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LETTER TO THE EDITOR

Field dependence of the magnetic susceptibility of an Fe/Cr(211) superlattice: effect of discreteness and chaos

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Abstract. Recent experimental measurements on Fe/Cr(211) superlattices (Wang R W *et al* 1994 *Phys. Rev. Lett.* 72 920) show several peaks in the field dependence of magnetic susceptibility. We show that the presence of these peaks is due to the chaotic nature of the two-dimensional area-preserving map by which we determine the equilibrium configuration of the system. We find a large number of metastable states and that the ground state changes abruptly with increasing field, showing the fundamental role played by the discreteness of the lattice.

Multilayers composed of ferromagnetic (FM) films antiferromagnetically coupled across non-magnetic spacers have received great attention since the discovery of the giant magnetoresistance effect, which is characterized by an electrical resistance in which the parallel orientation of the magnetic films is found to be up to 60% lower than the orientation pertinent to the antiparallel orientation [1, 2]. These two collinear configurations are the only ones commonly considered in the theoretical analysis, even if, for magnetic fields lower than the saturation value, one can have non-uniform ground states owing to the lack of translational invariance; on the other hand, all the experimentally measurable quantities are strongly dependent on the spin configuration, so that an exact knowledge of the ground state is of great importance.

Recently, Fe/Cr(211) single-crystal samples with an uniaxial in-plane anisotropy have been grown using a sputtering technique. For a suitable choice of the Cr thickness ($t = 11 \text{ \AA}$), the interlayer coupling is antiferromagnetic (AF) and the system becomes isomorphic to a classical two-sublattice antiferromagnet (like MnF_2) with FM planes antiferromagnetically coupled [3]. Very recently, magnetic measurements performed on such a system with an even number, N , of Fe films have shown two main peaks in the magnetic susceptibility in correspondence to the so-called surface (H_S) and bulk (H_B) spin-flop transitions and other minor peaks in the intermediate region [4]. This behaviour was explained using a numerical self-consistent approach, due to jumps at a Bloch wall which forms at the surface at $H = H_S$, and proceeds, for growing fields, in the interior of the sample until the symmetric bulk spin-flop phase (BSF) is reached [4].

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In this letter we study the same problem in terms of a two-dimensional area-preserving map, where the surfaces are introduced as appropriate boundary conditions. This approach was introduced in [5]; it allows rapid, exact, calculation of the ground state, and a complete description of the equilibrium configurations space, which turns out to be essential in the present case, in order to obtain a clear comprehension of the observed data.

The Fe/Cr(211) crystal is characterized by a high value of the uniaxial anisotropy H_A with respect to the exchange interaction H_E (at variance with the situation described in [5]), which determines a clearly chaotic behaviour of the map phase portrait in the interesting field region; as a consequence, a large number of metastable configurations exist and we find a non-continuous evolution of the ground state at the varying of the field H . The existence of these metastable states and the non-continuous H dependence of the ground state emphasizes the effect of the discreteness of the lattice. In fact, owing to the high value of the ratio H_A/H_E , the spin configuration of all these states, obtained for a fixed H , is well represented by a lattice-pinned narrow π -domain wall whose centre is always located in between the sites of the crystal, in order to minimize the anisotropy energy.

Let us start from the expression of the energy of the system in the mean field approximation. This energy reduces because of the strong ferromagnetic coupling and the translational invariance in the plane of the Fe films, to the energy of a classical spin chain given by

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

where $H_E = zJS$, $H_A = 2KS$, $n = 1, \dots, N$ is the index plane, J and K are the exchange and anisotropy constants, respectively, and z is the number of nearest neighbours; we assume $H_E = 2.0$ kG as in [4] and $H_A = 0.5$ kG, so that the system presents a high value of the ratio H_A/H_E with respect to other uniaxial antiferromagnets [6]. The limit $H_A/H_E \ll 1$ has been studied in [5] and—as it will be shown—a completely different behaviour of the susceptibility is found.

Now, let us explain very briefly the method, referring to [5] for more details. All the equilibrium configurations are obtained by the condition $\partial E/\partial \phi_n = 0$, which gives, for an N -plane film

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

where $\xi = H/H_E$ and $\zeta = H_A/H_E$. Introducing the quantity $s_n = \sin(\phi_n - \phi_{n-1})$, the previous equations can be rewritten as a two-dimensional area-preserving map

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

$$s_{n+1} = s_n - 2\xi \sin \phi_n - \zeta \sin 2\phi_n \quad (3b)$$

and $s_1 = s_N = 0$ as boundary conditions.

