Field and Surface Effects on the Ground State of Antiferromagnetic Systems

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The field dependence of the ground state of semi-infinite uniaxial antiferromagnets and films is investigated in terms of a two-dimensional area-preserving map, where the surfaces are introduced as appropriate boundary conditions. For the film, the ground state is calculated for any value of the number of planes in a very rapid and accurate way. For the semi-infinite system, we show that the so-called surface spin-flop state does not exist; by energetic arguments, we find that the instability of the antiferromagnetic configuration leads to an interchange of the sublattices. A nonhomogeneous ground state is instead found in the bulk spin-flop phase.

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It is well known [1] that when a magnetic field applied along the easy axis of an infinitely extended antiferromagnet exceeds a critical value, $H_{\rm BSF}$, there occurs a sudden nearly $\pi/2$ rotation of the sublattices vectors that drives the system into the so-called bulk "spin-flop" phase [Fig. 1(a)]. On the other hand, films and semi-infinite systems can have an inhomogeneous ground state owing to the presence of surfaces. The uniaxial antiferromagnet with a field H along the easy axis has been largely studied since the prediction of a surface spin-flop (SSF) state in the semi-infinite system [2–4] [Fig. 1(b)]. A surface phase transition was suggested [5,6] by the full softening, when H tends to a critical field H_S, of the surface mode calculated assuming an antiferromagnetic ground state with the surface spins antiparallel to H ($\mathrm{AF}_{\uparrow\downarrow}$) [Fig. 1(c)]. In the SSF state, the spins turn by roughly $\pi/2$ close to the surface, and asymptotically reach the $\mathrm{AF}_{\uparrow\downarrow}$ configuration in the bulk. According to Keffer and Chow [3], for $H \to H_{\rm BSF} \simeq \sqrt{2}H_S$, the SSF region expands occupying continuously stable equilibrium states, until the usual bulk spin-flop (BSF) state is reached. A doubtful point of this description is that the BSF phase is achieved in different ways according to whether the surface spins are antiparallel or parallel ($\mathrm{AF}_{\uparrow\uparrow}$) [Fig. 1(d)] to H. In the latter case the only instability is announced for $H = H_{\rm BSF}$, like in the infinite system where a bulk mode softens [5,6]. An analogous behavior of the excitations is found for a film with an even number of planes N: one has two surface modes and for $H = H_S$ only the one localized at the surface with the spins antiparallel to H shows a complete softening [7]. For such a system, the nonuniform ground state was previously investigated [8] by means of a numerical self-consistent approach in the absence of anisotropy, for which $H_S = H_{\rm BSF} = 0^+$. Even though the obtained results are well founded, this technique presents some drawbacks: there is no reliability to obtain the absolute minimum and in proximity of phase transitions the convergence is very slow [9].

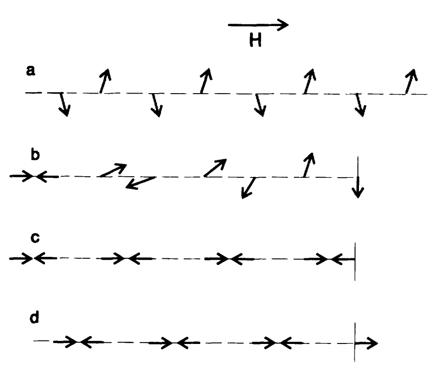

FIG. 1. (a) The (uniform) bulk spin-flop stucture, stable for $H > H_{\rm BSF}$, in a translationally invariant system. (b) The surface spin-flop structure, which was suggested to be metastable for $H_S < H < H_{\rm BSF}$ in Refs. [2,3]. (c) The metastable structure $\mathrm{AF}_{\uparrow\downarrow}$ for $H < H_S$. (d) The ground state $\mathrm{AF}_{\uparrow\uparrow}$ for $H < H_{\rm BSF}$.

