Magnetostriction-induced anisotropy in the exchange biased bilayers

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The exchange bias at ferromagnetic (FM)/antiferromagnetic (AF) interfaces strongly depends upon the state of antiferromagnetic layer which, due to strong magnetoelastic coupling, is sensitive to mechanical stresses. In the present paper we consider magnetoelastic effects that arise at FM/AF interface due to lattice misfit and magnetic ordering. We show how magnetostriction affects mutual orientation of AF and FM vectors and easy-axis direction in thin AF layer. The results obtained could be used for tailoring exchange biased systems.

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I. INTRODUCTION

Antiferromagnetic (AF) materials are widely used in spintronic devices as auxiliary elements for pinning of ferromagnetic (FM) magnetization through the effect of exchange bias (see, e.g.[1]). The possibility to control the state of coupled AF/FM bilayers requires investigation of the magnetic mechanisms that could be responsible for bias effect. Many researchers [2–7] emphasize the important role of the AF domain structure in the establishing of the exchange bias. The problem of AF domains is intimately related with magnetoelastic coupling [8] and can strongly depend upon the mechanical stress that appears at the FM/AF interface due to the lattice misfit. Magnetostriction can also provide additional coupling between FM and AF layers and affect orientation of AF moments in the near-surface region [9, 10]. Widely studied epitaxial films consisting of FM and nonmagnetic materials [11–14] show strong correlation between magnetoelastic coupling and magnetic properties. Analogous and even more striking phenomena could be expected in the systems which combine FM with AF that possesses large magnetostriction.

In the present paper we show that magnetostriction of AF produces uniaxial anisotropy in the plane of the adjacent FM layer and thus causes strong surface magnetic anisotropy in AF itself.

II. UNIAXIAL ANISOTROPY OF FERROMAGNET

Epitaxial ferromagnetic film deposited on top of AF inherits the crystallographic structure of the substrate. If the substrate has a certain anisotropy induced by magnetoelastic strains, then, this anisotropy in atomic arrangement will be reproduced by the FM layer. Thus, additional contribution to the magnetic energy of the film should be proportional to magnetoelastic coupling in both FM and AF materials. Phenomenological expression for such type of uniaxial in-plane anisotropy can be deduced from the magnetoelastic energy of FM which for a cubic-symmetry crystal follows as

\[ f_{\text{me}}^{F} = b_1^F [u_{xx} \alpha_2^2 + u_{yy} \alpha_2^2 + u_{zz} \alpha_2^2] + 2b_2^F \{ \alpha_x \alpha_y u_{xy} + \alpha_y \alpha_z u_{yz} + \alpha_z \alpha_x u_{zx} \}. \]  

Here \( u_{ik} \) are strain tensor components which we calculate with respect to the bulk nonmagnetic reference state, \( b_{1,2}^F \) are magnetoelastic coupling coefficients. Magnetisation vector \( M_F \) of FM is described by the direction cosines \( \alpha_k \), \( k = x, y, z \). In the relaxed state of FM/AF system an equilibrium strain \( u_{ik} \) includes deformations produced by lattice mismatch \( \varepsilon_{\text{MF}} \) and spontaneous strain \( \varepsilon_{\text{mag}} \) induced by magnetic ordering in the AF substrate. For a symmetric (001) surface the misfit-induced strains are isotropic and can influence only out-of-plane anisotropy of FM. In contrast, magnetostrictive contribution, though small as compared with the misfit strain, has nontrivial shear components, \( u_{xx}^{AF} - u_{yy}^{AF} \) and/or \( u_{xy}^{AF} \) which can remove degeneracy between different in-plane directions. Thus, uniaxial contribution into magnetocrystalline energy of FM film takes a form

\[ J_{\text{ua}}^{F} = \frac{1}{2} K_{1ua}^{F} \rho_j (\alpha_x^2 - \alpha_y^2) + K_{2ua}^{F} \rho_j \alpha_x \alpha_y, \]  

with anisotropy constants

\[ K_{1ua}^{F} = b_{1}^{F} (u_{xx}^{AF} - u_{yy}^{AF}), \quad K_{2ua}^{F} = 2b_{2}^{F} u_{xy}^{AF}. \]  

Variable \( \rho_j = \pm 1 \) distinguishes between the different AF domains.

