogoliubov transformation for spin operators expressed in Fourier transformed forms with wavenumber $\boldsymbol{k}$. The detailed discussion of this procedure has been reported in other piblication.[7]

§4. Calculated Results

Figure 2 presents the calculated results[7,8] of the dispersion in 3-dimensional illustration, where $|J_F/J_{AF}| = 0.5$. The first Brillouin zone is also shown in Fig.2. We would like to emphasize that each group is composed of three modes, which are assigned by α, β and γ modes according to the Watabe's analysis[9,10] proposed for the discussion of magnons in triangular antiferromagnets. The α and β modes of lower-group are not degenerate as well as those of other groups. It should be noted that the γ mode in each group has no dispersion in the Brillouin zone. This appearance of three "dispersionless modes" is the remarkable behavior in this system.

In particular, the γ mode for the lower-group has no excitation energy. This mode corresponds to "the flat mode" of the antiferromagnetic system on Kagomé lattice.[11] Characteristic features of these dispersionless modes has been discussed in connection with the semi-local symmetry in this system, as well as the quantum effect expressed by the zero-point energy E_Q of magnons.[7,8]

§5. Monte Carlo Simulation

Furthermore, we made Monte Carlo simulation for this system in order to discuss spin dynamics[12] in connection with the behavior of the averaged energy and the susceptibility. We adopt classical rotator model for spins. Each "spin" can changed its angle for the unit of $\pi/6$. The averaged energy, specific heat and susceptibility can be obtained from the partition function

$$Z \equiv \sum_i e^{-\beta E_i + \beta HM}, \qquad \beta \equiv \frac{1}{k_B T}, \quad (2)$$

For example, the energy is obtained by

$$\langle E \rangle \equiv \frac{\sum_i E_i \, e^{-\beta E_i}}{\sum_i e^{-\beta E_i}} \quad (3)$$

$$= -\frac{1}{Z(HM=0)} \frac{\partial Z(HM=0)}{\partial \beta}. \quad (4)$$

The specific heat is expressed as

$$C \equiv \frac{\partial \langle E \rangle}{\partial T} = \frac{\langle E^2 \rangle - \langle E \rangle^2}{k_B T^2}. \quad (5)$$

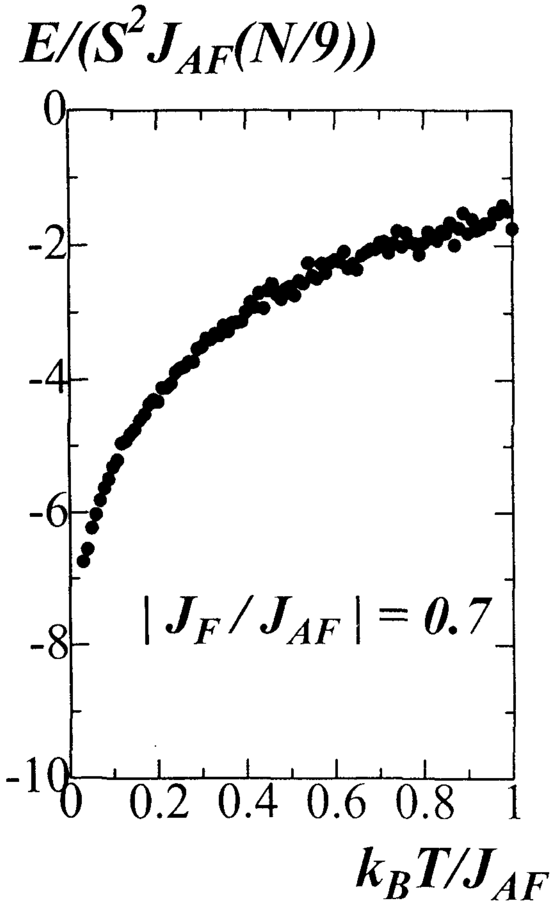

Fig.3 The obtained averaged energy vs. temperature for $|J_F/J_{AF}| = 0.7$. The gradient, i.e. specific heat has the peak around $k_\mathrm{B}T/J_{AF} = 0.4 \sim 0.8$.

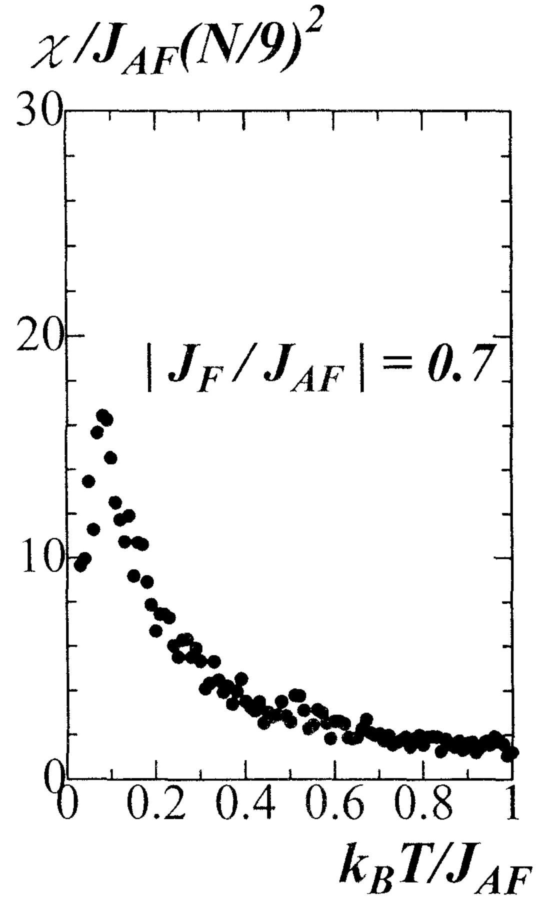

Fig.4 The susceptibility vs. temperature for $|J_F/J_{AF}| = 0.7$. The peak at $k_\mathrm{B}T/J_{AF} = 0.08$ is obtained.