Let us recall the values of H which define the different phases. For an infinitely extended antiferromagnet, the application of a sufficiently strong field produces a first-order phase transition which drives the system in the so-called bulk spin-flop phase. The AF and BSF phases have the same energy at the field $H = H_B = (2H_A H_E - H_A^2)^{1/2}$, and the lower and upper boundaries of the metastability region are $H = 2H_E H_A \pm H_A^2 / (2H_A H_E + H_A^2)^{1/2}$ [7]. On the other hand, for a film with an even number of layers, we know that the AF configuration is no more stable for $H > H'_S = (H_A H_E + H_A^2)^{1/2}$ and that the system also goes to a non-collinear ground state via a first-order phase transition. The field of energetic equivalence between the AF and the canted configuration is $H_S < H'_S$, and for our parameters it is numerically found to be $H_S = 0.93$ kG.

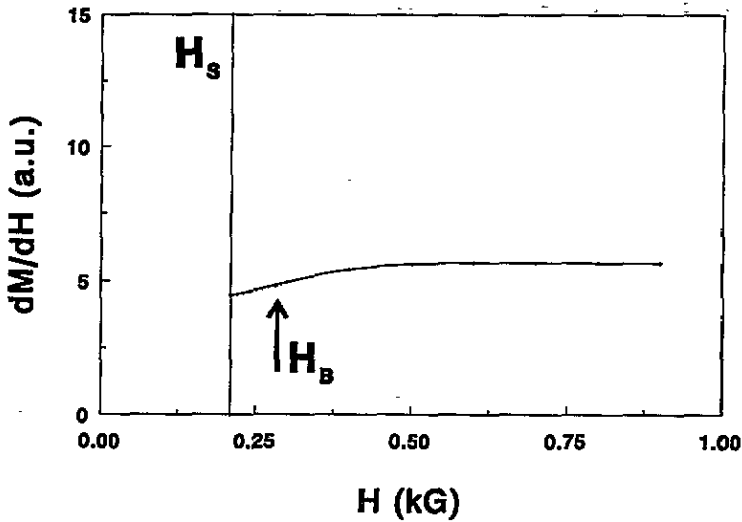


Figure 1. Field dependence of the magnetic susceptibility for $H_E = 2$ kG, $H_A = 0.02$ kG, $N = 22$. Only one peak at $H = H_S$ ($H_S = 0.20$ kG) is present.

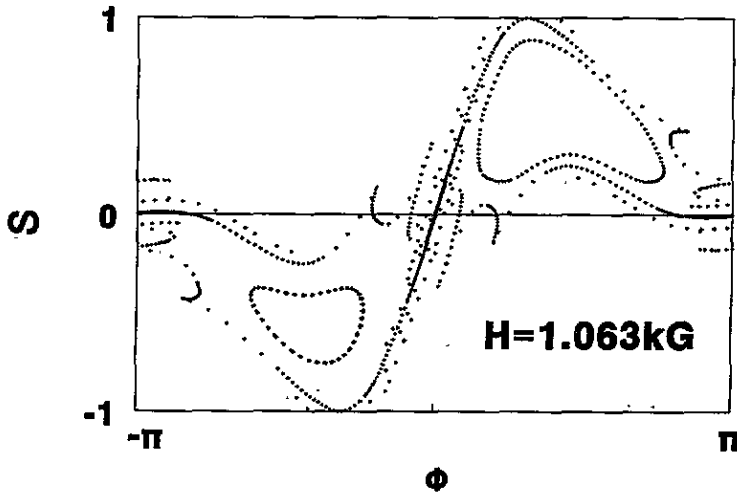


Figure 2. Map phase portrait for values of the parameter pertinent to the Fe/Cr(211) superlattices ($H_E = 2$ kG, $H_A = 0.5$ kG).

To elucidate the non-chaotic case, let us refer again to [5]. The phase diagram is very regular and this is reflected in the behaviour—shown in figure 1—of the susceptibility $\chi(H) \equiv dM/dH$. Non-collinear configurations are found by a sampling of the axis $s = 0$ and searching for the initial condition ϕ_1 which, through iteration of the map, gives $s_{N+1} = 0$. It is important to remark that the canted configuration is always unique and symmetric with respect to the middle of the sample, and it evolves in a continuous way at the increasing of the field. As a consequence, only one peak in the susceptibility is present, corresponding to the transition at $H = H_S$. For $H > H_S$, $\chi(H)$ goes with continuity to the constant value of the infinitely extended system. Thus, the behaviour of the system is

