Orientational phase transition in a two-dimensional four-sublattice antiferromagnet $(NH_3)_2(CH_2)_3MnCl_4$ subjected to an inclined magnetic field

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Experimental and theoretical investigations were made of an orientational spin-flopping transition in a four-sublattice antiferromagnet $(NH_3)_2(CH_2)_3MnCl_4$. The antiferromagnetic resonance and differential magnetic susceptibility methods were used to construct the (H,φ) diagram of this ferromagnet when an external magnetic field was inclined in the easy magnetization plane. The coordinates of the critical points of a first-order phase transition were $H_{cr} = 25.0 \pm 0.1$ kOe and $\varphi_{cr} = 7 \pm 0.25°$. The experimental results were described well by a theory developed by the present authors.

1. INTRODUCTION

In a recent paper[1] we considered high-frequency and static properties of a quasitwo-dimensional four-sublattice antiferromagnet $(NH_3)_2(CH_2)_3MnCl_4$. We found that the characteristics of the magnetoresonance properties of this Heisenberg antiferromagnet are primarily due to the formation of a noncollinear four-sublattice structure of the magnetic moments in the ground state. The feasibility of a new type of noncollinear structure with a large noncollinearity angle in low-dimensional Heisenberg magnetic materials was predicted in Ref. 2 on the following basis. A weak coupling between the separate layers (or chains) forming the magnetic structure in such compounds means that the orientation of the antiferromagnetic vectors is determined by a competition between the interlayer (interchain) exchange and intralayer (intrachain) relativistic interactions. This competition establishes a noncollinear magnetic structure of relativistic origin, which we shall call a relativistic noncollinear structure (RNS). The strong noncollinearity of the structure arises because the strengths of the weak exchange and relativistic interactions are comparable. This mechanism of formation of noncollinear structures is manifested most clearly by low-dimensional magnetic materials for which the symmetry class of layers or chains is less than the symmetry class of the crystal as a whole.

A surprising manifestation of the special nature of the static properties of an antiferromagnet with an RNS has been the observation in $(NH_3)_2(CH_2)_3MnCl_4$ of a first-order phase transition[1] due to collective motion of magnetic sublattices belonging to neighboring magnetic layers coupled by a weak exchange interaction. In contrast to the spin-flopping transition in a two-sublattice antiferromagnet, which involves rotation of the antiferromagnetic vector by 90°, the phase transition in $(NH_3)_2(CH_2)_3MnCl_4$ involves rotation of one of the antiferromagnetic vectors of a layer through an angle close to 180°. The total antiferromagnetic vector rotates by 90°, i.e., although the external features of the transition remain as before (rotation of the principal antiferromagnetic vector of the system), the physics of this effect is essentially different.

We shall consider the nature and characteristics of such a spin-reorientation transition in inclined fields when an external field is oriented at an angle φ to the easy axis **b** of the system and is within the easy plane. In the case of three-dimensional uniaxial antiferromagnets the range of existence of a spin-flopping transition when **H** deviates from the easy axis in the easy plane lies within a narrow range of angles $\Delta\varphi \sim H_A/H_e$, where H_A and H_e are the anisotropy and exchange fields. This problem was first considered theoretically in Refs. 3–5 and experimental studies were reported for MnF_2 (Ref. 6), α-Fe_2O_3 (Ref. 7), $CuCl_2 \cdot 2H_2O$ (Refs. 8 and 9), and $GdAlO_3$ (Ref. 10). It is natural to expect low-dimensional antiferromagnets with an RNS to be affected much less by a deviation of the field from the easy axis of a crystal. However, no investigations of this topic have yet been made.

We shall report the results of an experimental and theoretical study of the spin-flopping transition in $(NH_3)_2(CH_2)_3MnCl_4$ subjected to inclined fields when **H** was oriented in the easy plane of the system.

2. EXPERIMENTAL RESULTS

We constructed the (H,φ) phase diagram of $(NH_3)_2(CH_2)_3MnCl_4$ in inclined fields using the results obtained by two methods: measurements of the differential susceptibility and antiferromagnetic resonance (AFMR). In both cases a magnetic field was created by a pulsed Helmholtz solenoid which was rotated in a horizontal plane. The angle of rotation of the solenoid was measured using a special vernier scale; this was done with an error of less than 0.5°.

