Ground-state crossover in the quasi-two-dimensional classical Heisenberg model with dipolar-type interaction

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We have studied the quasi-two-dimensional Heisenberg model with nearest-neighbor dipolar interaction by using a Monte Carlo simulation. By varying the dipolar coupling magnitude D or the anisotropy ratio λ of the interlayer exchange interaction to the intralayer one, crossovers between different ground-states were observed. The phase boundaries in the (D, λ) space were calculated from the ground-state energies, which are in agreement with the Monte Carlo results. Our Monte Carlo results were compared with experimental data on $(C_nH_{2n+1}NH_3)_2MnCl_4$.

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I. INTRODUCTION

Molecule-based magnets, such as $(C_nH_{2n+1}NH_3)_2MnCl_4$ (CnMn) (Ref. 1) and $Cu_2(OH)_3(C_mH_{2m+1}CO_2)$,[2] have been regarded as representative quasi-two-dimensional (quasi-2D) Heisenberg magnets. They consist of alternating magnetic and nonmagnetic layers. The nonmagnetic layers are constituted of organic chains, whose length can be easily controlled. Thus, the interlayer separation and the interlayer magnetic interaction between the magnetic layers can be systematically controlled. Because the two-dimensional Heisenberg model cannot have a long-range order at any finite temperature, it can be expected that the magnetic ordering temperature will decrease down to zero with increasing interlayer separation, i.e., with increasing organic chain length. This expectation was confirmed for short chain lengths[3] but failed for long chain lengths:[1,2] Even for very large interlayer separations, finite temperature magnetic orderings were observed. An Ising type of anisotropy can be an origin of the magnetic ordering, since a magnetic ordering is allowed in the quasi-2D Ising model at a critical temperature close to that of the 2D Ising model even for an infinitesimal interlayer interaction, as is well known. Renormalization-group[4] and spin-wave[5] approaches for the quasi-2D Heisenberg model with Ising type of anisotropy have also been reported. On the other hand, a dipolar interaction between the effective dipoles of correlated spin clusters was suggested as an origin of the magnetic ordering,[6,7] since a high-temperature ferromagnetism observed for a very large interlayer separation was not easily understood with only an anisotropy. A dipolar effect on the quasi-2D Heisenberg model deserves a systematic study in order to understand the magnetic ordering at a sizable temperature even with a very small interlayer exchange interaction.

The model Hamiltonian for our quasi-2D system is described by

$$H=-\sum_{i\neq j} J_{ij}S_i\cdot S_j+D\sum_{i\neq j}\,[S_i\cdot S_j-3(S_i\cdot\hat{r})(S_j\cdot\hat{r})]f(r). \tag{1}$$

The first term describes a nearest-neighbor exchange interaction, where $J_{ij}=J$ when S_i and S_j belong to the same layer, and $J_{ij}=\lambda J$ otherwise. When $D=0$, Eq. (1) describes a stack of noninteracting 2D Heisenberg layers with $\lambda=0$, and describes a 3D Heisenberg system with $\lambda=1$. The second term indicates a dipolar interaction, where $\vec{r}$ denotes the vector connecting the two spins S_i and S_j. $D(\geqslant 0)$ is the magnitude of the dipolar coupling and λ is the ratio of the interlayer exchange interaction to the intralayer one. When considering only the short-range interactions, the function $f(r)$ can be restricted to the nearest-neighboring pairs $[f(1)=1]$.

