Thermally induced weak terromagnetism in an anisotropic material with competing interactions

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The range of temperatures and exchange interaction strengths in which a weak ferromagnetic moment is induced thermally in a magnetic material with competing interactions is calculated by the Monte Carlo method for a square lattice. The calculations are carried out for the second coordination sphere and an anisotropic distribution of tensor interactions of opposite signs in the first sphere. The temperature dependences of the susceptibility, specific heat, and magnetization in an external field are determined.

Competition between the exchange interactions gives rise to a variety of magnetic structures.[1,2] In a square lattice such an exchange competition is due to the mismatch between the signs of the exchange interactions in the first (I_1) and second (I_2) coordination spheres and the nature of spin ordering. This situation has already been considered for an isotropic antiferromagnet with the negative sign of the exchange interaction in the nearest two coordination spheres.[3-5] Rigorous calculations carried out for small samples demonstrate that there is no spin liquid state for $S = 1/2$ spins[3,4] and that a paramagnetic state appears for classical spins.[5] If the ratio of the interaction strengths is $I_2/I_1 = 0.5$, the resultant magnetic state is degenerate and it may change in the presence of an anisotropy of the exchange interactions and when the distribution of interactions in the lattice is anisotropic. For example, the anisotropy of the exchange interactions in a triangular lattice lifts the continuous degeneracy and gives rise to a new type of chiral symmetry.[6]

The model just described is not a theoretical abstraction, but it reflects the signs and strengths of the exchange interactions in crystals with the structure characterized by the $Pb2_{1m}$ (C^2_{2v}) symmetry exhibited by antiferromagnets $CuGeO_3$ (Ref. 7) and $MgCu_2O_3$ (Ref. 8) with an anisotropic distribution of the couplings. The interaction between the planes is very weak and the magnetic structure is established by the exchange interactions in one plane. Neutron-diffraction studies of $MgCu_2O_3$ (Ref. 8) reveal a weakly canted structure, whereas the temperature dependences of the susceptibility and magnetization obtained for $CuGeO_3$ (Ref. 7) in the presence of a field cannot be described by the standard model with an antisymmetric exchange.

Our aim will be to determine the magnetic structure and to study changes in the spin degeneracy state as well as characteristic thermodynamic singularities of a magnetic material as a function of three factors:

1) an anisotropic distribution of the exchange interactions with opposite signs in a square lattice;

2) different parameters of the exchange interaction between the longitudinal and transverse components of the spins;

3) the magnitude of the frustrated interactions in the second coordination sphere.

We shall tackle this task by the method of numerical modeling using the Monte Carlo procedure.

1. MODEL

We shall consider a two-dimensional magnetic material with different parameters of the exchange between the components of the spins distributed anisotropically over a square lattice in the classical Heisenberg model. The Hamiltonian is of the form

$$H=-\sum_{\alpha=1}^{3}\Big\{\sum_{i=1}^{N} K^{\alpha\alpha}_{i,i+1}S_i^\alpha S^\alpha_{i+1}+\sum_{j=1}^{N}(I_1^{\alpha\alpha})_{j,j+1}S_j^\alpha S^\alpha_{j+1}+\sum_{\langle ij\rangle}(I_2)_{i,j}S_i^\alpha S_j^\alpha+\sum_{i=1}^{N}h_i^\alpha S_i^\alpha\Big\},$$

where $K^{\alpha\alpha}<0$ corresponds to the antiferromagnetic (AFM) exchange interaction between the spin components $\alpha = x, y$, and z along the y axis (Fig. 1), when the summation is carried out over the index i; $I_1^{\alpha\alpha}>0$ corresponds to the ferromagnetic (FM) exchange along the x axis when the summation is carried out over the index j; $I_2^{\alpha\alpha}>0$ corresponds to the ferromagnetic exchange in the second sphere; $h^\alpha = H^\alpha|K^{zz}|[S_0(S_0+1)]^{1/2}$ is the external magnetic field applied along the x, y, and z axis; $S^\alpha = S_0^\alpha/[S_0(S_0+1)]^{1/2}$.