The differential susceptibility measurements were carried out by an induction method. A sample was placed in a measuring coil which could be rotated in a vertical plane together with the sample. A compensation coil, connected in opposition to the measuring one, had a number of turns sufficient to reduce the signal proportional to $\partial H/\partial t$ by a factor of 10^4 in the absence of a sample. The quantity $\partial M/\partial H = \chi$ was determined as a function of the applied field using an S8-11 storage oscilloscope. A special circuit limiting the signal proportional to H made is possible to observe an extended part of the $\chi(H)$ dependence. There was a susceptibility peak in the region of the spin-flopping transition. For the **H** orientation along the easy axis the width of the peak was ≈ 100 kOe and an increase in the angle of inclination increased **H** and reduced χ_{max}. The peak could no longer be observed for $\varphi > 9°$. The dependence of H_m, deduced from

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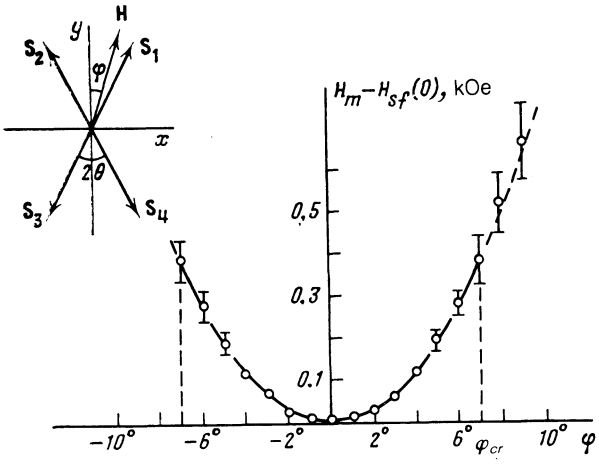

FIG. 1. Angular dependence of the field H_m corresponding to the susceptibility maximum $[\chi(H_m) = \chi_{max}]$. The spin-flopping field is $H_{sf}(0) = 24.6$ kOe. The continuous curve represents a first-order phase transition described by Eq. (11). The critical point of this phase transition has the coordinates $H_{cr} = 25 \pm 0.1$ kOe and $\varphi_{cr} = 7 \pm 0.25°$. The inset shows schematically the magnetic configuration of $(NH_3)_2(CH_2)_3MnCl_4$ and the geometry of inclination of an external magnetic field.

the position of χ_{max}, on the angle of inclination of the external field was determined (Fig. 1). Unfortunately, this dependence was insufficient to determine reliably an important characteristic of the (H,φ) diagram, the critical angle for a first-order phase transition.

As shown in Refs. 11 and 12, it was possible to determine φ_{cr} by the AFMR method. We therefore investigated the AFMR in inclined fields. The angular dependences of the AFMR were investigated by a method basically similar to that described in Ref. 1. A measuring cell in the form of a cavity resonator was used in conjunction with an rf spectrometer and a pulsed magnetic field. Microwave radiation was provided by klystrons operating in suitable ranges. A sample could be rotated in a vertical plane inside the resonator.

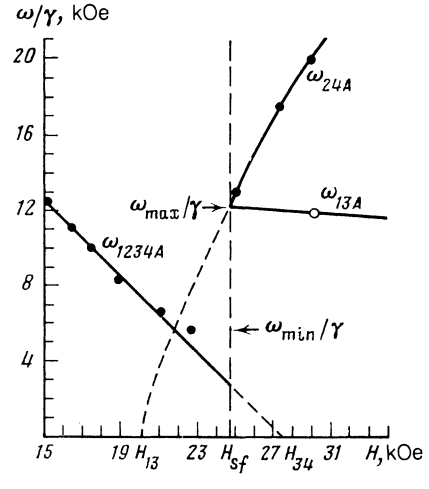

FIG. 2. Frequency-field phase diagram of an antiferromagnet in the vicinity of a spin-flopping transition in a field of the $\mathbf{H}\|\mathbf{b}$ ($\varphi = 0$) orientation.[1] The frequencies ω_{min} and ω_{max} define the interval within which the angular dependences of the antiferromagnetic resonance spectrum were investigated.

