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Random-field model of exchange anisotropy at rough ferromagnetic-antiferromagnetic interfaces

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A field-asymmetric offset of the hysteresis loop in ferromagnetic-antiferromagnetic sandwiches, one of the manifestations of so-called exchange anisotropy, can be predicted from the presence of random interface roughness giving rise to a random field acting on the interface spins. The antiferromagnet breaks up into domains of size determined by the competition of exchange and an additional uniaxial in-plane anisotropy, and this size sets the scale for averaging of the random field.

A ferromagnetic film exchange-coupled to an antiferromagnetic film exhibits a complex of unusual properties such as a hysteresis loop whose center can be shifted away from zero field. These properties have been interpreted as arising from an effective "exchange field" H_E or, equivalently, from a "unidirectional" anisotropy energy $K_E \sin\theta$, where θ is the polar angle of the ferromagnet's magnetization M_F with respect to the anisotropy axis, and where $K_E = H_E M_F$. This form contrasts with the more conventional time-reversal-symmetric uniaxial anisotropy $K \sin^2\theta$, and it is usually called "exchange anisotropy" because it arises from the exchange coupling between the two layers. Although exchange anisotropy has been intensively studied both theoretically and experimentally,[1-12] no theory has yet succeeded in predicting its order of magnitude.[13]

Here I propose a novel mechanism for exchange anisotropy which postulates a randomness in the exchange interactions at the ferromagnetic-antiferromagnetic (F-AF) interface, arising, for example, from interface roughness. These random exchange interactions act like a random field on the antiferromagnet (AF) and form domains reminiscent of the so-called "Imry-Ma domains" of the random-field problem.[10] While consistent with the original suggestions of AF domains by Kouvel[2] and Neel,[4] this mechanism goes a step further to give the first semiquantitative predictions for the size of the exchange anisotropy and related phenomena in agreement with experiment. The model also opens up a rich new class of problems in disordered magnetism where interfacial or two-dimensional disorder interacts with neighboring order in the bulk. In other words, these are problems involving a mixture of two- and three-dimensional effects. By contrast, earlier theories in surface or interfacial magnetism[14] have, by and large, been limited to one-dimensional models, i.e., to the assumption of uniformity along the interface and of perpendicular variations only.

The context for calculating the exchange anisotropy field is shown in Fig. 1, where a domain wall in a uniaxial ferromagnet is driven by an applied in-plane field H. If the interfacial energy with the AF differs for the two domains, then the exchange field is determined by the balance of the applied field pressure $2HM_F t_F$ and the effective pressure from the interfacial energy difference $\Delta\sigma$:

$$H_E = \Delta\sigma / 2M_F t_F \ . \quad (1)$$

Here M_F and t_F are the magnetization and thickness of the ferromagnet. Thus the problem reduces to calculating the interfacial energy difference between the two ferromagnetic orientations.

Several general materials requirements for obtaining exchange anisotropy can already be deduced at this point. There must be some initial breaking of time-reversal symmetry. This is usually obtained by cooling the AF through its ordering temperature while the ferromagnet with a higher-ordering temperature is single-domained by an applied field. Further, once this process is complete and a domain wall is dragged through the ferromagnet, the underlying AF structure, at least away from the immediate interface, must remain fixed. Otherwise the interfaces of both ferromagnetic domains can relax to their lowest-energy states, which are the same by time-reversal symmetry, and the effect disappears. Thus we must calculate the energy difference between two opposite ferromagnetic configurations with the *same* bulk AF configuration.

Another important requirement is on the orientation of the bulk AF moments. If they are colinear and point *perpendicular* to the interface, then no matter what the spin structure at the interface, an equivalent energy configuration can be generated by keeping the same perpendicular spin components but inverting all the planar components. Again the effect disappears. So we must have either a noncolinear antiferromagnet or an anisotropy which holds the bulk AF moments in the plane. This anisotropy must have components in the plane to prevent simple rotation and keep the bulk fixed as required above. In what follows, an in-plane bulk AF orientation will be assumed. Further details on spin flop to the perpendicular orientation and on the interesting case of noncolinear antiferromagnetism will be considered elsewhere.

