Phase diagram of quasi-one-dimensional hexagonal antiferromagnets

S. I. Abarzhi

Moscow Physicotechnical Institute
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The phase diagram of an easy-plane hexagonal quasi-one-dimensional antiferromagnet is studied on the basis of the theory of symmetry. It is shown that near the Néel temperature Θ for three-dimensional ordering this transition splits into two second-order transitions, and the degree of splitting depends on the direction of the field. It is shown that ordering occurs through an intermediate collinear phase and that in the field-exchange approximation strongly nonlinear transverse magnetization exists in the spiral phase.

Antiferromagnets with a triangular lattice are frustrated systems and are characterized by strong degeneracy. This is manifested in a rich diversity of structures, which arise when the temperature and the external field are varied, as well as in the unusual critical properties of these compounds.[1] The object of the present work is to study the phase diagram of ABX_3 hexagonal antiferromagnets in an arbitrarily oriented external magnetic field. In these compounds A is a large univalent cation, B is a magnetic ion, and X is a halogen ion; in addition, the A and X ions form a hexagonal close-packed structure and the B ions are located inside octahedra of halide ions. The octahedra, adjoining at the faces, form infinite chains along the C_6 axis (Fig. 1). Since the distance between magnetic ions along the principal axis is shorter than in a perpendicular direction, exchange being direct along the principal axis and indirect in the perpendicular direction, these compounds exhibit quasi-one-dimensional properties.[2,3] At temperatures $T<\Theta\approx 10$ K, where Θ is the Néel temperature, they change into a noncollinear "triangular" ordered state: The spins alternate antiferromagnetically along the principal axis and make angles of 120° between neighboring spins in the basal plane.[3] The space group of most of these compounds is D_{6h}^4. The unit cell is chosen so that the origin of coordinates is a c position [$\bar{6}m2$ symmetry, coordinates (2/3, 4/3, 1)] and the magnetic ions occupy two-fold a position [$\bar{3}m$ symmetry, coordinates (0, 0, 0) according to Kovalev's handbook[4]] (see Fig. 2). The transition into a "triangular" antiferromagnetic structure is associated with the double-ray star K 13 of the wave vector $\mathbf{k}=\pm(1/3, 1/3, 0)$ (the point K of the Brillouin zone) and tripling of the unit-cell volume. The primitive translations of the magnetic and crystallographic cells are determined by the relations

$$\mathbf{a}_1'=\mathbf{a}_1-\mathbf{a}_2,\quad \mathbf{a}_2'=\mathbf{a}_1+2\mathbf{a}_2,\quad \mathbf{a}_3'=\mathbf{a}_3.$$

The pair of vectors $\mathbf{S}^+$ and $\mathbf{S}^-$ corresponds to the irreducible representation associated with this transition (Fig. 3a). The representation determined by the vectors $\mathbf{S}^+$ and $\mathbf{S}^-$ is irreducible. The components of these vectors are distributed as follows over the irreducible representations of the symmetry group of the paramagnetic phase:

$$(S_z^+, S_z^-) \text{ and } (S_x^+, S_y^+, S_x^-, S_y^-).$$

Since the spiral "triangular" structure is commensurate, there exists a relation between the vectors $\mathbf{S}^+$, $\mathbf{S}^-$, and the spin moments S_k ($k=1, 2, \ldots, 6$) of the sublattices. Then this structure can be viewed as a collection of three intercalated two-sublattice antiferromagnets characterized by the antiferromagnetism vectors ($\mathbf{L}_i=\mathbf{S}_i-\mathbf{S}_{i+3}$, $i=1, 2, 3$). In the absence of a magnetic field the angles between $\mathbf{L}_1$, $\mathbf{L}_2$, and $\mathbf{L}_3$ are all equal to $2\pi/3$. We underscore the fact that due to the quasi-one-dimensionality, at least in magnetic fields which are weak compared with the main exchange field, it can always be assumed that $\mathbf{S}_{i+3}=-\mathbf{S}_i$, $i=1, 2, 3$.