The dipolar models $(J=0)$ have long been studied, and magnetic ordering at finite temperatures was rigorously proven for the 3D short-range dipolar model.[8] A spin-wave theory also predicts a magnetic ordering for the 3D long-range dipolar model.[9] The critical exponents of the 3D short-range dipolar model were reported to be numerically close to the 3D Heisenberg values.[10] The ground state of long-range dipolar models was predicted to be antiferromagnetic for the simple cubic lattice and ferromagnetic for a bcc or fcc lattice.[11] The dipolar ferromagnetism predicted was observed in a recent experiment on a fcc rare-earth salt, $Cs_2NaR(NO_2)_6$.[12] The 3D Heisenberg model $(\lambda=1)$ with long-range dipolar interaction has been extensively studied. For $J<0$, the critical exponents are not affected by the dipolar interaction.[13] For $J>0$, the critical exponents were found to be numerically very close to the 3D Heisenberg values.[14] The linear spin-wave theory was employed for the 2D and quasi-2D Heisenberg models with dipolar interaction,[15,16] where the dipolar interaction was shown to act as an anisotropy enabling a long-range order even in a pure 2D system. The effect of anisotropic exchange interactions has also been considered, especially in the extreme case of uniaxial (Ising-like) dipoles. A crossover behavior from the dipolar to the Ising magnet was expected at $(T/T_c-1)\sim(D/J)^{1/\phi}$, where T_c is the Ising critical temperature and ϕ is the crossover exponent.[17] In a 2D uniaxial long-range dipolar model with $J>0$, the ground state was found to be a striped phase.[18]

In this work, ground-state crossover in systems described by Hamiltonian (1) have been investigated. Phase boundaries were calculated from the ground-state energy, and Monte Carlo simulations were employed to study crossovers be-

TABLE I. Ground states (g.s.) in the Heisenberg model.

J	λ	g.s.	E
+	+	F	$-(2+\lambda)J=-(2+\lvert\lambda\rvert)\lvert J\rvert$
-	+	AF	$(2+\lambda)J=-(2+\lvert\lambda\rvert)\lvert J\rvert$
+	-	AF_L	$-(2-\lambda)J=-(2+\lvert\lambda\rvert)\lvert J\rvert$
-	-	DP_l	$(2-\lambda)J=-(2+\lvert\lambda\rvert)\lvert J\rvert$

tween the ground states. Finally, the results were compared with experiments on $(C_nH_{2n+1}NH_3)_2MnCl_4$ (CnMn).

II. GROUND STATE

The 3D Heisenberg model on a simple cubic lattice can have four different spin configurations in the ground state, with an energy $-3|J|$, depending on the signs of J and λ ($|J|=|\lambda|=1$). Also, the quasi-2D or quasi-1D Heisenberg models can have the same spin configurations as those in the 3D case. The spin configurations and ground-state energies are shown in Table I, in which $|\lambda|=1$, $|\lambda|<1$, and $|\lambda|>1$ correspond to the 3D, the quasi-2D, and the quasi-1D cases, respectively. In the ferromagnetic (F) and antiferromagnetic (AF) phases, all the nearest-neighbor spins are aligned parallel and antiparallel, respectively. In the layered antiferromagnetic (AF_L) phase, neighboring spins are aligned parallel in the same layer and antiparallel between the adjacent layers. The DP_l phase is one of the dipolar ground states. The spin configurations of the dipolar ground state are defined by[8,10]

$$\vec{S}(h,k,l)=(-1)^k(-1)^l S_x\hat{x}+(-1)^h(-1)^l S_y\hat{y}+(-1)^h(-1)^k S_z\hat{z}, \quad |\vec{S}_i|=1, \tag{2}$$

with the ground-state energy of $-4D$. For convenience, it was assumed that the spin axes coincide with the lattice axes: $\hat{x}=\hat{h}$, $\hat{y}=\hat{k}$, and $\hat{z}=\hat{l}$. The spin configuration of the DP_l phase corresponds to Eq. (2) with $S_x=S_y=0$ and $S_z=1$, where the spins are aligned parallel in the same chain and antiparallel between the adjacent chains.