Because the magnetization is given by

$$\langle M\rangle = \frac{\partial \ln Z}{\partial(\beta H)} = \frac{1}{Z}\frac{\partial Z}{\partial(\beta H)}, \tag{6}$$

we can estimate the susceptibility as follows;

$$\begin{aligned}\chi &= \frac{\partial\langle M\rangle}{\partial H} \\ &= \frac{\partial}{\partial H}\left[\frac{1}{Z}\frac{\partial Z}{\partial(\beta H)}\right] \\ &= \beta\frac{\partial Z}{\partial(\beta H)}\frac{-1}{Z^2}\frac{\partial Z}{\partial(\beta H)} + \frac{1}{Z}\beta\frac{\partial^2 Z}{\partial(\beta H)^2} \\ &= \beta\left[\frac{1}{Z}\frac{\partial^2 Z}{\partial(\beta H)^2} - \left(\frac{1}{Z}\frac{\partial Z}{\partial(\beta H)}\right)^2\right] \\ &= \frac{1}{k_\mathrm{B}T}\left[\langle M^2\rangle - \langle M\rangle^2\right].\end{aligned} \tag{7}$$

Considering that we have no external field, we use the magnetization for some direction, for example, the direction of spin of A site. This method on the basis of the estimation for the fluctuation is characteristic for the Monte Carlo simulation.

5.1 Results of Monte Carlo simulation

Figure 3 presents the obtained averaged energy vs. temperature for $|J_F/J_{AF}| = 0.7$, in which the gradient, i.e. specific heat has the peak around $k_\mathrm{B}T/J_{AF} = 0.4 \sim 0.8$. The obtained susceptibility vs. temperature for $|J_F/J_{AF}| = 0.7$ is shown in Fig.4, where the peak at $k_\mathrm{B}T/J_{AF} = 0.08$ is obtained. The peak structure is clear in comparison with that of specific heat. For the purpose of investigating the patterns for spins around the peak of the susceptibility at $k_\mathrm{B}T/J_{AF} = 0.08$, we show averaged distributions of angles between spins for this spin system in Fig.5. As for spins A,B and C, angles are distributed mainly to 120° and 150°. On the other hand, angles between spins of small triangles and the original Kagomé-lattice point has the tendency of 30°, rather than that of 60°. Furthermore, we can see the parallel spins.

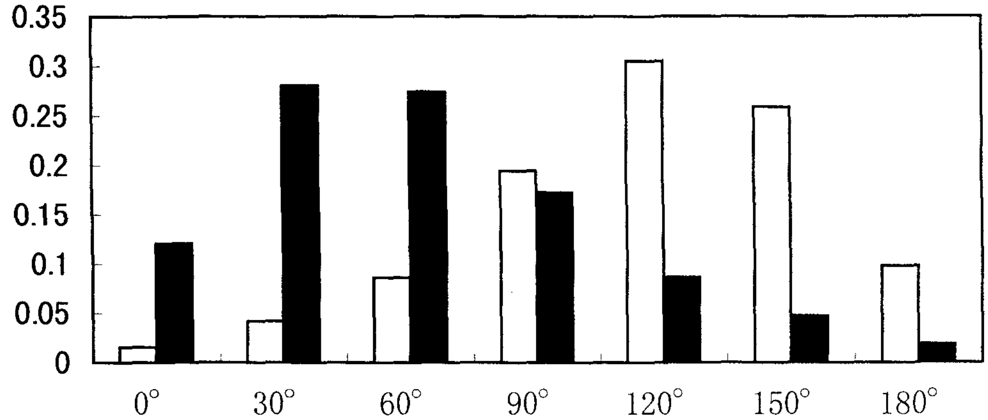

Fig.5 Averaged distributions of angles between spins. The white bars present distributions of angles among A, B and C spins, while the black bars denote angles between spins of small triangles and the original Kagome-lattice point (AP, PB, BQ, QC, CR, RA). As for spins A,B and C, angles AB, BC, CA are distributed mainly to 120° and 150°. On the other hand, angles between spins of small triangles and the original Kagome-lattice point have the tendency of 30° and 60°.

Accordingly we would like to point out that the spin moves significantly by the effect of each magnetic bond (J_{AF}, J_F) remaining the pattern of Néel structure.

§6. Conclusion

We made clear the dispersion of magonons for Heisenberg spin state in triangulated Kagomé lattice. This Heisenberg model with nine-sublattice system had been proposed for the compound $Cu_9Cl_2(cpa)_6 \cdot xH_2O$. Because obtained modes has both characters of triangular and Kagomé antiferromagnetc lattices, remarkable features can be found in the behavior of magnons. In addition to this calculation for magnons, we made Monte Carlo simulation for this systems in order to discuss spin dynamics in connection with the behavior of the averaged energy and susceptibility. In particular, characteristic features of spin dynamics analyzed from averaged distributions of angles between spins.

We wish to thank Professor Mekata of Fukui University for stimulating suggestions on the basis of his experimental work.

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(Received December 11, 1998; accepted February 28, 1999)