Theory of the Magnetization Reversal of Ultrathin Fe Films on a Cr Substrate

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A micromagnetic theory based on the thickness-averaged Landau-Lifshitz equation is proposed to describe the magnetization behavior of ultrathin Fe films on Cr. The calculations predict the appearance of an effective uniaxial anisotropy of Fe below the Cr spin-flip transition temperature when its magnetic state is characterized by a longitudinal spin density wave. This anisotropy results in a *perpendicular* coupling of Fe and Cr spins, suggesting the modification of the coercive behavior. The calculations provide a possible explanation for recently discovered anomalous magnetic properties of Fe films grown on Cr(100). [S0031-9007(97)05178-8]

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The behavior of mixed ferro/antiferromagnetic systems is a subject of interest since the discovery forty years ago of the exchange anisotropy phenomenon [1] and more recently of a variety of intriguing properties of layered magnetic sandwiches, including oscillatory exchange coupling through a nonmagnetic spacer [2–5], biquadratic coupling [6–8], and giant magnetoresistance [9]. It is well established that magnetic and transport properties of ultrathin layered systems are in a great extent governed by interface phenomena. However, an outstanding problem in the thin-film magnetism is to understand the mechanism of the influence of microscopic interfacial interactions on the macroscopic magnetic properties as exchange shift in the hysteresis loop, coercive force, and remanent magnetization. Theoretical models for exchange coupling in ferro/antiferromagnetic systems so far have concentrated on attempts to account for exchange shift and paying no attention to the coercivity and remanent magnetization behavior [10,11].

Recently, an anomalous temperature behavior of the coercivity and of the remanent magnetization of thin Fe layers grown on Cr(100) substrate has been reported [12]. Among layered materials Fe/Cr systems demonstrate a unique set of unusual physical properties such as extreme magnetoresistance [9,13], two periods in the bilinear interlayer coupling [2–5], and non-Heisenberg biquadratic coupling [6,8,14]. The most intriguing problem for Fe/Cr systems is to understand the correlation in the magnetic behavior of Fe and Cr. At the Néel temperature $T_N = 311$ K, bulk Cr is magnetically ordered as a transverse spin density wave (SDW), while below the spin-flip transition temperature $T_{SF} = 123$ K a phase transition to a longitudinal SDW is observed [15]. Temperature anomalies of macroscopic magnetic properties reported in Ref. [12] have been observed around T_{SF} and qualitatively have been associated with the change of the Cr magnetic ordering. In this Letter we propose a quantitative micromagnetic theory of the magnetization behavior of Fe films on Cr substrate. Some conclusions of the theory can be applied not only to the Fe/Cr system, but also to other layered magnetic materials.

First we offer an approach to average the Landau-Lifshitz equation over the thickness of a thin magnetic sandwich, taking into account interface/surface boundary conditions. Second, for Fe on Cr, a mechanism of the appearance of an effective uniaxial anisotropy below T_{SF} is proposed, similar to a change of the effective potential of a pendulum in a rapidly oscillating field. The calculations give the energy minimum when the wave vector of the longitudinal SDW is *perpendicular* to the Fe magnetization. The change of the ground state of Fe/Cr system below T_{SF} alters the magnetization reversal mechanism and results in a drastic increasing in the coercivity. Third, to be specific, we calculate nonuniform nucleation modes localized near the film edge and at Fe/Cr interface steps to find the coercive field below and above T_{SF}. The thickness dependence of nucleation fields allows a detailed comparison with the experiment [12].

To derive the thickness-average Landau-Lifshitz equation, we write the three-dimensional micromagnetic energy functional as the sum of volume and surface parts

$$E = \int [A(\nabla\phi)^2 + K \sin^2\phi \cos^2\phi]\, dV + \int J \cos(\phi - \psi)\, dS, \tag{1}$$

where the Fe and Cr in-plane magnetization is characterized by ϕ and ψ angles with respect to the x axis in the film plane [Fig. 1(a)], A and K are the Fe exchange and the cubic anisotropy constants, and $J > 0$ is the Fe/Cr interface exchange coupling per unit area which is expected to be antiferromagnetic. The spin distribution of Cr is assumed to be "frozen". Below we discuss this restriction.

