Discommensurate and inhomogeneous states induced by a strong magnetic field in low-dimensional antiferromagnets

B. A. Ivanov,[1] C. E. Zaspel,[2,*] and A. Yu. Merkulov[1]
[1]*Institute of Magnetism, NASU, Vernadskii Avenue 36-B, 03142 Kiev, Ukraine*
[2]*Department of Physics, Montana State University-Bozeman, Bozeman, Montana 59717, USA*
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Anisotropic antiferromagnetic systems of dimensionality greater than 1 in an external field are shown to exhibit a complicated array of ground states depending on the spin structure of the surface. The simplest structure that exhibits these effects is the spin ladder with the surface being the ladder end, which can be either compensated or noncompensated spins. The structure with the compensated end has a surface spin-flop phase, the noncompensated end has a discommensurational phase, and the transition to these phases can be either first or second order with a tricritical point.

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Spin-flop transitions in antiferromagnetic (AFM) systems induced by magnetic fields have been studied for more than 50 years,[1–3] and this area still generates much interest. Using the classical semi-infinite spin chain model with single-ion anisotropy, Mills mentioned[3] that spin-flop states could be localized at the surface of an AFM system at a critical field that is lower than the bulk critical field. This surface spin-flop (SSF) state recently was observed[4,5] in Fe/Cr multilayer systems, which stimulated renewed interest in the nature of localized surface states.[4–8] In all of these references the one-dimensional model has been used since it is obviously adequate[3] for multilayer systems, as well as for AFM systems with a simple surface consisting of spins from one sublattice only.

In this paper the nature of the surface states and the transitions to these states are investigated for systems of dimensionality greater than 1. Even for the simple spin ladder, which is intermediate between one- and two-dimensional systems, there are unexpected and interesting effects absent from the simple spin-chain model considered previously.[3,6–8] These effects originate in the more complicated surfaces that are possible in ladder structures and in two-dimensional (2D) arrays. In general there can be two types of surfaces (compensated, with an equal number of spins from different sublattices on the surface and zero surface magnetization in the AFM state, and noncompensated, with nonzero surface magnetization). For the last case, with the noncompensated end spin antiparallel to the external field, the discommensuration state with a 180° domain wall common to that considered for spin chains[6–8] appears. In contrast, for compensated surfaces the SSF phase, as proposed by Mills,[3] is found. In this state the surface spins rotate to about 90° from the external field with a net magnetic moment at the surface, and there is uniform decay to the AFM phase moving into the bulk. For both cases a critical field is lower than the bulk critical field.

In past investigations of chain models only first-order transitions were reported in the literature.[6–8] However, for these more complicated structures it is shown here that the transition to the SSF phase or discommensurational states can be either first or second order depending on the type of anisotropy (exchange or single-ion anisotropy) as well as on the character (compensated or noncompensated) of the surface spins. For the second-order transition the amplitude of the nonuniform spin distribution goes to zero at the transition point. In the vicinity of this point the state is neither SSF nor discommensurational; rather, it is a slightly broken Néel state with the deviation from the Néel state decaying into the bulk with oscillations. Moreover, with both types of anisotropy, a tricritical point can be present.

Both numerical and analytical methods are used to investigate the nature of the ground state with different surfaces and anisotropies. We begin with the discrete Hamiltonian of the uniaxial spin ladder with classical spins $\mathbf{S_i}$ at sites $\mathbf{i}$ of a dimer lattice and an AFM interaction between nearest neighbors,

$$W=\sum_{\mathbf{i},\delta}\ (J\mathbf{S_i}\mathbf{S_{i+\delta}}+\kappa S_{z,\mathbf{i}}S_{z,\mathbf{i}+\delta})-k\sum_{\mathbf{i}}\ S_{z,\mathbf{i}}^2-H\sum_{\mathbf{i}}\ S_{z,\mathbf{i}}. \tag{1}$$