In the 3D isotropic Heisenberg model with a short-range dipolar interaction, for which $\lambda=1$, it is expected that the ground state is (anti)ferromagnetic with $D\lll|J|$ or dipolar with $D\ggg|J|$. Thus, a crossover of the ground states is expected at $D=D_c$. Table II shows the ground-state energy in each ground-state. At $D=D_c$, the ground-state energy in the dipolar ground state should be equal to that in the (anti)ferromagnetic ground state, $J-4D_c=-3|J|$, so that

$$D_c=(J+3|J|)/4. \tag{3}$$

Then, $D_c/J=1$ for $J>0$, or $D_c/J=1/2$ for $J<0$. In the quasi-2D Heisenberg model with a short-range dipolar interaction, the ground states are quite complicated depending on the values of λ and D, and the sign of J. Table III shows the ground-state energies in several spin configurations, where $\lambda=1$ corresponds to the ground-state energy in the 3D isotropic case which is shown in Table II. Our goal is to find the phase boundaries in the (λ, D/J) space starting from the limiting cases in Tables I and II. The spin configuration with the lowest energy is assigned to be a ground-state, and the ground-state energies are the same at the phase boundary. The energy in the dipolar ground-state DP has a minimum at $S_z=0$ for ($\lambda<1$, $D/J>0$) and ($\lambda>1$, $D/J<0$), or one at $S_z^2=1$ for ($\lambda<1$, $D/J<0$) and ($\lambda>1$, $D/J>0$). At the phase boundary between (A)F and DP_l, $-4D+(2-\lambda)J=-(2+\lambda)|J|$, so that

$$\lambda=\frac{2(J+|J|)-4D}{J-|J|}. \tag{4}$$

For $J<0$, $\lambda=2D/J$, where a crossover from AF to DP_l occurs. For $J>0$, the phase boundary is independent of λ and is equal to $D/J=1$. At the phase boundary between (A)F and AF_L, $-2D-J(2-\lambda)=-|J|(2+\lambda)$, and thus

$$\lambda=\frac{2(J-|J|)+2D}{J+|J|}. \tag{5}$$

For $J>0$, $\lambda=D/J$, at which a crossover from F to AF_L occurs. For $J<0$, the phase boundary between AF and AF_L appears to be at $D/J=-2$. However, neither AF nor AF_L are a ground-state in the region, where the ground state is DP_l. In a similar manner, we can acquire a phase diagram with $|\lambda|<2$, as shown in Fig. 1.

TABLE II. Ground state in the 3D isotropic Heisenberg model with a short-range dipolar interaction, where $\lambda=1$ and $D_c=(J+3|J|)/4$. E_{ex} and E_d are the exchange energy and the dipolar energy, respectively.

D	g.s.	E_{ex}	E_d	E
$D<D_c$	F	$-3\lvert J\rvert$	0	$-3\lvert J\rvert$
$D<D_c$	AF	$-3\lvert J\rvert$	0	$-3\lvert J\rvert$
$D>D_c$	DP	J	$-4D$	$-4D+J$

TABLE III. Ground states and energies in the quasi-2D Heisenberg model with a short-range dipolar interaction.

g.s.	E
F	$-(2+\lambda)\lvert J\rvert$
AF	$-(2+\lambda)\lvert J\rvert$
AF_L	$-2D-(2-\lambda)J$
DP	$-4D+[\lambda+2(1-\lambda)S_z^2]J$
DP_l	$-4D+(2-\lambda)J$
DP_{hk}	$-4D+\lambda J$

III. MONTE CARLO SIMULATION

Monte Carlo simulations were employed for the classical Heisenberg spins, $|s|=1$, placed on a L^3 simple cubic lattice with periodic boundary conditions (if not specified, $L=10$). The conventional Metropolis algorithm was employed to update the spin configurations. All the measurements were carried out by decreasing the temperature from

