Strain Induced Anisotropy Change in Ultrathin Fe Films Grown on MnAs(110)/GaAs(001)

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Mechanical stress due to a misfit between a thin film and its substrate induces strains which can strongly modify the unstrained thin film properties. One good and interesting example to study strain effects is given by ultrathin films of Fe epitaxially grown on MnAs(110)/GaAs(001). The MnAs(110) films show, at room temperature, coexistence of two structural phases, which organize themselves forming a striped pattern. The Fe epilayer senses the strain effects due to lattice mismatch and to the border constraints given by the striped substrate. In this work, we are concerned with the consequences that this strain has on the magnetic anisotropy of the Fe thin film and try to explain recent experimental results. These experiments indicate an easy axis rotation of the film Fe atoms sitting on one of the striped phases. In order to have an approach to the understanding of the observed phenomenon, we make use of *ab initio* calculations and of the magnetoelastic model. We find that both the magnetoelastic model and the *ab initio* calculated spin orbit coupling point towards the strain effects as the most important contribution to the observed easy axis rotation.

***Index Terms*— Magnetic anisotrophy, magnetic devices, magnetoelasticity, nanomaterials.**

I. Introduction

MANGANESE ARSENIDE (MnAs) thin films have been studied quite intensively in recent years due to the good compatibility of this material with standard semiconducting substrates and due to its high spin polarization; both characteristics make the system well suited for the development of spintronic devices. MnAs is, in fact, a metallic material which presents an interesting variety of polymorphic transformations as a function of temperature. In bulk, the low temperature hexagonal structure of the NiAs-type (α phase) is ferromagnetic. At 40 °C a first order transition takes place and the system goes into a paramagnetic orthorhombic MnP-type structure (β phase) [1], [2]. At 127 °C it undergoes a second order phase transition transforming again into the low temperature hexagonal structure, which is now paramagnetic (γ phase). Most of the recent works on MnAs have been done on thin films grown on GaAs(100), within a range of temperatures near room temperature for which there exists phase coexistence of the α and β phases, due to the strain induced by the GaAs substrate. In this coexistence region both phases order forming a striped pattern. Near this temperature range MnAs/GaAs could be used as a magnetic active template [1] to change the orientation of the magnetization of a deposited magnetic epilayer, without using an external magnetic field, but just by changing the substrate's temperature[2].

Fe epilayers grow on MnAs(110) thin films, along the [2-11] direction, as it has been shown in different contributions [1], [2]. The lattice misfit of Fe for this stacking is around 13% along the [11-1] direction of bcc-Fe and -8% along the [110] direction. Experimental FMR measurements presented in [2] indicate that there exist three well-defined temperature regions for this system. Below and above phase coexistence, the easy axis of the Fe epilayer is along the [011] direction, as expected for an unstrained Fe layer. In a temperature window, within the phase coexistence region, it is found that as the β phase grows, there is a critical width of the α-MnAs stripes, below which the Fe easy axis abruptly switches from the [011] to the [11-1] direction. That is, a 90° in-plane rotation [2] of the easy axis of Fe on α-MnAs takes place. The authors of [2] and [3] suggest that the morphology of MnAs could underlie this change, while they disregard any influence originated at the magnetic interaction between substrate and epilayer.

Within the phase coexistence region, each magnetic phase of the striped pattern displays a different height, and the system offers a corrugated surface. It is well known that α-MnAs stripes have a larger height than β-MnAs ones, given by $h \approx 0.01t$, where t is the MnAs film thickness. This implies that if there are 100 nm of MnAs, the difference in height of the α and β phases is around 1 nm. This could give rise to a large stress at the borders of the stripes, constraining the Fe region on top of the α phase. If we deposit a few layers of Fe, let us say 5 nm on it, then, at the coexistence temperature range, it is possible that between Fe on α-MnAs and Fe on β-MnAs there appears a domain-wall structure separating both phases [4].

In this contribution we are going to address the reasons for the change observed in the magnetic anisotropy of the Fe film on α-MnAs for stripes' widths below the critical value. We assume that the width of the α-MnAs stripes, together with the different heights of the Fe film on the two phases, pose strong enough constraints that prevent Fe on α-MnAs to relax. These constraints imply a large strain on the involved Fe regions due to the large lattice mismatch with MnAs. We study, then, the influence that this strain has on the anisotropy of the thin Fe epilayer. We find that the change in magnetic anisotropy is not due to the magnetic coupling with the substrate, but that it mainly concerns the strain, as is had been suggested in a previous experimental work [3].