In this Letter we present a study where the determination of the ground state is formulated as a two-dimensional area-preserving map [10–12] and the surfaces are introduced with appropriate boundary conditions [13]. The mapping is characterized by the fixed points and the orbits of the infinite system, but the spin structures relevant for our problem are only those which satisfy the previous conditions, a very selective constraint. In this way we are able to calculate the nonuniform ground state configuration very rapidly and with great precision, getting new and very interesting results. In particular, for the semi-infinite system we find that the SSF state does not exist, and the instability for $H \geq H_S$ is ascribed to a Bloch wall nucleation with a consequent interchange of the sublattices, making the surface spins parallel to H. On the contrary, we show that a nonuniform ground state is present in the BSF phase. Finally, by our method we also get the nonuniform ground state for a film, and in the zero anisotropy limit we recover the results obtained in Ref. [8].

Following Mills [2] and Keffer and Chow [3], the energy is given by

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$$\frac{E}{N_{\parallel}S} = \sum_n [H_E \cos(\phi_n - \phi_{n-1}) - H_A \cos^2\phi_n - 2H\cos\phi_n], \tag{1}$$

where $H_E = zJS$, $H_A = 2KS$. J and K are the exchange and anisotropy costants, respectively; n is the plane index, with $n \in \mathbb{Z}$ for the infinite system, $n \in \mathbb{N}$ for the semi-infinite one, and $n = 1, 2, \ldots, N$ for a film. The energy can be written as Eq. (1) provided that the system can be subdivided in planes parallel to the surfaces with all the $N_{\parallel}$ spins ferromagnetically aligned. Introducing $\xi = H/H_E$ and $\zeta = H_A/H_E$, the equilibrium configurations are given by

$$\sin(\phi_{n+1} - \phi_n) + \sin(\phi_{n-1} - \phi_n) + 2\xi\sin\phi_n + \zeta\sin 2\phi_n = 0 \quad \text{(infinite)}, \tag{2a}$$

$$\sin(\phi_{n+1} - \phi_n) + (1 - \delta_{1,n})\sin(\phi_{n-1} - \phi_n) + 2\xi\sin\phi_n + \zeta\sin 2\phi_n = 0 \quad \text{(semi-infinite)}, \tag{2b}$$

$$(1 - \delta_{N,n})\sin(\phi_{n+1} - \phi_n) + (1 - \delta_{1,n})\sin(\phi_{n-1} - \phi_n) + 2\xi\sin\phi_n + \zeta\sin 2\phi_n = 0 \quad \text{(film)}. \tag{2c}$$

We first consider Eq. (2a), valid for the infinite system. Introducing [12] $s_n = \sin(\phi_n - \phi_{n-1})$, it can be written as a two-dimensional mapping

$$\phi_{n+1} = \phi_n + \sin^{-1}(s_{n+1}), \tag{3a}$$

$$s_{n+1} = s_n - 2\xi\sin\phi_n - \zeta\sin 2\phi_n. \tag{3b}$$

Solutions to Eqs. (3) can be obtained choosing a pair (ϕ_i, s_i) and iterating the equations [14]. This mapping is area preserving (because the Jacobian is $J \equiv 1$) and invariant with respect to the transformation $\phi \to -\phi$, $s \to -s$. The fixed points correspond to uniform ground states and, owing to the antiferromagnetic nature of the system, are second-order ones. They are $P_-^{\rm AF} = (0, 0)$, $P_+^{\rm AF} = (\pi, 0)$, $P_-^{\rm BSF} = (-\overline{\phi}, -\sin\overline{\phi})$, $P_+^{\rm BSF} = (\overline{\phi}, \sin\overline{\phi})$, where $\cos\overline{\phi} = \xi/(2-\zeta)$. Carrying out a linear stability analysis, it results that the AF fixed points are hyperbolic for $\xi^2 \le 2\zeta + \zeta^2 \equiv (H_{\rm BSF}/H_E)^2$ and elliptic otherwise. On the contrary, the BSF ones are hyperbolic for $\xi^2 \ge 2\zeta - 3\zeta^2$, and elliptic for lower fields [15]. In general, from the mapping (3) we obtain phase portraits (see Fig. 2) characterized by inflowing and outflowing orbits connecting the hyperbolic fixed points, elliptic orbits which encircle the homonymous fixed points, and, finally, nonhomotopic to zero curves. We remind that energetically stable states corresponds to topologically unstable (i.e., hyperbolic) fixed points [10,11].

