Field-induced magnetic order in the singlet-ground-state magnet $CsFeCl_3$

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The field-induced magnetic order in the singlet-ground-state system $CsFeCl_3$ ($H\|c$) has been studied by the measurements of the magnetization $M_\|$ (the ferromagnetic component along the c axis) and the magnetic neutron scattering from $M_\perp$ (the antiferromagnetic component in the c plane). The field dependence of $M_\perp$ has clearly shown that the field-induced ordered phase is described by the order parameter $\langle S_x\rangle$ ($M_\perp = g_\perp \mu_B \langle S_x\rangle$). The development of the spin correlation and the incommensurate-commensurate phase transition have been observed in the field region of 5 T$<H<$6 T, and the comparison has been made with the phase boundary at $H\sim 10$ T ($H>H_c$). Two possible magnetic configurations have been suggested for the commensurate three dimensional long-ranged order phase. The frustration and the magnetostriction effects have been discussed.

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I. INTRODUCTION

Recently, many researchers have reviewed the field-induced magnetic order in the singlet-ground-state system with great interests. Especially, the field-induced magnetic order in the dimer spin system have been impressively investigated in the context of a Bose-Einstein condensation (BEC) of magnetic excitations (magnons).[1] A well-known singlet-ground-state system in the hexagonal compound $CsFeCl_3$ is constituted by the ground state $|S_z=0\rangle$ and the first excited states $|S_z=\pm 1\rangle$ of a fictitious spin $S=1$ of an Fe^{2+} ion, each state being separated by single-ion anisotropy energy D, owing to a trigonal symmetrical distortion of Cl^- ions along the c axis ($c\|z$). Since the ferromagnetic exchange interaction ($J_1/k_B \approx 4K$) along the c axis is relatively larger than the antiferromagnetic exchange interaction in the c plane ($J_2/k_B \approx -0.2$ K, triangular lattice antiferromagnet), the spin system is quasi-one dimensional. The effect of D is predominant over the effect of the exchange interaction energies, therefore the ground state is nonmagnetic at zero magnetic field. On the other hand, applying the external magnetic field along the c axis around the level crossing field ($H_c=7.5$ T) of $|S_z=0\rangle$ and $|S_z=+1\rangle$ states, the three dimensional long-range ordered (3D-LRO) state appears[2] in the field region of $4\ \text{T} \leqslant H \leqslant 11\ \text{T}$ at $T_N \leqslant 2.6$ K. The theoretical study[3] has shown that the phase transition is considered to be of the second order and the ordered state is characterized by the nonvanishing value of $\langle S_x\rangle$.

Magnetic excitations and the magnetic phase transition in $CsFeCl_3$ have been extensively investigated experimentally by the neutron scattering,[4–7] electron spin resonance[8–10] (ESR), and NMR[11–13] experiments so far. The excitation energy decreases with lowering temperature, and development of the short range order has been found at the wave vector $\boldsymbol{q} \approx (1/3\ 1/3\ 0)$ (K point).[4] Suzuki described these magnetic behaviors by the dynamical correlated effective field approximation (DCEFA) theory,[14] which takes into account the effect of a fluctuation and correlation. The nuclear magnetic relaxation time T_1 of ^{133}Cs has been measured, and the relaxation mechanism through the magnetic excitations has been clarified.[11–13] The characteristics in the temperature dependence of T_1 are different in the field region below and above 4 T. This fact suggests[12,13] that the large numbers of magnetic excitons are created and a life time of these excitons becomes short in the critical region at $H>4$ T. Magnetization measurements of $M_\|$ (the ferromagnetic component along the c axis) have been performed up to 35 T ($H\|c$), and the anomalous magnetization around 33 T has been observed.[11,15] High-field ESR experiments have been performed up to 40 T by use of a pulse magnet and gyrotron.[16–18]

As for the 3D-LRO state around H_c, an incommensurate-commensurate phase transition has been already studied[7] by the neutron diffraction experiments up to 5.5 T. However, the research has been limited in the vicinity of the phase boundary owing to the performance limitations of cryostats. Invoked by the development of the steady-state magnet available for the neutron scattering experiment, in this paper, we report the neutron diffraction and magnetization experiments which have been performed under the magnetic fields up to 10 T ($H\|c$, $H>H_c$).