Here, the first term describes the interaction of neighboring spins connected by the vector δ, J is the exchange integral, κ is the measure of the exchange anisotropy, k is the single-ion anisotropy constant, and H is the external field along the easy z axis in energy units $g\mu_B$, where g is gyromagnetic ratio and μ_B is the Bohr magneton. For a description of the ladder system it is natural to take a single dimer as the magnetic unit cell and use the net magnetization $\mathbf{m}_n$ and the antiferromagnetic vector, $\mathbf{l}_n$ for the nth dimer,

$$\mathbf{m}_n=(\mathbf{S}_1+\mathbf{S}_2)/2S, \mathbf{l}_n=(\mathbf{S}_1-\mathbf{S}_2)/2S. \tag{2}$$

It is sufficient to consider the spins confined to one plane and to express $\mathbf{m}_n$ and $\mathbf{l}_n$ through the angular variable θ_n and the length of the magnetization, m_n:

$$l_z=l\cos\theta, \quad l_x=l\sin\theta, \quad m_z=m\sin\theta,$$

$$m_x=-m\cos\theta, \quad l=\sqrt{1-m^2}. \tag{3}$$

Elimination of m for the infinite antiferromagnet without a surface gives the energy $W(\theta)$ as a function of θ. In the lowest approximation in the small parameters k/kJ and κ/J, one can find the effective magnetic anisotropy per dimer, $W(\theta)=K\sin^2\theta$, $K=S^2(2k+Z\kappa)$, which is the easy axis for

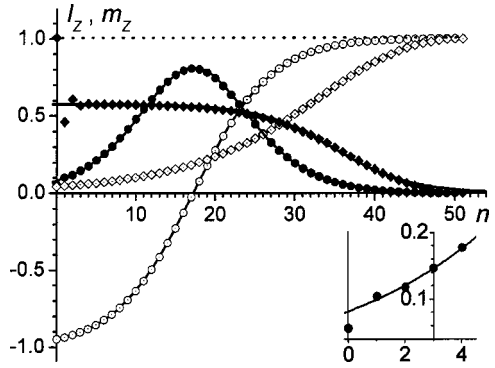

FIG. 1. The spin distribution for different ladder systems with single-ion anisotropy for magnetic fields (in units of J) far below the critical field. Circles: regular ladder, $H=0.848$, $k=0.06$. Diamonds: NC ladder, $H=0.24$, $k=0.01$. Open symbols represent l_z; solid full symbols represent m_z, multiplied by 4 for the NC ladder and by 20 for the RL. For the last case the end details are shown in the inset.

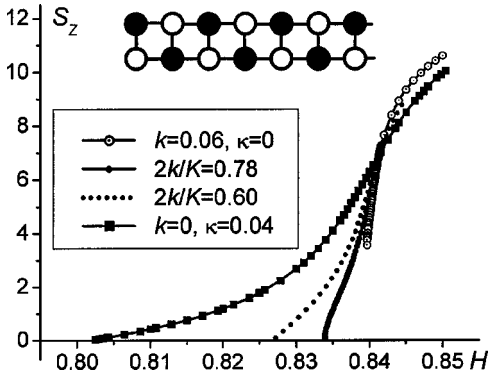

FIG. 2. The $S_z(H)$ dependence for a regular ladder with single-ion anisotropy, exchange anisotropy, and two types of combined anisotropy. For the spin ladder structure shown here and in Fig. 3, open and solid circles present up and down spins in the Néel state, respectively.

$K>0$, and Z is the coordination number. (In the following we put $S=1$.) It follows that the collinear Néel state, in which $\theta=0$ or π is stable for $H<H_1$, where

$$H_1=\sqrt{K(2ZJ+K)}, \tag{4}$$

and a spin-flop phase, with $\theta=\pm\pi/2$, of lower energy than the Néel state is stable for $H>H_{SF}=\sqrt{K(2ZJ+Z\kappa-2k)}$. If $k>0$, then $H_{SF}<H_1$ and the spin-flop transition is of first order. As will be shown below, the second-order transition is the more common case for the surface spin-flop transition.