The paper is organized as follows, in the next section we give the *ab initio* calculation details, then the theoretical results for the elastic and magnetoelastic energy. Finally, magnetic anisotropy energy results are shown together with a brief discussion. Conclusions are drawn in Section IV.

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II. Calculation Details

Electronic calculations are performed using *ab initio* density functional theory as implemented in the VASP [5] code. PAW pseudopotentials are used [6], together with the generalized gradient approximation as parameterized by Perdew *et al.* [7] for the exchange and correlation potential as it has been shown to give the proper magnetic and structure properties for MnAs [8]. The spin-orbit interaction is included inside each atomic sphere as described in [9]. We consider a plane wave basis set with kinetic energy values up to 350 eV. Since the magnetic anisotropy energies (MAEs) are below the meV range, a convergence of the total energy within 10^{-6} eV is demanded during the self-consistency procedure, and a density in-plane mesh of 1200 k-points in the Brillouin zone is used. This sampling is enough to attain convergence of the MAE in these 3-D transition metal bidimensional systems as it has also been pointed out by other authors [10], [11].

As we are interested on the effects of α-MnAs, we assume that the stripes' borders just impose on Fe the substrate lattice parameters and that other border effects are negligible. At the temperature range of interest the width of the stripes is small enough to produce the mentioned constraint but large enough (≈200 nm) to consider a bidimensional α-MnAs substrate in the calculations. We simulate, then, the α-MnAs substrate with 12 layers' 2-D slabs separated by a vacuum region of 10 Å and do periodic slab calculations. No inversion symmetry is imposed in order to have full formula units of MnAs in the slab's unit cell. Fe is grown on only one of the surfaces, the one with As termination due to the reported experimental conditions [12]. The internal coordinates are fully relaxed until forces are smaller than 0.04 eV/Å, and the in-plane lattice constants of bulk MnAs are kept.

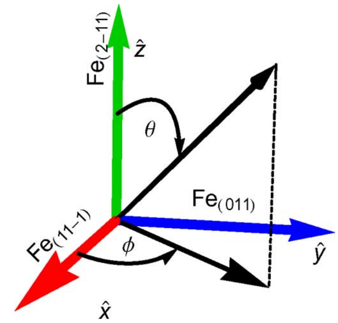

Fig. 1. Reference frame referred to as the Fe(2-11) film in-plane and out-of-plane directions.

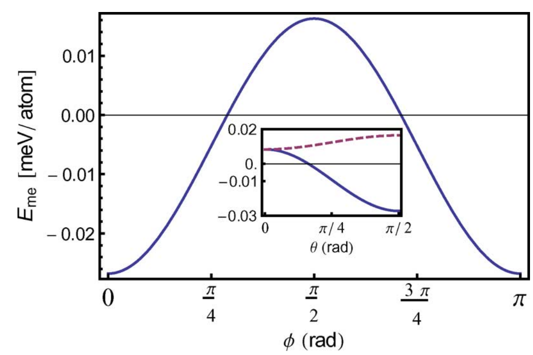

Fig. 2. Angular dependence of the magnetoelastic energy [from eq. (1)] using Fe-bulk parameters $B_1 = -3.43$ MJ m^{-3} and $B_2 = 7.83$ MJ m^{-3} for the strain given by α-MnAs. The in-plane magnetoelastic energy reaches a minimum at $\phi = 0$, which corresponds to the $Fe_{[11-1]}$ direction. In the inset, the magnetoelastic energy angular variation following two different paths starting from the out-of-plane direction ($Fe_{[2-11]}$) to the in-plane $Fe_{[11-1]}$ and to the in-plane $Fe_{[011]}$, solid/dashed lines, respectively. In-plane $Fe_{[11-1]}$ is always favored. Angles are given according to the reference frame of Fig. 1.

III. Results and Discussion

We assume that the Fe epilayer on α-MnAs follows pseudomorphically the underlying substrate. To disentangle the effects of strain from the interfacial contact ones, we perform calculations for pure Fe slabs as well as for the mentioned Fe/MnAs bilayers. We consider a perfectly unreconstructed interface, just to sense how bonding effects influence the epilayer anisotropy in this limiting growing case, where no alloying is being taken into account.