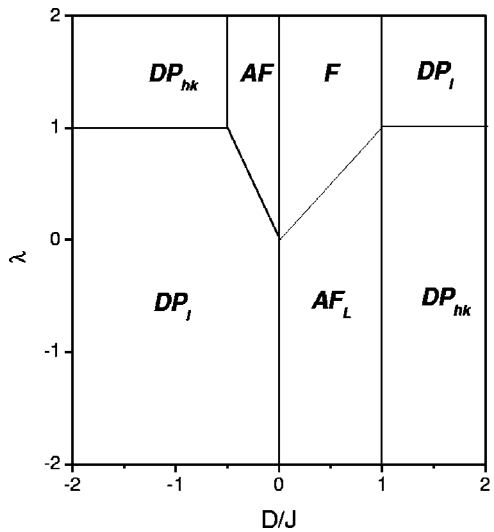

FIG. 1. Phase diagram in the quasi-2D Heisenberg model with a short-range dipolar interaction, calculated from the ground-state energies.

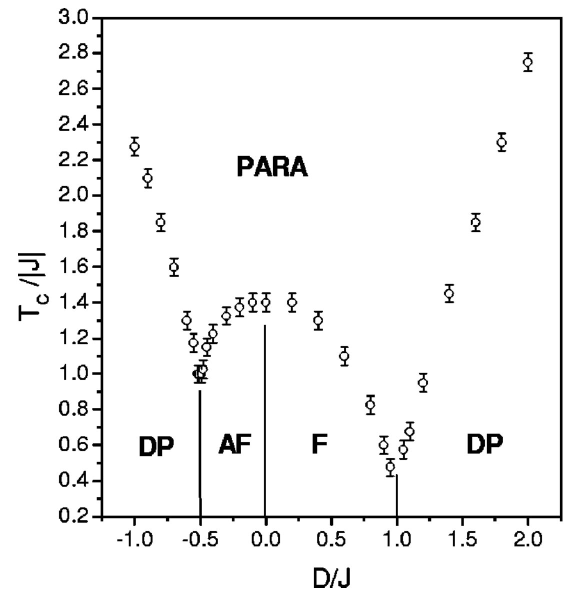

FIG. 2. $T_c/|J|$ vs D/J for $\lambda=1$. Ground-state crossovers occur at $D_c/J=1$ $(J=+1)$ and 1/2 $(J=-1)$. PARA stands for a paramagnetic phase.

an infinite temperature. 5000–20 000 MCS's (Monte Carlo steps) were used for thermal equilibration, and 30 000–120 000 averages for a physical quantity were taken. For a small λ, the system size affects the effective dimensionality of the system.[19] In a quasi-2D system with a small size, the growth of the correlation length in a layer is much faster than that along the anisotropy axis. Then, the spins in the same layer may act as a single spin and the system as a 1D one. In order to conserve the 3D nature of the system, λ was confined to the range $0.1\leqslant\lambda\leqslant1$.

The energy and the specific heat were measured following common definitions:

$$E=\frac{1}{L^3}\langle H\rangle, \tag{6}$$

$$C_v=\frac{1}{T^2}\left[\frac{1}{L^3}\langle H^2\rangle-L^3E^2\right]. \tag{7}$$

The exchange energy E_{ex} and the dipolar energy E_d were also measured independently, in the same manner. The temperature of maximum specific heat was adopted as an effective critical temperature. The effective critical temperature of the 3D short-range dipolar model was measured to be $T_c/D=1.86(0.02)$, which is compatible with a previous report.[10] The (staggered) magnetization M_f and the (staggered) susceptibility χ_f were also measured, which are defined by

$$M_f=\frac{1}{L^3}\left\langle\left[\left(\sum f_i\vec{S}_i\right)^2\right]^{1/2}\right\rangle, \tag{8}$$

$$\chi_f=\frac{1}{T}\left[\frac{1}{L^3}\left\langle\left(\sum f_i\vec{S}_i\right)^2\right\rangle-L^3M^2\right], \tag{9}$$