Previous understanding of exchange anisotropy is based

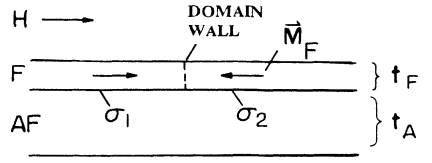

FIG. 1. Schematic side view of ferromagnetic-antiferromagnetic sandwich with ferromagnetic domain wall driven by an applied field H.

largely on the assumption of idealized planar "compensated" or "uncompensated" interfaces.[1,3] If, as shown in Fig. 2(a), the first AF plane next to the interface is compensated, that is, if the AF sublattice structure is such as to give equal numbers of the two opposite spin directions at the interface, then the net field exerted on the ferromagnet by an assumed ferromagnetic exchange J_i across the interface is zero. (Here we define $E_{kl}=-J_i\mathbf{S}_k\cdot\mathbf{S}_l$ per pair of nearest-neighbor spins kl at the interface.) On the other hand, if the first AF plane is uncompensated, as shown in Figs. 2(b) and 2(c), with all interfacial spin directions the same, there is an interfacial energy difference $\Delta\sigma$ per unit area favoring one ferromagnetic orientation over the other. This is just $2J_i/a^2$, assuming a simple cubic structure with lattice parameter a. Equation (1) then gives $H_E=J_i/a^2M_Ft_F$.

Unfortunately, reasonable estimates of the relevant parameters give predictions of H_E which are two orders of magnitude too large.[2,7] For example, in the system of ferromagnetic $Ni_{0.8}Fe_{0.2}$ (permalloy) on antiferromagnetic FeMn,[6-12] it is plausible that the interfacial exchange interaction should be comparable to the effective exchange interactions of FeNi or FeMn, namely, of order 10^{-14} erg. But observed[7] hysteresis loop offsets H_E of order 50 Oe in sandwiches with permalloy thickness 400 Å, magnetization $4\pi M=10000$ G, and nearest-neighbor distance 2.5 Å, imply J_i of only 10^{-16} erg. To explain this factor-of-100 discrepancy, one could try to invoke a "pinhole" effect[2,3] in which some unspecified surface contamination breaks 99 out of 100 exchange interactions, but the consistency of experimental results points to a more intrinsic mechanism. Drastically reduced interfacial exchange J_i is also not plausible, nor is any unit cell in the real crystal structures sufficiently complex to permit a 99% compensated AF plane.

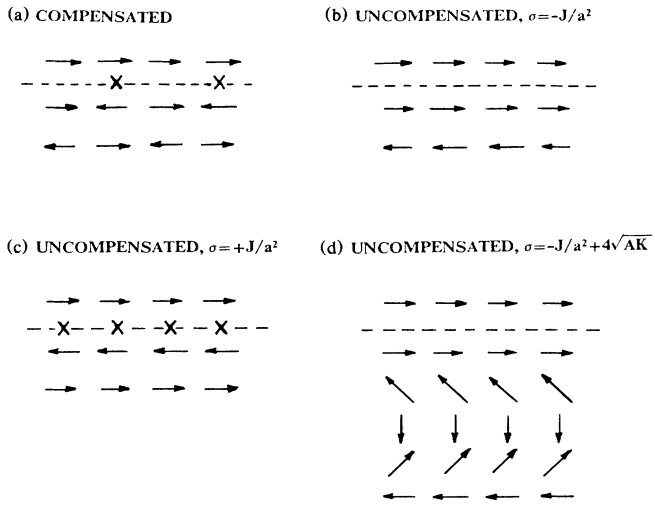

FIG. 2. Schematic side view of possible atomic moment configurations in a ferromagnetic-antiferromagnetic sandwich with a planar ferromagnetically coupled interface indicated by the dashed line. Frustrated bonds are indicated by crosses. The unfavorable configuration of (c) can reduce its energy by forming a planar antiferromagnetic domain wall as in (d).