A. Magnetoelastic Effects

In the presence of strain, the equilibrium out-of-plane lattice constant of the grown epilayer should change so as to keep the equilibrium bulk volume of Fe. For the Fe film grown in the [2-11] direction, the in-plane directions are [11-1] and [011]. In this case, the unstrained Fe in-plane parameters are 4.95 Å and 4.05 Å, respectively. When a few layers of Fe epitaxially grow on the MnAs(110) substrate, the previously mentioned in-plane parameters go over to 5.69 Å and 3.73 Å, respectively. The resulting stress is the origin of the biaxial strain, which is reflected in the strain tensor components. These tensor components are $\varepsilon_{1,1} = 0.131$ ($Fe_{[11-1]}$) and by $\varepsilon_{2,2} = -0.079$ ($Fe_{[011]}$). The misfit produces, then, a considerable elastic energy of 39 meV/atom, which is a factor 10^4 larger than the magnetic uniaxial anisotropy energy (k) of bulk Fe (4 μeV/atom [13]) and 10^2 larger that the uniaxial magnetic anisotropy energy reported for ultrathin Fe-films grown on MnAs, which is 0.62 meV/atom for temperatures outside the coexistence region [2]. This implies that the large interfacial strain could induce a modification on the Fe anisotropy properties. The magnetoelastic energy density, $E_{\rm me}$, can give us an insight into how the magnetization directions are influenced by strain, giving rise to changes in the magnetic anisotropy energy. Following Kittel [14], we use for the magnetoelastic energy the expression

$$E_{\rm me} = B_1\left(\epsilon_{1,1}\alpha_1^2 + \epsilon_{2,2}\alpha_2^2 + \epsilon_{3,3}\alpha_3^2\right) + B_2\left(\epsilon_{2,3}\alpha_2\alpha_3 + \epsilon_{3,1}\alpha_1\alpha_3 + \epsilon_{1,2}\alpha_1\alpha_2\right) \quad (1)$$

where B_1 and B_2 are the magnetoelastic coupling coefficients, α_i refers to the direction cosines of the magnetization and $\epsilon_{i,j}$ are the elements of the strain tensor. α_i and $\epsilon_{i,j}$ are referred to as the orthogonal directions which define the film plane, see Fig. 1. If we use bulk values for B_1 and B_2 [13] and the values for the strain tensor elements given above, the angle dependence of the magnetoelastic energy is the one shown in Fig. 2. Note that the magnetoelastic energy is just one order of magnitude smaller than the magnetic anisotropy energy reported for Fe-thin films [2]. This implies a competition between the magnetic anisotropy due to the crystal field and the magnetoelastic anisotropy due to surface strain. From Fig. 2, it can be observed that for Fe grown on MnAs(110), the magnetoelastic energy contribution favors the [11-1] direction ($\phi = 0$) within the plane. The in-plane biaxial strain induces a perpendicular strain because it tends to keep the Fe-volume constant. If we use the $\epsilon_{1,1}$ and $\epsilon_{2,2}$ given above for the case of the MnAs substrate, the perpendicular strain for the Fe epilayer is $\varepsilon_{3,3} = -0.052$. We employ strain values to obtain the angular dependence of the magnetoelastic energy. The inset of Fig. 2 shows the magnetoelastic energy dependence of the film with the azimuthal angle of the magnetization going from the out-of-plane to two different in-plane directions. It is observed that the in-plane direction [11-1] is always favored. Thereafter, one would expect in the case under study that the in-plane anisotropy should overcome the out-of-plane one, at least when the number of layers is large enough to consider bulk magnetoelastic constants.

As reported by Sander [13], in ultrathin films the magnetoelastic constants can present values very different from the bulk ones. In order to make an approach to the extreme case of ultrathin Fe layers on MnAs, we report in the next sections the *ab initio* results obtained for supported and unsupported Fe layers with and without strain.

B. Spin Orbit Coupling Contribution to MAE

As we want to disentangle the effects on the MAE produced by iron thin film width, interface and strain, different situations are considered. Calculations are done, then, for 1/3/6/12 unsupported and supported Fe layers (on MnAs). For the unsupported cases we consider strained and unstrained Fe films. For one Fe-layer on MnAs with As termination, we find that Fe occupies Mn positions following the MnAs structure. After relaxation of the internal coordinates the Fe and As end up lying in the same plane and exhibiting a strong covalent bonding. When three Fe layers are deposited on MnAs, there is no mixing with the substrate after relaxation. For thicker Fe-layers all calculations are done for unsupported strained and unstrained films.