II. EXPERIMENT

Samples have been prepared in the laboratory at the Institute of Advanced Energy (IAE), Kyoto University. The

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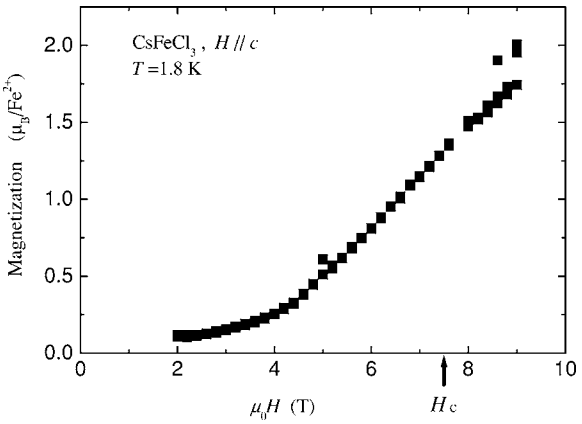

FIG. 1. Magnetization data of $M_\parallel$ as a function of external magnetic field ($H \| c$) at 1.8 K. Dissipation of the data at $H \approx 9$ T might be the experimental error.

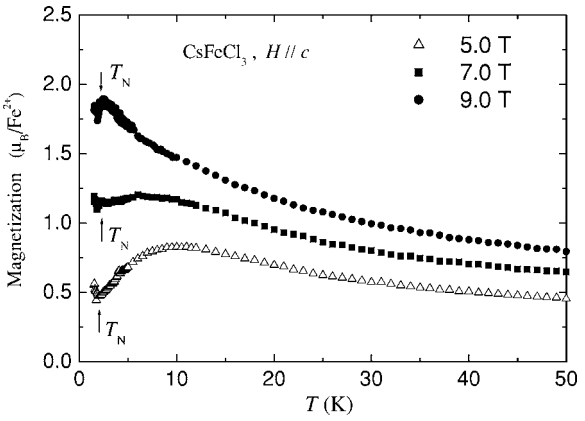

FIG. 2. Magnetization data of $M_\parallel$ as a function of temperature at 5, 7, and 9 T ($H \| c$).

equimolar 99.9% CsCl and $FeCl_2$ powders mixed in the ampoule were dehydrated using HCl gas,[19] and single crystals of a volume of 2–3 cm^3 large were separated out by the Bridgman method. Clear hexagonal cleavage planes were seen, and the color of the small crystal (~ 0.2 cm^3) was transparent dark green. The mosaic width was about $\sim 0.5°$, and the hexagonal lattice parameter as used is $a = 7.174$ Å at 1.8 K (the lattice parameter of the c axis is not measured) in the neutron experiments.

The magnetization measurements have been performed using a 9 T superconducting quantum interference device (SQUID) magnetometer at Berlin Technical University. The measured sample was the needle shaped crystal of sample mass 0.61 mg. The field dependent signal resolution is in the range of 5×10^{-5} emu.

The neutron diffraction experiments were carried out on the triple-axis spectrometer E1 at the Berlin Neutron Scattering Center (BENSC), Hahn Meitner Institute (HMI), Berlin. The wavelength $\lambda = 2.4153$ Å was selected by Bragg reflection at a pyrolytic graphite monochromator for the incoming beam, and the analyzer was adjusted to measure the elastic part of the scattering. The used collimation was 70′-80′-60′-60′. The sample was mounted in the high-field superconducting magnet VM1 built by Oxford Instruments, which with a maximum field of 17 T is actually the strongest steady-state magnet available for the neutron scattering experiment. The preliminary experiments have been carried out on the triple-axis spectrometer PONTA at Institute for Solid State Physics (ISSP), Japan Atomic Energy Research Institute (JAERI), Tokai.

III. RESULTS AND DISCUSSION

A. Magnetization

First we report the results of the magnetization measurements. Figure 1 shows the data of $M_\parallel$ ($M_\parallel = g_\parallel \mu_B \langle S_z \rangle$) measured at $T = 1.8$ K, plotted as a function of external magnetic field ($H \| c$).