A preferable direction of FM magnetisation \( M_F \), which depends upon the sign of the coefficients \( K_{1ua}^{F} \), is defined by correlation between self-striction of the FM and external striction imposed by the AF. If, for example, magnetostriction of FM in the direction of magnetisation is positive (elongation, \( b_{1}^{F} < 0 \)), then, \( M_F \) will tend to align...
In the direction of maximal elongation of the AF, i.e., for positive $u_{AF}$ value (elongation) $K^F_{\psi\psi}$ is negative, as can be easily checked from equation (3).

Magnetostriction-induced uniaxial anisotropy (3) competes with the anisotropy arising from the FM/AF exchange in a thin near-surface region of thickness $\xi$. For a compensated AF surface this contribution depends upon the exchange integral between the atoms of F and AF $J^{F\rightarrow AF}$ and susceptibility of AF $\chi_{AF} \equiv 1/J_{AF}$ (Koon’s model, [15]):

$$f_{exch} = -\frac{1}{2} \chi_{AF} J^{2}_{F\rightarrow AF}/M_{F} \times L_{S}]^{2}.$$  

The AF vector $L_{S}$ describes orientation of spins at the surface of the AF substrate (which in principle can differ from that in the bulk, as will be shown later).

To elucidate the effect of both contributions let us consider a simple case when one of the in-plane easy axis (say, $x$) of FM coincides with in-plane $L_{S}$ direction and $u^{F}_{xy} = 0$. For the in-plane FM ordering ($\alpha_z = 0$) we set $\alpha_x = \cos \psi$, $\alpha_y = \sin \psi$. The effective energy is thus

$$f^{F}_{eff} = \frac{1}{4} K_4 \sin^2 2\psi + \frac{1}{2} [K_{ua} \rho_j + \frac{\xi}{2l_F} \chi_{AF} J^{2}_{F\rightarrow AF}] \cos 2\psi.$$  

Constant $K_4 > 0$ is magnetostrictive constant, and we suppose the FM film to be homogeneously ordered throughout the thickness $l_F$.

Equilibrium value $\psi = \psi^{eq}$ minimizes effective energy (5), so, it satisfies the relations

$$\sin 2\psi^{eq} (K_4 \cos 2\psi^{eq} - [K_{ua} \rho_j + \frac{\xi}{2l_F} \chi_{AF} J^{2}_{F\rightarrow AF}]) = 0$$

$$K_4 \cos 4\psi^{eq} - [2K_{ua} \rho_j + \frac{\xi}{2l_F} \chi_{AF} J^{2}_{F\rightarrow AF}] \cos 2\psi^{eq} > 0.$$  

In the absence of FM/AF interaction, FM has two equivalent easy directions in (001) plane, $\psi^{(0)}_1 = 0$ and $\psi^{(0)}_2 = \pi/2$. Antiferromagnetic substrate removes this degeneracy. If exchange coupling is not too large, $\xi \chi_{AF} J^{2}_{F\rightarrow AF} \leq K_4 l_F$, both solutions $\psi^{(1,2)}$ satisfy equations (6), but have different energies, the difference being

$$f^{F}_{eff}(\psi^{(1)}_{eq}) - f^{F}_{eff}(\psi^{(2)}_{eq}) = K_{ua} \rho_j + \frac{\xi}{2l_F} \chi_{AF} J^{2}_{F\rightarrow AF}.$$  

It can be easily seen from (7) that the FM/AF exchange coupling makes favourable the solution with $M_{F} \perp L_{S}$ ($\psi^{eq}_{2} = \pi/2$) for any sign of the exchange constant $J_{F\rightarrow AF}$. In turn, magnetostriction-induced anisotropy $K_{ua}$ may oppose this tendency and make preferable in-plane orientation of $M_{F}$ and $L_{S}$ ($\psi^{eq}_{1} = 0$). It should be stressed that these two mechanisms have different origin and the system can switch from one easy-axis to another with variation of FM thickness. Exchange mechanism ties together mutual orientation of FM magnetisation and AF spins in the near-surface layer. This mechanism is important for very thin films where factor $\xi/l_F$ is not vanishingly small. Magnetostriction-related mechanism is a long-range one, it depends upon orientation of AF moments in the bulk which can be different from $L_{S}$. Moreover, in some AFs widely used in FM/AF systems (e.g., NiO, CoO, LaFeO$_3$, KCoF$_3$) magnetostriction originates from the strong spatial dependence of the exchange integral and is insensitive to exact orientation of AF spins. In this very important case uniaxial anisotropy of FM is defined mainly by the domain structure of AF.