The vectors $\mathbf{S}^+$ and $\mathbf{S}^-$ can be expressed in terms of the spin moments of the sublattices as follows:

$$\mathbf{S}^+=2\mathbf{L}_1-\mathbf{L}_2-\mathbf{L}_3=2\mathbf{S}_1-\mathbf{S}_2-\mathbf{S}_3-2\mathbf{S}_4+\mathbf{S}_5+\mathbf{S}_6,$$

$$\mathbf{S}^-=3^{1/2}(\mathbf{L}_2-\mathbf{L}_3)=3^{1/2}(\mathbf{S}_2-\mathbf{S}_3-\mathbf{S}_5+\mathbf{S}_6).$$

We expand the thermodynamic potential near the phase-transition point $|T-\Theta|\ll\Theta$, taking into account fourth-order invariants as well as anisotropic terms:[5]

$$\Phi=\frac{1}{2}A_0[(\mathbf{S}^+)^2+(\mathbf{S}^-)^2]+\frac{1}{4}B[(\mathbf{S}^+)^2+(\mathbf{S}^-)^2]^2+\frac{1}{2}\gamma[\mathbf{S}^+\mathbf{S}^-]^2+\frac{1}{2}d[(S_z^+)^2+(S_z^-)^2]+\frac{1}{2}\beta[\mathbf{S}^+\mathbf{S}^-]_z^2, \quad (1)$$

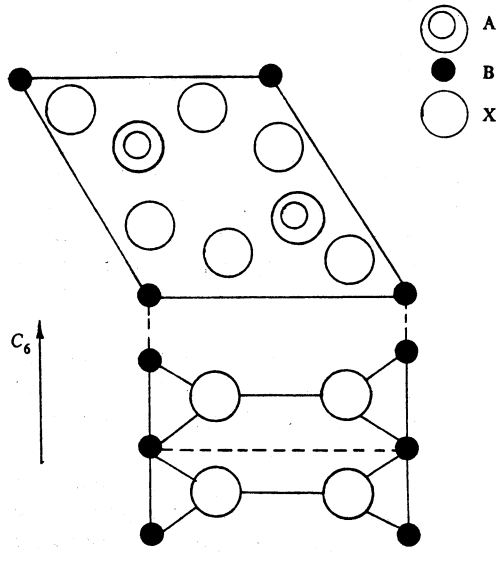

FIG. 1. Arrangement of the atoms in the crystallographic unit cell of ABX_3.

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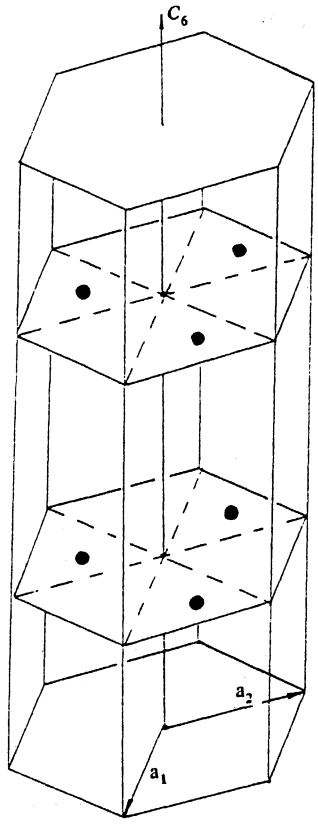


FIG. 2. a-type position occupied by magnetic B ions (●) in a crystal with D^4_{6h} symmetry.