In order to study the semi-infinite system and the film, we have to take into account the presence of surfaces given by the terms with the Kroneker's δ in Eqs. (2b) and (2c). For the semi-infinite system, this is equivalent to introducing a fictitious plane for $n = 0$, so that

$$s_1 = \sin(\phi_1 - \phi_0) = 0 \tag{4}$$

becomes the boundary condition. For the film, two fictitious planes must be introduced for $n = 0$ and $n = N+1$, so that the boundary conditions are now given by

$$s_1 = \sin(\phi_1 - \phi_0) = 0, \qquad s_{N+1} = \sin(\phi_{N+1} - \phi_N) = 0. \tag{5}$$

Among all trajectories obtained from (3), only those which satisfy (4) or (5) represent equilibrium configurations. For a film, the physical trajectories must have two intersections separated by exactly N steps of the recursive mapping, a very selective condition.

For $H < H_S$ (i.e., $\xi < \sqrt{\zeta + \zeta^2}$), the phase portrait shows nothing special; the ground state for all systems is the AF one.

More interesting is the analysis for $\sqrt{\zeta + \zeta^2} < \xi < \sqrt{2\zeta - 3\zeta^2}$, the relevant region for the SSF state. In terms of mapping (3) this state exists if and only if the inflowing orbits (converging to $P_-^{\rm AF}$ or $P_+^{\rm AF}$) cross the

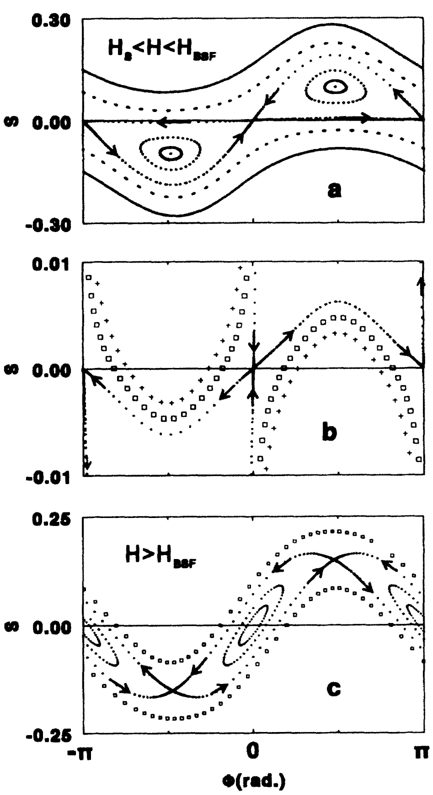

FIG. 2. Phase portraits obtained from mapping (3) for $\zeta = 0.009$. The arrows denote the inflowing and outflowing (IO) orbits associated to the hyperbolic fixed points. (a) $\xi = 0.0975$. (b) Enlargement of (a) for $s \simeq 0$. Note that the IO orbits do not intersect the boundary conditions line $s = 0$, i.e., the SSF state cannot exist. In addition, two nonhomotopic to zero curves are shown: the one with square symbols satisfies Eq. (5) for an $N = 26$ film. (c) $\xi = 0.15$. In this case the IO orbits do intersect the $s = 0$ line, giving rise to a nonuniform surface configuration. The nonhomotopic to zero curve pertinent to an $N = 26$ film is also reported.