The role of magnetostriction-induced mechanism can be illustrated by some experimental examples. Simultaneous observation of the FM and AF spins in Co/LaFeO$_3$ [16] and Co/NiO [17] systems revealed that FM magnetisation is aligned parallel or antiparallel to the in-plane projection of the AF axis in contrast to the usually observed perpendicular coupling consistent with the Koon’s model [15]. Uniaxial anisotropy was also detected after deposition of Fe on top of KCoF$_3$ [18, 19]. All these AFs are known to have rather large magnetostriction of the exchange nature (see Table I). Using the values of magnetoelastic constants for ferromagnets (Table II), one can calculate from equation (3) the expected value of uniaxial anisotropy in different FM/AF combinations (see Table III).

As can be seen from Table III, uniaxial anisotropy in the Fe film constitutes only 10% from the “pure” magnetic anisotropy. Nevertheless, this value can be enough to choose preferable axis of magnetisation as was clearly observed in the experiments [18, 19]. More pronounced effect is expected in Co films which have rather high magnetostriction and small bulk magnetic anisotropy. Predicted value of the uniaxial anisotropy is of the same order as $K_4$ or even one order of magnitude larger, as in the case of Co/CoO. It should be noted, that in calculation we started from the bulk values of magnetoelastic coefficients for Fe and Co. In the case of ultrathin Co films these values need to be ascertained because of the large potential misfit between FM and AF lattices (nearly 10%). Depending on the growth mode this mismatch can either relax through the formation of dislocations or produce strong internal stresses in the Co film which, in turn, can give rise to a crucial change of the value and even the sign of magnetoelastic coefficient (see, e.g.,[13, 25]).

### Table I. Magnetostriction (spontaneous deformations) of typical AFs calculated the experimentally observed lattice constants above and below Neel temperature

| AF      | Magnetostriction |
|---------|------------------|
| NiO     | -2.6·10$^{-5}$   |
| LaFeO$_3$| -4.76·10$^{-4}$  |
| KCoF$_3$| -2.0·10$^{-3}$   |
| CoO     | -2.10·10$^{-2}$  |
III. SURFACE ANISOTROPY OF ANTIFERROMAGNET

It is widely recognised that lattice misfit strongly influences the magnetic and magnetoelastic properties of the film (see, e.g. [29]). On the other hand, epitaxial misfit may equally induce large stress in the substrate (this phenomenon is used to measure stress in the film [11]). In the case when the substrate is rather thick, stress exerted by the film relaxes over a small distance $\Sigma_{AF}$ in the near-surface layer of AF. For AFs with large magnetoelastic coupling this surface stress can produce an additional magnetic anisotropy which we will call a surface anisotropy.

Phenomenological description of this effect is based on the analysis of the Helmholtz free energy potential $G$ which includes elastic $f_e$ and magnetoelastic $f_{me}^{AF}$ energy of AF using antiferromagnetic vector $\mathbf{L}$ and components of stress tensor $\sigma$ as the internal parameters:

$$G = \int_{AF} (f_{me}^{AF} + f_e) dV. \tag{8}$$

In the simplest case of a cubic crystal the elastic energy density $f_e$ takes a form

$$f_e = \frac{1}{2} s_{11} [\sigma_{xx}^2 + \sigma_{yy}^2 + \sigma_{zz}^2] + s_{12} [\sigma_{xx} \sigma_{yy} + \sigma_{yy} \sigma_{zz} + \sigma_{zz} \sigma_{xx}] + 2 s_{44} [\sigma_{xy}^2 + \sigma_{yz}^2 + \sigma_{zx}^2], \tag{9}$$

where we turned from strains to stresses using the Hook’s law. Compliances $s_{ik}$ are expressed through the elastic modulae $c_{ik}$ in a usual way:

$$s_{11} = \frac{c_{11} + c_{12}}{(c_{11} - c_{12}) (c_{11} + 2 c_{12})},$$

$$s_{12} = -\frac{c_{12}}{(c_{11} - c_{12}) (c_{11} + 2 c_{12})},$$

$$s_{44} = \frac{1}{c_{44}}. \tag{10}$$

Density of magnetoelastic energy $f_{me}$ can be written as

$$f_{me}^{AF} = f_{exch} + \frac{b_{AF}^{2}}{c_{11} - c_{12}} [\sigma_{xx} L_x^2 + \sigma_{yy} L_y^2 + \sigma_{zz} L_z^2]$$