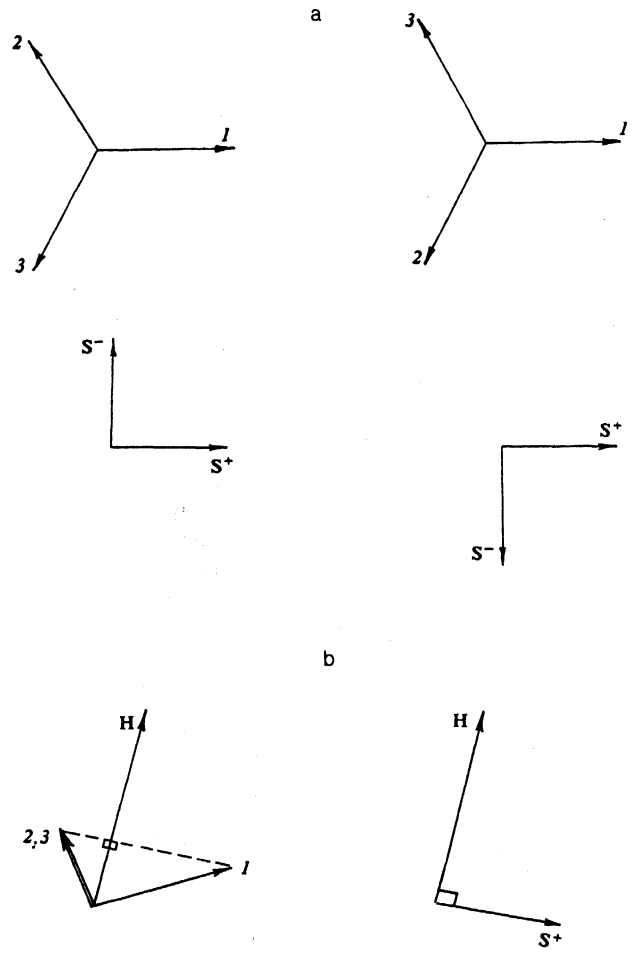


FIG. 3. a—Ground state—spiral 120-degree structure and its discrete degeneracy. The chirality vector is $\mathbf{K} = [\mathbf{S}^+\mathbf{S}^-]$. b—Collapsed "collinear" phase. The arrows mark the antiferromagnetic vectors $\mathbf{L}_1$, $\mathbf{L}_2$, and $\mathbf{L}_3$, respectively. The spin moments of the sublattices $\mathbf{S}_1$, $\mathbf{S}_2$, and $\mathbf{S}_3$ are parallel, and the spin moments of the sublattices $\mathbf{S}_4$, $\mathbf{S}_5$, and $\mathbf{S}_6$ are antiparallel to the vectors $\mathbf{L}_1$, $\mathbf{L}_2$, and $\mathbf{L}_3$, respectively.

where $A_0 = \alpha(T - \Theta)$, $\alpha > 0$.

In the exchange approximation the "coarse" spiral 120-degree structure ($\mathbf{S}^+ \perp \mathbf{S}^-$, $|\mathbf{S}^+| = |\mathbf{S}^-|$) will appear if $\gamma < 0$ and $B + \gamma > 0$. Anisotropic relativistic interactions distort the "coarse" structure and orient it with respect to the crystallographic axes. We confine our attention to the easy-plane case ($d > 0$) when the only role of anisotropy is to orient the "coarse" structure. Then the ground state

$$\mathbf{S}^+ \perp \mathbf{S}^-, \quad |\mathbf{S}^+| = |\mathbf{S}^-| = S, \quad S_z^{\pm} = 0,$$

$$\mathbf{S} = \frac{A_0}{2B + \beta + \gamma}, \quad B + \beta + \gamma > 0$$

arises at the second-order transition point $T = \Theta$. We note that the ground state is continuously degenerate with respect to simultaneous rotation of the vectors $(\mathbf{S}^+, \mathbf{S}^-)$ around the C_3 axis and discretely degenerate ("chirally") with respect to a change in sign of the chirality vector $\mathbf{K} = [\mathbf{S}^+\mathbf{S}^-]$ (Fig. 3a).[6]

A magnetic field will change the symmetry of the system and distort the ground state, and the field component perpendicular to the principal axis will remove the continuous degeneracy.

Since magnetization appears only in the presence of a magnetic field $\mathbf{H}$, we expand the thermodynamic potential Φ with respect to $\mathbf{S}^+$, $\mathbf{S}^-$, and $\mathbf{H}$. Taking into account only exchange invariants of the interaction of the system with the field, we write the expansion of Φ for $|T - \Theta| \ll \Theta$ as follows:

$$\Phi = \frac{1}{2}(A_0 + b\mathbf{H}^2)[(\mathbf{S}^+)^2 + (\mathbf{S}^-)^2] + \frac{1}{2}a[(\mathbf{H}\mathbf{S}^+)^2 + (\mathbf{H}\mathbf{S}^-)^2] - \frac{1}{2}\chi_p\mathbf{H}^2 + \frac{1}{2}d[(S_z^+)^2 + (S_z^-)^2] + \frac{1}{2}\beta[\mathbf{S}^+\mathbf{S}^-]_z^2 + \frac{1}{4}B[(\mathbf{S}^+)^2 + (\mathbf{S}^-)^2]^2 + \frac{1}{2}\gamma[\mathbf{S}^+\mathbf{S}^-]^2. \quad (2)$$