Variational procedure for Eq. (1) leads to the volume and surface static equilibrium conditions

$$-2\delta^2\nabla^2\phi + \sin 2\phi \cos 2\phi = 0, \tag{2a}$$

$$\phi_z|_{z=0} = \frac{J}{2A}\sin(\phi - \psi), \quad \phi_z|_{z=d} = 0. \tag{2b}$$

where $\delta = \sqrt{A/K}$, and d is the Fe thickness. We seek solutions of Eq. (2a) in the form $\phi(x, y, z) = \tan^{-1}[f(\xi, \eta)g(\zeta)]$, where $\xi = x/\delta$, $\eta = y/\delta$, and

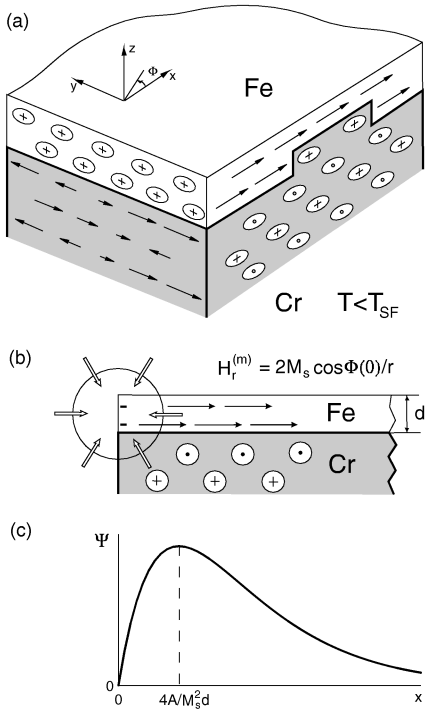

FIG. 1. (a) The ground state of Fe/Cr system below the Cr spin-flip transition temperature. 90° coupling between Fe and Cr spins results in an effective uniaxial anisotropy. (b) The magnetostatic field at the edge of a thin magnetic film. (c) The nucleation mode near the film edge.

$\zeta = z/\delta$. Then Eqs. (2) are rewritten as

$$[(f_{\xi\xi} + f_{\eta\eta})g + fg_{\zeta\zeta}](1 + f^2g^2) - 2fg[(f_\xi^2 + f_\eta^2)g^2 + f^2g_\zeta^2] - fg(1 - f^2g^2) = 0, \tag{3a}$$

$$fg_\zeta|_{\zeta=0} = \frac{J\delta}{2A}[fg\cos\psi - (1 - f^2g^2)\sin\psi],$$

$$g_\zeta|_{\zeta=d/\delta} = 0. \tag{3b}$$

Because of Lamb's remark [16] we imply $g_\zeta^2 = \beta g^2 - \alpha$ where α and β are arbitrary constants. The solution of the equation satisfying the boundary condition at the free ferromagnet surface is given by $g = \sqrt{\alpha/\beta}\cosh[\sqrt{\beta}\,(\zeta - d/\delta)]$. For thin films ($d/\delta \ll 1$) $g(0) \approx \sqrt{\alpha/\beta}$, $g_z|_{\zeta=0} \approx -\sqrt{\alpha\beta}\,d/\delta$. Integrating Eq. (3a) over the ferromagnet thickness d taking into account the boundary condition at ferro/antiferromagnet interface and introducing the variable $\Phi = \tan^{-1}[f \cdot g(0)]$, the following equation is obtained:

$$2A(\Phi_{xx} + \Phi_{yy}) - \frac{K}{2}\sin 4\Phi + \frac{J}{2d}\sin(\Phi - \psi) = 0. \tag{4}$$

This equation describes the thickness-averaged in-plane magnetization behavior of the cubic ferromagnet in contact with an antiferromagnet. Interface spins of the antiferromagnet play the role of an effective in-plane magnetic field $H_e = J/2M_sd$. In principle, this particular result might be obtained from a qualitative consideration of the equilibrium condition of a fictitious domain wall in the ferromagnet. In our case the effective magnetic field results from a ferromagnet spin variation along the z axis due to different boundary conditions at free surface and at ferro/antiferromagnetic interface. Although we concentrate here on the case of a ferro/antiferromagnetic bilayer with "frozen" spins in the antiferromagnet, this procedure can be extended for other magnetic sandwiches with flexible spin structure in all layers.