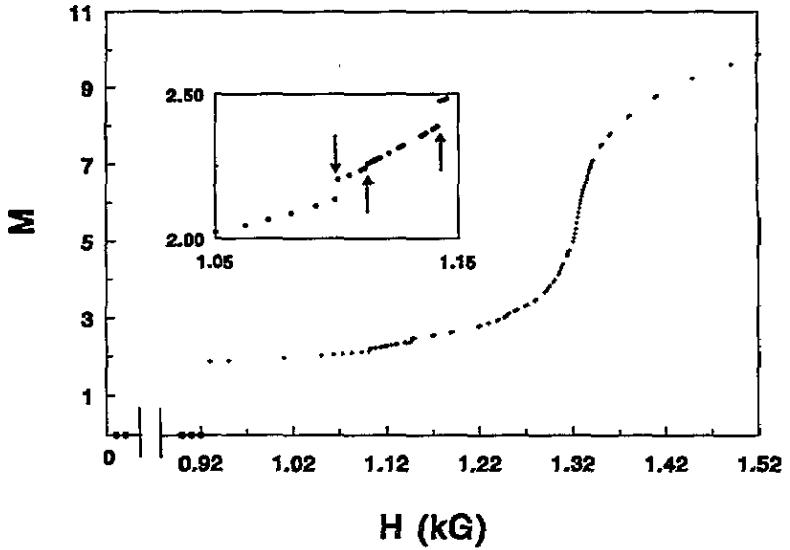


Figure 3. Field dependence of the magnetization for a $N = 22$ Fe/Cr(211) superlattice. The inset shows more clearly some of the jumps in the intermediate region between H_S and H_B . The saturation value, reached for $H = 3.5$ kG, is $M = M_{sat} = 22$.

fully analogous to the one of an infinitely extended antiferromagnet, the only significant difference being the position of the peak at H_S rather than at H_B .

The situation is completely different for values of the parameters relevant to the experimental system under study. The map phase portrait (see figure 2) clearly shows chaotic behaviour: the inflowing orbit (i.e. the stable manifold) in the fixed point $P_{AF}^+ = (-\pi, 0)$ does not coincide with the outflowing orbit (i.e. the unstable manifold) from the other fixed point $P_{AF}^- = (0, 0)$, but they cross in an infinite number of so-called heteroclinic points. Because of the infinite number of oscillations of the inflowing orbit, there are several intersections with the boundary condition axis $s = 0$. So, these intersections define distinct regions which—in principle—the angle ϕ_1 of an equilibrium configuration can belong to. This is exactly what happens: several stable equilibrium states coexist and the ground state is determined by the comparison of their energies. Moreover, at the growing of the field some intersections of the inflowing orbit with the $s = 0$ axis disappear, or, a previously metastable state now becomes the ground state. *This causes abrupt changes in the ground state configuration and consequently in $M(H)$* (see figure 3). These discontinuities are made more apparent in the derivative function $\chi(H)$, which is shown in figure 4, so that the chaotic nature of the mapping has a direct and visible consequence on this measurable quantity.

Many spikes are observed in correspondence with the steps of $M(H)$. We would like to stress that the peak at H_B has a finite width [8], because it is associated with a very sharp variation of the convexity of the magnetization profile rather than to a jump as in the other peaks, and that the *shoulder* for slightly higher fields than H_B has been experimentally observed in Kerr measurements [4]. The spikes in the intermediate region between H_S and H_B should be, in principle, Dirac- δ functions, and their height and width is only due to the finiteness of the increment (10^{-3} kG) we use to calculate $M(H)$.

We would like to remark that the map method allows us to have a clear picture of the phase-space complexity of the system and to foresee *a priori* the possibility of the

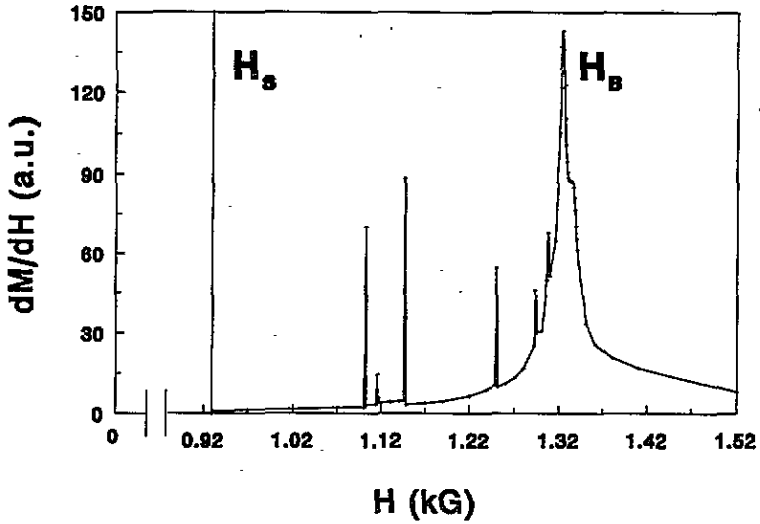


Figure 4. Field dependence of the magnetic susceptibility for an $N = 22$ Fe/Cr(211) superlattice, obtained by numerical derivation of the magnetization data reported in figure 3.