A more reasonable estimate for H_E comes from allowing a planar domain wall to form at the interface with the unfavorable ferromagnetic orientation,[13] in analogy to well-known phenomena in ferromagnetic sandwiches.[15,16] This domain wall could be either in the AF, as illustrated schematically in Fig. 2(c), or in the ferromagnet, wherever the energy is lower. For example, if the AF has an in-plane uniaxial anisotropy energy K_A and exchange stiffness $A_A\simeq J_A/a$, the AF domain-wall energy has the well-known value[16] $4\sqrt{A_AK_A}$, provided the AF is assumed to be infinitely thick. Since this energy represents the difference between the interfacial energies of the two ferromagnetic orientations, Eq. (1) now gives

$$H_E=2\sqrt{A_AK_A}/M_Ft_F\ . \tag{2}$$

This prediction is significantly smaller than the previous estimate by the factor $2a^2\sqrt{A_AK_A}/J_i$ and thus, for the first time, offers a possible micromagnetic origin for exchange anisotropy. If we assume all exchange parameters are about the same and take $A\simeq J/a$, then the reduction factor is just $2a/\sqrt{A/K}$, that is, twice the ratio of the lattice parameter to the standard micromagnetic domain-wall width parameter[16] $\sqrt{A/K}$. Since the wall can be either in the ferromagnet or in the AF, K is the lesser of the two anisotropies. A typical permalloy planar anisotropy of 1.6×10^4 erg/cm^3 would give exactly the factor-of-100 reduction needed.

Nevertheless, this model suffers from the assumption of the atomistically perfect uncompensated boundary exchange, which is unlikely in practice. Any monatomic step in the surface will change the sign of the interactions and create a situation similar to a compensated interface. Therefore, naively one might expect the interface energy to decrease to zero as the number of defects increases. As we shall see next, consideration of a randomly disordered surface actually can lead to a finite value for H_E and one which has essentially the same parameter dependences as Eq. (2). Let us then consider a rough (nonplanar) interface, starting with a single monatomic bump in the compensated simple cubic interface as shown in Fig. 3(a). One ferromagnetically oriented nearest-neighbor pair across the interface is now replaced by five AF-aligned pairs, for a net *antiferromagnetic* deviation of 6 away from perfect compensation. A bump shifted by one lattice spacing as shown in Fig. 3(b), which corresponds by symmetry to the opposite ferromagnetic or AF domain, gives, similarly, a net *ferromagnetic* deviation of 6 away from perfect compensation. Thus a net energy difference of z_iJ_i, with $z_i=12$, acts as the interface, favoring one domain orientation over the other. This simple example suggests then how interface roughness can give rise to breaking of time-reversal symmetry, provided the AF configuration remains fixed.

A more detailed model reduces somewhat the estimate of this local energy difference. For example, it is easy to see that by inverting the spin in the bump of Fig. 3(a), the interfacial energy difference is reduced by $5\times2J_i$ at the cost of generating one frustrated pair in the AF layer just under the bump, as shown in Fig. 3(c). This AF frustrated pair increases the energy difference by $2J_A$, where J_A is

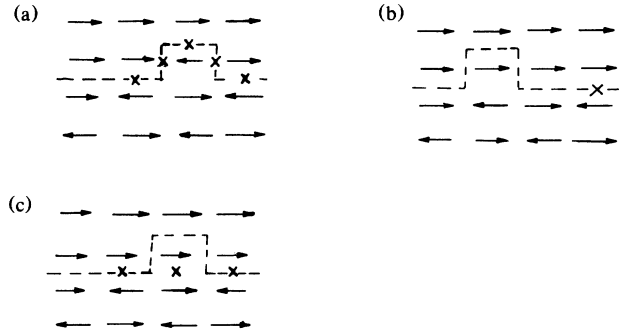

FIG. 3. Schematic side view of possible atomic moment configurations in the ferromagnetic-antiferromagnetic sandwich with a nonplanar interface. The bump should be visualized on a *two*-dimensional interface. Configuration (c) represents a lower energy state of (a). The shifted bump of (b) is energetically equivalent to flipping the ferromagnetic spins of (a).