Further, we use Eq. (4) to analyze the magnetic behavior of Fe on Cr. For simplicity, we assume the Cr magnetic surface structure is given by the bulk terminated magnetic configuration. Below T_{SF} the surface Cr spins in the longitudinal SDW vary as $S = S_0\cos kx$ or $S = S_0\cos ky$ [Fig. 1(a)] where the wave vector $k = 2\pi/\lambda$ with $\lambda \simeq 50$ Å [15]. The spin variations result in a periodic spatial dependence of the exchange coupling magnitude $J = J_0\cos ky$. Because $\lambda \ll \delta$ further simplifying can be achieved by averaging Eq. (4) over rapid spatial spin oscillations, similar to a pendulum in a rapidly oscillating field [17]. Consider a single-domain Cr state with the longitudinal SDW along the y axis ($\psi = \pi/2$). Let $\Phi(x, y) = \tilde{\Phi}(x, y) + \chi(y)$, where $\tilde{\Phi}(x, y)$ describes averaged over the rapid oscillations magnetization "motion", and $\chi(y)$ represents small oscillations of Fe magnetic moment around $\tilde{\Phi}(x, y)$. Averaging Eq. (4) yields

$$2A(\tilde{\Phi}_{xx} + \tilde{\Phi}_{yy}) - \frac{K}{2}\sin 4\tilde{\Phi} - K_u^{\text{eff}}\sin 2\tilde{\Phi} = 0, \tag{5}$$

where $K_u^{\text{eff}} = J_0^2/32Ak^2d^2$ is the additional effective uniaxial anisotropy constant, and J_0 is the amplitude of the interface exchange interaction between Fe and Cr. The density of the effective anisotropy energy may be written as the sum of cubic and uniaxial terms

$$w_a = K\sin^2\tilde{\Phi}\cos^2\tilde{\Phi} + K_u^{\text{eff}}\sin^2\tilde{\Phi}. \tag{6}$$

Thus, below T_{SF} the fourfold symmetry of Fe is broken by interactions at the Fe/Cr interface and the energy minimum is achieved by 90° coupling between Fe and Cr spins. Note that the same result has been found by numerical calculations for the case of the ordinary antiferromagnet with fully compensated interface spin structure [18]. It has been pointed out that the 90° coupling is similar to the spin-flop state of an antiferromagnet in an external magnetic field. In the case of Fe on Cr we are able to give an analytic treatment of this phenomenon.

Further, we calculate the coercive force below T_{SF}. The reversal behavior of ferromagnets is often discussed in terms of the coherent spin rotation. In the case of ultrathin extended layers that mechanism seems to be extremely unlikely. Because of the presence of such defects as film edges and interface steps which serve as nucleation centers, we consider incoherent reversal mechanisms. It is very

important to note that the interfacial roughness plays no essential role in the reversal at $T < T_{SF}$ when Fe and Cr spins are perpendicular in the magnetized state [Fig. 1(a)]. Below we suggest that the coercive force below T_{SF} is determined by the instability of a nonuniform mode that in the absence of great volume defects is assumed to be localized at the film edge. The magnetostatic field due to Fe edge for thin films can be determined as being caused by a charged line with the linear "charge" density $-M_s d\cos\Phi(0)$, where M_s is the Fe saturation magnetization [Fig. 1(b)]. The integration of the Maxwell equation yields the radial component of the magnetostatic field $H_r^{(m)} 2\pi r = -4\pi M_s d\cos\Phi(0)$ ($r \gg d$). The field component along x direction when $|x| \gg d$ may be written as $H_x^{(m)} \approx -2M_s d\cos\Phi(0)/|x|$. For the stability analysis we consider the equation of motion of the Fe magnetization in the limit of a large damping α

$$-\frac{\alpha M_s}{\gamma}\tilde{\Phi}_t = -2A\tilde{\Phi}_{xx} + \frac{K}{2}\sin 4\tilde{\Phi} + K_u^{\text{eff}}\sin 2\tilde{\Phi} - \frac{M_s^2 d}{x}\cos\tilde{\Phi}(0)\sin\tilde{\Phi} + HM_s\sin\tilde{\Phi}\,, \quad (7)$$