Then the magnetization

$$\mathbf{M} = -\left(\frac{\partial\Phi}{\partial\mathbf{H}}\right) = \{\chi_p - b[(\mathbf{S}^+)^2 + (\mathbf{S}^-)^2]\}\mathbf{H} - a[\mathbf{S}^+(\mathbf{H}\mathbf{S}^+) + \mathbf{S}^-(\mathbf{H}\mathbf{S}^-)] \quad (3)$$

in the general case with arbitrary orientation of the field is not parallel to $\mathbf{H}$.[7] The signs of a and b cannot be determined on the basis of purely thermodynamic considerations. Actually, however, the jump in the derivative of the susceptibility

components parallel ($\chi_\parallel$) and perpendicular ($\chi_\perp$) to the magnetic field at the transition into the paramagnetic phase is always negative,[7-9] which means that $a>0$ and $b>0$.

The behavior of the system (2) is determined by the competition between exchange and anisotropic terms and the field. In standard non-quasi-one-dimensional antiferromagnets the energy of the "exchange" fields H_c, in which a transition from the ordered into the paramagnetic phase occurs at zero temperature, is comparable to the Néel temperature.[9] A peculiarity of the quasi-one-dimensional problem considered here is that the energy of the "exchange" fields ($H_c \approx 10^3$ kOe) is much higher than the ordering temperature Θ, i.e., a and b are such that the exchange "field" terms $a[(\mathbf{HS}^+)^2 + (\mathbf{HS}^-)^2]$ and $b\,\mathbf{H}^2[(\mathbf{S}^+)^2 + (\mathbf{S}^-)^2]$ in fields which are weak compared with the "exchange" fields ($H \ll H_c$) can always be assumed comparable in energy to the second-order anisotropic terms. It is the quasi-one-dimensionality that is responsible, as we shall see below, for the significantly nonlinear transverse magnetization before the transition of the system into the collinear phase. We also note that taking into account the fourth-order anisotropy will not change the qualitative results, but it will merely introduce small corrections. For this reason, everywhere below we neglect the fourth-order anisotropy energy compared with the energies of the exchange "field" terms and the second-order anisotropy terms (i.e., we set $\beta = 0$).

It is obvious from the expansion of the thermodynamic potential that in the general case the second-order phase transition at $T=\Theta$ into the paramagnetic phase splits into two transitions, which are associated with the fact that the vectors $\mathbf{S}^+$ and $\mathbf{S}^-$ do not both vanish simultaneously. Vanishing of one of the vectors for some value of H means that the spiral phase vanishes and the "collapsed" collinear phase appears (Fig. 3b).[8] The solution of the equations $\partial\Phi/\partial\mathbf{S}^\pm = 0$ with stability conditions

$$\left\| \frac{\partial^2\Phi}{\partial \mathbf{S}^+ \partial \mathbf{S}^-} \right\| > 0$$

will determine the points of the phase transitions $T = T(\mathbf{H})$.

The field $\mathbf{H}$ strives to orient the vectors $\mathbf{S}^+$ and $\mathbf{S}^-$ so that $\mathbf{HS}^\pm = 0$ and the anisotropy strives to orient the vectors $S^\pm$ in a plane perpendicular to the principal axis. The competition between these two phenomena determines the ground state of the system. For $\mathbf{H}=0$ the vectors $\mathbf{S}^+$ and $\mathbf{S}^-$ lie in a plane perpendicular to the principal axis. Minimizing the potential Φ (2) with respect to $\mathbf{S}^+$ and $\mathbf{S}^-$ for $\mathbf{H}\neq 0$ gives a ground state in which one of the vectors $\mathbf{S}^+$ (where $\mathbf{S}^+ \perp \mathbf{S}^-$) will be oriented perpendicular to the field and the z axis ($\mathbf{S}^+ \perp \mathbf{H}$, $\mathbf{S}_z^+ = 0$) and the second vector $\mathbf{S}^-$ will lie in the plane formed by the vector $\mathbf{H}$ and the z axis. If φ_0 is the angle by which $\mathbf{H}$ tilts away from the z axis ($0 \leqslant \varphi_0 \leqslant \pi/2$), then the tilt angle φ of $\mathbf{S}^-$ from the z axis in the (z, x) plane (see Fig. 4) will satisfy the condition $\pi/2 \leqslant \varphi \leqslant \pi$. This ground state is described by the system of equations

$$(S^+)^2 = -\frac{A}{B(2+x)} - \frac{\Delta(1+x)}{Bx(2+x)},$$

$$(S^-)^2 = -\frac{A}{B(2+x)} + \frac{\Delta}{Bx(2+x)},$$

$$\operatorname{tg} 2\varphi = \frac{\varkappa \sin 2\varphi_0}{\varkappa \cos 2\varphi_0 + 1}. \qquad (4)$$

