New types of structures in a stochastic easy-plane antiferromagnet with a hexagonal lattice

S. S. Aplesnin

L. V. Kirienskiĭ Institute of Physics

(Submitted 8 August 1990; in revised form 11 January 1991)
Zh. Eksp. Teor. Fiz. **100**, 2068–2073 (December 1991)

New magnetic states have been found by means of numerical modeling in a stochastic magnet with competing interactions and easy-plane anisotropy: antiferromagnets with domain structure, having periodic and nonperiodic solutions with respect to the transverse spin components, and two spin-glass states with respect to the chiral and spin order parameters. Two temperature-dependent transitions, associated with the breaking of the chiral and spin symmetry and the region of magnetic fields with induced long-range chiral order were calculated.

Antiferromagnets with a triangular lattice are frustrated spin systems and are characterized by additional chiral symmetry[1] and strong degeneracy. This is manifested in the rich diversity of structures which arise both when the temperature and the external field are changed[2,3] and when the antiferromagnetic bonds are randomly replaced by ferromagnetic bonds with fluctuating strength.

The aim of this work is to determine the new types of magnetic states and the sequence of phase transitions that do not occur in the theory developed for spin glasses[4] and disordered magnets.[5,6] A complete analysis of the ground state and thermodynamics of a stochastic magnet with a hexagonal lattice can be performed by means of numerical modeling.

1. MODEL

We shall study a stochastic model with fluctuating exchange in a hexagonal lattice with easy-plane anisotropy in the classic Heisenberg model. In order to be able to neglect quantum effects we shall study the case when the anisotropy field is much weaker than the exchange field. The Hamiltonian has the form

$$\mathcal{H}=-\sum_{ij}J_{ij}\mathbf{S}_i\mathbf{S}_j-\sum_i D_i(S_i^z)^2-\sum_i h_iS_i^z, \tag{1}$$

where $D<0$ ($d=D/K$) is the easy-plane uniaxial anisotropy constant, $h=H/|K|$ is the magnetic field along the anisotropy axis, $\mathbf{S}=\mathbf{S}_0\,[S_0(S_0+1)]^{-1/2}$, and J_{ij} is the exchange interaction between the nearest neighbors and satisfies the distribution law

$$P(J_{ij})=\nu\delta(J_{ij}-I)+\tilde{\nu}\delta(J_{ij}-K), \qquad \tilde{\nu}=1-\nu,\quad I>0,\quad K<0,\quad \lambda=|K|/I. \tag{2}$$

In the Monte Carlo (MC) calculations[7] periodic and mirror boundary conditions on a $18\times18\times18$ and $24\times24\times24$ lattice are employed. All quantities employed, i.e., the energy ε, the heat capacity C, the temperature T, the magnetization m, the susceptibility χ, and the distance $r_c=\tilde{r}/c$ along the hexagonal c axis and $r_a=\tilde{r}/a$ in the basal plane, are given in dimensionless units. In addition, the following are calculated: the Edwards-Anderson parameter

$$q^\alpha=\frac{1}{N}\sum_i^N\langle S_i^\alpha\rangle^2,$$

the spin-spin correlation function $\langle S_0^\alpha S_r^\alpha\rangle$ ($\alpha=x,y,z$) in the direction of the crystal axes, the distribution function of the local fields, and the Fourier transform of the spin

$$S^\alpha(\mathbf{Q})=\frac{1}{N}\sum_r e^{i\mathbf{Q}\mathbf{r}}S_r^\alpha,$$

corresponding to the wave vector $\mathbf{Q}$ of the structure.