where γ is the gyromagnetic ratio, and H is the external magnetic field along x axis. Linearization of Eq. (7) $\tilde{\Phi} = \tilde{\Phi}_0 + \tilde{\psi}(x,t)$ around the reference state $\tilde{\Phi}_0 = 0$ and substitution $\tilde{\psi} + \Psi(x)\exp(\nu t)$ [19] yields the equation

$$\Psi_{xx} + \left[-p^2 + \frac{2p\kappa}{x}\right]\Psi = 0\,, \quad (8)$$

where $p^2 = (2K + 2K_u^{\text{eff}} + HM_s + \alpha M_s\nu/\gamma)/2A$, $\kappa = M_s^2 d/4pA$. This is well known "radial Schrödinger equation" for the hydrogen atom with zero orbital moment. The localized solutions are given by $\Psi \sim x\exp(-px)_1F_1(1-\kappa, 2; 2px)$, where ${}_1F_1()$ is the degenerated hypergeometric function, and $\kappa = 1, 2, \ldots$. The reference state $\tilde{\Phi}_0 = 0$ becomes unstable when $\nu = 0$ [19]. The largest (the smallest in absolute value) magnetic field of the instability is determined by the condition $\kappa = 1$. This yields the nucleation field and the associated nucleation mode

$$H_n^W = H_c^W = -\frac{2(K + K_u^{\text{eff}})}{M_s} + \frac{M_s^3 d^2}{8A}\,, \quad (9a)$$

$$\Psi \simeq x\exp\left(-\frac{M_s^2 d}{4A}x\right). \quad (9b)$$

Evidently, this result is valid not only for Fe/Cr system, but for any thin magnetic film. It is clear that the linear nucleation mode (9b) shown schematically in Fig. 1(c) develops eventually into a domain wall. The first term of the expression (9a) would give the coercive force for the case of the coherent magnetization rotation. In contrast, the appearance of the second term is entirely due to an incoherent spin rotation near the film edge. In accordance with an intuitive understanding of the nonuniform reversal picture, its magnitude is determined by the competition between magnetic and exchange energy. Spin deviations in the nucleation mode (9b) reach a maximum at the distance from the film edge of order of $4A/M_s^2 d$ [Fig. 1(c)]. Hence, for a particular case of Fe ($M_s = 1700$ emu/cm^3, $A = 2\times10^{-6}$ erg/cm) our representation of the film edge as a "charged" line remains valid for the film thickness of $d \lesssim 2\sqrt{A}/M_s \simeq 160$ Å. The second term in the expression (9a) is comparable with the first term even for the thinnest films and is increased crucially with the film thickness (see Fig. 3, curve 1), we assume for Fe $K = 4.6\times10^5$ erg/cm^3. In contrast, the effective uniaxial anisotropy constant K_u^{eff} is small as compared to the Fe cubic anisotropy and does not substantially contribute to the coercivity. Actually, assuming $J_0 \simeq 1.5$ erg/cm^2 (see below) we obtain $K_u^{\text{eff}} \simeq 10^4$ erg/cm^3 for $d = 20$ Å. Altogether, this anisotropy establishes the perpendicular orientation of Fe and Cr spins below T_{SF}, making the role of interface steps in the reversal insignificant [Fig. 1(a)].

The situation is radically changed above T_{SF} when Fe and Cr spin suggest 0° or 180° coupling [Fig. 2(a)]. In this case we should take into account a possibility of the nucleation at interface steps. In the simplest version of our model, the exchange coupling $J(x)$ per unit area is presented by the step function with period $2L$. The fluctuations in coupling due to monoatomic terraces at Fe/Cr interface are shown in Fig. 2(b). Further, we approximate the step function as $J(x) = J_0\cos\pi x/L$. In this case the instability of the reference state $\Phi = 0$ is determined by the existing of periodic solutions of Mathieu's equation

$$\Psi_{xx} + \left(a - 2q\cos\frac{\pi x}{L}\right)\Psi = 0\,, \quad (10)$$

where $a = -(2K + HM_s)/2A$, $q = J_0/4Ad$. The bottom of zero zone of periodic solutions of Mathieu's equation can be approximated for $q \lesssim 1.5$ by $a \approx -q^2/2$ [20]