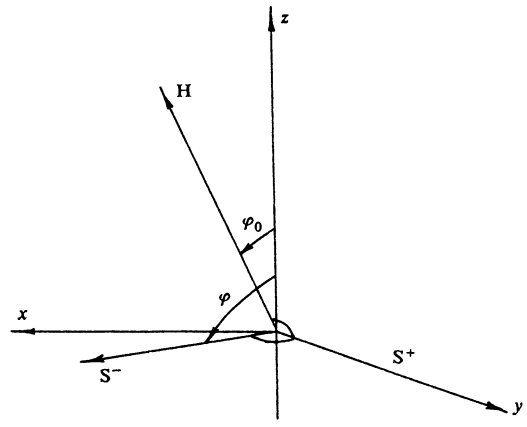


FIG. 4. Structure of the ground state of the spiral in an arbitrarily oriented external magnetic field.

Here $A = A_0 + bH^2$, $\Delta = d[\varkappa \cos^2(\varphi - \varphi_0) + \cos^2\varphi]$, $\varkappa = aH^2/d$ and $x = \gamma/B$. Since $\gamma < 0$, we have $x < 0$. According to Eqs. (4), the case $\varphi_0 = \pi/2$ is special, since in the admissible range of values of φ it corresponds to two solutions $\varphi = \pi$ and $\varphi = \pi/2$ in some nonzero field H_1 in which the energies of these states are equal to one another. If, however, $\varphi_0 = 0$, then the state with $\varphi = \pi/2$ has the lowest energy and is the only state for any value of H.

The instability and, therefore, the points of the phase transitions of the system (2) are determined by the vanishing of the determinant $\|\partial^2\Phi/\partial\mathbf{S}^+\partial\mathbf{S}^-\|$ for some values of $\mathbf{S}^+$ and $\mathbf{S}^-$. In the absence of a field this occurs for $\mathbf{S}^+ = \mathbf{S}^- = 0$ at the point of a second-order phase transition $T=\Theta$. For $\varphi_0 \neq 0, \pi/2$ the determinant vanishes twice: for $\mathbf{S}^- = 0$, determining the second-order transition into the collinear nonspiral phase (Fig. 3b), and for $\mathbf{S}^+ = 0$, determining the second-order transition into the paramagnetic phase.

The equation relating the magnetic field H to the temperature T_1 of the transition from the spiral to the collinear phase has the form

$$\left(\frac{-A_1}{2B}\right)^2 \gamma^2 + \left(\frac{-A_1}{2B}\right)(d + aH^2)\gamma + aH^2 d \sin^2\varphi_0 = 0, \qquad (5)$$

where $A_1 = \alpha(T_1 - \Theta) + bH^2$. The discriminant of this quadratic equation for the easy-plane structure ($d>0, \gamma<0$, $a>0$) is always positive. From the two roots of Eq. (5) we choose the one corresponding to the condition ($\mathbf{S}^\pm = 0$ for $\mathbf{H}=0$ and $T=\Theta$):

$$\frac{\alpha(\Theta - T_1) - bH^2}{2B} = \frac{-(d + aH^2) + [(d + aH^2\cos 2\varphi_0)^2 + (aH^2)^2\sin^2 2\varphi_0]^{1/2}}{2\gamma} \qquad (6)$$

The transition from the collinear into the paramagnetic phase occurs when the vector $\mathbf{S}^+$, whose magnitude in the collinear phase is determined by the relation $(S^+)^2 = -A/B$ (in this phase $\mathbf{S}^- \equiv 0$), vanishes at the temperature

$$T_2 = \Theta - \frac{b}{\alpha} H^2 \qquad (7)$$

and does not depend on the direction of the field **H**.

We now write the limiting expressions (5) and (6) for the temperatures T_1 of the transition from the spiral into the collinear phase.