At such low temperature, most of spins should be distributed at the ground state; therefore, $M_\parallel$ is nearly zero at low fields. The field variation of $M_\parallel$ shows explicit increase above 4 T in the ordered phase, and increases linearly up to 9 T. The value of magnetization is ~ 1.3 μ_B at H_c, which is approximately half the magnetic moment of $g_\parallel \mu_B S = 2.54 \mu_B$.[14]

Figure 2 shows the results of the magnetization measurements as a function of temperature at 5, 7, and 9 T. At 5 T, the curve exhibits a broad maximum at $T \approx 10$ K, which is explained by the term of the thermal activation type, $\exp(-E_1/k_B T)$, where E_1 is the gap energy between the ground state $|S_z = 0\rangle$ and the first excited state $|S_z = +1\rangle$. On the other hand, this broad maximum disappears at 9 T, where the ground state is magnetic $|S_z = +1\rangle$. At the transition temperature T_N, a kink appears in the temperature dependences of magnetization under the magnetic fields between 5 and 9 T.

The various temperature dependences of $M_\parallel$ are observed in the 3D-LRO state. The similar feature has been found in the dimer spin system $TlCuCl_3$.[1] In the molecular field approximation (MFA), however, the magnetization should not depend on temperature in the ordered phase.[3] We expect that such temperature dependences result from the many body effect, which is neglected in the MFA. Considering that the magnetic order is derived from the softening of magnetic excitation, the coherent state of $|S_z = 0\rangle$ and $|S_z = +1\rangle$ will develop following the freezing of magnetic excitons, which gives temperature dependence of $M_\parallel$ in the ordered phase.

B. Neutron Diffraction

Next we show the results of the high field neutron diffraction experiments. The single crystal of a volume of ~ 1 cm^3 was attached to the sample holder, and combined into the microgoniomator (Kohzu-Seiki Co.), which controls the inclination of the c axis below $\sim 0.3°$. This configuration was essential to observe the incommensurate state.[20] The former NMR experiments[21] have clarified the dependence of the phase diagram on the crystal orientation in the magnetic field.

Figure 3 shows the dependence of integrated intensity on external magnetic field ($H \| c$) for the magnetic reflection at $T = 1.8$ K. At first, we confirmed the appearance of the in-

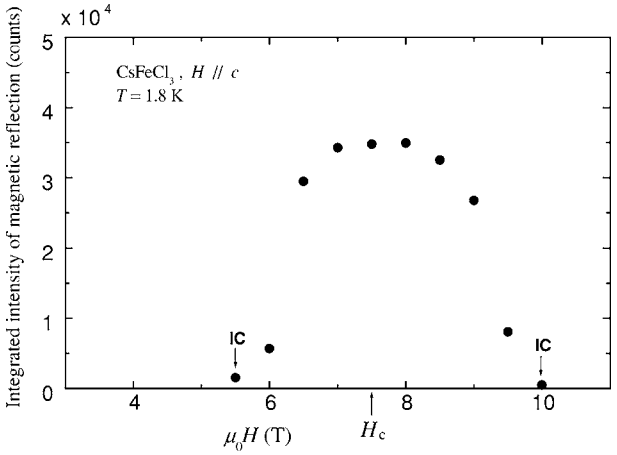

FIG. 3. The dependence of integrated intensity on external magnetic field ($H\|c$) for the magnetic reflection at $T=1.8$ K. "IC" indicates that the measurements have been performed in the incommensurate phase.

commensurate state at $H\approx 5$T, and then by increasing the magnetic field we brought the magnetic state to the commensurate state. The intensity of magnetic (1/3 1/3 0) reflection ($I_{\frac{1}{3}\frac{1}{3}0}$) is directly related to $M_\perp$ (the antiferromagnetic component in the c plane), and the experimental results show that $M_\perp$ increase and decrease almost symmetrically with respect to H_c. Comparison with the phase diagram[2] clearly indicates that the LRO is described by the order parameter $\langle S_x\rangle$ ($M_\perp = g_\perp \mu_B \langle S_x\rangle$). It is noted that even a slight tilt of the c axis yielded asymmetry of the scattering intensity with respect to H_c.[20]

Figure 4 shows the dependences of integrated intensities on temperature for the magnetic reflections at 7.0, 7.5, 8.0, and 8.5 T ($H\|c$). The critical exponent β for the order parameter has been estimated from the following equation:

$$\sqrt{I_{\frac{1}{3}\frac{1}{3}0}} \propto (T_N - T)^\beta. \tag{1}$$

The values of β varied with field, each being 0.30±0.03 (7.0 T), 0.22±0.02 (7.5 T), 0.27±0.03 (8.0 T), and 0.33±0.02 (8.5 T). For reference, the value of β for the Bose Einstein condensation in the 3D-XY system is 0.35,[1] which is near to the value of β at 8.5 T.

