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Spin dynamics in classical and quantum Kagome lattice magnets studied by NMR

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Abstract

Jarosite, $KFe_3(OH)_6(SO_4)_2$, is a typical example of a classical frustrated antiferromagnet with $s = 5/2$ Fe^{3+} ions on the Kagome lattice, and a metallic complex, $[Cu_3(titmb)_2(CH_3CO_2)_6]\cdot H_2O$, abbreviated as Cu-titmb, is considered to be a candidate material for a quantum frustrated magnet with $s = 1/2$ on the Kagome lattice. Jarosite has a Weiss temperature of -530 K and shows magnetic ordering below 65 K due to weak anisotropy. The magnetic structure in the ordered phase has been determined, using neutron scattering and NMR, to be a $q = 0$ type 120° spin structure with + chirality. The nuclear spin–lattice relaxation rate T_1^{-1} and the spontaneous magnetization indicate the existence of the spin wave in the ordered phase. A small number of substitutions or deficiencies induce successive phase transitions, which represent two-dimensional ordering on the Kagome planes and three-dimensional ordering between the planes. For the quantum spin magnet, Cu-titmb, the nuclear spin relaxation at low temperatures shows the existence of anomalous low-lying excited states in the magnetic field. The relaxation rates at high temperatures give much smaller exchange interaction compared to the value estimated from the specific heat. Several interesting properties, such as double peaks of the specific heat, a 1/3 magnetization plateau for the pulse field and a ferromagnetic transition at 56 mK have been reported. We review the experimental results of Cu-titmb.

1. Introduction

An antiferromagnet on the Kagome lattice is a typical geometrically frustrated magnet, and is expected to show large frustration effects. The ground state of classical Heisenberg Kagome lattice antiferromagnets has a 120° spin structure, where the vector sum of three spins on the basic triangle is zero. However, the spin plane formed by the three spins has the freedom of twist with respect to the spin planes of the neighbouring triangles. Therefore, the ground state

has an infinite and continuous degeneracy, and no long range ordering can be expected even at zero temperature. However, theoretical studies have predicted that ordering may occur by thermal disorder, called order by disorder, and coplanar 120° spin structures, such as $q = 0$ and $\sqrt{3} \times \sqrt{3}$ structures, are favoured [1, 2]. On the other hand, small perturbations can resolve the degeneracy and induce magnetic phase transitions at finite temperature.

In the case of quantum spin systems, quantum effects are expected to introduce exotic magnetic properties in the frustrated Kagome lattice antiferromagnets. Theoretical studies predict that the system has no long range Néel-type ordering, and that the ground state is disordered [3]. Spin liquid states, such as the resonating valence bond (RVB) state, singlet dimers and the chiral spin liquid, where the spin correlations are very short, have been discussed [4, 5]. For the low-lying excitations, one theoretical study predicts the existence of an energy gap between the singlet ground state and the triplet excited states [5]. Another theory predicts that the energy gap between the singlet ground state and the triplet magnetic excited state is filled with continuous nonmagnetic excitations [6].

Several model materials of the Kagome lattice antiferromagnets have been studied experimentally. First, the second layer of ^{3}He absorbed on graphite was considered to be an example of the spin 1/2 Kagome antiferromagnet, and attracted theoretical studies [7, 8]. However, the anomalous properties were found to be induced by multiple exchange interactions of ^{3}He [9]. A Kagome bilayer compound, $SrCr_{8-x}Ga_{4+x}O_{19}$, with spin 3/2 Cr^{3+} ions, has been studied extensively as a candidate material for a Kagome lattice antiferromagnet. The spin-glass-like transition occurs at 3 K, which is much lower than the Weiss temperature of −515 K [10, 11]. Anomalous properties have been observed in this compound; however, the compound is now considered not to be a good model of the Kagome antiferromagnet since it is composed of the stacking of Kagome and triangular lattices with non-negligible interactions, and deficiencies of Ga and Cr atoms on the Kagome planes give rise to several effects on the magnetic properties and destroy the perfect Kagome lattice [12]. Recently, a similar Kagome bilayer material, $Ba_2Sn_2ZnCr_{7p}Ga_{10-7p}O_{20}$, has been studied and compared with $SrCr_{8-x}Ga_{4+x}O_{19}$ [13]. The jarosites, $AM_3(OH)_6(SO_4)_2$, where A denotes the alkali ions K, Rb, Na or NH_4, and M denotes the magnetic ions Fe^{3+} or Cr^{3+}, are good model materials of the classical Kagome lattice antiferromagnets with spins 5/2 or 3/2. Our experimental studies for these materials are reviewed in the next section [14–19].