All the quantities used here including the temperature of a sample $T=\widetilde{T}/[k_B|K^{zz}|\cdot S_0(1+S_0)]$, the specific heat $C=\widetilde{C}k_B/N=dE/dT$, the susceptibility $\chi^\alpha = \widetilde{\chi}^\alpha|K^{zz}|/N = m^\alpha/H^\alpha$, and the magnetization m^α are given in dimensionless units. We shall calculate additionally the Edwards–Anderson parameter

$$q^\alpha=\sum_{i=1}^{N}\langle S_i^\alpha\rangle^2/N,$$

the spin–spin correlation functions of the longitudinal and transverse spin components $\langle S_0^\alpha S_r^\alpha\rangle$, the distribution functions of the local energies $P(E_i)$ and spin projections $P(S_i^z)$, and the Fourier spectrum of the spin $S^\alpha(\mathbf{Q})$ corresponding to the wave vector $\mathbf{Q}$ of the structure.

2. DISCUSSION OF RESULTS

An isotropic magnetic material with the exchange interactions satisfying the condition $\lambda = I_2/I_1 = 0.5$ is a paramagnet in the limit $T\to 0$. If $\lambda < 1/2$, a two-sublattice state of the Néel type is obtained, whereas for $\lambda > 1/2$ the x axis is the

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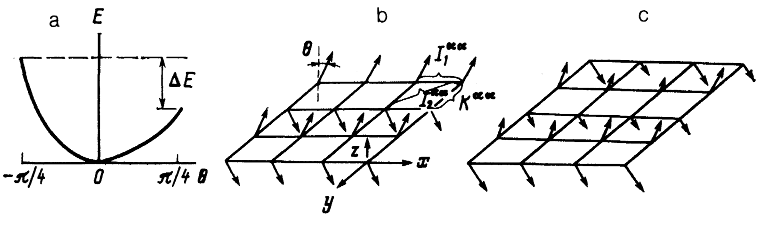

FIG. 1. Energies of excited states of an anisotropic antiferromagnet in the case of cophasal ($\theta > 0$) and antiphasal ($\theta < 0$) vibrations of K-coupled chains (a), a canted structure of an antiferromagnet characterized by $I_1/I_1 < 0.5$ (b), and a randomly modulated structure in the case when $\lambda = 0.5$ (c).

ferromagnetic direction and the y axis is the antiferromagnetic direction.[5] Inclusion of the exchange anisotropy in the Hamiltonian (1), for example $K^{zz} \neq K^{xx} = K^{yy}$ and $I_1^{zz} \neq I_1^{xx} = I_1^{yy}$, gives rise to a long-range order at finite temperatures and the magnetic exchange along the x axis induces at finite temperatures a weak canting of the magnetic moments of the sublattices, i.e., when an anisotropic magnetic material is heated it is found that higher energy levels, whose spin configurations have a weak ferromagnetic moment in the basal plane, become mixed with the ground state. The distribution function of the local energies becomes strongly broadened and the distribution function of the longitudinal components of the spin remains symmetric relative to zero. If we consider the energy of vibrations of AFM chains relative to one another, we find that it is asymmetric relative to the z axis along which an external magnetic field is applied (Fig. 1). The energy splitting is proportional to $\Delta E = 2(I_2 + I_1)$. As temperature is increased, it is found that coherent tilts of the spins in chains with the K coupling become favorable from the energy point of view.

The induced ferromagnetic moment increases on increase in the antiferromagnetic exchange anisotropy $(K^{zz} - K^{\perp\perp})/K^{zz}$ and on increase in the ferromagnetic coupling in the first and second coordination spheres (Fig. 2). There is no induced ferromagnetic moment in an isotropic magnetic material. An anisotropic magnetic material characterized by $I_2^{zz}/I_1^{zz} = 0.5$ undergoes a transition, during heating to a temperature T_{c1}, from a paramagnetic phase to a nonperiodic structure modulated in respect of the longitudinal spin components. The Fourier spectrum of the spin is $S^z(\mathbf{Q}) \ll q^z$ (Fig. 2), the component of the wave vector along the K-coupled chain is $Q^y = \pi/a$, whereas the component along the orthogonal axis is random. The transverse spin components have the FM order. Therefore, the frustrated interaction in the second coordination sphere destroys the long-range AFM order at $\lambda = 0.5$, but a weaker magnetic order is retained. The resultant structure is shown schematically in Fig. 1c.