The description of surface phase transitions begins with a numerical minimization of the discrete Hamiltonian for the two most interesting cases: the regular ladder (RL) structure that is a semi-infinite spin ladder having a regular dimer at the end and the ladder with the single noncompensated (NC) spin on the end. (The configurations of atoms for these cases are present in the insets in the figures below.) The energy minimization has been performed through a Seidel-like algorithm, for spin ladders as long as 100 dimers, which is much larger than the size of a local state. The spins on one end of the ladder are free, with spins on the other end are fixed in the Néel state, corresponding to the bulk nonperturbed state. The distributions of m_z and l_z as a function of the distance from the end of the ladder are qualitatively different for the RL and NC structures as can be seen in Fig. 1. For the RL structure, which is a model for the spin-flop transition in AFM systems with a compensated infinite surface, the l_z data show a surface spin-flop state described by Mills[3] where l rotates approximately 90° with a magnetization that decays to zero into the ladder. For the NC ladder, the discommensuration state[6–8] containing a 180° domain wall appears. The $l_{z,n}$ data for both cases are well described by an AFM domain wall in the continuum approximation:[2] a 90° domain wall with $\tan\theta_n=\exp[-(n-n_0)/\Delta_{\pi/2}]$ or a 180° domain wall with $\tan(\theta_n/2)=\exp[-(n-n_0)/\Delta_\pi]$. Numerical data illustrate the nonregular behavior of m_z near the end of the ladder.

The numerical analysis also shows the presence of two types of behavior for the dependence of the z component of total spin S_z on the magnetic field. For both systems with single-ion anisotropy, the nonuniform phases have a finite value of S_z at the transition point, where the energies of collinear and nonuniform phases coincide. However, for the case of exchange anisotropy the value of S_z goes to zero at the transition point as shown in Fig. 2. This can be interpreted as the presence of first- and second-order transitions, respectively.

To explain these features, consider the stability problem of the collinear phase, having the values of $\theta_n=0$ and $m_n=0$. The stability of this state can be investigated using the quadratic approximate Hamiltonian with small variables θ_n and m_n written as $W=W^{(r)}+\Delta W$, where $W^{(r)}$ is, for the regular semi-infinite ladder,

$$W^{(r)}=\sum_{n=0}^{\infty}[(3J+2k+3\kappa)m_n^2+2Jm_nm_{n+1}+(J+\kappa)m_{n+1}^2 - 2H\theta_nm_n+J(\theta_n-\theta_{n+1})^2+2(k+3\kappa)\theta_n^2+\kappa\theta_{n+1}^2], \tag{5}$$

with the value $n=0$ indicating the end dimer, and in the presence of extra spins, ΔW describes their interaction with the end dimer. The connections between variables θ_n and m_n for any value of n can be found from the equations $\partial W/\partial\theta_n=0$, $\partial W/\partial m_n=0$. For $n>0$, there is an infinite set of equations having the same structure as those for the infinite ladder:

$$J(2\theta_n-\theta_{n+1}-\theta_{n-1})+(2k+3\kappa)\theta_n-Hm_n=0,$$

$$(4J+2k+3\kappa)m_n+J(m_{n+1}+m_{n-1})-H\theta_n=0. \tag{6}$$

These equations can be solved using the exponential ansatz

$$\theta_n=\theta_pe^{-np}+(-1)^n\theta_qe^{-nq}, \tag{7a}$$

$$m_n=m_pe^{-np}+(-1)^nm_qe^{-nq}, \tag{7b}$$

where p and q are determined by substitution of Eqs. (7) into Eqs. (6) to give $2\cosh p=3\tilde{J}/J-1$ and $2\cosh q=3\tilde{J}/J+1$,