The noncollinear arrangement of the spins is characterized by the chiral vector

$$\mathbf{k}=2\cdot3^{-1/2}([\mathbf{S}_1\mathbf{S}_2]+[\mathbf{S}_2\mathbf{S}_3]+[\mathbf{S}_3\mathbf{S}_1]), \tag{3}$$

where $\mathbf{S}_1$, $\mathbf{S}_2$, and $\mathbf{S}_3$ are unit spin vectors, arranged among the vertices of elementary triangles. We determine the projection of the vector $\mathbf{k}$ perpendicular to the basal plane as

$$k^z=2\cdot3^{-1/2}[\sin(\varphi_2-\varphi_1)+\sin(\varphi_3-\varphi_2)+\sin(\varphi_1-\varphi_3)], \tag{4}$$

where φ_i is the angle between the spin vector and the x-axis. At $T=0$ $k^{(z)}=\pm1$ is a pseudoscalar variable analogous to the Ising spin, and at finite temperatures it takes on values in the interval $[-1,1]$. With respect to the new variable the magnet forms a two-sublattice structure, for which we shall calculate the average chiral order parameter

$$m^k=\frac{2}{N}\sum_{i=1}^{N/2}\langle k_i^z\rangle^\alpha$$

(where $\alpha=1$ and 2 are the sublattice numbers), the Edwards–Anderson chiral parameter

$$q^k=\frac{1}{N}\sum_{i=1}^{N}\langle k_i^z\rangle^2,$$

and the binary correlation function $\langle k_0^z k_r^z\rangle$ of the chiral parameter over the sublattices in the basal plane and along the hexagonal axis.

2. DISCUSSION

When the antiferromagnetic K-coupling is replaced by ferromagnetic coupling is an isolated triangle the spins at its vertices become aligned parallel to one another and the local chiral parameter satisfies $k_i=0$, though the spin order is preserved. For small concentrations of I-bonds ($\nu\ll1$) the angle between the spins on the impurity bond decreases from

 0038-5646/91/121147-04$03.00

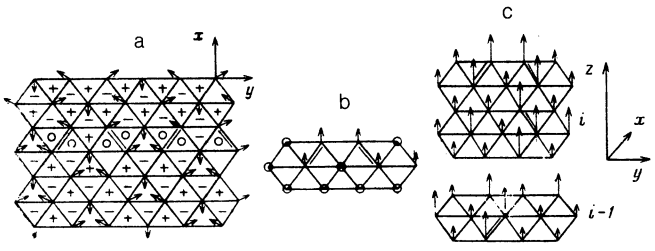

FIG. 1. Domain structure of a disordered AFM with ferromagnetic exchange (double lines) and chirality parameter $k^z = 1(+)$, $-1(-)$, and $0(\bigcirc)$ (a), the z-component of spins in the basal plane in a magnetic field $H < H_{c1}$ (b) and in neighboring planes (c).

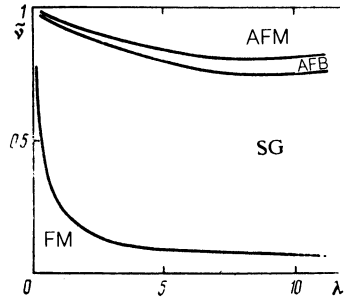

FIG. 3. The phase diagram of a stochastic hexagonal magnet with easy-plane anisotropy: disordered antiferromagnet (AFM), antiferromagnetic with domain (block) structure (AFB), chiral and spin glass (SG), disordered ferromagnet (FM) in the plane (concentration of AFM bonds, exchange ratio $\lambda = |K|/I$).

120° to 0° as the quantity $0 \leqslant \lambda \leqslant 3/2$ increases. Thus, for $\lambda = 0$ the angle decreases by 19° and for $\lambda = 3/2$ the spins become aligned parallel to one another, i.e., $k_i^z = 0$ holds near an impurity bond. When the concentration of I-bonds reaches a critical value, $\nu = \nu_{c1}$, order with respect to one spin component vanishes. Domains which correspond to the symmetry of the Hamiltonian exceed the size of the lattice being modeled and have equally probable order with respect to only the x- or y- spin component form. In the Monte Carlo calculations using different starting spin configurations in the initial conditions we obtain, correspondingly

$$|\langle S_0^{x(y)} S_{r=12}^{x(y)} \rangle| \gg 0, \quad |\langle S_0^{y(x)} S_{r=12}^{y(x)} \rangle| \to 0.$$

