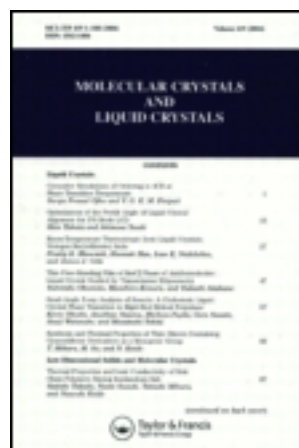


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## ROLE OF DIPOLE–DIPOLE INTERACTION IN PHASE TRANSITION OF ORGANIC ANTIFERROMAGNET MOTMP

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**Abstract** The role of the dipole-dipole (DD) interaction in organic molecular antiferromagnet MOTMP is elucidated. The anisotropy energy due to the DD interaction can explain well the experimental values of the spin-flop field and the saturation field along each crystal axis if we use the nearest neighbor (n.n.) interchain exchange coupling  $J_c = -0.0087$  K and if we introduce small anisotropy for the intrachain exchange coupling. The uniform magnetic susceptibility in the paramagnetic state has been calculated by taking into account the DD interaction on the basis of the dynamical correlated-effective-field approximation. The observed susceptibility are well reproduced by assuming the n.n. intrachain exchange coupling  $J_a = 0.45$  K and the next n.n. interchain exchange coupling  $J_b = 0$  K. The Néel temperature  $T_N$  is determined to be 0.15 K, which agrees with the observation.

### INTRODUCTION

Recently many purely-organic molecular crystals have been found to show long range magnetic ordering.<sup>1</sup> In the sense that the molecular magnets thus far discovered can be described in terms of Heisenberg Hamiltonian of  $S = \frac{1}{2}$  spins their magnetic properties are essentially the same as those of transition-metal compounds. But we can expect some peculiar features characteristic to the molecular organic magnets. One is the origin of anisotropy. For  $S = \frac{1}{2}$  spin systems the single-ion type anisotropy energy is absent because  $S_\beta^2 \equiv \frac{1}{4}$  ( $\beta = x, y, z$ ). Further the anisotropy of the exchange coupling in molecular magnets is expected to be small compared with that in transition-metal compounds because of smallness of the spin-orbit interaction. Then, it is naturally expected that the dipole-dipole interaction will dominantly contribute to the anisotropy energy in molecular magnets.

MOTMP (4-methacryloyloxy-2,2,6,6-tetramethyl-piperidin-1-oxyl) molecule has a pendant nitroxyl radical N-O on which one electron is localized. The paramagnetic susceptibility of the MOTMP crystal from 2.5 K to 300 K follows the Curie-Weiss law with a positive Weiss constant.<sup>2</sup> From measurements of the heat capacity down to 0.07 K a phase transition to a long range ordering has been observed at  $T_N = 0.15$  K.<sup>3</sup>

By measurements of magnetic field dependence of ac-susceptibility and magnetization at  $T=30$  mK the spin-flop transition has been observed when the field is parallel to the  $a$ -axis.<sup>4</sup> The critical field at which the spin moment saturates has been also determined for each crystal axis.

Kawamoto and Suzuki<sup>5</sup> have already demonstrated that the observed spin easy axis is explained certainly by the DD interaction. It has been shown also that the anisotropy energy due to the DD interaction can explain fairly well the experimental values of the spin-flop field and the saturation field along each crystal axis. It is noted, however, that there remain some discrepancies, which may indicate the necessity of introducing the anisotropic exchange coupling. It should be noted also that the strength of the DD interaction is the same order as the interchain exchange coupling. This fact suggests that the DD interaction cannot be neglected in discussing quantitatively the magnetic susceptibility and the transition temperature.

The purpose of this paper is first to analyze more quantitatively the transition fields of MOTMP by introducing the anisotropic exchange coupling, and secondly to calculate the magnetic susceptibility and the transition temperature of MOTMP by taking account of the DD interaction.