Several Kagome materials with small spin values of $s = 1/2$ or 1 have been synthesized to study the quantum effects on the frustrated Kagome lattice magnets. The organic compound m-MPYNN–BF_4 has radicals with $s = 1/2$, and they couple strongly with ferromagnetic interactions and form the $S = 1$ Kagome lattice antiferromagnet. The susceptibility shows the existence of an energy gap from the nonmagnetic ground state [20]. A metallic complex $[Cu_3(titmb)_2(CH_3CO_2)_6]\cdot H_2O$ has Cu^{2+} ions with spin 1/2 which form a Kagome lattice on the c-planes [21]. The double peaks of specific heat that have been predicted theoretically for the quantum Kagome lattice antiferromagnets were reported. We investigated this material by means of NMR, and present the experimental results in section 3. Volborthite, $Cu_3V_2O_7(OH)_2\cdot H_2O$, also contains Cu^{2+} ions with $s = 1/2$ and has been studied as a candidate material for the quantum Kagome lattice [22]. The Kagome lattice is a little deformed in this material; however, no magnetic ordering has been observed down to 2 K in spite of the large Weiss temperature of −100 K. Recently herbertsmithite, $ZnCu_3(OH)_6Cl_2$, was synthesized as a quantum antiferromagnet with spin 1/2 Cu^{2+} ions on the Kagome lattice [23]. No magnetic ordering has been observed for this material down to 2 K, which is much lower than the Weiss temperature of −314 K.

In this paper, we summarize the magnetic properties of the classical Kagome lattice antiferromagnets, jarosites, studied by means of NMR, magnetization and neutron diffraction,

and then review the experimental studies for Cu-titmb as a candidate material for the quantum Kagome magnet.

2. Classical Kagome lattice antiferromagnets, jarosites

Jarosite family compounds, $AM_3(OH)_6(SO_4)_2$, are nearly two-dimensional magnets. Magnetic ions Fe^{3+} with spin 5/2 or Cr^{3+} with spin 3/2 form the Kagome lattice on the c-planes. Alkali ions K, Rb, Na or NH_4 are located between the Kagome planes. The protons observed by NMR are located nearly on the Kagome c-plane. We used powder samples for our experiments.

The temperature dependence of susceptibility of $KFe_3(OH)_6(SO_4)_2$ shows a clear, sharp cusp at 65 K, which indicates a magnetic phase transition. Magnetic susceptibility of the Heisenberg Kagome lattice antiferromagnet has been calculated using high temperature expansion up to the eighth order [24]. By fitting the experimental data with the theory, the Weiss temperature and the exchange interaction are estimated to be −530 and 23 K, respectively [14]. These values give the frustration factor as $65/530 = 0.12$.

The ^{1}H-NMR spectrum for $KFe_3(OH)_6(SO_4)_2$ in the paramagnetic phase is very sharp with a half width of about 10 G, while the spectra in the ordered phase are broadened and the width reaches 4.1 kG at 1.4 K, indicating the development of sublattice magnetization [15]. The spectrum in the ordered phase shows the typical pattern for powder antiferromagnets, where all protons in the jarosite feel the same magnitude of the internal dipolar fields.

The widths of spectra increase sharply below the transition temperature T_N as the temperature decreases, which indicates the development of sublattice magnetization. These measurements give the critical exponent β as 0.19 for $KFe_3(OH)_6(SO_4)_2$ and 0.21 for $NH_4Fe_3(OH)_6(SO_4)_2$ [15]. The theoretical critical exponents calculated for nonfrustrated three-dimensional spin systems yield values larger than 0.32, and for the two-dimensional Ising system 0.125 [25]. For frustrated antiferromagnets on the triangular lattice, the value of 0.253 has been obtained theoretically for the $Z_2 \times S_1$ system [26]. The experimental value of 0.25 in $CsMnBr_3$, which is a model material of the XY antiferromagnet on the triangular lattice, agrees well with the theoretical value [27]. The exponents obtained for jarosites are small and do not coincide with the theoretical values. The Kagome lattice antiferromagnet jarosites thus might belong to a new universality class.