We investigated the part of the spectrum of acoustic AFMR modes in the range between ω_{max} and ω_{min} (Fig. 2). We assumed that a first-order phase transition should have a transparency window in the frequency-field dependence of AFMR modes in the investigated frequency range and that the angle of inclination of the external field corresponding to disappearance of the transparency window in the frequency-field dependence could be identified with the critical angle of a first-order phase transition.[11,12] The results of these experiments are plotted in Fig. 3. We can see from this figure that the investigated interval of frequency-field dependence of the AFMR modes was filled asymmetrically relative to ω_{max} and ω_{min}. An increase of the inclination of the external field to angles equal to φ_{cr} reduced the transparency window because of bending of the dependence $\omega_{13A}(\varphi)$ in the low-frequency range. In this situation the value of φ_{cr} was governed by the angle corresponding to $\omega_{13A}(\varphi) = \omega_{min}$ ($\omega_{min}/\gamma = 5.85$ kOe) and the value of twice this angle was $2\varphi_{cr} = 14 \pm 0.5°$. Comparing these results with the data deduced from the dependence $H_m(\varphi)$, we concluded that the dependence of $H_{sf} - H_{sf}(0)$ on the angle φ was identical with the dependence $H_m - H_{sf}(0)$ in Fig. 1 in the angular interval $\pm 7°$. The point with the coordinates $H = 25 \pm 0.1$ kOe and $\varphi = 7 \pm 0.25°$ was the critical point of the first-order phase transition under investigation. In the range $\varphi > \varphi_{cr}$ the magnetization disappeared abruptly although the susceptibility singularity was retained up to angles $\varphi = 9°$ (Fig. 1).

3. THEORY; COMPARISON WITH THE EXPERIMENTAL RESULTS

We shall recall briefly the crystal stucture of the investigated compound.[13] A crystallochemical cell of $(NH_3)_2(CH_2)_3MnCl_4$ contains four formula units and represents a practically quadratic layer of Mn^{2+} atoms in an octahedral environment of chlorine ions between which there are NH_3–$(CH_2)_3$–NH_3 organic molecules. The distance between the nearest Mn^{2+} ions in a layer is 5.2 Å and the distance between the atoms in different layers is 9.5 Å. The superexchange is weak because of the large distance between the spins in neighboring layers, and it is responsible for the quasi-two-dimensional magnetic behavior of the system. It is shown in Ref. 1 that all the experimental data on the field dependence of the AFMR frequencies and the observed spin-reorientation phase transitions can be explained

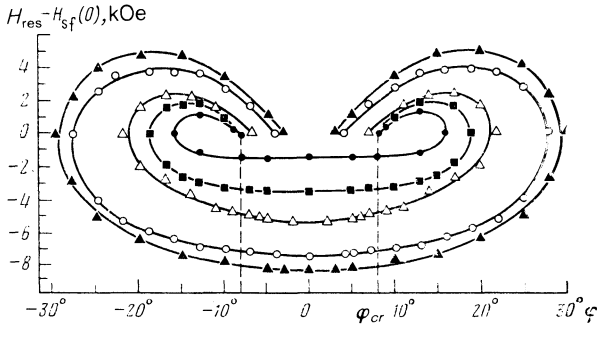

FIG. 3. Angular dependence of the resonance field of acoustic antiferromagnetic resonance modes in the vicinity of a spin-flopping transition in $(NH_3)_2(CH_2)_3MnCl_4$: ▲) $\omega/\gamma = 11.3$ kOe; ○) $\omega/\gamma = 10.55$ kOe; △) $\omega/\gamma = 8.24$ kOe; ■) $\omega/\gamma = 6.75$ kOe; ●) $\omega/\gamma = 5.85$ kOe.

in a nonconflicting manner by ascribing the space group *Pnma* (D_{2h}^{16}) to this compound.

In accordance with the layer structure of a crystal, we shall introduce the antiferromagnetic and ferromagnetic vectors for the first and second layers, respectively:

$$\begin{aligned} &\mathbf{L}_1=\mathbf{s}_1-\mathbf{s}_3\equiv 2s\mathbf{l}_1, \quad \mathbf{L}_2=\mathbf{s}_2-\mathbf{s}_4\equiv 2s\mathbf{l}_2, \\ &\mathbf{F}_1=\mathbf{s}_1+\mathbf{s}_3=2s\mathbf{m}_1, \quad \mathbf{F}_2=\mathbf{s}_2+\mathbf{s}_4\equiv 2s\mathbf{m}_2, \end{aligned} \tag{1}$$

where s is the atomic spin. Then, linear combinations of spin operators representing irreducible representations of the symmetry group of the paramagnetic phase are

$$\begin{aligned} &\mathbf{F}=\mathbf{F}_1+\mathbf{F}_2, \quad \mathbf{C}=\mathbf{L}_1+\mathbf{L}_2, \\ &\mathbf{G}=\mathbf{F}_1-\mathbf{F}_2, \quad \mathbf{A}=\mathbf{L}_1-\mathbf{L}_2. \end{aligned} \tag{2}$$

In the ground state of $(NH_3)_2(CH_2)_3MnCl_4$ the magnetic configuration is A_x, C_y, F_z with the principal antiferromagnetic vector **C** (representing the easy axis y).[1] The inset in Fig. 1 shows schematically the sublattice spin orientations in the neighboring layers s_1 and s_3; s_2 and s_4 are the orientations in a magnetic cell.