### FIELD-INDUCED PHASE TRANSITION

The structure of MOTMP crystal is monoclinic (space group  $P2_1$ ) and the primitive unit cell contains two molecules. An  $S=\frac{1}{2}$  spin is localized on the N-O radical of each molecule. The spins in the same sublattice align ferromagnetically, but the spin directions on different sublattices are opposite (see Figure 1). Judging from the

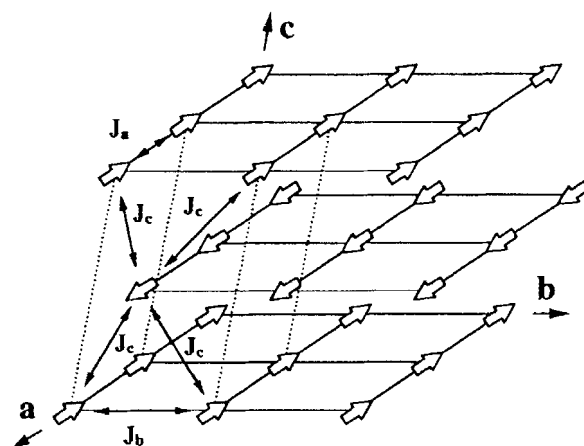


FIGURE 1 The spin structure of MOTMP.  $J_a$  and  $J_b$  are intra-sublattice exchange coupling, and  $J_c$  is inter-sublattice coupling.

lattice constants,<sup>4,6</sup> this system is regarded as a pseudo-one-dimensional system consisting of linear spin chains along the  $a$ -axis.

We express the relevant Hamiltonian for MOTMP as follows ( $S=\frac{1}{2}$ ):

$$\begin{aligned} \mathcal{H} = & - \sum_{ij} \sum_{\mu\nu} \sum_{\beta} J_{i\mu,j\nu}^{\beta} S_{i\mu}^{\beta} S_{j\nu}^{\beta} \\ & + \frac{1}{2} (g\mu_B)^2 \sum_{ij} \sum_{\mu\nu} \sum_{\beta\gamma} S_{i\mu}^{\beta} D_{i\mu,j\nu}^{\beta\gamma} S_{j\nu}^{\gamma} - g\mu_B \sum_{i\mu} \mathbf{S}_{i\mu} \cdot \mathbf{H}, \end{aligned} \quad (1)$$

$$\text{with } D_{i\mu,j\nu}^{\beta\gamma} = \frac{\delta_{\beta\gamma}}{|\mathbf{R}_{i\mu} - \mathbf{R}_{j\nu}|^3} - \frac{3(\mathbf{R}_{i\mu} - \mathbf{R}_{j\nu})^{\beta}(\mathbf{R}_{i\mu} - \mathbf{R}_{j\nu})^{\gamma}}{|\mathbf{R}_{i\mu} - \mathbf{R}_{j\nu}|^5},$$

where  $i, j$  denote the unit cell number,  $\mu, \nu=1,2$  specify the sublattice,  $\beta, \gamma=x, y, z$ ,  $\mathbf{R}_{i\mu}$  represents the position vector of  $\mathbf{S}_{i\mu}$  and  $\mathbf{H}$  is the external uniform magnetic field. We take the  $x$ - and  $y$ -axis parallel to the crystal  $a$ - and  $b$ -axis, respectively, and the  $z$ -axis perpendicular to the  $xy$ -plane, *i.e.* parallel to the  $c^*$ -axis. As to the exchange coupling we have adopted the anisotropic form.

Throughout this section we consider only the case of  $T=0$  K and treat the spin as a classical vector. Then the expressions of the spin-flop field  $H_{sf}$  and the saturation field  $H_c^{\beta}$  along the each crystal axis are obtained straightforwardly even in the case of anisotropic exchange coupling:

$$\begin{aligned} H_c^x = & -\frac{2S}{g\mu_B} \{J_{11}^x(0) - J_{11}^y(0)\} - \frac{2S}{g\mu_B} \{J_{12}^x(0) + J_{12}^y(0)\} \\ & + g\mu_B S [\{D_{11}^{xx}(0) + D_{12}^{xx}(0)\} - \{D_{11}^{yy}(0) - D_{12}^{yy}(0)\}] \end{aligned} \quad (2)$$

$$\begin{aligned} H_c^y = & \frac{2S}{g\mu_B} \{J_{11}^x(0) - J_{11}^y(0)\} - \frac{2S}{g\mu_B} \{J_{12}^x(0) + J_{12}^y(0)\} \\ & + g\mu_B S [\{D_{11}^{yy}(0) + D_{12}^{yy}(0)\} - \{D_{11}^{xx}(0) - D_{12}^{xx}(0)\}] \end{aligned} \quad (3)$$

$$\begin{aligned} H_c^z = & \frac{2S}{g\mu_B} \{J_{11}^x(0) - J_{11}^z(0)\} - \frac{2S}{g\mu_B} \{J_{12}^x(0) + J_{12}^z(0)\} \\ & + g\mu_B S [\{D_{11}^{zz}(0) + D_{12}^{zz}(0)\} - \{D_{11}^{xx}(0) - D_{12}^{xx}(0)\}] \end{aligned} \quad (4)$$