The spin structure in the ordered phase determined from our NMR and neutron experiments is the 120° spin structure of the $q = 0$ type with a chirality of +1 in the c-plane, as shown in figure 1 [14, 15]. The magnetic unit cell along the c-axis is double the chemical unit cell due to ferromagnetic interplane interactions. On the other hand, neutron diffraction for $KCr_3(OD)_6(SO_4)_2$ revealed that the spin structure in the ordered phase below $T_N = 4.0$ K is also the 120° spin structure in the c-plane; however, the magnetic unit cell along the c-axis coincides with the chemical unit cell due to the antiferromagnetic interplane interactions [16]. The long-range magnetic ordering at finite temperature in $KFe_3(OH)_6(SO_4)_2$ is realized due to the weak anisotropy [14, 15]. The antisymmetric Dzyaloshinskii–Moriya interaction has been discussed as the source of the anisotropy [28, 29].

The temperature dependence of nuclear spin–lattice relaxation rates T_1^{-1} of $KFe_3(OH)_6(SO_4)_2$ is shown in figure 2. The rates show a cusp at the transition temperature of 65 K. The rates in the paramagnetic phase is almost temperature independent above about 100 K, while the rate in the ordered phase decrease sharply with decreasing temperature. The experimental rates in the ordered phase can be explained by the two-magnon process of spin waves with an energy gap of 15 K, which is caused by the anisotropy. The agreement between the experimental data and theoretical values is fairly good over four decades of the relaxation rates, as seen in figure 2. The temperature dependence of the sublattice magnetization at low

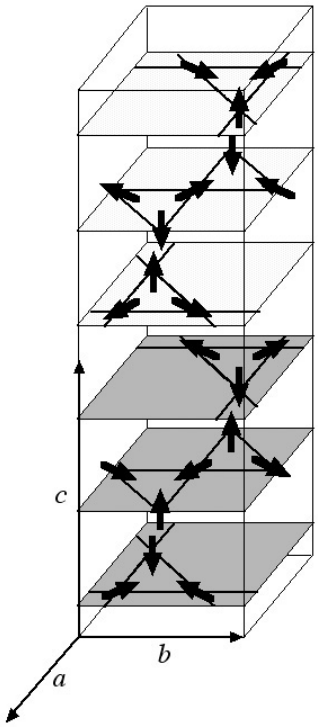

Figure 1. Spin structure of jarosite $KFe_3(OH)_6(SO_4)_2$ in the ordered phase.

temperatures also supports the existence of the spin wave. These results show that the low-energy excitation in the frustrated classical Kagome lattice antiferromagnet is described by the spin wave on the three-sublattice system [15].

In addition to the magnetic transition, some jarosite samples show the second transition at a lower temperature, which was found through the susceptibility and NMR measurements. The NMR experiment for these samples suggests the existence of the partial substitution of alkali ions by H_3O^+ and the deficiency of Fe^{3+}. The successive orderings can be attributed to the two-dimensional ordering in the domains at T_N and the three-dimensional ordering between the Kagome planes at the lower temperature, which are caused by the substitutions, deficiencies and weak interplane interactions [17].

3. Quantum Kagome lattice magnets, Cu-titmb

A metallic complex, Cu-titmb, contains Cu^{2+} ions with spin 1/2, which form the Kagome lattice on the *c*-plane [21]. As the Kagome planes are separated by large titmb molecules, the exchange interactions between the planes are considered to be very weak, and good two-dimensionality is expected. Therefore, this material is expected to be a model material of the quantum frustrated magnet on the Kagome lattice.