$$+ 2 \frac{b_{AF}^{2}}{c_{44}} [L_x L_y \sigma_{xy} + L_y L_z \sigma_{yz} + L_z L_x \sigma_{zx}], \tag{11}$$

where $b_{AF}^{2}$ are magnetoelastic coupling coefficients of a cubic AF, and from the Néel temperature AF vector can be normalised, so $|L| = 1$. The first term in (11) describes a possible nonisomorphic contribution which arises from the space dependence of the exchange interactions described by a coefficient $B_0^{AF}$. It depends upon the specific type of AF, for example, for a single-domain NiO it can be expressed as

$$f_{exch} = P_{0}^{AF} \frac{c_{44}}{c_{11}} (\sigma_{xy} + \sigma_{yz} + \sigma_{zx}) L^2. \tag{12}$$

In the presence of the FM coverage, the AF substrate exerts a surface stress $\hat{\tau}^{AF}$ opposite to the surface stress in the FM film $\hat{\tau}^{F}$:

$$\hat{\tau}^{AF} \equiv \int \hat{\sigma} dz = -\hat{\tau}^{F}. \tag{12}$$

is over the AF thickness. For a (001) cubic surface $\hat{\tau}^{F}$ can be estimated from the misfit value $\epsilon_{MF}$ as follows:

$$\hat{\tau}_{xx}^{F} = \hat{\tau}_{yy}^{F} = t_F \left[ (c_{11} + c_{12} - \frac{c_{12}^2}{c_{11}}) \epsilon_{MF} \right], \tag{13}$$

where $t_F$ is the FM film thickness. Substituting (12) into free energy (8) we obtain a contribution from the FM/AF misfit as

$$G_{eff} = \int_S \frac{b_{AF}^{2}}{(c_{11} - c_{12})} \hat{\tau}^{F} L^2 dS = \frac{1}{2} \int_S k_{S}^{AF} L^2 dS, \tag{14}$$

Table II. Magnetoelastic coupling coefficients for FM’s [11], in erg/cm³

| Co, fcc | Fe, bcc |
|---------|---------|
| $b_1$   | $9.2 \cdot 10^9$ | $-3.43 \cdot 10^9$ |
| $b_2$   | $7.7 \cdot 10^7$  | $7.83 \cdot 10^7$  |

Table III. Magnetic anisotropy of FM/AF systems, erg/cm³. $K_4$ (2nd column) is the 4-th order magnetocrystalline anisotropy observed in the bulk Fe and Co crystals. Theoretical values of $K_{ua}$ (2nd column) are calculated from equation (3). Experimental values of $K_{ua}$ (the last column) are extracted from from measurement of hysteresis loops (for Co) and ferromagnetic resonance (for Fe).

| System      | $K_4$, erg/cm³ | $K_{ua}$, erg/cm³ |
|-------------|----------------|------------------|
| Co/NiO      | -2.3 \cdot 10^9 [26] | 2.0 \cdot 10^5 | 1.8 \cdot 10^5 [27] |
| Co/LaFeO₃   | 0.37 \cdot 10^5 | 1.4 \cdot 10^5 [16] |
| Co/CoO      | 6.0 \cdot 10^6 | 1.2 \cdot 10^6 [28] |
| Fe/NiO      | 8.5 \cdot 10^2 [12, 19] | 0.9 \cdot 10^5 | - |
| Fe/KCoF₃    | 0.7 \cdot 10^5 | 0.8 \cdot 10^5 [19] |
which could be associated with the surface/interface energy of AF. Effective constant

\[ K_{\text{AF}}^\delta = 2 b_1^{\text{AF}} \epsilon_{MF} \left( 1 + \frac{2 c_{12}}{c_{11}} \right) t_F \]  

is proportional to the product of magnetoelastic coupling coefficient of AF and misfit (or effective stress) in the FM layer.