line $s = 0$ at $\phi \neq 0, \pi$. From Figs. 2(a) and 2(b), where a typical phase portrait is shown, it is apparent that this does not happen. Consequently, *the SSF state is not an equilibrium one and it can never be the ground state* [16]. Another characteristic useful to stress is that an infinite number of nonhomotopic to zero curves cross the $s = 0$ line. These trajectories present a fundamental relevance for the films, since they are the only ones crossing the $s = 0$ line in two different points which are not fixed points [see Fig. 2(b)]. Furthermore, there is only one trajectory which, starting from $s_1 = 0$, is able to satisfy the condition $s_{N+1} = 0$, being always confined in the gain energy region of the map [17]. If N is even, the corresponding energy is smaller than the AF one and we obtain the new ground state which is reported in Fig. 3. For $\xi > \sqrt{2\zeta + \zeta^2}$ the fixed points $P_{\pm}^{\rm AF}$ are elliptic while $P_{\pm}^{\rm BSF}$ are hyperbolic. The latter ones do not lie on the boundary condition line and therefore the uniform BSF configuration cannot be an equilibrium one neither for the semi-infinite system, nor for the film. In fact, from Fig. 2(c), where the results obtained for a moderate field ($\xi = 0.15$) are reported, it is possible to note that in the semi-infinite system we have a nonuniform surface configuration, because inflowing orbits intersect the $s = 0$ line. Again, the nonhomotopic to zero curves provide the ground state for films [see Fig. 2(c)]. We note that increasing N we must consider trajectories closer and closer to $P_{-}^{\rm BSF}$ and consequently we have that the middle planes present a configuration very similar to the BSF one (see Fig. 3). In the zero anisotropy limit, and for $\xi = 0.1925$, we recover the results of Ref. [8]. It is worthwhile to note that our method is able to determine the ground state in a very rapid and accurate way (within machine double precision) [18].

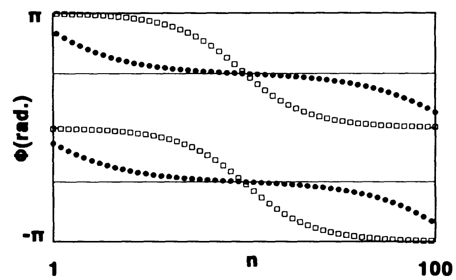

FIG. 3. Ground state configuration for an $N = 100$ film with $\zeta = 0.009$. Open squares refer to $\xi = 0.0975$, full circles to $\xi = 0.15$. The solid horizontal lines show the canting angles for an infinite system in the bulk spin-flop phase for $\xi = 0.15$.

A further increase of the field makes the analysis of the phase portait more intricate because there is evidence for chaotic behavior [7], and the problem will be discussed in a future paper.

At this point, for $H > H_S$, we have determined the ground state for a film with a generic number of planes N, but the problem is still open for the semi-infinite system. We know from the analysis of the excitations [5,6] that in this range of magnetic field the $\mathrm{AF}_{\uparrow\downarrow}$ state is no more a stable configuration, but we have also shown that the predicted SSF state, or any other nonuniform configuration, cannot exist. This will be confirmed below by an energetic, though approximate, argument.

The last term in the bracket of Eq. (1) is difficult to handle in a semi-infinite system because it is an infinite sum, the general term of which does not go to a limit at infinity. To solve this problem, one can rewrite Eq. (1) as

$$\frac{E}{N_{\|}S} = \sum_{n=1}^{\infty}[H_E\cos(\phi_n - \phi_{n-1}) - H_A\cos^2\phi_n] - H\sum_{n=1}^{\infty}[\cos\phi_n + \cos\phi_{n+1}] - H\cos\phi_1. \tag{6}$$

Only systems in which $\cos\phi_n + \cos\phi_{n+1}$ goes to 0 when n goes to ∞ will be considered. It is so both in the $\mathrm{AF}_{\uparrow\downarrow}$ state and in the $\mathrm{AF}_{\uparrow\uparrow}$ state, since $\cos\phi_n + \cos\phi_{n+1}$ is identically zero for any n in both cases. Therefore, in both states, the Zeeman energy reduces to $-H\cos\phi_1$ which is minimum in the $\mathrm{AF}_{\uparrow\uparrow}$ state, but maximum in the $\mathrm{AF}_{\uparrow\downarrow}$ state.