The temperature and field dependence of the magnetic specific heat with double peaks has been reported for Cu-titmb [30]. The higher peak at 13 K is field independent, while the lower peak around 2 K is field dependent. These experimental results correspond to theoretical predictions of the double peaks at temperatures of 0.7 J and 0.1 J for the quantum Kagome lattice antiferromagnet [31, 32], and the exchange interaction was estimated to be 19 K.

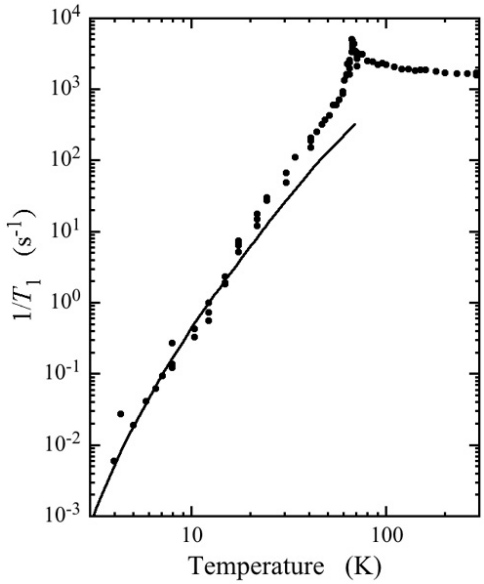

Figure 2. Temperature dependence of the spin–lattice relaxation rates T_1^{-1} of $KFe_3(OH)_6(SO_4)_2$. The NMR frequency is 75.1 MHz in a field of 1.7 T. The solid curve indicates the relaxation rates calculated based on the two-magnon process with an energy gap of 15 K.

In spite of the several tens of kelvin of exchange interactions, our measurement of the magnetic susceptibility in the 50 mT field fits the Curie–Weiss law with a small Weiss temperature of −1.2 K [33]. The exchange interaction is thus estimated to be 1.2 K from the molecular field theory for the Kagome lattice, and this value is one order of magnitude smaller than the value estimated through the specific heat.

The temperature dependence of the spin–lattice relaxation rates T_1^{-1} at various field is shown in figure 3. The rates T_1^{-1} are almost temperature and field independent above 20 K, and are 1.5×10^3 s^{-1}. The rates T_1^{-1} above 20 K are explained by the paramagnetic fluctuation of the Cu^{2+} spins in the magnets. In this case, the rate is proportional to χT, and therefore becomes constant due to the susceptibility χ with the small Weiss temperature. This relaxation rate can also be expressed theoretically as [34]

$$\frac{1}{T_1} = \sqrt{\frac{\pi s(s+1)}{3z}} \frac{A^2}{\hbar |J|}, \tag{1}$$

where z is the number of the nearest neighbours and A is a coupling constant between the Cu^{2+} spin and proton nuclear spin. Using the experimental values of T_1^{-1}, $z = 4$ and $A = 5.7 \times 10^{-28}$ J derived from the spectral width, the exchange constant is estimated to be 0.1 K. This value is remarkably small compared to the values estimated from the specific heat measurement and the Weiss temperature.

The relaxation rates below 10 K show a sharp decrease by lowering the temperature, as is found in figure 3. The relaxation rates can be described by an activation-type exponential function below about 3 K as $T_1^{-1} = B \exp(-\Delta / k_B T)$, where the gap energy Δ depends on the magnetic field [35]. Some measurements were performed below 1 K down to 180 mK by means of a dilution refrigerator in order to precisely obtain the gap energy. The field dependence of the obtained gap energies is shown in figure 4. The field dependence of the gap energies Δ below 3 T fits well with the values of $\Delta = g\mu_B H$ with $g = 2.20$; however,