The sign of \( K_{\text{AF}}^\delta \) and hence, the character of the induced surface anisotropy, is defined by the relation between the sign of AF spontaneous striction and that of external stress. Suppose, FM lattice constant is smaller than that of AF \( (t_F > 0) \). Then, AF surface exerts a compressive stress. According to the general Le-Chatelier principle, AF vector at the surface will rotate in a way which reduces the external influence. In the case of positive striction (AF spontaneously elongates in spin direction) in-plane orientation of AF spins will be preferable \( (K_{\text{AF}}^\delta > 0) \). It worth to mention that the analogous, magnetoelastic, mechanism related with the rotation of AF moments in near-surface region is responsible for the shape-induced magnetic anisotropy in AF nanoparticles [30].

IV. DISCUSSION

The misfit-induced surface anisotropy can produce a noticeable rotation of AF spins in the vicinity of interface region. The most pronounced effect can be expected for NiO, CoO, and LaFeO\(_3\) AFs in which the bulk AF vector makes some angle with (001) surface. Particularly, in NiO and CoO the AF spins are ordered in (111) planes (with small deflection in the case of CoO [31–33]) in which they can be easily rotated. An easy-axis is directed along (211) in NiO and (311) in CoO, thus, for a cleaved (001) surface AF moments have nonzero component perpendicular to the surface plane, as was observed for a NiO crystal [34, 35].

Deposition of Fe and Co on NiO, and Fe\(_3\)O\(_4\) and Co on CoO produces compressive surface stress in AF (see Table IV), which gives the values of interatomic distance for different FM and AF at (001) surface of fcc lattice (2nd column) calculated from the bulk lattice parameters (1st column)).

For NiO and CoO the magnetoelastic constant \( b_1^{\text{AF}} \) is positive (as deduced from the data [24, 36]), so, as it follows from (14), (15), for all the mentioned FM/AF combinations the preferable orientation of AF vector \( \mathbf{L} \) is in the interface (001) plane. A compromise between the strong dipole-dipole anisotropy which tends to keep AF moments close to (111) plane and strain-induced surface anisotropy in (001) plane is the direction [110]. So, depending upon the balance between the bulk magnetic anisotropy and the induced surface anisotropy (15), AF moments may rotate from the bulk easy direction to [110] to a smaller or larger angle. The effect should be obviously stronger for Co FM because of the large misfit value.

Experimentally this phenomenon was observed in [17], where deposition of 2 nm Co film on the (001) surface of NiO resulted in the total reorientation of NiO spins to [110] direction. An observed collinear alignment of Co and NiO spins in this system arises from both misfit-induced realignment of AF moments and magnetostriiction-induced uniaxial anisotropy in the FM layer.

An analogous effect was observed in the Fe\(_3\)O\(_4\)/CoO multilayers [3] where an influence of the surface stress is much more pronounced. In this system all of the AF Co moments lie along [110] or [110] directions (depending on the AF domain type). This orientation does not vary with temperature, magnetic field and thickness of CoO layers.

Misfit-induced anisotropy of AF layer depends upon the internal stresses \( t_F \) in the adjacent ferromagnet which could relax in the course of field cycling. Variation of stress, in turn, affects the domain structure of AF. So, magnetoelastic mechanism can explain training effect (irreversible changes in configuration of AF domains) frequently observed in bilayers with multidomain state of AF in the as-cast sample (see, e.g. [2, 37]).

In summary, we have studied the effect of magnetostriction on the properties of FM/AF coupled system. Spontaneous striction which appears in antiferromagnet due to AF ordering can cause uniaxial in-plane anisotropy in the ferromagnetic film and set preferential easy axis of FM either along with or perpendicular to the orientation of AF vector. Competition between uniaxial anisotropy induced by long-range magnetostriction and short-range exchange mechanism results in different orientation of the FM easy-axis depending on the thickness of FM layer. Lattice misfit between FM and AF is a source of a magnetic surface anisotropy in AF substrate which can cause rotation of AF moments in the near-surface region compared with their bulk orientation.

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Table IV. Bulk lattice parameters (2nd column) and calculated interatomic distances at (001) surface of fcc lattice (3d column) different FM and AF, in Å.

|       | Bulk | (001) surface |
|-------|------|---------------|
| Fe, bcc | 2.866 | 4.053          |
| Co, fcc | 3.544 | 3.544          |
| Fe₃O₄ | 8.398 | 4.199          |
| NiO    | 4.177 | 4.177          |
| CoO    | 8.508 | 4.254          |

*Magnetic unit cell*  

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