where the Boltzmann constant is set to unity and f_i is a weighting function of the lattice site coordinate. Let the lattice site coordinates be expressed as $\vec{S}_i=\vec{S}(h,k,l)$ and $f_i=f(h,k,l)$. Then, if $f(h,k,l)=1$, $M_f=M$ and $\chi_f=\chi$ are the magnetization and the susceptibility for the ferromagnetic (F) ground-state. If $f(h,k,l)=(-1)^{h+k+l}$, $M_f=M_S$ and $\chi_f=\chi_S$ are the staggered magnetization and susceptibility for the antiferromagnetic (AF) ground-state, respectively. If $f(h,k,l)=(-1)^{h+k}+(-1)^{k+l}+(-1)^{h+l}$,[10] $M_f=M_D$ and $\chi_f=\chi_D$ are the staggered magnetization and susceptibility for the DP ground-state, respectively. If $f(h,k,l)=(-1)^l$, $M_f=M_L$ and $\chi_f=\chi_L$ are the staggered magnetization and susceptibility for the AF$_L$ ground-state, respectively.

IV. RESULTS AND DISCUSSION

Figure 2 shows the critical temperatures as a function of D/J with $\lambda=1$, which shows a dip at $D_c/J=1$ $(J=+1)$ and at $D_c/J=1/2$ $(J=-1)$, in agreement with Eq. (3). When $0\leqslant D<D_c$, the ground-state is a 3D (anti)ferromagnet and above D_c/J the ground-state is a dipolar one, which was confirmed by measuring the (staggered) magnetization M, M_S, and M_D. At D_c/J, a crossover from the (anti)ferromagnetic to the dipolar ground-state occurs, which is accompanied by a sharp dip in $T_c/|J|$. Except for the absolute value of D_c/J, the overall crossover behaviors are similar regardless of the sign of J.

Figure 3 displays the critical temperatures as a function of λ with $J=-1$ and $D=0.2$, showing a crossover from AF to DP$_l$ at λ_c. The crossover behavior is very similar to that in the 3D case, where the critical temperature, shows a dip at the crossover. With λ decreasing below λ_c, the critical temperature increases, in contrast to the expectation that the critical temperature should decrease in the quasi-2D Heisenberg model. When $\lambda\geqslant\lambda_c$, the ground-state is AF. Just below

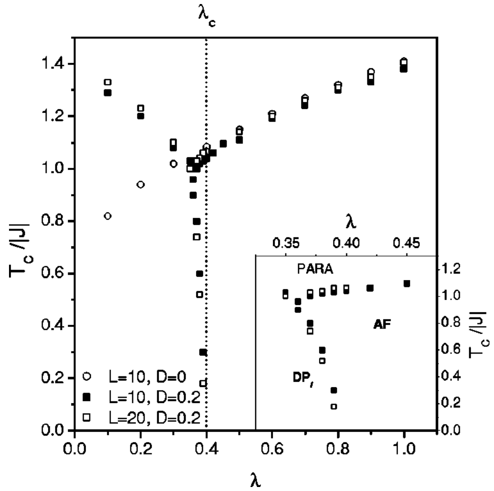

FIG. 3. $T_c(\lambda)/|J|$ vs λ for $J=-1$ and $D=0.2$. The open and solid squares correspond to $L=20$ and $L=10$, respectively. The open circle corresponds to $D=0$. In the inset the crossover region is shown, where two successive phase transitions occur with decreasing temperature: a second-order transition from PARA to AF phase and a first-order one from AF to DP_l phase.

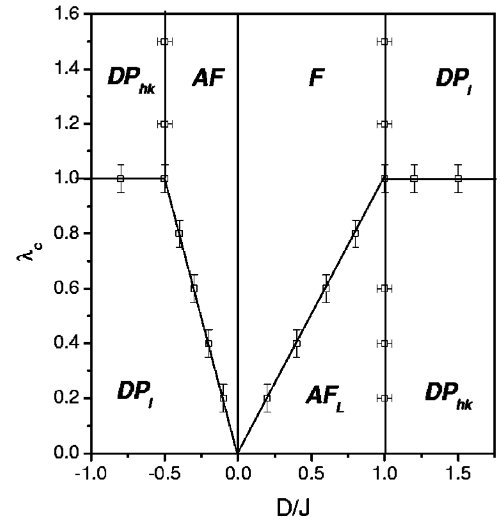

FIG. 4. Ground-state phase diagram in the $(\lambda, D/J)$ space, measured by the Monte Carlo simulations with $J=+1$ and -1 (symbols), resulting in good agreement with that (solid lines) calculated from the ground-state energy. Crossover regions are included in the error bars.