The local SOC expectation value, obtained from $\mathrm{E}_{SO} = \langle (1/c^2)(1/r)(d\mathrm{V}/dr)l.s \rangle$, is integrated inside each atomic sphere, whose radius is defined by the used PAW pseudopotentials. In E_{SO}, c is the velocity of light, r is the radial distance within each atomic sphere, V is the effective potential as a function of r; l and s are the orbital and spin operators. The local magnetic anisotropy energy (LMAE) due to each atom is given by

$$\mathrm{LMAE} = \Delta E_{SO} = E_{SO}(\theta_j, \phi_j) - E_{SO}(\theta_k, \phi_k) \qquad (2)$$

where θ and ϕ are the angles defining a given quantization axis. The reference frame to define these angles is the one in Fig. 1. For supported and unsupported films of one/three strained Fe-layers, we obtain that the in-plane easy axis given by LMAE agrees with the one favored by the magnetoelastic energy as calculated in Section III-A $(\phi = 0)$. This is different from what happens for the same Fe-films when they are unstrained.

In Fig. 3, we show the in-plane evolution of the LMAE for an unsupported Fe monolayer when it evolves from the unstrained situation to the MnAs strained one. Note that when the monolayer is unstrained the minimum LMAE is attained for $\phi = \pi/2$ $(\mathrm{Fe}_{[011]})$, while for the strained situation this direction becomes the hard magnetic anisotropy one. This implies that the strain induces a rotation of the in-plane easy axis of the Fe-film. Moreover, the modulus of the in-plane LMAE is reduced by almost 50% of its unstrained value. This LMAE intensity reduction due to the MnAs induced strain points towards a tendency to a more isotropic in-plane anisotropy. For the thicker unsupported Fe-films the same trends are obtained.

We now include the effect of the substrate, not only the effect of its induced strain, and calculate the LMAE for one/three

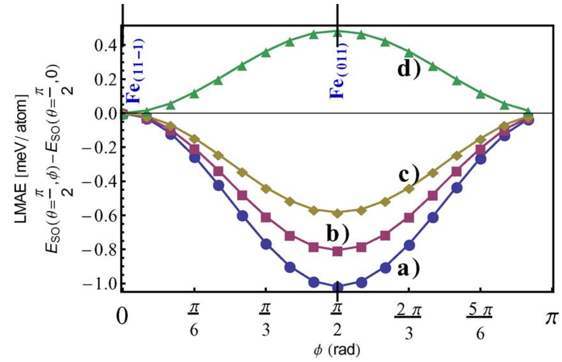

Fig. 3. In-plane LMAE versus angle for an unsupported Fe-monolayer. Different curves correspond to increasing strains. (a) Unstrained monolayer. (b) and (c) Intermediate strain cases. (d) Corresponds to the largest strain considered, equal to the one given by the MnAs substrate.

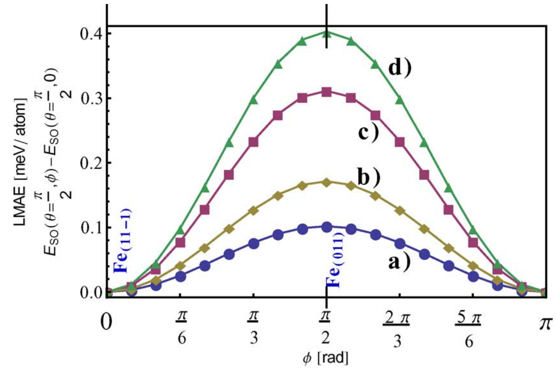

Fig. 4. Local MAE versus in-plane angle for MnAs supported one/three Fe-layers. LMAE contribution per atom of each film layer is given. (a) Interface Fe-layer. (b) Outermost Fe layer. (c) Middle Fe layer. (d) Fe-monolayer. The easy axis is the same for the monolayer as for the trilayer and the same as for the strained unsupported films.

layers of Fe supported on MnAs. In Fig. 4, we give the in-plane layer by layer contribution to the MAE for these two cases. The results are to be compared with the curve shown in Fig. 3(d) for the strained Fe unsupported monolayer. It is clear that the main effect of the MnAs substrate is to provide the strain that triggers the magnetic easy axis rotation of the Fe-film. It is interesting to remark that the easy axis of the α-MnAs(110) substrate is perpendicular to the easy axis of the strained Fe-film, as it has been experimentally reported by Steren *et al.* [15]. In Fig. 5, we show the calculated in-plane LMAE, layer by layer for the Mn atoms of the substrate, which agrees with the previously mentioned experiments. These results mean that the magnetic anisotropy of the Fe epilayers is not coupled to the magnetic anisotropy of the MnAs substrate, as it was suggested by previous experimental results [3]. The nonstrain effect of the substrate, that is the interface effect, is to lower the value of the anisotropy energy with respect to the unsupported Fe-films.