Since the $\mathrm{AF}_{\uparrow\downarrow}$ state is not the ground state, but it is stable with respect to spin waves for $H < H_S$, we conclude that it is metastable for $H < H_S$.

However, for $H_S < H < H_{\rm BSF}$ the situation is different. We are going to argue that the $\mathrm{AF}_{\uparrow\downarrow}$ state is unstable with respect to the formation of a Bloch wall which forms at the surface and sinks into the bulk, thus transforming the system in its $\mathrm{AF}_{\uparrow\uparrow}$ state. We start from Eq. (6). As explained in Ref. [19], it is possible to eliminate the even spins $\mathbf{S}_{2n}$ by a decimation procedure, which in the present case is to be performed at zero temperature. For small H_A and H, and to lowest order in H_A and H, the result is

$$\frac{E}{N_{\|}S} = \sum_{n=1}^{\infty}\left[\frac{H_E}{4}(\phi_{2n-1} - \phi_{2n+1})^2 - \left(2H_A - \frac{H^2}{H_E}\right)\cos^2\phi_{2n-1}\right] - H\cos\phi_1, \tag{7}$$

where the last term is due to the existence of a free surface at $n = 1$. The other terms can be derived as in Ref. [19]. Equation (7) has the well-known sine-Gordon form. However, this is only correct to lowest order. This approximation is questionable near the bulk spin-flop transition, where the coefficient of $\cos^2\phi$ becomes small, so that higher order terms should be introduced. In order to investigate the transformation of the $\mathrm{AF}_{\uparrow\downarrow}$ state into the $\mathrm{AF}_{\uparrow\uparrow}$ state, and also the stability of the SSF state, it is of interest to minimize this energy for a given value of ϕ_1 and for fixed

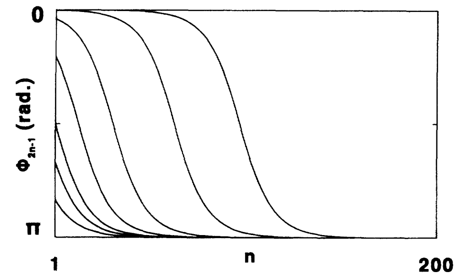

FIG. 4. Succession of nonequilibrium configurations for the semi-infinite system, starting from the unstable $AF_{\uparrow\downarrow}$ state. The energy decreases as $\phi_1 \to 0$. The maximum energy gain is achieved when the Bloch wall is pushed to an infinite distance from the surface, thus accomplishing the stable $AF_{\uparrow\uparrow}$ ground state.

$\cos\phi_{2n+1} = -1$ at $n = \infty$. The solution of this problem is given by the Euler-Lagrange equation of a domain wall

$$\frac{H_E}{2}[2\phi_{2n+1} - \phi_{2n+3} - \phi_{2n-1}] + \left[2H_A - \frac{H^2}{H_E}\right]\sin 2\phi_{2n+1} = 0, \quad (8)$$

which in the continuum approximation reduces to the pendulum equation. Inserting its solution into (7), one obtains, after some algebra, the energy as

$$\frac{E}{N_{\parallel}S} = \sqrt{(2H_AH_E - H^2)}(\cos\phi_1 + 1) - H\cos\phi_1. \quad (9)$$

We now want to minimize this energy with respect to ϕ_1. For weak fields, the coefficient of $\cos\phi_1$ is positive, so that the energy presents a minimum for $\cos\phi_1 = -1$ or $\phi_1 = \pi$. Thus, the $AF_{\uparrow\downarrow}$ state is a metastable one, in agreement with the spin wave argument. In other words, if a Bloch wall is introduced into the $AF_{\uparrow\downarrow}$ state from the surface, the system expels it through the surface. This occurs if $H^2 < H_AH_E$, i.e., if $H \lesssim H_{\rm BSF}/\sqrt{2}$. On the other hand, if $H_{\rm BSF}/\sqrt{2} \lesssim H < H_{\rm BSF}$, the energy (9) is a decreasing function of ϕ_1 and it is therefore minimum for $\phi_1 = 0$. In this case, if a Bloch wall is introduced into the $AF_{\uparrow\downarrow}$ state from the surface, the system swallows it and transforms progressively into the $AF_{\uparrow\uparrow}$ state (see Fig. 4). In particular, the SSF state corresponds to $\cos\phi_1 = 0$, and it is unstable, in agreement with the mapping argument. This description is corroborated by the results obtained considering the case of a surface anisotropy smaller than the bulk one [7]. We have for $H \gtrsim H_S$ a metastable surface canted state but increasing H this configuration disappears when its energy exceeds the energy of the Bloch wall.