$$\begin{aligned} H_{sf} = & \left[ H_c^x \times \left\{ \frac{2S}{g\mu_B} \left( \{J_{11}^x(0) - J_{11}^y(0)\} - \{J_{12}^x(0) - J_{12}^y(0)\} \right) \right. \right. \\ & \left. \left. + g\mu_B S \left( \{D_{11}^{yy}(0) - D_{12}^{yy}(0)\} - \{D_{11}^{xx}(0) - D_{12}^{xx}(0)\} \right) \right\} \right]^{1/2} \end{aligned} \quad (5)$$

where  $J_{\mu\nu}^{\beta}(0)$  and  $D_{\mu\nu}^{\beta\gamma}(0)$  represent the  $q=0$  component of Fourier transform of  $J_{i\mu,j\nu}^{\beta}$  and  $D_{i\mu,j\nu}^{\beta\gamma}$ , respectively. Here it is noted that  $J_{11}^{\beta}(0) = J_{22}^{\beta}(0)$  and  $D_{11}^{\beta\gamma}(0) = D_{22}^{\beta\gamma}(0)$ . If we consider the three kinds of exchange coupling as shown in Fig. 1, we have  $J_{11}^{\beta}(0) = 2J_a^{\beta} + 2J_b^{\beta}$  and  $J_{12}^{\beta}(0) = 4J_c^{\beta}$ . Calculations of  $D_{\mu\nu}^{\beta\gamma}(q)$  have been done by the Ewald method with use of the following lattice parameters:<sup>4,6</sup>  $a = 6.071 \text{ \AA}$ ,  $b = 9.770 \text{ \AA}$ ,  $c = 11.828 \text{ \AA}$ ,  $\gamma = 101.82 \text{ deg}$ ,  $\mathbf{r}_1 = 0$ ,  $\mathbf{r}_2 = 0.86\mathbf{a} + 0.5\mathbf{b} + 0.52\mathbf{c}$  where  $\mathbf{r}_1$  and  $\mathbf{r}_2$  denote the position vectors of two N-O radicals in the unit cell.

If we neglect the anisotropy of the exchange coupling, the transition fields depend only on the inter-sublattice coupling  $J_{12}(0) = 4J_c$ . We have determined the value of  $J_c$  so as to reproduce the observed  $H_c^z = 755 \text{ Oe}$ .<sup>4</sup> The result is  $J_c = -0.0087 \text{ K}$ . Then the other transition fields have been evaluated as shown in Table I. Agreement between the theory and the experiment is fairly well. Compared with the experimental data in more detail, however, the theoretical values of  $H_{sf}$  and  $H_c^y$  are a little bit larger, and that of  $H_c^x$  is smaller. By looking into the expressions of  $H_c^x$ ,  $H_c^y$  and  $H_{sf}$  (eqs.(2), (3) and (5)) we become aware that these small differences may be removed if we introduce anisotropy to the intra-sublattice exchange coupling  $J_{11}(0)$ . In fact, if we assume  $J_{11}^x(0) - J_{11}^y(0) = -0.0026 \text{ K}$  and  $J_{11}^z(0) = J_{11}^x(0)$ , we can reproduce the observed transition fields quite well (see Table I).

TABLE I The transition fields of MOTMP (in unit of Oe).

	$H_{sf}$	$H_c^x$	$H_c^y$	$H_c^z$
Theory: Isotropic Exchange	310	380	770	755
Theory: Anisotropic Exchange	300	400	750	755
Experiment <sup>4</sup>	300	400 ~ 450	750	755

## MAGNETIC SUSCEPTIBILITY AND NÉEL TEMPERATURE

In this section we calculate the magnetic susceptibility in the paramagnetic state by taking into account the DD interaction on the basis of the dynamical correlated-effective-field approximation (DCEFA).<sup>7</sup> Originally the DCEFA was formulated for systems with exchange coupling only, but its extension to systems with the DD interaction is straightforward. The essence of the DCEFA lies in the following decoupling:

$$S_{i\mu}^{\beta} S_{j\nu}^{\gamma} \rightarrow S_{i\mu}^{\beta} (\langle S_{j\nu}^{\gamma} \rangle - \alpha \langle S_{i\mu}^{\beta} \rangle) + S_{j\nu}^{\gamma} (\langle S_{i\mu}^{\beta} \rangle - \alpha \langle S_{j\nu}^{\gamma} \rangle), \quad (6)$$

where  $\langle S_{i\mu}^{\beta} \rangle$  denotes the spontaneous or the field-induced spin moment, and  $\alpha$  represents the correlation parameter which can be determined by a self-consistency

condition required from the fluctuation-dissipation theorem. If we put  $\alpha=0$ , this approximation is reduced to the molecular-field approximation (MFA).