λ_c, a crossover region exists, where two phase transitions occur as shown in the inset: with decreasing temperature from the paramagnetic phase, a second-order antiferromagnetic transition occurs and then a first-order transition from AF to DP_l occurs near the zero temperature. With further decrease in λ, the first-order transition temperature increases and the first-order nature weakens. It then becomes equal to the antiferromagnetic transition temperature, where the antiferromagnetic transition is smeared out, and the system undergoes a single second-order transition from a paramagnetic state to a dipolar ground-state.

While the overall behavior is similar to the 3D case, the quasi-2D Heisenberg model undergoes a crossover from AF to DP_l for $J<0$ or a crossover from F to AF_L for $J>0$. The crossover anisotropy λ_c is a function of D/J and is shown in Fig. 4: $\lambda_c=2D/J$ for $J=-1$ or $\lambda_c=D/J$ for $J=+1$, in agreement with Eqs. (4) and (5). In Fig. 4 is shown the phase diagram measured by Monte Carlo simulations, which is in good agreement with that calculated, shown in Fig. 1.

While we have assumed that the dipolar interaction is isotropic in spite of the anisotropic exchange interaction, a structural anisotropy as that in CnMn leads to an anisotropic dipolar interaction as well as an anisotropic exchange one. Then, the magnitude of the dipolar coupling D in the model Hamiltonian [Eq. (1)] should be redefined: $D=D_o$ when S_i and S_j belong to the same layer and $D=\delta D_o$ $(0\leqslant\delta\leqslant 1)$ otherwise. The energy in the dipolar ground-state is

$$E=-D_o[3+\delta-(1-\delta)S_z^2]+J[\lambda+2(1-\lambda)S_z^2], \quad (10)$$

where the dipolar energy (first term) was modified, but not the exchange energy (second term), in comparison to Table III. The energy does not change in different ground-states.

Assuming $0\leqslant\lambda\leqslant 1$, $J<0$, and $\delta\geqslant\lambda$, Eq. (10), has a minimum at $S_z=1$ (DP_l phase) or at $S_z=0$ (DP_{hk} phase). At the phase boundary between AF and DP_l, $-2D_o(1+\delta)+J(2-\lambda)=J(2+\lambda)$, so that

$$-\frac{D_o}{J}=\frac{\lambda}{1+\delta}. \quad (11)$$

Along the anisotropy axis, both the dipolar and the exchange interactions are functions of the interlayer distance, and the dipolar anisotropy δ can be reduced to a function of the exchange anisotropy λ. Assuming $\delta=\lambda^{\alpha}$ $(0\leqslant\alpha\leqslant 1)$, Eq. (11) becomes

$$-\frac{J}{D_o}\lambda_c=1+\lambda_c^{\alpha}. \quad (12)$$

Because λ_c is a monotonic function of α, the maximum reduction of λ_c is

$$\frac{\lambda_c(\alpha=1)}{\lambda_c(\alpha=0)}=\frac{1}{2(1-D_o/|J|)}. \quad (13)$$

The reduction in λ_c due to the dipolar anisotropy is at best 1/2 when $D_o/|J|\rightarrow 0$ and $\alpha\rightarrow 1$.

Figure 5 shows $T_c/|J|$ measured in CnMn, which is a well-known quasi-2D Heisenberg antiferromagnet with spin canting, as a function of the carbon number n. In the case of C3Mn, the lattice constants are about 7 Å in the layer and about 26 Å between the layers.[20] The dipolar interaction between spins is much smaller in different layers than in the same layer, by about 4^3 times, i.e., $\delta\sim 1/64$. The lattice anisotropy increases further with increasing carbon number. The approximation of dipolar interaction as being short ranged is believed to be fairly suitable at least for the dipolar interaction between different layers.