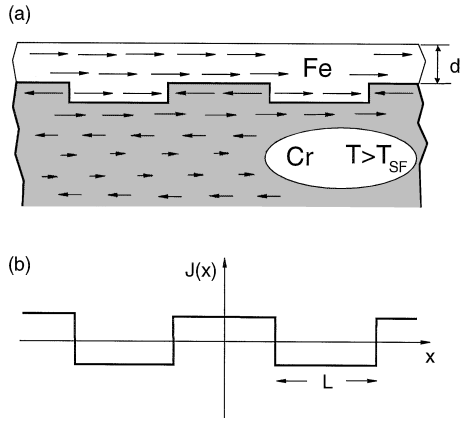

FIG. 2. (a) The arrangement of Fe and Cr spins at the interface with atomic steps above the Cr spin-flip transition temperature. (b) The variations of the exchange coupling between Fe and Cr in the presence of the interface roughness.

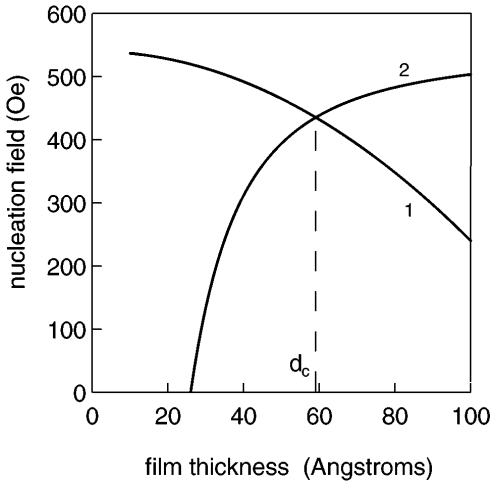

FIG. 3. The thickness dependence of the nucleation field (in absolute value) of both the domain wall near the Fe film edge (curve 1) and of the ripple structure of Fe/Cr interface steps (curve 2). Exchange coupling per unit area $J_0 = 1.5$ erg/cm^2; the lateral size of monoatomic terraces $L = 300$ Å.

which yields the nucleation field above T_{SF}

$$H_n^R = H_c^R = -\frac{2K}{M_s} + \frac{1}{16\pi^2}\frac{J_0^2}{M_s A}\frac{L^2}{d^2}. \qquad (11)$$

In contrast to the case $T < T_{SF}$ when the reversal is governed by the nucleation of the domain wall near the film edge, here we expect the occurrence of a specific ripple structure associated with the interface roughness. The typical thickness dependence of the ripple nucleation field is shown in Fig. 3 (curve 2) (we assume $J_0 = 1.5$ erg/cm^2, $L = 300$ Å). The nucleation field alters the sign below the film thickness of about $d \approx 25$ Å, resulting in decreasing the remanent magnetization. This fact is in good agreement with the experiment [12]. Note that no decrease in the remanent magnetization of the thinnest Fe film has been observed below T_{SF}.

It is clear that the expression (11) can be applied not only for Fe on Cr but also for other magnetic layered structures. A ferromagnet grown onto a conventional antiferromagnet (NiFe/NiO), two ferromagnetic films separated by a nonmagnetic layer (Co/Cu/Co) and spin valves provide some examples. For such structures, we would predict a crossover in the hysteresis behavior at a critical film thickness d_c using calculated thickness dependence of the nucleation field for the domain wall and for the ripple structure (Fig. 3). Namely, the reversal proceeds by nucleation and motion of the domain wall above $d > d_c \approx 60$ Å, whereas below d_c the occurrence of ripple structure should be observed. The magneto-optical indicator film imaging technique (see, for example, [21,22]) study of that phenomenon is in progress.

For the particular case of Fe on Cr such a crossover occurs with the temperature variation due to the spin-flip transition in Cr. We believe that this crossover leads to the abrupt change of the coercive field reported in Ref. [12].

Strictly speaking, we find the expressions for the coercivity of Fe on Cr above and below T_{SF} in the frames of a micromagnetic theory assuming "frozen" Cr spin distribution. A more realistic picture would have the frustrated magnetic order located near the Fe-Cr interface but within the Cr layer. Microscopic calculations of stepped Fe/Cr interface demonstrate that nodes in the Cr spin density wave could be moved toward the interior of the Cr layer during the Fe magnetization. Electronic structure calculations [23] are not in contradiction with the micromagnetic description while we study the instability condition of the magnetized Fe state. The exchange coupling constant in (11) can simply be thought as a phenomenological parameter which should be determined from the experiments.

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