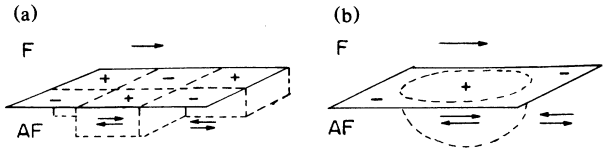

FIG. 4. Schematic perspective view of antiferromagnetic domains (dashed lines) stabilized by a net (locally averaged) interface field surrounded by regions of net oppositely directed interface field. In contrast to Figs. 2 and 3, this figure is macroscopic, with the arrows indicating not the atomic but only the global ferromagnetic and antiferromagnetic moments. (a) represents the small and (b) the large domain-size limit.

the AF exchange constant. Thus the energy difference between the two domains becomes only $2J_i+2J_A$ or roughly $4J$ if $J_i \sim J_A \sim J$.

If one further allows localized canting of the spins, one can expect the energy difference to be reduced somewhat further. The detailed solution of this problem is complex and depends on the specific parameters J_i and J_A, as well as the crystal structure (e.g., fcc rather than simple cubic) and the ferromagnetic exchange J_F since the ferromagnetic layer may also cant at the interface. For our purposes it will be sufficient to assume that associated with each interface irregularity is a local energy difference between the two domains whose sign depends on the particular location of the irregularity and whose local magnitude is on the average $2zJ$. Here z is a number of order unity as suggested by the above model, and J is the magnitude of the exchange constant assumed to be similar at and on either side of the interface.

The next key physical insight is that for an interface which is random on an atomic scale, the local unidirectional interface energy $\sigma_l = \pm zJ/a^2$ will also be random, much as in the Imry-Ma random-field problem,[10] and its average σ in a region of area L^2 will go down statistically as $\sigma \sim \sigma_l/\sqrt{N}$, where $N = L^2/a^2$ is the number of sites projected onto the interface plane. This argument is quite general[10] and so, in principle, not only interface roughness but also other mechanisms such as random alloy effects[2] could do. Here also is the key difference with Kouvel's earlier model[2] of AF domains: He simply averaged over an unknown distribution of domain sizes and parameters rather than using the randomness to determine the domain size, as we do next.

Given the random field and assuming a region with a single domain of the ferromagnet, it will now be energetically favorable for the AF to break up into domainlike regions, as pictured schematically in Fig. 4(a), to minimize the net random unidirectional interfacial anisotropy. The situation is analogous to the classic Imry-Ma domains[10] of the random-field problem, except that here the random field is at an interface while the competing domain-wall exchange (and possible uniaxial anisotropy) energies are in the bulk. Of course, we have assumed that the AF configuration in the bulk is fixed; so in effect we are considering here the initialization of an AF domain pattern as the AF is cooled through its ordering temperature.

Since approximately half the AF interface exchange field points in each direction, a simple model is a square grid with lateral dimension L and depth h, as in Fig. 4(a). Then the approximate interfacial energy (per area of the F-AF interface) is

$$\sigma = -\frac{2zJ}{\pi aL} + \tfrac{1}{2}J\left[\frac{1}{2}\left(\frac{\pi a}{h}\right)^2\frac{h}{a^3} + 2\left(\frac{\pi a}{L}\right)^2\frac{h}{a^3}\right]. \quad (3)$$

Here the first term represents the stabilizing (negative) net random interface anisotropy energy, with a factor $2/\pi$ from averaging the spin component along the interfacial exchange field in a one-dimensional linear-rotation approximation $\theta = \pi x/L$ to the domain structure. The second term represents the competing exchange energy $J(\Delta\theta)^2/2$ with its vertical and horizontal components estimated in the same linear approximation. Minimizing the energy with respect to h, we find $h = L/2$, showing that the domain height is comparable to its lateral dimensions. Substituting this result back into Eq. (3), we find $\sigma = (\pi^3 - 2z)J/\pi aL$. Thus, both interface anisotropy and exchange terms go inversely with L. Even with the factor-of-2 accuracy of the above approximations, the π^3 term will certainly be larger than $2z$, and then it will be favorable for the domain size L to expand to lower the energy. This case corresponds to the case of critical dimensionality in the Imry-Ma random-field problem,[10] and small additional energy terms can now play a dominant role.