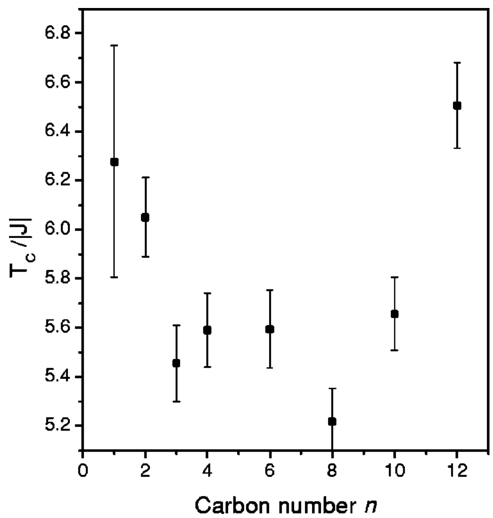

FIG. 5. $T_c/|J|$ measured in $(C_nH_{2n+1}NH_3)_2MnCl_4$.

The ordering temperature and the exchange energy in CnMn were reported in our previous work.[1] The ordering temperature was determined from the weak magnetic moment originating from the canted spin component. The exchange energy $|J|$ was calculated from the temperature of maximum susceptibility, $k_BT_{\chi max}=1.2625JS(S+1)$ with $S=5/2$. The temperature of maximum susceptibility was measured again within 1 K in this work. The exchange energy was calculated to be about 7 K. The usual estimate of the dipolar energy scale is $(\mu_o/4\pi)g^2\mu^2/a^3$, where μ_o, g, μ, and a are the magnetic permeability, g factor, the magnetic moment, and the lattice constant, respectively.[21] The dipolar energy D_o was calculated to be about 0.5 K, so that $D_o/|J| \sim 0.07$. Our simulation was limited to $0.1 \leqslant D_o/|J| \leqslant 0.5$, corresponding to $0.1 \leqslant \lambda_c \leqslant 1$, in order to conserve the 3D nature of our model, as discussed in Sec. III. $D_o/|J|$ calculated in CnMn is not far from our simulation range.

With increasing carbon number n, the interlayer separation increases, whereas the interlayer exchange interaction decreases. For $n<8$, $T_c/|J|$ decreases with increasing n, as expected for the quasi-2D Heisenberg model. However, for $n>8$, $T_c/|J|$ increases again with increasing n. $T_c/|J|$ shows a typical behavior of the ground-state crossover in the presence of dipolar interaction. Unfortunately, the magnetic ground-states for the long-chain compounds are not known and the dipolar ground-state also appears antiferromagnetic. Nevertheless, it appears that the turning up of $T_c/|J|$ observed with decreasing interlayer exchange interaction cannot be understood without a crossover from the AF to the DP_l ground-state, since $T_c/|J|$ of the quasi-2D Heisenberg model decreases monotonously with decreasing λ, even in the presence of an additional anisotropy.[4,5] A quasi-2D Heisenberg magnet with $\lambda<\lambda_c$ is believed to provide a good example of a dipolar magnet.

In summary, the quasi-2D classical Heisenberg model with a short-range dipolar interaction was studied. Phase boundaries were calculated from the ground-state energy, which was confirmed by Monte Carlo simulations, showing that the ground-state crossover appears as a sharp dip of $T_c/|J|$. The observed turning up of $T_c/|J|$ in $(C_nH_{2n+1}NH_3)_2MnCl_4$ with increasing interlayer separation is in contrast to the expectation that the critical temperature should decrease to zero with decreasing interlayer interaction in the quasi-2D Heisenberg model. The turning up is believed to be due to a crossover from the antiferromagentic to the dipolar ground-states.

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