The magnetic structure of a single domain is complicated and shown schematically in Fig. 1. Such a structure can appear only if the sign of $S_{i+r}^y = -S_i^y$ changes along the x-axis while order with respect to the components S_i^x is preserved. Numerical calculations of the corresponding characteristics by the Monte Carlo method confirm these estimates (Fig. 2). Thus for $\nu = \nu_{c1}$ the chiral-order parameter approaches zero and the correlation functions of the chiral-order parameter $\langle k_0^z k_r^z \rangle$ and the spin order parameter $\langle S_0^y S_r^y \rangle$ exhibit damped oscillations as a function of distance with different period along the hexagonal axis and in the basal plane, since the substitution of a ferromagnetic I-bond for a K-bond in the basal plane and along the c-axis is not symmetric and results in a different change in the energy and angles of rotation of the nearest-neighbor spins.

The Fourier spectrum of the y-component of the spin $S^y(\mathbf{Q})$ for $\mathbf{Q}(\pi/10 \cdot 3^{1/2}a,\ 4\pi/3a,\ \pi/c)$ is much less than the thermodynamic average value of the spin at a site. It is possible that there exists a nonperiodic (stochastic) or incommensurate phase with respect to S^y for $\nu > \nu_{c1}$ with incommensurateness vector $\mathbf{q} = \mathbf{Q} - \mathbf{Q}_0$, where $\mathbf{Q}_0 = (0,\ 4\pi/3a,\ \pi/c)$.

Further increase of the ferromagnetic bonds destroys the spin order for $\nu \geqslant \nu_{c2}$, and at intermediate concentrations freezing of the spins in the lattice occurs, and it is characterized by two order parameters: the chiral parameter q^k and spin parameters $q^{x,y}$, determined on an interval of 5000 steps/spin after dropping 5000 steps/spin. As the temperature increases these parameters decrease logarithmically. The computed regions of the corresponding phases are shown in Fig. 3.

A weakly disordered AFM ($\nu < \nu_{c1}$) with easy-plane anisotropy has one transition as a function of the temperature. The breaking of the discrete (chiral) symmetry coincides with the breaking of the continuous (spin) symmetry as a function of the temperature. The heat capacity has a strong maximum, slightly smeared out as a result of impurity ferromagnetic bonds.

A disordered AFM with domain structure ($\nu_{c1} \leqslant \nu \leqslant \nu_{c2}$) has two transitions as a function of the temperature. The first transition is associated with the destruction of the inhomogeneous (stochastic) structure with respect to the $y(x)$ component of the spin. This gives rise to a local maximum of the heat capacity at $T = T_{N1}$. The second transition, occurring at $T = T_{N2}$, is caused by destruction of the periodic spin order with respect to the X-component of

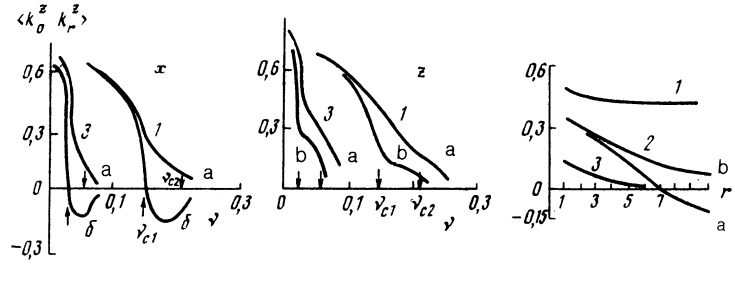

FIG. 2. Correlation function of the chiral parameter $\langle k_0^z k_r^z \rangle$ at distances $r = 1$ (a) and 8 (b) along the z and x axes as a function of the concentration of FM bonds with $\lambda = 1/4$ (1) and 1(3) as a function of the distance along the axes $X(1,\ 2a,\ 3)$ and $z(1,\ 2b,\ 3)$ for $\lambda = 1/4$, and $\nu = 0.12$ (1), 0.15 (2), and 0.22 (3).