where $3\tilde{J}=\sqrt{9J^2+H_1^2-H^2}$. The connections between the amplitudes $\theta_{p,q}$ and $m_{p,q}$ can be written as $\theta_p=A\ m_p$, $\theta_q=m_q/A$, where $A\approx H/6J\ll 1$. Thus, for a slowly decaying exponent with $p\ll 1$ one obtains $m_p\ll\theta_p$, and for the staggered exponent with fast decay the situation is the opposite. Next minimization of ΔW with respect to the parameters describing the nonregular part of ladder with given values of θ_0 and m_0 for the dimer with $n=0$ gives this energy as a quadratic form with variables θ_0 and m_0. Then the analysis of stability can be done using the equation for $n=0$, which by use of the relations $\theta_0=\theta_p+\theta_q$ and $m_0=m_p+m_q=A\ \theta_p+\theta_q/A$ can be written as

$$(Je^p-J-\kappa)\theta_p-(Je^q+J+\kappa)\theta_q+\partial\Delta W/2\partial\theta_0=0,$$

$$(Je^p+J+\kappa)A\theta_p-(Je^q-J-\kappa)\theta_q/A-\partial\Delta W/2\partial m_0=0. \tag{8}$$

These equations have nontrivial solutions only for some particular value of the magnetic field, which defines the instability field H_c. Finally the critical field, as well as the connections between variables, θ_p and θ_q, can be obtained through the solvability condition for Eqs. (8).

Using this procedure for the regular ladder ($\Delta W=0$), the critical field to a first approximation in the small parameters is

$$H_c^2=H_1^2-2(K+\kappa\sqrt{3})^2. \tag{9}$$

Notice that this value is smaller than the volume critical field, where the difference between H_c and H_1 is proportional to the small parameter $(\kappa,k)/J$ and for a given effective anisotropy K it is an increasing function of κ. This is in good agreement with the numerical data illustrated in Fig. 2. It is also noticed that $\theta_q\ll\theta_p$, but m_p and m_q are comparable, $m_q\approx 2/(1+\sqrt{3})m_p\approx 0.73m_p$, implying that the oscillatory part of m is quickly decaying, but observable as seen in Fig. 1.

Next, this method is used to analyze the NC ladder. The minimization of ΔW with respect to the extra spin will give the simple expression for this energy in terms of θ_0 and m_0:

$$\Delta W=-\alpha(\theta_0-m_0)^2/2,\quad \alpha=H-2k-2\kappa+H^2/J. \tag{10}$$

Then the solvability condition for Eqs. (8) gives the critical field $H_c^2=2H_1^2/5$, which is significantly below H_1. For this case again $\theta_q\ll\theta_p$ and $m_p\approx m_q$, but with opposite signs, $m_q\approx -m_p/(1+\sqrt{3})\approx -0.37m_p$, which is also seen in the inset to Fig. 1.

Previously only first-order spin-flop transitions were known to exist for the model Hamiltonian given by Eq. (1) and only first-order transitions were previously reported[6–8] in the spin-chain literature. The order of the transition can be clearly demonstrated through a plot of S_z versus H, which goes to zero near the critical field for the second-order transition. As was mentioned above, the first-order transition appears for the RL and NC ladders with pure single-ion anisotropy (see Fig. 2); however, for the case of exchange anisotropy the transition becomes second order and the amplitude of the nonuniform distribution goes to zero at the transition point. Therefore, for some finite value of the ratio of the exchange and single-ion anisotropy constants there is a tricritical point. Taking into account that the magnetization per dimer, $m_{z,n}=\theta_n\ m_n$, is quadratic over linear variables and that the value of $S_z=0$ in the Néel state, it is remarked that S_z is the square of the order parameter. For a description of the phase diagram with a tricritical point one needs a Landau expansion of the system energy containing the order parameter to the sixth power:

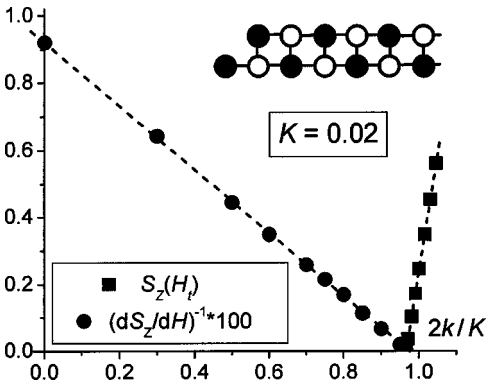

FIG. 3. Plots of S_z and $(dS_z/dH)^{-1}$ vs k/K for $K=0.02$ on a NC ladder shown in the inset.

$$F=-(H-H_c)S_z+\frac{1}{2}\beta S_z^2+\frac{1}{3}\gamma S_z^3, \tag{11}$$

where the first term has been obtained analytically from the linear approximation, with β and γ being phenomenological coefficients. If the coefficients are known, then further analysis is simple: for $\beta>0$ the transition is second order with an S_z magnetic field dependence $S_z=(H-H_c)/\beta$. For $\beta<0$ and $\gamma>0$ the transition will be first order with nonzero value of total spin at the transition point given by $S_z^t=3|\beta|/4\gamma$. Therefore, both quantities $(dS_z/dH)^{-1}=\beta$ for $\beta>0$ and S_z^t at $\beta<0$ are proportional to the value of $|\beta|$. Near the tricritical point $\beta\to 0$ and plots of these quantities versus k/K at a constant value of the effective uniaxial anisotropy $K=2k+3\kappa$ can be extrapolated to zero to obtain the anisotropy value at the tricritical point. This graphical analysis for the NC ladder is illustrated in Fig. 3 and gives a tricritical point at the anisotropy ratio $2k/K\approx 0.966$. For the RL structure the tricritical point is determined to be at $2k/K\approx 0.78$, and the characteristic square root dependence is also seen in Fig. 2. Thus, the second-order transition takes place for almost all values of anisotropies with $k>0$ and $\kappa>0$. For a small AFM particle bounded on all sides the situation becomes more complicated. It is more or less obvious that the edge-noncompensated spins are the main sources of pinning for nonuniform states. Using numerical methods only for square particles with NC edge spins, it is remarked that the transition is always second order for both single-ion and exchange anisotropy.

These analytical and numerical calculations show that there is a rich array of ground-state structures in AFM systems of dimensionality greater than 1. In addition to the bulk

spin-flop state there are localized surface states of both the spin flop and discommensuration type depending on whether or not the surface is compensated. The transition to these states can be either first or second order, which can be determined from the $S_z(H)$ behavior near the transition point. The second-order transition is realized for all systems considered with exchange anisotropy, as well as for the 2D model of a square particle with NC edge spin. As the second-order transition is approached, the amplitude of the nonuniform spin distribution goes to zero at the transition point, producing a state which is neither SSF nor discommensuration, but rather a slightly broken Néel state. For spin ladder models, RL and NC structures with combined anisotropy, a tricritical point on the $(H,k/K)$ plane is present.

The two-dimensional square planar structure can be realized in layered classical AFM-like Mn(II)-halide compounds which can be approximately modeled by a classical spin.[9] Another interesting possibility is provided by arrays of nanostructures such as magnetic dots,[10] which can be synthesized in square planar or ladder structures. Owing to the magnetostatic interaction between the dots, these have AFM ordering of the dot's magnetic moment at zero field.[11] These effects could possibly be observed in these magnetic dot arrays, where the effective anisotropy for one dot as well as the anisotropy of the dot interaction (the analog of single-ion or exchange interaction) can be adjusted by modification of the geometries of the dot and lattice, respectively.

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*Corresponding author. FAX: 406-994 4452. Electronic address: zaspel@physics.montana.edu

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