In this paper we consider only the paramagnetic state. Then, the effective single-ion Hamiltonian is simply the Hamiltonian of an  $S=\frac{1}{2}$  free spin. In order to calculate the susceptibility tensor  $\tilde{\chi}(\mathbf{q}, \omega)$  of the paramagnetic state in the spirit of DCEFA, we apply fictitious magnetic fields  $h_{i\mu}^\beta e^{-i\omega t}$  which is dependent on time and position. Then, the effective Zeeman interaction for the  $i\mu$ -th spin within the DCEFA is expressed as

$$\mathcal{H}_{i\mu, \text{Zeeman}}^{\text{eff}} = - \sum_{\beta} \left[ h_{i\mu}^\beta e^{-i\omega t} + 2 \sum_{j, \nu, \gamma} C_{i\mu, j\nu}^{\beta\gamma} (\langle S_{j\nu}^\gamma \rangle - \alpha \langle S_{i\mu}^\gamma \rangle) \right] S_{i\mu}^\beta, \quad (7)$$

$$\text{with } C_{i\mu, j\nu}^{\beta\gamma} = \delta_{\beta\gamma} J_{i\mu, j\nu} - \frac{1}{2} (g\mu_B)^2 D_{i\mu, j\nu}^{\beta\gamma}, \quad (8)$$

where  $\langle S_{i\mu}^\beta \rangle$  denotes the field-induced spin moment. In this section we assume the isotropic exchange coupling. Now, within the linear response approximation we get the following relations:

$$\langle S_{i\mu}^\beta \rangle = \phi(\omega) \left[ h_{i\mu}^\beta e^{-i\omega t} + 2 \sum_{j, \nu, \gamma} C_{i\mu, j\nu}^{\beta\gamma} (\langle S_{j\nu}^\gamma \rangle - \alpha \langle S_{i\mu}^\gamma \rangle) \right]. \quad (9)$$

Here  $\phi(\omega)$  represents the dynamical susceptibility of a free  $S=\frac{1}{2}$  spin and is expressed as

$$\phi(\omega) = \delta_{\omega,0} \frac{S(S+1)}{3k_B T} = \delta_{\omega,0} \frac{1}{4k_B T}. \quad (10)$$

Performing the spatial Fourier transformation of eq.(9) we obtain

$$\sum_{\nu\gamma} M_{\mu\nu}^{\beta\gamma}(\mathbf{q}, \alpha) \langle S_\nu^\gamma(\mathbf{q}) \rangle = \phi(\omega) h_\mu^\beta(\mathbf{q}), \quad (11)$$

where  $\langle S_\nu^\gamma(\mathbf{q}) \rangle$  and  $h_\mu^\beta(\mathbf{q})$  represent the Fourier transforms of  $\langle S_{i\nu}^\gamma \rangle$  and  $h_{i\mu}^\beta$ , respectively, and  $M_{\mu\nu}^{\beta\gamma}(\mathbf{q}, \omega)$  is defined by

$$M_{\mu\nu}^{\beta\gamma}(\mathbf{q}, \omega) = \delta_{\beta\gamma} \delta_{\mu\nu} - 2\phi(\omega) \left[ C_{\mu\nu}^{\beta\gamma}(\mathbf{q}) - \delta_{\mu\nu} \alpha \{ C_{11}^{\beta\gamma}(0) + C_{12}^{\beta\gamma}(0) \} \right] \quad (12)$$

with

$$C_{\mu\nu}^{\beta\gamma}(\mathbf{q}) = \delta_{\beta\gamma} J_{\mu\nu}(\mathbf{q}) - \frac{1}{2} (g\mu_B)^2 D_{\mu\nu}^{\beta\gamma}(\mathbf{q}). \quad (13)$$