Concerning the relevance of our results for real systems, we remember that many different materials can be described by Eq. (1). This depends on the crystal structure and surface orientation: fcc lattices like NiO, MnO, and CoO [20] with (111) surfaces and bcc lattices as MnF_2 and FeF_2 [21] with (001) surfaces. Also, superlattices made of ferromagnetic films which are antiferromagnetically coupled [8,22] appear to be suitable for the purpose.

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[1] L. Néel, Ann. Phys. (Paris) **5**, 232 (1936).
[2] D.L. Mills, Phys. Rev. Lett. **20**, 18 (1968).
[3] F. Keffer and H. Chow, Phys. Rev. Lett. **31**, 1061 (1973).
[4] S.P. Vernon, R.W. Sanders, and A.R. King, Phys. Rev. B **17**, 1460 (1978).
[5] D.L. Mills and W.M. Saslow, Phys. Rev. **171**, 488 (1968); **176**, 760(E) (1968).
[6] R.E. De Wames and T. Wolfram, Phys. Rev. **185**, 752 (1969).
[7] L. Trallori, thesis (unpublished).
[8] F.C. Nörtemann *et al.*, Phys. Rev. B **46**, 10 847 (1992).
[9] J.G. LePage and R.E. Camley, Phys. Rev. Lett. **65**, 1152 (1990).
[10] S. Aubry, in *Solitons and Condensed Matter Physics*, edited by A.R. Bishop and T. Schneider (Springer, New York, 1979).
[11] P. Bak, Phys. Rev. Lett. **46**, 791 (1981).
[12] P.I. Belorov *et al.*, Zh. Eksp. Teor. Fiz. **87**, 310 (1984) [Sov. Phys. JETP **60**, 180 (1984)].
[13] R. Pandit and M. Wortis, Phys. Rev. B **25**, 3226 (1982).
[14] The presence of the $\sin^{-1}$ function implies that $\phi_{n+1} = \phi_n - \nu\pi + (-1)^\nu\psi$ with $\nu = 0, 1$. A local minimization criterion implies that ν is an invariant of the mapping and for $\zeta \ll \xi \ll 1$ it is $\nu = 1$ (see Ref. [12]).
[15] The narrow window of ξ where all the fixed points are hyperbolic is connected with the metastability region in the first order BSF phase transition.
[16] The prediction of the SSF state in Refs. [2,3] is not due to the continuum approximation, but to an inconsistent procedure for the boundary conditions. The correct treatment requires that the solution of the equation for $n > 1$ satisfies the equation for $n = 1$, while in Ref. [3] the condition $\phi_1 = \pi/2$ was imposed.
[17] The analysis of the energy of the nonuniform configurations shows that there is a gain with respect to the $AF_{\downarrow\uparrow}$ state only in the region of the map with $\phi_{2n-1} \in [0, \pi]$ and $s_{2n-1} < 0$.
[18] Such a high precision turns out to be necessary in order to obtain in the spectrum of excitations a zero frequency Goldstone mode within at least six significant figures [L. Trallori *et al.* (unpublished)].
[19] J. Villain and J.M. Loveluck, J. Phys. (Paris) **38**, L77 (1977).
[20] J.A. Borchers *et al.*, Phys. Rev. Lett. **70**, 1878 (1993).
[21] C.A. Ramos *et al.*, Phys. Rev. Lett. **65**, 2913 (1990).
[22] Z.Q. Qiu *et al.*, Phys. Rev. B **45**, 2252 (1992).