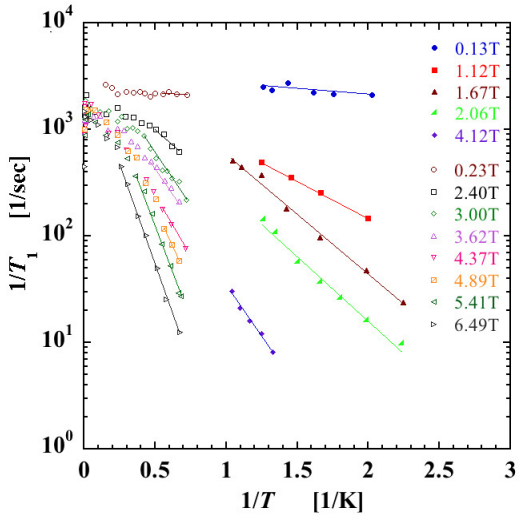

Figure 3. Temperature dependence of the spin–lattice relaxation rates T_1^{-1} at various fields for Cu-titmb. The solid lines indicate the fitting lines for $T_1^{-1} \propto \exp(-\Delta/k_B T)$.
(This figure is in colour only in the electronic version)

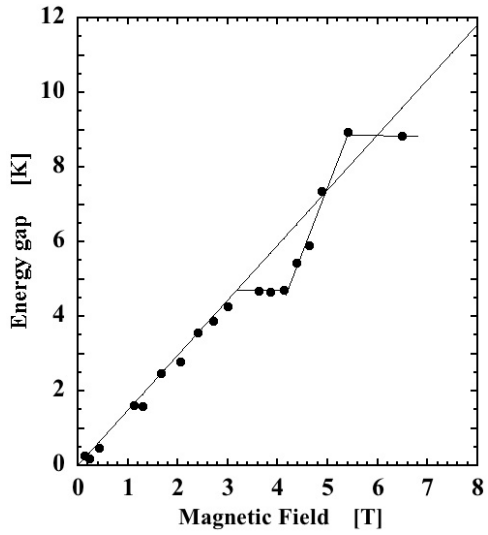

Figure 4. Field dependence of the gap energies obtained from the relaxation rates at low temperatures.

the data seem to deviate from the values above 3.2 T and show structures. The experimental data of the relaxation rates depend on the pulse interval between the first and second pulses of the searching pulse sequence, when the intervals are shorter than about 40 μs. The dependence is caused by the distribution of spin-spin relaxation rates at different ^{1}H nuclear sites. The spin–lattice relaxation rates were measured with pulse intervals longer than 50 μs, for which the rates are independent of the intervals. The rates T_1^{-1} also have the distribution of values at different nuclear sites; however, the rates measured at the same NMR conditions show the

curious structure of the energy gaps in the magnetic fields above 3.2 T. This fact is considered to indicate the existence of anomalous low-energy states in the magnetic field. Recent ESR experiments show that the anomalous absorption lines, which have round shape, appear above 3 T and 5 T below 20 K [36]. These observations just correspond to the anomaly of energy gaps observed through NMR. In addition, it should be noted that no energy gap seems to exist at zero field, as is found in figure 4.

For the low-lying energy states in the field, interesting pulse field magnetization curves have been reported by Narumi *et al* [37]. The magnetization curves at 0.1 K show the 1/3 plateau for the increasing field; however, for the decreasing field this plateau has not been observed. Also, the magnetization curve depends on the sweeping rate of the field. The static magnetization curve at the same temperature increases steeply with increasing fields from zero and no plateau was found. The unstable magnetization behaviour in the pulse fields was explained by the instability of the magnetic state in the spin system with two exchange interactions that have frustration.

The J_1–J_2 model on the Kagome lattice has been investigated by Domenge *et al* [38]. When the nearest neighbour exchange interaction J_1 is antiferromagnetic, the ground states have a 120° spin structure of $q = 0$ type or $\sqrt{3} \times \sqrt{3}$ type depending on the next nearest neighbour interaction J_2 being antiferromagnetic or ferromagnetic, respectively. On the other hand, when J_1 is ferromagnetic, J_2 is antiferromagnetic and $|J_2/J_1| > 1/3$, a complex spin structure with 12 sublattices is realized. The anomalous low-energy states in the magnetic field observed by NMR, the pulse field magnetizations and ESR may be related to these competing exchange interactions and metastable states.

Recently, the ferromagnetic transition at 56 mK was found for this material by the measurements of the specific heat and magnetization at ultra-low temperatures [39]. The specific heat shows a clear cusp at 56 mK and the low-field static magnetization shows the ferromagnetic ordering.