Applying the symmetry operations of the space group *Pnma* to the components of the vectors in Eq. (2), we can classify them in accordance with the irreducible representations of this group and write down an invariant expansion of the Hamiltonian of the system (the necessary expressions are given in Ref. 1). An analysis of a spin-reorientation transition in an inclined field will be made using a Hamiltonian in which the Dzyaloshinskiĭ interaction is ignored.

This interaction may have a considerable influence on the nature of a spin-reorientation transition and in some cases it may change it from a first- to a second-order phase transition.[14,15] Where the nature of the phase transition does not change, the Dzyaloshinskiĭ interaction may influence the sequence of phase transitions in a system.[16–18] For example, in $(NH_3)_2(CH_2)_3MnCl_4$ because of the Dzyaloshinskiĭ interaction of the $D_{2z}(L_{1z}F_{1y}+L_{2z}F_{2y})$ type it is found that when the field $\mathbf{H}\|\mathbf{y}$ reaches the spin-flopping value H_{sf}, the reorientation of the principal antiferromagnetic vector from the y axis does not occur strictly to the x axis but results in some deviation toward the z axis. Moreover, the same interaction operating in the range of fields $H_{sf}<H<H_c$ causes rotation of the pincipal antiferromagnetic vector from a direction parallel to the x axis toward the z axis. Our neglect of the Dzyaloshinskiĭ interaction in a theoretical analysis of a spin-flopping transition in an inclined magnetic field is justified by the following considerations. Firstly, the investigated crystal exhibits first-order phase transitions. Secondly, the transition fields H_{sf} and H_c are separated quite widely in the case of $(NH_3)_2(CH_2)_3MnCl_4$ ($H_{sf}=24.6$ kOe, $H_c=98$ kOe—see Ref. 1), so that an admixture of a layer component l_z can be regarded as small. Moreover, as shown below, this model describes satisfactorily the experimental results. The free energy system is

$$\begin{aligned} \Phi=&-{}^1/_2J_x(L_{1x}^2+L_{2x}^2)-{}^1/_2J_y(L_{1y}^2+L_{2y}^2)+{}^1/_2I_x(F_{1x}^2+F_{2x}^2) \\ &+{}^1/_2I_y(F_{1y}^2+F_{2y}^2)-\beta(L_{1x}L_{1y}-L_{2x}L_{2y}) \\ &-J_3\mathbf{L}_1\mathbf{L}_2+I_3\mathbf{F}_1\mathbf{F}_2-g\mu_B\mathbf{H}(\mathbf{F}_1+\mathbf{F}_2). \end{aligned} \tag{3}$$

The constants of the magnetic interactions in Eq. (3) can be described linearly in terms of the intrasublattice and intersublattice exchange interaction constants and in terms of the anisotropy. It is then found that J_α and I_α ($\alpha=x,y$) include the isotropic exchange interaction and part of the anisotropic interaction between spins within a layer; β is the intralayer anisotropic interaction of monoclinic symmetry; only the isotropic exchange from among the interlayer interactions is included in Eq. (3). We shall assume that the following inequalities apply:

$$J_\alpha,\ I_\alpha \gg J_3,\ I_3,\ |J_\alpha-J_{\alpha'}|,\ \beta, \tag{4}$$

which is in agreement with the experimental results on the investigated crystal.[1,19]

The free energy (3) describes a four-sublattice antiferromagnet with a structure in the form of a planar cross. Since the Dzyaloshinskii interaction is ignored, when an external magnetic field is inclined in the easy plane of the system, the magnetic moments of the sublattices are rotated in the same plane.