1) $\varphi_0 = 0$:

$$T_1 = \Theta - \frac{b}{\alpha} H^2. \qquad (8a)$$

A field applied along the z axis preserves the C_3 axis:

$$\mathbf{S}^+ \perp \mathbf{S}^-, \quad |\mathbf{S}^+| = |\mathbf{S}^-| = S, \quad S_z^{\pm} = 0,$$

$$S = \frac{-A_0 - bH^2}{2B + \gamma}.$$

For this reason, in this case $T_1 = T_2$ holds and the transition from the paramagnetic phase occurs directly into the spiral phase, bypassing the collinear phase.

2) $\varphi_0 = \pi/2$:

$$T_1 = \Theta - \frac{b}{\alpha} H^2 + 2\frac{aH^2}{\gamma}\frac{B}{\alpha} = \Theta - \frac{b}{\alpha} H^2 - 2\frac{aH^2}{|\gamma|}\frac{B}{\alpha}, \qquad (8b)$$

which agrees with the results of Ref. 10. A magnetic field applied perpendicular to the C_3 axis (group D_{3h}) leaves only a two-fold axis directed along the field and splits the transition at $T = \Theta$ into two second-order transitions.

3) In weak magnetic fields, such that $\varkappa = (aH^2/d) \ll 1$ holds, the expression (6) becomes

$$T_1 = \Theta - \frac{b}{\alpha} H^2 + 2\frac{aH^2 \sin^2\varphi_0}{\gamma}\frac{B}{\alpha}. \qquad (9a)$$

In strong magnetic fields ($\varkappa \gg 1$) the expression (6) becomes

$$T_1 = \Theta - \frac{b}{\alpha} H^2 + 2\frac{d \sin^2\varphi_0}{\gamma}\frac{B}{\alpha}. \qquad (9b)$$

For an isotropic magnet ($d = 0$) we obtain

$$T_1 = \Theta - \frac{b}{\alpha} H^2 = T_2,$$

since the C_3 axis of the spiral in this case is always directed along the field. The qualitative phase diagram for $T \simeq \Theta$ and arbitrary orientation of **H** is displayed in Fig. 5. In the case of an arbitrarily oriented field the expression for the magnetization has the form

$$\mathbf{M} = \{\chi_p - b[(\mathbf{S}^+)^2 + (\mathbf{S}^-)^2]\}\mathbf{H} - [a\mathbf{H}(\mathbf{S}^-)^2\cos(\varphi - \varphi_0)]\mathbf{n}, \qquad (10)$$

where **n** is a unit vector in the direction $\mathbf{S}^-$. The vector **M** has a component

$$M_\perp = \frac{1}{2} aH(\mathbf{S}^-)^2 \sin[2(\varphi - \varphi_0)], \qquad (11a)$$

perpendicular to the field. This component vanishes at the point $T = T_1$ of the transition into the collinear phase (Fig. 6a). The longitudinal magnetization $M_\parallel$ has a kink at this transition. These results agree with the results of Ref. 7. At higher temperatures $T_1 < T < T_2$

$$\mathbf{M} = \mathbf{M}_\parallel = [\chi_p - b(S^+)^2]\mathbf{H}. \qquad (11b)$$

Since at temperatures $T \gg T_2$ the magnetization is $\mathbf{M} = \chi_p \mathbf{H}$, the magnetization also has a break at the point T_2 of the second-order transition (Fig. 6b).