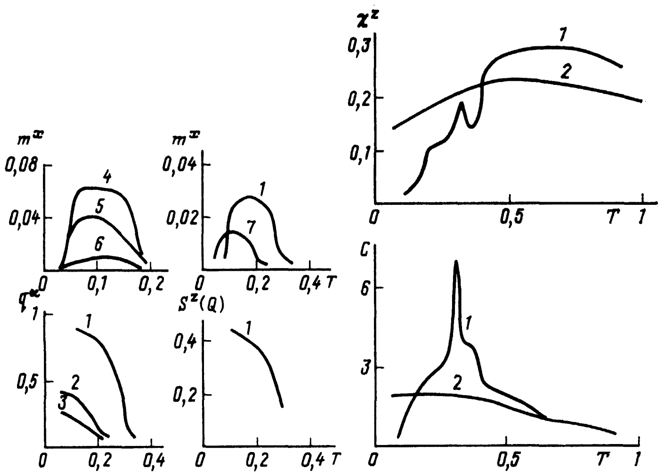

FIG. 2. Temperature dependences of a weak ferromagnetic moment m^x in a plane, of the longitudinal Fourier component $S^z(Q)$ of the spin, of the Edwards–Anderson parameter q^α for the cases when $\alpha = z$ (curves 1 and 2) and x, y (curve 3), of the longitudinal susceptibility χ^z, and of the specific heat C of an anisotropic magnetic material characterized by $\lambda = 0.5$ and by the following exchange parameters: $K^{\perp}/|K^{zz}| = -0.2$, $I_1^{zz}/|K^{zz}| = 0.3$, $I_1^{\perp}/|K^{zz}| = 0.3$ (*1*); $K^{\perp}/|K^{zz}| = -1$, $I_1^{zz}/|K^{zz}| = 0.4$ (*2, 3*); $K^{\perp}/|K^{zz}| = -0.8$, $I_1^{zz}/|K^{zz}| = 0.8$ (*4*); $I_1^{zz}/|K^{zz}| = 0.6$ (*5*); $I_1^{zz}/|K^{zz}| = 0.3$ (*6*); $K^{xx}/|K^{zz}| = -0.2$, $K^{yy}/|K^{zz}| = -0.1$, $I_1^{zz}/|K^{zz}| = 0.3$ (*7*).

At the temperature T_{c1} the specific heat and the susceptibility have an inflection point (Fig. 2). An increase in temperature destroys the induced ferromagnetic moment at T_{c2} and this gives rise to anomalies in the temperature dependences of the suceptibility and specific heat. The specific heat has a sharp maximum typical of destruction of the long-range order, which is different from a thermodynamic nonequilibrium state of a spin glass.[9] A wide susceptibility maximum is due to the appearance of a short-range order in the K-coupled chain because $|K| \gg I_{1,2}$. Clearly, the appearance of the weak ferromagnetic moment in a finite range of temperatures is a thermodynamically stable effect. For example, an increase in the number of steps in the Monte Carlo method from 5000 to 50 000 per spin and of the size of the lattice from 30×30 to 60×60 does not alter the magnitude of this moment. The stability of a weak FM moment is supported also by the sharp specific heat maximum. In the case of an isotropic magnetic material the temperature dependences of the susceptibility and specific heat are similar to the corresponding dependences exhibited by a one-dimensional AFM chain.[10]

A weak ferromagnetic moment is induced thermally in an anisotropic magnetic material at some critical value of the FM interaction I_{c1}, which decreases on increase in the anisotropy of the exchange between the spins along the Y axis. When the FM interaction between the K chains exceeds the value $I_{c2} = (K^{zz} + K^{\perp\perp})/2$, a ferromagnet with an easy-plane anisotropy is obtained.

The appearance and disappearance of this weak moment in a plane within an anisotropic magnetic material, characterized by a weak frustrated interaction $\lambda < 0.5$, is also accompanied by anomalies in the temperature dependences of the susceptibility and the specific heat, whereas the induced ferromagnetic moment disappears simultaneously with destruction of the long-range AFM order.