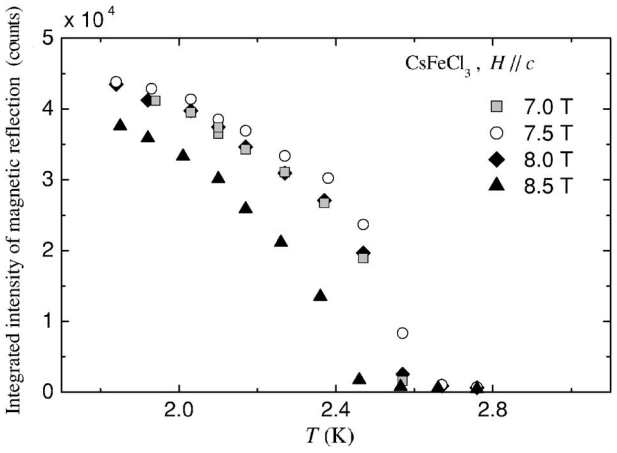

FIG. 4. The dependences of integrated intensities on temperature for the magnetic reflections at 7.0, 7.5, 8.0, and 8.5 T ($H\|c$).

Now, we estimate the value of the order parameter $\langle S_x\rangle$ by comparing the intensities of the nuclear reflection with the magnetic reflection. Although the magnetic configuration in $CsFeCl_3$ in the ordered state has not been determined, we assume a cone structure model (ferromagnetic along the c axis and 120° periodic in the c plane), referring to the isomorphous compound $RbFeCl_3$.[22]

The magnetic structure factors from each component are described as

$$|F_{hkl^\pm}|^2 = \tfrac{1}{4} p_0^2 f^2 M_\perp^2 (1+\cos\phi^2)|G_{hkl}|^2, \tag{2}$$

$$|F_{hkl}|^2 = p_0^2 f^2 M_\parallel^2 \sin\phi^2 |G_{hkl}|^2, \tag{3}$$

where p_0 is 0.269×10^{-12} cm, f is the form factor, $|G_{hkl}|^2 = |\Sigma_i e^{i\mathbf{q}_{hkl}\cdot\mathbf{r}_i}|^2$ is the geometrical structure amplitudes of magnetic ions for the (hkl) reflection, and ϕ is the angle between the scattering vector and the c axis. The notation of $|F_{hkl^\pm}|^2$ denotes the fact that the magnetic scatterings from the $M_\perp$ component appear at $\vec{s}=\vec{B}_{hkl}\pm\vec{\tau}$, where $\vec{B}_{hkl}$ is the reciprocal lattice vector and $\vec{\tau}$ is the modulation vector for the magnetic structure.

Because the experiments have been performed at very low temperatures, we may approximate the Debye-Waller factor as 1, and the value of $M_\perp$ can be determined experimentally by comparing the intensities of nuclear (1 1 0) reflection at zero field (I_{110}; nuclear) with $I_{\frac{1}{3}\frac{1}{3}0}$ as following:

$$\frac{I_{\frac{1}{3}\frac{1}{3}0}}{I_{110}} \approx \frac{\sin^{-1}(2\theta_{\frac{1}{3}\frac{1}{3}0})\frac{1}{4}p_0^2 f^2 M_\perp^2 |G_{110}|^2}{\sin^{-1}(2\theta_{110})|F_{110}^n|^2}, \tag{4}$$

where $|F_{110}^n|^2 = |\Sigma_i b_i e^{i\mathbf{q}_{110}\cdot\mathbf{r}_i}|^2$ is the square of the nuclear structure amplitude factors of all ions. For example, values of $M_\perp$ and $\langle S_x\rangle$ at $T=1.55$ K and $H=7.5$ T are derived as $1.83\mu_B$ and 0.477, respectively ($g_\perp = 3.84$).[14]

The calculation of $\langle S_x\rangle$ in the MFA[3] gives the value of 0.703 at the same condition, where the Hamiltonian parameters are chosen as $J_1=4.4$ K and $D=17.4$ K. The discrepancy between 0.477 (experimental) and 0.703 (MFA) is large. Probably, the spin fluctuation is not easily stabilized because of the magnetic frustration in the triangular lattice antiferromagnet.