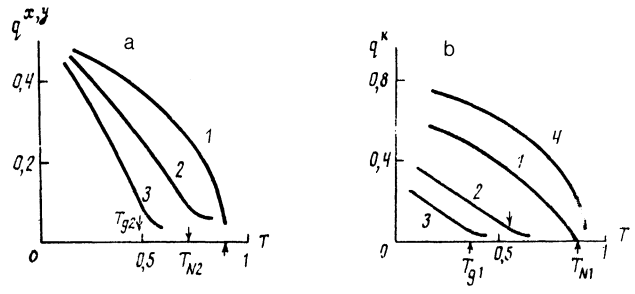

FIG. 4. The spin $q^{x,y}$ (a) and chiral q^k (b) Edwards–Anderson parameters, chiral order parameter (c) in a disordered AFM with $\lambda = 1$, $\nu = 0.02$ (1, 4), in a block AFM with $\lambda = 1$, $\nu = 0.05$ (2), and in a spin glass $\lambda = 0.25$ and $\nu = 0.3$ (3) as a function of the temperature.

the spin. The heat capacity is a maximum at this temperature. The correlation functions of the chiral order parameter vanish at a distance $r = 8$ at $T = T_{N1}$ while the spin-spin correlation functions $\langle S_0^x S_{r=8}^x \rangle$ vanish at $T = T_{N2}$. Disclinations in the domain contain many triangles with $k_i = 0$ (see, for example, Fig. 1a). As a result of the random distribution of bonds the local field at a lattice site is strongly nonuniform, and as the temperature increases the spins in weak local fields become paramagnetic. For this reason, a low concentration of paramagnetic sites, formed at some critical temperature, is sufficient for onset of percolation along sites in the effective lattice, where each site corresponds to the chirality parameter. The spin order with respect to one component is preserved.

Two transitions associated with breaking of local discrete symmetry are also observed at intermediate concentrations ($\nu_{c2} < \nu < \nu_{c3}$ [FM]): one at $T = T_{g1}$ and a local spin transition at $T = T_{g2}$. Correspondingly, at these temperatures the Edwards-Anderson parameter vanishes with respect to the chiral order parameter $q^k \to 0$ ($T = T_{g1}$) and with respect to the spin order parameter $q^{x,y} \to 0$ ($T = T_{g2}$) (Fig. 4). The temperature behavior of the heat capacity at $T = T_{g2}$ exhibits a diffuse maximum. Since in spin glasses short-range order, which can be limited to the first coordination sphere, is preserved at low temperatures, a transition of two spins into the paramagnetic state at $T = T_{g1}$ suffices to make the short-range chiral vanish. The spin order approaches zero if all nearest-neighbor spins are paramagnetic.

In a stochastic AFM with domain (block) structure it is possible to identify three transitions in an external magnetic field directed along the hexgonal axis. In weak fields ferromagnetic order with a three-sublattice structure, shown in Figs. 1b and c for $H \ll H_{c1}$, is induced with respect to the longitudinal components of the spin. Here the z-components of the spins are coupled by the minimum number of K-bonds along all directions of the crystal axes.