The different strengths of the exchange interactions between the x, y, and z components of the spins are responsible for the dependence of the temperature $T_{\rm CW}$ at which the Curie–Weiss law begins to be satisfied on the direction of the external magnetic field. If the anisotropies of the exchange antiferromagnetic and ferromagnetic interactions differ, for example, if $K^{\perp\perp}/K^{zz} = 0.8$ and $I_1^{\perp\perp}/I_1^{zz} = 2$, the tempera-

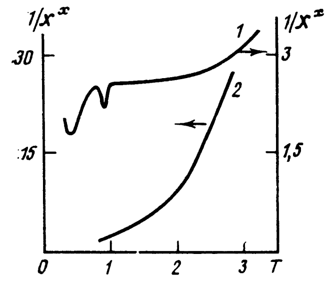

FIG. 3. Temperature dependences of the reciprocal of the susceptibility of a magnetic material with the following parameters: $K^{\perp}/|K^{zz}| = -0.2$, $I_1^{zz}/|K^{zz}| = 0.5$, $I_2/I_1 = 0.5$ (*1*); $K^{\perp}/|K^{zz}| = -0.8$, $I_1^{zz}/|K^{zz}| = 0.1$, $I_1^{\perp}/|K^{zz}| = 0.2$, $I_2 = 0$ (*2*). These dependences are calculated for a field $H^x = 0.05$.

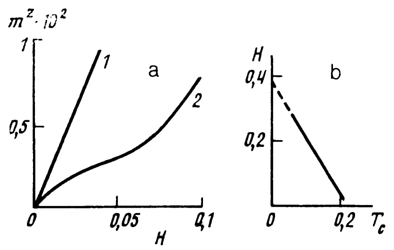

FIG. 4. a) Dependences of the magnetization H along the direction of an external field (along the Z axis) on the value of this field at temperatures $T = 0.15$ K (curve 2) and 0.35 K (curve 1). b) Dependence of the critical temperature T_c of the disappearance of the magnetic moment on the external field applied to a magnetic material characterized by $I_2/I_1 = 0.5$, $K^{\perp}/|K^{zz}| = -0.8$, $I_1/|K^{zz}| = 0.3$.

tures at which the linear temperature dependences of the longitudinal and transverse susceptibilities are observed differ by a factor of 2.5. An additional anomaly of the susceptibility in the paramagnetic phase, predicted for the same parameters (Fig. 3), is due to a strongly anisotropic distribution of the interactions.

In an external magnetic field directed parallel to the antiferromagnetic vector a nonperiodic modulated spin structure without any long-range AFM order, applicable in the case when $\lambda = 0.5$, is gradually transformed at $T > 0$ to a collinear antiferromagnetic order. Therefore, the dependence of the magnetization on the external field is nonlinear when low fields are applied to an anisotropic magnetic material characterized by $\lambda = 0.5$ (Fig. 4a). A further increase in the field rotates the antiferromagnetic vector at right-angles to the field and the dependence $m(H)$ becomes typical of an antiferromagnet. Consequently, the temperature dependence of the susceptibility changes qualitatively and a low-temperature dip disappears. The range of existence of a thermally induced weak ferromagnetic moment becomes narrower on increase in the field and the moment disappears at some critical value of the field (Fig. 4b).

These results can account for the canted magnetic structure in $MgCu_2O_3$ (Ref. 8) and can be used to carry out purposeful measurements of the susceptibility and magnetization of this compound, and also of the same properties of $CuGeO_3$ (Ref. 7) at low temperatures in the range $T < 4$ K.

CONCLUSIONS

A magnetic material with different strengths of the interaction between the x, y, and z components of the spins and an anisotropic distribution of the antiferromagnetic and ferromagnetic interactions in the square lattice exhibits a thermally induced weak ferromagnetic moment in a finite range of temperatures and exchange interaction strengths.

If a frustrated ferromagnetic interaction exists in the second coordination sphere and the constant of this interaction is half that in the first, an anisotropic magnetic material exhibits a structure modulated nonperiodically in respect of the longitudinal components of the spin and a weak ferromagnetic ordering of the transverse components.

An increase in the field lowers the critical temperature at which the weak moment is destroyed. The Curie–Weiss law appears at different temperatures in the three orthogonal directions because of the anisotropic nature of the interactions.

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