So far, we have focused on the symmetry in the field dependence of $\langle S_x\rangle$ with respect to H_c in the commensurate ordered region. On the other hand, the field dependence of the incommensurate phase is asymmetric with respect to H_c as shown previously.[2] The origin of the incommensurate state is attributed to the magnetic dipole-dipole interaction by Shiba,[24] and to the spin fluctuation by Lindgård.[23] To clarify the behavior of the incommensurate state, we have investigated the incommensurate state in the low field region ($H<H_c$) and in the high field region ($H>H_c$).

Figure 5 shows the field variation of the scattering vector $\boldsymbol{q}$ along (h h 0) at $T=1.8$ K in the incommensurate phase (5 T$<H<$6 T), where bars denote the linewidth [Δh: full width at half maximum (FWHM)] determined by fitting to a

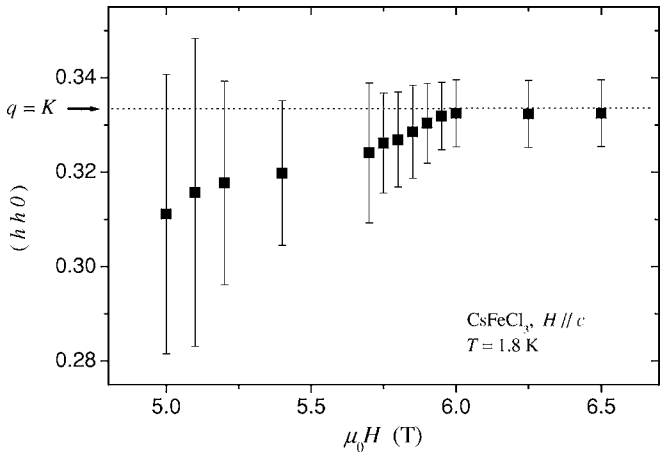

FIG. 5. The field variations of the scattering vector $\boldsymbol{q}$ and the line width along (h h 0) at $T=1.8$ K in the incommensurate phase (5 T$<H<$6 T). Bars are the linewidths (Δh; FWHM) determined by fitting to a Gaussian curve.

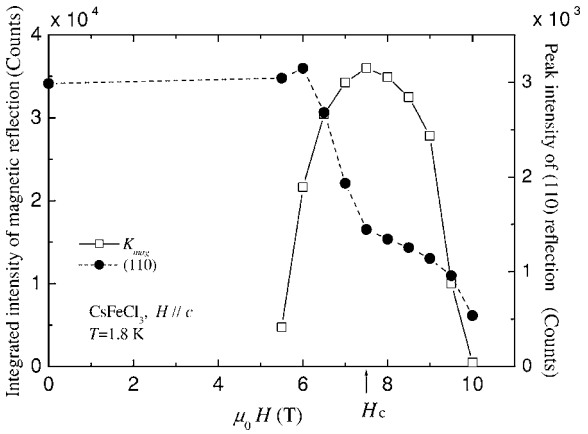

FIG. 6. The field variations of the intensities of the magnetic (1/3 1/3 0) reflection and the nuclear (1 1 0) reflection at $T=1.8$ K.

Gaussian curve. Increasing from zero field, we first found the magnetic reflection around 5 T at $\boldsymbol{q}\approx(0.311\ 0.311\ 0)$. For the further increasing of the magnetic field, $\boldsymbol{q}$ gradually moved toward the K point, accompanied by narrowing of the linewidth and increasing of the scattering intensity. Development of the coherent state or the spin correlation has been observed in the incommensurate state. We can estimate the numbers of correlated spins in the c-plane (N) by the relation $N=\xi/a\sim(1/\Delta h)/a$ (ξ: the correlation length, a: the lattice parameter). We find that the numbers of correlated spins is relatively small, and it increases approximately from 3 to 10 in the incommensurate phase. It seems that the effect of spin fluctuation is large in the incommensurate phase.