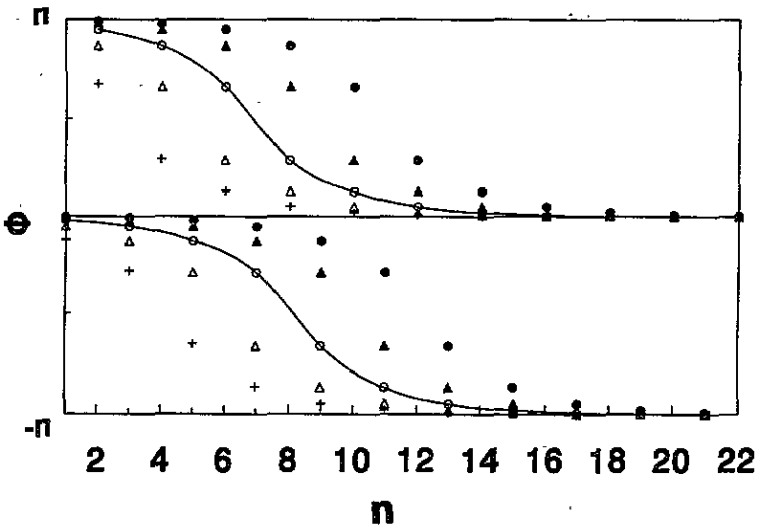


Figure 5. Spin configuration on both the sublattices of all the stable equilibrium states for $H = 1.116$ kG. The solid line (a guide for the eye) refers to the ground state. The configuration with full circles is the one symmetric with respect to the middle of the sample.

existence of a large number of metastable configurations. Other self-consistent recursive methods do not allow clear visualization of such complexity [4, 8], and they have to be handled with great care in such a situation because they can converge to a metastable configuration rather than to the real ground state, without giving any hint of the presence of other equilibrium configurations. Moreover, the map method also permits calculation of the equilibrium configurations with great rapidity and precision for values of the number of planes (N) higher than the one investigated in the present case ($N = 22$), without demanding

calculations from the perspective of numerical precision [8], even if, due to the presence of chaos (i.e. of a positive Lyapunov exponent), the amplification of an uncertainty on the initial condition ϕ_1 makes the solution of the problem difficult for very high values of N .

Let us now proceed to show the importance of the lattice discreteness, in the case of high anisotropy. The relationship between the description obtained from the map phase portrait and the discreteness of the lattice can be understood from figure 5, where all the stable equilibrium spin configurations for $H = 1.116$ kG are reported. It is evident that each of these non-uniform stable states looks like a π -Bloch wall whose centre is always located in between the sites of the crystal. In the case considered in [5], due to the low value of H_A , a Bloch wall nucleating at the surface is much broader than the lattice constant; in other words the anisotropy energy barrier around $\pm\pi/2$ is very low and the domain wall can continuously get into the sample, accomplishing a symmetric canted ground state, with a net energy gain with respect to the AF configuration due to the Zeeman interaction. In contrast, in the present case, the high value of H_A makes the Bloch wall very narrow and, at the same time, enhances the anisotropy energy barrier, so that continuous evolution is forbidden and a metastable configuration is found each time this barrier is located in between two sites of the crystal. Which of these metastable states results to be the ground state is a consequence of an extremely sensitive balancing between the Zeeman energy gain and the anisotropy and exchange cost. Increasing the magnetic field, the ground state evolution can be thought of as assuming successive positions of a π -Bloch wall penetrating into the sample, separated by the anisotropy energy barrier.

In conclusion, in this letter we have shown that the surprising experimental results [4] obtained for the Fe/Cr(211) susceptibility can be easily understood as a consequence of the presence of chaos in the equilibrium phase space of the system. A large number of metastable configurations exist and the ground state changes abruptly at the growing of the field. This behaviour is strictly linked to the high value of the anisotropy energy, so that the discrete nature of the lattice turns out to play a fundamental role.

We believe that further experimental investigations of systems with different ratios H_A/H_E and with a variable number of planes would be of extreme interest and would contribute to an even clearer vision of the magnetic properties of this system.

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