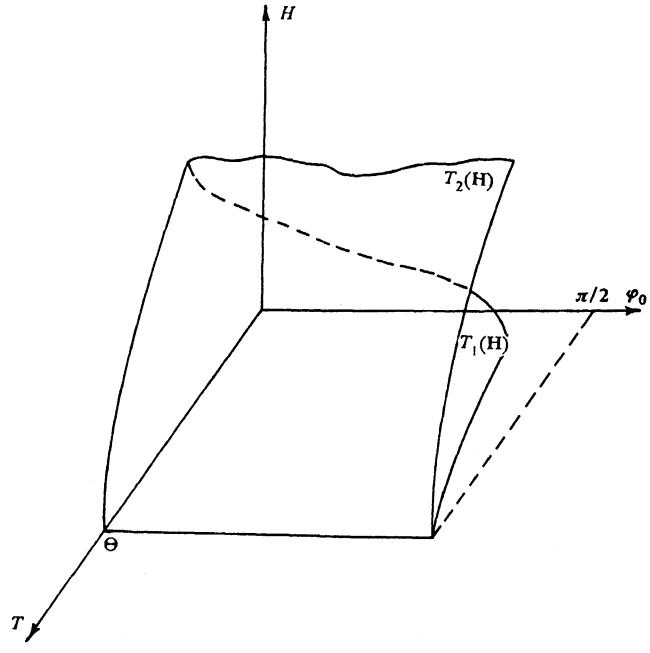


FIG. 5. Phase-transition surfaces $T_1(\mathbf{H})$ and $T_2(\mathbf{H})$; $H_c \approx 10^3$ kOe, $H \ll H_c$, $\Theta \approx 10$ K, $0 \leqslant \varphi_0 \leqslant \pi/2$.

According to the expressions (10) and (11a), the magnitude of the transverse magnetization depends strongly on the direction of the field and on the ratio $\varkappa$. We underscore the fact that, neglecting the relativistic (in the field) interactions, the magnetization $\mathbf{M}_\perp$ is zero when the field has the orientations $\varphi_0 = \pi/2$ and $\varphi_0 = 0$ as well as after a transition into the collinear phase for any orientation of the field (Fig. 6a).

If the field **H** is oriented strictly perpendicular to the crystallographic axis C_3, then the quasi-one-dimensionality introduces additional features into the phase diagram. If the anisotropy is so large that it can confine the spins (and therefore also the vectors $\mathbf{S}^+$ and $\mathbf{S}^-$) in the basal plane before the transition into the collinear phase, i.e., $\varkappa \leqslant 1$, then, in agreement with Refs. 8 and 10, the lines of second-order phase transitions in the HT plane are determined by the expressions (7) and (8b). If, however, the anisotropy is weak ($\varkappa \gg 1$), then there exists a field H_1 satisfying $H_1/H_c \ll 1$ (where H_c is the exchange field), in which the system-field interaction energy is equal in magnitude to the anisotropy energy. In the field H_1 a first-order spin-flop transition occurs from the state with $\mathbf{S}^- \| \mathbf{H}$, $S_z^- = 0$ into a conical phase with $\mathbf{S}^- \perp \mathbf{H}$, $S_x^- = S_y^- = 0$ (see Fig. 7b).[11] In this phase all spins make the same angle with the field (close to $\pi/2$ in fields which are weak compared with H_c, $H/H_c \ll 1$), while the ordering of the spin projections on a plane orthogonal to the field remains triangular. This transition does not depend on temperature and occurs only for a strict orientation of the field with $\varkappa = 1$. The latent heat of this transition is very small. Both second-order transitions into the collinear and paramagnetic phases are preserved and occur at the temperatures

$$T_1 = \Theta - \frac{b}{\alpha} H^2 + 2\frac{aH^2}{\gamma}\frac{B}{\alpha}, \qquad T_2 = \Theta - \frac{b}{\alpha} H^2$$

for $H < H_1$ and

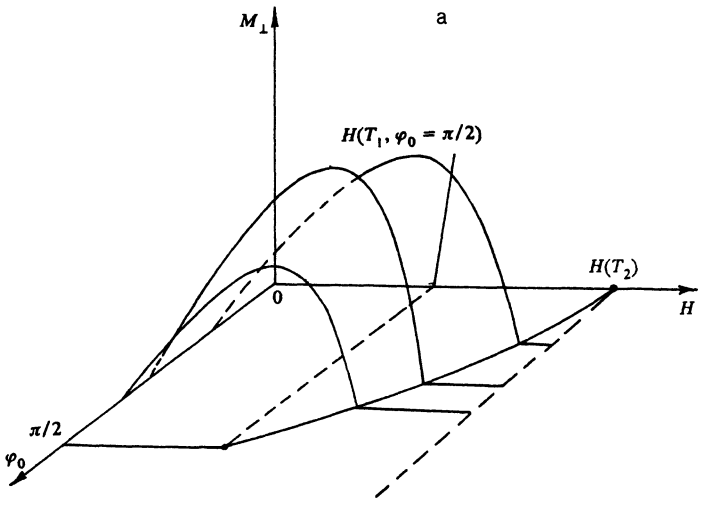
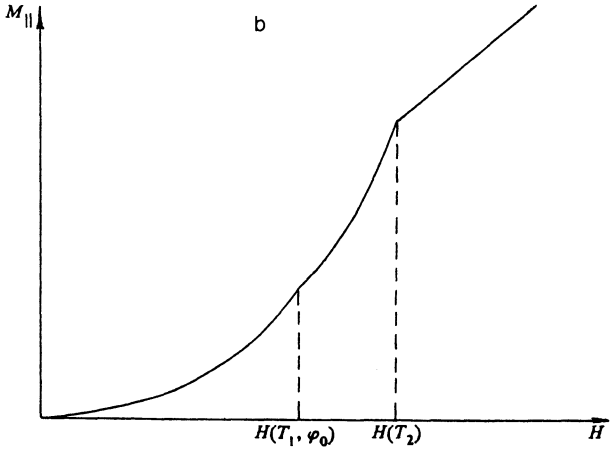