At the phase boundary around 10 T ($H>H_c$) at $T=1.8$ K, a satellite reflection from the incommensurate ordered state was found in coexistence with the magnetic (1/3 1/3 0) reflection. The presence of the commensurate ordered state was not excluded even in the incommensurate phase.

Figure 6 shows the experimental evidence of the lattice deformation induced by the magnetic ordering. Unexpectedly, the intensity of the nuclear (1 1 0) reflection decreased drastically at the occurrence of the magnetic ordering. For a possible magnetic configuration, we may suggest 120° periodic magnetic structure in the c plane as shown in Fig. 7(a). In this case, the mechanism of the lattice deformation can be interpreted as a result of releasing the magnetic frustration in the c plane. To determine the details, we have to examine other nuclear reflections systematically, and that is the work remained to be done in the future.

Before the conclusion, we here introduce another magnetic structure, which have already been derived from the group theory.[25,26] Varying the ratio of the trigonal field to the spin-orbit coupling, Suzuki[27] examined the metamagnetism in $RbFeCl_3$ and $CsFeCl_3$, and it is found difficult to explain in a unified way the observed magnetic behavior results at low fields and high fields. In order to explain this metamagnetism, Zavorotnev[25,26] investigated the magnetic configuration on the basis of the group theory. Figure 7(b) shows the candidate of the spin configuration suggested for the intermediate field region from 10 to 33 T, where 1/3 of the Fe^{2+} ions are paramagnetic (induced-ferro) and 2/3 are easy-plane type spins.

The benefit of the idea is that the metamagnetic step at $\sim 2.6\mu_B$ could be explained as 2/3 plateau of the $J=2$ multiplet. However, as observed in the neutron diffraction experiments, the ordered state clearly disappears at 10 T, and this result neglects possibility of the easy-plane type ordering at higher fields. Although, such magnetic configuration might appear in the ordered state around H_c, and it explains distortion of the lattice in a simple way. If such magnetic ordering occurs, then the crystal lattice could be deformed under the action of the exchange forces, and it will change to C_{2v} symmetry. In this case, the intensity of the nuclear (1 1 0) reflection will decrease owing to the lowering of the lattice symmetry in the c plane.

IV. CONCLUSIONS

We have investigated the field-induced magnetic order in the singlet-ground-state system $CsFeCl_3$ ($H\|c$) by the mea-

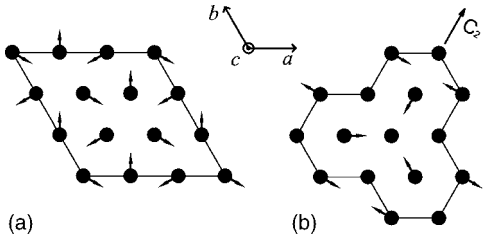

FIG. 7. Possible two magnetic structures suggested for the ordered state. The arrows show the directions of spins of corresponding ions. The magnetic scattering vectors of both configurations appear at the K point. (a) 120° periodic magnetic structure in the c plane. (b) C_{2v} symmetric magnetic configuration, derived from the group theory analysis (see Ref. 26). The magnetic moment of ions without arrow are paramagnetic. The occurrence of the lattice deformation is intuitively clear in Fig. 7(b), owing to the appearance of the symmetry axis C_2 in the c plane.

surements of the magnetization $M_{\parallel}$ and the magnetic neutron scattering from $M_{\perp}$. Values of the critical exponent β have been estimated for the temperature dependences of $M_{\perp}$, and the field dependence of $M_{\perp}$ has shown that the ordered phase is described by the order parameter $\langle S_x \rangle$ ($M_{\perp}=g_{\perp}\mu_{\mathrm{B}}\langle S_x \rangle$).

Development of the spin correlation in the incommensurate phase and the incommensurate-commensurate phase transition have been clearly observed in the field region of $5\ \mathrm{T}<H<6\ \mathrm{T}$ at $T=1.8$ K. On the contrary at $H\sim 10$ T ($H>H_c$), the presence of the commensurate ordered state was not excluded even in the incommensurate phase.

Two possible magnetic configurations have been suggested for the ordered phase, one is 120° periodic structure in the c plane and another is the partially disordered state, to explain the observed magnetic behaviors and its effect on the crystalline lattice.