As discussed in Section III-A, the magnetoelastic energy of strained Fe favors the in-plane orientation of the easy axis (see the inset of Fig. 2). *Ab initio* calculations for Fe grown in the [2-11] direction show the same behavior in the case of one Fe-layer, see Table I. However, for the three layers Fe-films, the scenario is reversed, the magnetic easy axis lies out of plane. If we regard at the local anisotropy contribution of each Fe-layer, the one in contact with As favors the in-plane magnetic orientation while the other two layers have an out-of-plane contribution, which in the average results in an out-of-plane LMAE for

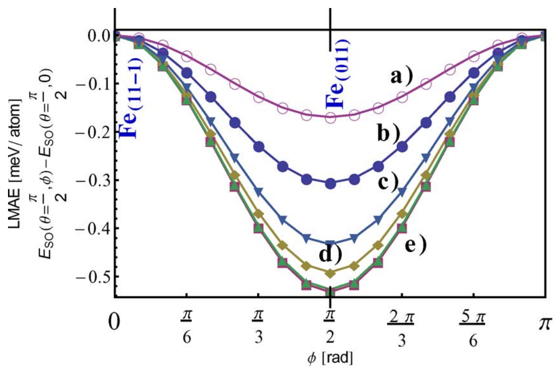

Fig. 5. Local MAE of Mn versus in-plane angle for an MnAs slab supporting a three Fe-layer epilayer. (a) LMAE on the outermost Mn-layer of the slab. (b) Mn-layer at the Fe/MnAs interface. (c), (d) and (e) Middle layer contribution to the MAE. Mn easy axis has a different direction that the on top strained Fe as it is shown in Fig. 4.

TABLE I
AVERAGED LOCAL OUT-OF-PLANE MAE ($E_{SO}((\pi/2),0)-E_{SO}(0,0)$) FOR DIFFERENT Fe FILM THICKNESSES. IN THE LABELS GIVEN IN PARENTHESIS, FIRST LETTER STANDS FOR UNSUPPORTED OR SUPPORTED (u/s) AND SECOND LETTER STANDS FOR UNSTRAINED OR STRAINED (u/s). STRAIN CONSIDERED IS THE ONE PROVIDED BY α-MnAs(110) SUBSTRATE (SEE TEXT)

# of Fe layers	LMAE [meV/atom]	# of Fe layers	LMAE [meV/atom]
1 (uu)	-0.17	3 (ss)	0.01
1 (us)	-0.36	6 (uu)	0.24
1 (ss)	-0.40	6 (us)	0.22
3 (uu)	0.89	12 (uu)	0.13
3 (us)	0.15	12 (us)	-0.07

the film. This means that the interface effect with the substrate induces an in-plane contribution along the [1-11] direction.

For the sake of completeness, we have calculated the perpendicular LMAE for unsupported strained and unstrained thicker films. In Table I, we summarize the results obtained for the different cases. Even if the magnetic easy axis changes from in-plane to out-of-plane for the three and six Fe-layer slabs, the shape anisotropy ends up inducing an in-plane easy axis already for the 12 layer Fe films, as obtained from the magnetoelastic energy contribution calculated using bulk constants. But in all strained cases the in-plane LMAE rotates 90° (pointing in the $Fe_{[1-11]}$) with respect to the unstrained easy axis.

IV. CONCLUSION

We have argued in this contribution that, at phase coexistence, the α zones of striped MnAs/GaAs(001) provide constraints strong enough so as to induce MnAs lattice parameters on deposited thin Fe epilayers. We have shown that the MnAs substrate induced strain gives rise to a change in the in-plane magnetic anisotropy of supported Fe films. This strain, namely, triggers a 90° rotation of the magnetic easy axis, in agreement with the experiments reported in [2].

We have also shown that the magnetic anisotropy of the Fe epilayers is not coupled to the magnetic anisotropy of the MnAs substrate, as it has been suggested by previous experimental results [3].

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