The spin correlation function between nearest neighbors satisfies the relation

$$\langle S_0^z S_{r=1}^z \rangle \ll \langle S_0^z S_{r=8}^z \rangle.$$

As the field increases the magnetic structure becomes noncoplanar, and all spins leave the basal plane. The Edwards-Anderson parameter with respect to the transverse components decreases and the parameter with respect to longitudinal components increases. In a field $H \geqslant H_{c1}$ the spins having weak local fields become aligned along the field and the local chirality parameter of approximately one-third of the triangles is zero. This is analogous to the XY-model with two-component order parameters and diamagnetic dilution. In this case the angle of inclination of the magnetization $\overline{m}(H)$ changes, and the correlation function of the chirality at the distance $r = 8$ becomes positive (Fig. 5), and for $H > H_{c1}$ it is virtually independent of the distance. The sublattice chirality parameter increases severalfold and for fields in the interval $H_{c1} < H < H_{c2}$ long-range order with respect to the chiral order parameter is induced. The second transition in fields $H > H_{c2}$ is caused by breaking of the discrete (chiral) symmetry of the Hamiltonian ($m^k = 0$, $\langle k_0^z k_{r=1}^z \rangle = 0$) and by breaking of the continuous global symmetry with respect to the transverse components of the spin, with respect to which long-range order vanishes: $\langle S_0^{x,y} S_{r=8}^{x,y} \rangle = 0$, but short-range order is preserved: $q^{x,y} > 0$, $\langle S_0^{x,y} S_{r=1}^{x,y} \rangle > 0$. In fields $H > H_{c3}$ short-range order vanishes: $q^{x,y} \to 0$, and the dependence $m(H)$ saturates.

The magnetic structures studied above can be realized in a solid solution with a hexagonal lattice $Fe(Cl_x Br_{1-x})_2$.[8] Crystals of halide compounds ABX_3, which have planes with a triangular lattice, are also promising materials.

Thus the existence of two types of symmetries in a stochastic hexagonal antiferromagnet with easy-plane anisotropy leads to the formation of new types of magnetic states: AFM with domain (block) structure and different period of modulation with respect to the transverse spin components and two spin-glass states with respect to chiral and spin order parameters. These phases are characterized by two successive phase transitions as a function of the temperature. In strong fields the domain (block) structure vanishes and long-range chiral order is induced.

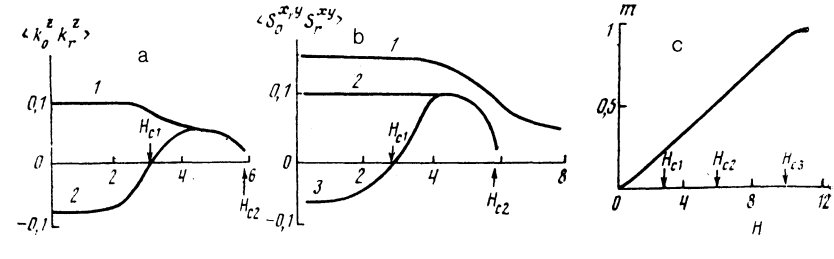

FIG. 5. The chiral $\langle k_0^z k_r^z \rangle$ (a) and spin $\langle S_0^\alpha S_r^\alpha \rangle$ ($\alpha = x(2)$ and $y(3)$) correlation functions at the distances $r = 1$ (1) and 8 (2, 3) (b) along the X-direction and the magnetization m (c) as a function of the external field H.

[1] J. Villain, Physique **38**, 385 (1977).
[2] R. S. Gekht, Usp. Fiz. Nauk **159**, 261 (1989) [Sov. Phys. Usp. **32**(10), 871 (1989)].
[3] S. S. Aplesnin and R. S. Gekht, Zh. Eksp. Teor. Fiz. **96**, 2163 (1989) [Sov. Phys. JETP **69**(6), 1224 (1989)].
[4] S. L. Ginzburg, *Irreversible Phenomena in Spin Glasses* [in Russian], Nauka, Moscow (1989).
[5] A. F. Andreev, Zh. Eksp. Teor. Fiz. **74**, 786 (1978) [Sov. Phys. JETP **47**(2), 411 (1978)].
[6] P. J. Ford, Contemporary Phys. **23**, 129 (1982).
[7] K. Binder, *Monte Carlo Methods in Statistical Physics*, Springer, New York (1979).
[8] K. S. Aleksandrov, N. V. Fedoseeva, and I. P. Spevakova, *Magnetic Phase Transitions in Halide Crystals* [in Russian], Nauka, Novosibirsk (1983), pp. 160–166.

Translated by M. E. Alferieff