We shall introduce angles θ_1 and θ_2 describing deviation of the antiferromagnetic vectors of the layers of the easy axis in the xy plane. Then we find that ($i=1,2$)

$$\begin{aligned} &l_{ix}=l_i\sin\theta_i, \quad l_{iy}=l_i\cos\theta_i, \\ &m_{ix}=-m_i\cos\theta_i, \quad m_{iy}=m_i\sin\theta_i. \end{aligned} \tag{5}$$

The relationships in the system (5) allow for the orthogonality conditions: $l_1\cdot m_1=0$ and $l_2\cdot m_2=0$; substituting them in Eq. (3) and using the normalization condition $l_i^2=1-m_i^2$, we obtain the free energy of the system as a function of the variables m_i and θ_i.

Later we shall use the inequalities (4) which allow us to conclude that in the range of fields investigated we have $m\ll l$ and hence we can simplify greatly the free energy of the system. We shall ignore above all the terms $J_3m_i^2$ and $I_3m_i^2$. Then, minimizing Φ with respect to m_i we find that

$$\begin{aligned} m_1&=\frac{H_y\sin\theta_1-H_x\cos\theta_1}{2H_e-(H_{A1}-H_{A2}-H_A')\sin^2\theta_1+{}^1/_2H_{A3}\sin 2\theta_1}, \\ m_2&=\frac{H_y\sin\theta_2-H_x\cos\theta_2}{2H_e-(H_{A1}-H_{A2}-H_A')\sin^2\theta_2-{}^1/_2H_{A3}\sin 2\theta_2}. \end{aligned} \tag{6}$$

The following notation is introduced here $H_e\equiv s(J+1)\gamma^{-1}$ is the field of the intralayer exchange interaction; $H_{A1}-H_{A2}\equiv 2s(J_y-J_x)\gamma^{-1}$ is the anisotropy field stabilizing the antiferromagnetic vector of a layer along the y axis; $H_A'\equiv 2s(I_y-I_x)\gamma^{-1}$ is the anisotropy field stabilizing the ferromagnetic vector of a layer along the same axis; $H_{A3}\equiv 2s\cdot 2\beta\gamma^{-1}$ is the monoclinic anisotropy field; $\gamma=g\mu_B$.

Using Eq. (6), we can represent the free energy as a function of just two variables, θ_1 and θ_2. However, an analysis of this energy still presents considerable difficulties.

We shall bear in mind that the angular dependences of the denominators in Eq. (6) are proportional to the small ratio H_A/H_e. In other words, the anisotropy of the susceptibility of the layers can be allowed for using perturbation theory. Then, expanding the denominators of Eq. (6) as a series up to the terms H_A inclusive, we find that the free energy of the system becomes

$$\begin{aligned} \Phi=&-sJ_x\gamma^{-1}-\frac{H^2}{4H_e}-\frac{1}{2}\left(H_{A1}-H_{A2}-\frac{H_y^2-H_x^2}{2H_e}\right)\cos x\cos y \\ &-\frac{1}{2}H_{A3}\sin y\cos x-H_e'\cos y+\frac{H_xH_y}{2H_e}\sin x\cos y. \end{aligned} \tag{7}$$

Here, $H_e' \equiv 2sJ_3\gamma^{-1}$ is the field of the interlayer exchange interaction; $x \equiv \theta_1 + \theta_2$, $y \equiv \theta_1 - \theta_2$, and the angle x represents the deviation of the principal antiferromagnetic vector **C** of the system from the easy axis.

Equation (7) is derived relying heavily on the circumstance that $H_e' \gtrsim H_A$ for $(NH_3)_2(CH_2)_3MnCl_4$. Therefore, Eq. (7) and the results that follow from it do not allow us to analyze the limiting case of the ultraweak interlayer exchange $H_e' \ll H_A$.

The rest of the analysis of Eq. (7) is conventional. The equilibrium values of x and y are related by the conditions

$$\operatorname{tg} x = -\frac{H_x H_y}{H_e}\left\{ H_{A1}-H_{A2}-\frac{H_y^2-H_x^2}{2H_e}+H_{A3}\operatorname{tg} y\right\}^{-1}, \quad (8)$$

$$\operatorname{tg} y\left\{ H_{A1}-H_{A2}-\frac{H_y^2-H_x^2}{2H_e}-\frac{H_xH_y}{H_e}\operatorname{tg} x +2H_e'\operatorname{sign}(\cos x)(1+\operatorname{tg}^2 x)^{1/2}\right\}=H_{A3}. \quad (9)$$