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[1] T. Nikuni, M. Oshikawa, A. Oosawa, and H. Tanaka, Phys. Rev. Lett. **84**, 5868 (2000).
[2] T. Haseda, N. Wada, M. Hata, and K. Amaya, Physica B & C **108B**, 841 (1981).
[3] T. Tsuneto and T. Murao, Physica (Amsterdam) **51**, 186 (1971).
[4] H. Yoshizawa, W. Kozukue, and K. Hirakawa, J. Phys. Soc. Jpn. **49**, 144 (1980).
[5] M. Steiner, K. Kakurai, W. Knop, B. Dorner, R. Pynn, U. Happek, P. Day, and G. McLeen, Solid State Commun. **38**, 1179 (1981).
[6] B. Schmid, B. Dorner, D. Petitgrand, L. P. Regnault, and M. Steiner, Z. Phys. B: Condens. Matter **95**, 13 (1994).
[7] W. Knop, M. Steiner, and P. Day, J. Magn. Magn. Mater. **31–34**, 1033 (1983).
[8] M. Motokawa, H. Ohta, K. Fukuda, N. Kitamura, T. Hirano, and K. Yamazaki, Physica B **155**, 340 (1989).
[9] H. Ohta, N. Makita, K. Yoshida, T. Nanba, and M. Motokawa, Int. J. Infrared Millim. Waves **13**, 457 (1992).
[10] H. Ohta and M. Motokawa, *Recent Advances in Magnetism of Transition Metal Compounds*, edited by A. Kotani and N. Suzuki (World Scientific, Singapore, 1993), p. 316.
[11] M. Chiba, Y. Ajiro, K. Adachi, and T. Morimoto, J. Phys. Soc. Jpn. **57**, 3178 (1988).
[12] M. Toda, T. Goto, M. Chiba, K. Adachi, and N. Suzuki, J. Phys. Soc. Jpn. **68**, 2210 (1999).
[13] M. Toda, T. Goto, M. Chiba, and N. Suzuki, J. Phys. Soc. Jpn. **71**, 930 (2002).
[14] N. Suzuki and J. Makino, J. Phys. Soc. Jpn. **64**, 2166 (1995).
[15] H. Hori, I. Shiozaki, M. Chiba, T. Tsuboi, and M. Date, Physica B **155**, 299 (1989).
[16] M. Chiba, Aripin, K. Kitai, S. Mitsudo, T. Idehara, S. Ueda, and M. Toda, Physica B **294–295**, 64 (2001).
[17] M. Chiba, K. Kitai, S. Mitsudo, T. Idehara, S. Ueda, and M. Toda, in *EPR in the 21st Century*, edited by A. Kawamori, J. Yamauchi and H. Ohta (Elsevier, New York, 2002), p. 779.
[18] M. Chiba, T. Higuchi, K. Kitai, S. Mitsudo, T. Idehara, M. Toda, and S. Ueda, Physica B **329–333**, 950 (2003).
[19] K. Hirakawa and K. Ubukoshi, Solid State Phys. **14**, 625 (1979).
[20] M. Toda, T. Goto, M. Chiba, S. Ueda, K. Nakajima, K. Kakurai, R. Feyerherm, A. Hoser, H. A. Graf, and M. Steiner, J. Phys. Soc. Jpn. **70**, Suppl. A, 154 (2001).
[21] M. Chiba, S. Ueda, T. Yanagimoto, M. Toda, and T. Goto, Physica B **284**, 1529 (2000).
[22] N. Wada, K. Sumiyoshi, T. Watanabe, and K. Amaya, J. Phys. Soc. Jpn. **52**, 1893 (1983).
[23] P. A. Lindgård and B. Schmid, Phys. Rev. B **48**, 13636 (1993).
[24] H. Shiba and N. Suzuki, J. Phys. Soc. Jpn. **51**, 3488 (1982).
[25] Yu. D. Zavorotnev, J. Magn. Magn. Mater. **135**, 30 (1994).
[26] Yu. D. Zavorotnev, J. Magn. Magn. Mater. **135**, 37 (1994).
[27] N. Suzuki and Y. Tagawa, Physica B **155**, 375 (1989).