By solving eq.(11) with respect to  $\langle S_\mu^\beta(\mathbf{q}) \rangle$  we obtain

$$\langle S_\mu^\beta(\mathbf{q}) \rangle = \sum_{\nu\gamma} \chi_{\mu\nu}^{\beta\gamma}(\mathbf{q}, \omega) h_\nu^\gamma(\mathbf{q}) e^{-i\omega t} \quad (14)$$

where the dynamical susceptibility  $\chi_{\mu\nu}^{\beta\gamma}(\mathbf{q}, \omega)$  is expressed by

$$\chi_{\mu\nu}^{\beta\gamma}(\mathbf{q}, \omega) = \frac{\Delta_{\nu\gamma, \mu\beta}(\mathbf{q}, \omega)}{D(\mathbf{q}, \omega)} \phi(\omega). \quad (15)$$

Here  $D(\mathbf{q}, \omega)$  denotes the determinant of a  $6 \times 6$  matrix  $\tilde{M}$  whose elements are given by  $M_{\mu\nu}^{\beta\gamma}(\mathbf{q}, \omega)$ , and  $\Delta_{\nu\gamma, \mu\beta}(\mathbf{q}, \omega)$  represents the  $(\nu\gamma, \mu\beta)$ -cofactor of  $\tilde{M}$ .

To determine the correlation parameter  $\alpha$  we impose the following self-consistency condition:

$$\langle \{S_{i1}^x, S_{i1}^x\} \rangle = \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \coth\left(\frac{\omega}{2k_B T}\right) \text{Im} \chi_{11}^{xx}(\mathbf{q}, \omega + i\eta) \quad (\eta \rightarrow +0). \quad (16)$$

Here  $\langle \{S_{i1}^x, S_{i1}^x\} \rangle = 2 \langle S_{i1}^x S_{i1}^x \rangle$  represents the on-site spin correlation calculated by using the effective single-ion Hamiltonian, and we have  $\langle \{S_{i1}^x, S_{i1}^x\} \rangle = \frac{2}{3} S(S+1)$ . By using eqs.(10) and (15) the above self-consistency equation is rewritten as

$$\frac{1}{N} \sum_{\mathbf{q}} \frac{\Delta_{1x, 1x}(\mathbf{q}, 0)}{D(\mathbf{q}, 0)} = 1. \quad (17)$$

From this equation  $\alpha$  is determined as a function of temperature.

The implication of the above self-consistency condition is that the on-site spin correlation calculated from the dynamical susceptibility (the right-hand side of eq.(16)) should be equal to that calculated from the effective single-ion Hamiltonian. It is noted that the self-consistency equation constructed by using  $\chi_{22}^{xx}(\mathbf{q}, \omega)$  gives the same results. It is also noted that the self-consistency equation constructed by using  $\chi_{11}^{yy}(\mathbf{q}, \omega)$  or  $\chi_{11}^{zz}(\mathbf{q}, \omega)$  gives almost the same results.

The uniform susceptibility per unit cell  $\chi_u^\beta$  along each crystal axis is given by

$$\chi_u^\beta = \chi_{11}^{\beta\beta}(0, 0) + \chi_{12}^{\beta\beta}(0, 0) + \chi_{21}^{\beta\beta}(0, 0) + \chi_{22}^{\beta\beta}(0, 0), \quad (18)$$

and the staggered susceptibility per unit cell  $\chi_s^\beta$ , which is the susceptibility for alternating field  $h_1^\beta(\mathbf{q} = 0) = -h_2^\beta(\mathbf{q} = 0)$ , is given by

$$\chi_s^\beta = \chi_{11}^{\beta\beta}(0, 0) - \chi_{12}^{\beta\beta}(0, 0) - \chi_{21}^{\beta\beta}(0, 0) + \chi_{22}^{\beta\beta}(0, 0). \quad (19)$$

Since the Fourier transform  $D_{\mu\nu}^{\beta\gamma}(\mathbf{q})$  is almost diagonal with respect to  $\beta$  and  $\gamma$  for  $\mathbf{q} = 0$ ,  $\chi_u^\beta$  and  $\chi_s^\beta$  are well approximated by

$$\chi_u^\beta = \frac{2\phi(0)}{1 - 2\phi(0) [C_{11}^{\beta\beta}(0) + C_{12}^{\beta\beta}(0)] + 2\phi(0)\alpha [C_{11}^{\beta\beta}(0) + C_{12}^{\beta\beta}(0)]}, \quad (20)$$

$$\chi_s^\beta = \frac{2\phi(0)}{1 - 2\phi(0) [C_{11}^{\beta\beta}(0) - C_{12}^{\beta\beta}(0)] + 2\phi(0)\alpha [C_{11}^{\beta\beta}(0) + C_{12}^{\beta\beta}(0)]}. \quad (21)$$

At the Néel temperature  $T_N$  the staggered susceptibility  $\chi_s^x$  diverges, that is

$$1 - 2\phi(0)[C_{11}^{xx}(0) - C_{12}^{xx}(0)] + 2\phi(0)\alpha[C_{11}^{xx}(0) + C_{12}^{xx}(0)] = 0. \quad (22)$$

The Néel temperature and the correlation parameter  $\alpha_N$  at  $T_N$  are determined simultaneously from eqs.(17) and (22).