The phase transition field is given by

$$2H_e(H_{A1}-H_{A2})=H_y^2-H_x^2 \quad (10)$$

or, introducing the angle φ of the deviation of the vector **H** from the easy axis, by

$$H_{sf}^2(\varphi)=H_{sf}^2(0)/\cos 2\varphi,\ H_{sf}^2(0)=2H_e(H_{A1}-H_{A2}). \quad (11)$$

Figure 1 shows the angular dependence of the spin-flopping transition field obtained experimentally for $(NH_3)_2(CH_2)_3MnCl_4$. The continuous curve represents the dependence (11) in the case when $\varphi \leqslant \varphi_{cr}$. We can see that the experimental results are described well by the expression obtained above. The critical phase transition angle is

$$\operatorname{tg} 2\varphi_{cr}=[(H_e'^2+H_{A3}^2)^{1/2}-H_e']/(H_{A1}-H_{A2}). \quad (12)$$

In the case of $(NH_3)_2(CH_2)_3MnCl_4$ we have $H_e' = 0.46$ kOe, $H_{A3} = 0.34$ kOe, $H_{A1} = 0.54$ kOe, and $H_{A2} = 0.09$ kOe from Ref. 1 and Eq. (12) yields $2\varphi_{cr} = 14°$, which is again in good agreement with the experimental results.

It therefore follows that the critical angle of the spin-flopping transition in a low-dimensional antiferromagnet with an RNS is fairly large and exceeds by two orders of magnitude the corresponding angle for the three-dimensional case.

We shall compare φ_{cr} with the noncollinearity angle θ of the system found in Ref. 1. We then have

$$\operatorname{tg} 2\theta=H_{A3}(H_{A1}-H_{A2}+2H_e')^{-1} \quad (13)$$

[we have reproduced Eq. (6) from Ref. 1 on the assumption that the Dzyaloshinskiĭ field is $H_{D1} = 0$]. Substituting the various parameters, we find that $2\theta = 14°$. The agreement between θ and φ_{cr} is accidental, although in the case of low-dimensional antiferromagnets with an RNS these parameters should be comparable.

4. CONCLUSIONS

Our investigation of an orientational spin-flopping phase transition in two-dimensional four-sublattice antiferromagnets with a relativistic noncollinear structure $(NH_3)_2(CH_2)_3MnCl_4$ subjected to inclined fields oriented in the easy plane of the system shows that, in contrast to the familiar case of three-dimensional collinear antiferromagnets, for which the range of existence of a first-order phase transition is governed by low values of the angles $\Delta\varphi \sim H_A/H_e$, many-sublattice antiferromagnets with a relativistic nonlinear structure are much less sensitive to deviations of the external field from the easy axis. The critical fields for these two cases differ by at least two orders of magnitude.

We found that a study of a spin-flopping transition in an antiferromagnet with a relativistic noncollinear structure in inclined fields provides a fairly direct and reliable method for the determination of the degree of noncollinearity of such structures.

It is worth noting that the high critical angle of the phase transition may be realized not only in low-dimensional antiferromagnets. For example, if $H_A \ll H_e' \ll H_e$, when a system is nearly three-dimensional, the critical angle of Eq. (12) is

$$\operatorname{tg} 2\varphi_{cr} \approx \frac{H_{A3}}{2H_e'}\frac{H_{A3}}{H_{A1}-H_{A2}}. \quad (14)$$

Equation (14) differs from the familiar expression for $\tan 2\varphi_{cr}$ in the three-dimensional case found in Refs. 3–5. It should be noted that if $H_{A3} \gg H_{A1} - H_{A2}$, the angle given by Eq. (14) exceeds the noncollinarity angle of the system which is

$$\operatorname{tg} 2\theta \approx H_{A3}/H_e' \ll 1.$$

In other words, even if the magnetic structure of a many-sublattice antiferromagnet in the ground state is nearly two-sublattice, its properties in the course of spin-reorientation phase transitions may differ considerably from the properties of a two-sublattice antiferromagnet. Consequently, spin-reorientation phase transitions cannot be generally described by a two-sublattice model even in the case of weakly noncollinear many-sublattice magnetic materials.

We shall conclude by noting that a spin-flopping transition in an external magnetic field inclined at a large angle to the easy axis has been observed in a one-dimensional four-sublattice antiferromagnet with a noncollinear magnetic structure $CuCl_2C_4H_8SO$ (Ref. 20). This behavior can naturally be explained by the ideas put forward above.

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