4. Conclusions

The jarosite family compounds, $AFe_3(OH)_6(SO_4)_2$ [A = K, NH_4, Na, Rb], are good model materials of the classical Heisenberg Kagome lattice antiferromagnet. Potassium jarosite, $KFe_3(OH)_6(SO_4)_2$, shows long-range magnetic ordering below $T_N = 65$ K. The magnetic structure in the ordered phase is the $q = 0$ type 120° spin structure with +1 chirality. Though the isotropic Heisenberg Kagome lattice antiferromagnet is considered theoretically to remain disordered down to zero temperature due to the continuous degeneracy of the ground state, long-range magnetic ordering at a finite temperature is realized in this compound due to resolution of the degeneracy by the anisotropy. The spin–lattice relaxation rate T_1^{-1} in the ordered phase decreases sharply with decreasing temperature, and is explained well by the two-magnon process with an energy gap of 15 K of the spin wave dispersion. The temperature dependence of the sublattice magnetization also supports the existence of the spin wave.

The metallic complex Cu-titmb represents spin 1/2 copper magnets on the Kagome lattice, and is expected to be a frustrated quantum Kagome lattice magnet. Experimental results observed by several techniques show curious properties and have a discrepancy between them. One possibility is that the system is a simple ferromagnet with a small exchange interaction, judging from the ferromagnetic transition at 56 mK, the small exchange interaction of 0.1 K estimated through T_1^{-1} at high temperatures and the small Weiss temperature. On the other hand, several anomalous behaviours have been observed. Double peaks of the specific heat have been reported at 13 and 2 K. These values give a large exchange constant of 19 K. The pulse high-field magnetization experiments show the 1/3 magnetization plateau for an

increasing field depending on the sweep rates of the magnetic field and hysteresis. This implies the existence of a quasi-stable state. NMR experiments also show the possibility of anomalous low-energy excitations in the magnetic field. A theoretical J_1–J_2 model is attractive for interpreting the experimental anomalies. The system might have exchange interactions of $|J_2/J_1|$ nearly equal to $1/3$, which corresponds to the boundary of the ferromagnetic phase and the 12-sublattice phase. The anomalous low-lying energy level of this compound might be related to the competing exchange interactions on the Kagome lattice. For the case where two exchange interactions exist, the experimental small Weiss temperature can be explained by the small effective value due to the sum of two exchange interactions $J_1 + J_2$ with opposite signs. However, even in this case, $|J_1|$ and $|J_2|$ cannot have large values to reproduce the relaxation rates T_1^{-1} at high temperatures, because the $\sqrt{z}|J|$ in the denominator in equation (1) is replaced by $\sqrt{z_1}|J_1| + \sqrt{z_2}|J_2|$, and not by $|\sqrt{z_1}J_1 + \sqrt{z_2}J_2|$, where z_1 and z_2 are the numbers of the nearest and next nearest neighbours, respectively, and $z_1 = z_2 = 4$ for the Kagome lattice. We have tried to compress the samples up to 0.87 GPa to change the exchange interactions; however, no change in the relaxation rates could be obtained at any temperature. Further experimental and theoretical studies are desired to understand the magnetic properties of this material.