We have performed actual calculations by fixing the value of  $J_c$  at  $-0.0087$  K which has been determined in the previous section. We have found that the observed uniform susceptibilities above  $0.5$  K are well reproduced if we assume  $J_{11}(0)=0.9$  K or  $J_a+J_b=0.45$  K. At lower temperatures, however, the susceptibility depends on the respective value of  $J_a$  and  $J_b$ , and we have found that the observed susceptibility are well reproduced both above and below  $0.5$  K if we assume  $J_a=0.45$  K and  $J_b=0$  K. For these parameters the Néel temperature is determined to be  $T_N=0.15$  K, which agrees with the observation.<sup>4</sup> In Fig. 2 we show the susceptibility  $\chi_u^y$  calculated by the DCEFA together with the experimental results.

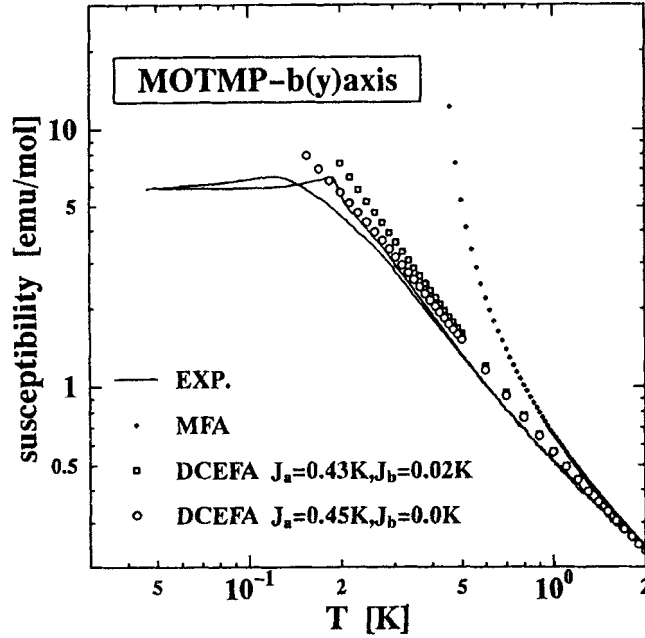


FIGURE 2 The temperature dependence of the uniform magnetic susceptibility along the  $y$ -axis of MOTMP. The full curve: experimental data,<sup>4</sup> the open circles: DCEFA ( $J_a=0.45$  K,  $J_b=0.0$  K), the open squares: DCEFA ( $J_a=0.43$  K,  $J_b=0.02$  K), the dots: MFA ( $J_a=0.45$  K,  $J_b=0.0$  K).

The correlation parameter  $\alpha$  determined self-consistently is a monotonously decreasing function of temperature:  $\alpha=0.758$  at  $T_N=0.15$  K and  $\alpha=0.06$  at  $T=4$  K. As



mentioned before the DCEFA is reduced to the MFA if we put  $\alpha=0$ . In Fig. 2 we show also the results obtained by the MFA. As expected, the MFA gives  $T_N$  much higher than the observation.

The anisotropy of the intra-sublattice exchange coupling estimated in the previous section is  $J_{11}^x(0) - J_{11}^y(0) = -0.0026$  K, and thus the ratio  $|J_{11}^x(0) - J_{11}^y(0)|/J_{11}(0) = 0.0026/0.9 = 0.003$  is quite small. Therefore, the results of the present section are almost unchanged even if we take into account the anisotropy of  $J_{11}(0)$ .

If we neglect the DD interaction completely, the observed susceptibility and the Néel temperature can be reproduced well by using  $J_b = 0.01$  K. As shown here, however, the value of  $J_b$  is estimated to be 0.0 K if we take account of the DD interaction. This fact clearly indicates that we must be careful in estimating the strength of weak interchain exchange coupling, namely we have to take into consideration the DD interaction quantitatively.

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