FIG. 6. Qualitative dependence of the transverse magnetization $M_\perp$ (a) and the longitudinal magnetization $M_\parallel$ (b) on the magnitude of the applied field with $T = \text{const}$ ($0 \leqslant \varphi_0 \leqslant \pi/2$); $M_\perp = 0$ for $\varphi_0 = 0$, $\pi/2$ and for $T_1 < T < T_2$.



$$T_1 = \Theta - \frac{b}{\alpha} H^2 + 2\frac{d}{\gamma}\frac{B}{\alpha}, \qquad T_2 = \Theta - \frac{b}{\alpha} H^2$$

for $H > H_1$. Note that for $H > H_1 = (d/a)^{1/2}$ at the transition point the magnetization $\mathbf{M} \| \mathbf{H}$ undergoes a jump

$$\Delta M = aH(S^-)^2 \approx (d)^{1/2}.$$

Thus we have constructed the H-T phase diagram of an easy-plane hexagonal quasi-one-dimensional antiferromagnet at $T \approx \Theta$. It was shown that in an arbitrarily oriented external magnetic field the transition to a three-dimensional ordered state at $T = \Theta \approx 10$ K in the general case splits into two second-order transitions from the paramagnetic to the collinear phase and then from the collinear to the spiral phase. It was shown in the field-exchange approximation that nonlinear magnetization perpendicular to the magnetic field exists in the spiral phase. If we neglect relativistic (in the field) interactions, this magnetization is zero when the field is oriented strictly along or perpendicular to the principal axis C_3. Similar results were obtained theoretically and

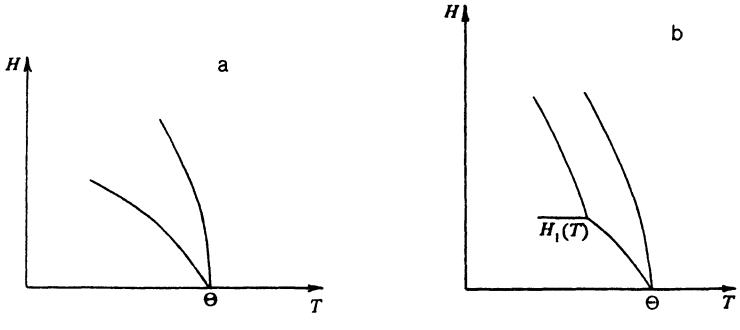


FIG. 7. Phase diagram with $\varphi_0 = \pi/2$ in the case of strong anisotropy $\varkappa \leqslant 1$ (a) and weak anisotropy $\varkappa \gg 1$ (b); $H_1/H_c \ll 1$. The line $H_1(T)$ is the line of first-order phase transitions from the state $\mathbf{S}^- \| \mathbf{H}$ into the state $\mathbf{S}^- \perp \mathbf{H}$.

experimentally in Ref. 7 (using the Heisenberg Hamiltonian) at temperatures close to zero. As the analysis performed above shows, the qualitative character of the transverse magnetization found in Ref. 7 also remains at high temperatures, right up to Θ.

We emphasize once again that taking into account the fourth-order anisotropy will not affect in the basic results, but will only result in some "linear" (in this anisotropy) change in the values of T_1 and T_2 and the magnitude of the field $H_1(T)$ of the first-order transition and will also increase the "latent" heat of this transition.

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