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References

[1] Ritchey I, Chandra P and Coleman P 1993 *Phys. Rev.* B **47** 15342
[2] Reimers J N and Berlinsky A J 1993 *Phys. Rev.* B **48** 9539
[3] Zeng C and Elser V 1990 *Phys. Rev.* B **42** 8436
[4] Kalmeyer V and Laughlin R B 1987 *Phys. Rev. Lett.* **59** 2095
[5] Zeng C and Elser V 1995 *Phys. Rev.* B **51** 8318
[6] Waldtmann Ch, Everts H-U, Bernu B, Lhuillier C, Sindzingre P, Lecheminant P and Pierre L 1998 *Eur. Phys. J.* B **2** 501
[7] Greywall D S 1990 *Phys. Rev.* B **41** 1842
[8] Elser V 1989 *Phys. Rev. Lett.* **62** 2405
[9] Fukuyama H, Okamoto T, Fukuda T, Akimoto H, Ishimoto H and Ogawa S 1991 *Phys. Rev. Lett.* **67** 1274
[10] Ramirez A P, Espinosa G P and Cooper A S 1990 *Phys. Rev. Lett.* **64** 2070
[11] Broholm C, Aeppli G, Espinosa G P and Cooper A S 1990 *Phys. Rev. Lett.* **65** 3173
[12] Lee S-H, Broholm C, Aeppli G, Perring T G, Hessen B and Taylor A 1996 *Phys. Rev. Lett.* **76** 4424
[13] Bono D, Mendels P, Collin G and Blanchard N 2004 *Phys. Rev. Lett.* **92** 217202
[14] Inami T, Nishiyama M, Maegawa S and Oka Y 2000 *Phys. Rev.* B **61** 12181
[15] Nishiyama M, Maegawa S, Inami T and Oka Y 2003 *Phys. Rev.* B **67** 224435
[16] Inami T, Morimoto T, Nishiyama M, Maegawa S, Oka Y and Okumura H 2001 *Phys. Rev.* B **64** 054421
[17] Nishiyama M and Maegawa S 2003 *Physica* B **329** 1065
[18] Maegawa S, Nishiyama M, Tanaka N, Oyamada A and Takano M 1996 *J. Phys. Soc. Japan* **65** 2776
[19] Morimoto T, Nishiyama M, Maegawa S and Oka Y 2003 *J. Phys. Soc. Japan* **72** 2085
[20] Awaga K, Okuno T, Yamaguchi A, Hasegawa M, Inabe T, Maruyama Y and Wada N 1994 *Phys. Rev.* B **49** 3975
[21] Liu H, Sun W, Tang W, Yamamoto T and Ueyama N 1999 *Inorg. Chem.* **38** 6313

[22] Hiroi Z, Hanawa M, Kobayashi N, Nohara M, Takagi H, Kato Y and Takigawa M 2001 *J. Phys. Soc. Japan* **70** 3377
[23] Shores A P, Nytko E A, Bartlett B M and Nocera D G 2005 *J. Am. Chem. Soc.* **127** 13462
[24] Harris A B, Kallin C and Berlinsky A J 1992 *Phys. Rev.* B **45** 2899
[25] Kawamura H 1992 *J. Phys. Soc. Japan* **61** 1299
[26] Mason T E, Gaulin B D and Collins M F 1989 *Phys. Rev.* B **39** 586
[27] Ajiro Y, Nakashima T, Unno Y, Kadowaki H, Mekata M and Achiwa N 1988 *J. Phys. Soc. Japan* **57** 2648
[28] Matan K, Groholm D, Nocera D G, Yildirim T, Harris A B, Lee S H, Nagler S E and Lee Y S 2006 *Phys. Rev. Lett.* **96** 247201
[29] Yildirim T and Harris A B 2006 *Phys. Rev.* B **73** 214446
[30] Honda Z, Katsumata K and Yamada K 2002 *J. Phys.: Condens. Matter* **14** L625
[31] Elstner N and Young A P 1994 *Phys. Rev.* B **50** 6871
[32] Nakamura T and Miyashita S 1995 *Phys. Rev.* B **52** 9174
[33] Kawahara S, Ueda M, Maegawa S, Fujita K and Yamaguchi R 2004 *J. Magn. Magn. Mater.* **272** e999
[34] Moriya T 1956 *Prog. Theor. Phys.* **16** 23
[35] Maegawa S, Yoshioka K, Kawahara S, Oyamada A, Fujita K and Yamaguchi R 2005 *Prog. Theor. Phys.* **48** S159
[36] Okubo S, Zhang W, Ohta H, Kaji R and Maegawa S 2006 *HFM2006* (presented at PS16-15)
[37] Narumi Y, Katsumata K, Honda Z, Domenge J-C, Sindzingre P, Lhuillier C, Shimaoka Y, Kobayashi T C and Kindo K 2004 *Europhys. Lett.* **65** 705
[38] Domenge J-C, Sindzingre P, Lhuillier C and Pierre L 2005 *Phys. Rev.* B **72** 024433
[39] Karaki Y, Ko M, Yamaguchi A, Ishimoto H, Honda Z, Katsumata K and Yamada K 2006 *HFM2006* (presented at PS16-31)