In particular, a term like a uniform in-plane uniaxial anisotropy energy K in the AF layer, will limit the domain size. To see this, we note that a large domain will no longer consist of uniform rotation dictated only by exchange, as in the small-domain limit treated above. Instead the domain wall is confined by the anisotropy to a width $\pi\sqrt{A/K}$, which is by assumption much smaller than the domain diameter L. As shown in Fig. 4(b), the surface tension $4\sqrt{AK}$ will then equalize curvature everywhere and create a hemispherical bubble domain, assuming vertical incidence of the wall on the F-AF interface. The total energy of the bubble is then the total interfacial stabilization energy $\simeq -\sqrt{\pi}zJL/2a$ plus the total hemispherical surface energy $2\pi\sqrt{AK}L^2$.

With a sea of these bubbles covering half the interface, the energy per area (that is, dividing by L^2) has a negative term going as $1/L$ and a positive constant term. Therefore the domains tend to *contract* until the size equals the wall width $\pi\sqrt{A/K}$. Squeezing the domains further causes exchange energy to mount over anisotropy energy and restore the limit of Eq. (3), where the domains prefer to *expand*. Thus the condition $L \simeq \pi\sqrt{A/K}$, which is also the crossing point between a volume and a surface type of domain exchange energy, gives equilibrium. It is noteworthy that $\sqrt{A/K}$ will be independent of temperature to a first approximation when both A and K go as the thermal average of S^2. Then $L \simeq \pi\sqrt{A/K}$ will give a good measure of the frozen-in AF domain size, even at room temperature.

Once these domains are fixed, flipping the ferromagnetic orientation causes an energy change per unit area of $\Delta\sigma = 4zJ/\pi aL$ with L now determined as above. Finally, then, Eq. (1) gives the predicted exchange anisotropy field

$$H_E = 2z\sqrt{AK}/\pi^2 M_F t_F \ . \tag{4}$$

This form is remarkably similar to Eq. (2) and therefore equally able to explain the order of magnitude of the exchange anisotropy effect, provided an appropriate uniaxial anisotropy K is present in the antiferromagnet.[12] This similarity reflects the fact that fundamentally both pictures have characteristic domain-wall energies at the interface. In principle, they could be differentiated if the various exchanges J_i, J_A, and J_F were substantially different, as can be easily shown by keeping track of these parameters in the above calculation.

Further differences between these pictures arise because the random-field model and its consequent lateral inhomogeneity more naturally explain the observed coercivity and remanent phenomena which appear as the ferromagnetic domain is cycled back and forth.[5] AF domain walls experience coercivity just as ferromagnetic domain walls do, except that in bulk antiferromagnets there is no easy handle to probe that coercivity. In the F-AF sandwich, coupling of the ferromagnetic domain wall to the underlying structure can put pressure on the AF walls and cause them to move slightly, generating coercive loss. But as in the classic ferromagnetic Rayleigh loop,[4] one must assume that the AF coercivity is still sufficient to hold most of the underlying domain pattern in place. This is, in fact, just Neel's original model,[4] in which he postulated an AF domain structure without being able to give a micromagnetic derivation for the size of the effect as I have done here.

In summary, I have given the first derivation of exchange anisotropy which plausibly estimates the size of the effect in terms of fundamental parameters of the component films and interface. The derivation involves extension of classic random-field concepts to an interesting new regime. Clearly further work is called for, both to extend the model and to test it by determining relevant parameters and looking for the antiferromagnetic interface structure directly.

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[13]In independent work, D. Mauri, H. C. Siegmann, P. S Bagus, and E. Kay have investigated the idea of an interfacial uniform planar domain and find a similar square-root anisotropy dependence.

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