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Fe/MnAs bilayers: Magnetic anisotropy and the role of the interface

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ABSTRACT

We have studied different aspects of the magnetic behavior of Fe(5 nm)/MnAs(100 nm) bilayers epitaxially grown on GaAs(1 0 0). Ferromagnetic resonance (FMR) measurements were performed in order to characterize the magnetic anisotropies of the films and the interlayer coupling between them. The chemical composition of the interface was investigated by X-ray photoemission spectroscopy (XPS).

The iron layer FMR spectrum is highly temperature dependent between 280 K and 320 K as its parameters are strongly altered in the α/β phase coexistence regime of the MnAs layer, revealing a strong interlayer coupling in this *T*-region. Additionally, the XPS experiments demonstrate the existence of an Fe–MnAs intermixed interface. This result would explain the appearance of an additional uniaxial anisotropy in the plane of the Fe films, together with a strong reduction in its magnetocrystalline anisotropy and a negligible direct exchange coupling in the MnAs ferromagnetic state.

MnAs thin films have been intensively studied due to its physical properties, specially profitable for spintronic applications [1]. MnAs grows epitaxially onto GaAs, a typical semiconductor component of electronic devices. Further, MnAs is ferromagnetic at room temperature, becoming a potential source of spin-polarized currents [2–4] in magnetoelectronic devices.

When MnAs films are deposited onto GaAs(1 0 0) substrates, they develop a phase coexistence regime [5,6] that occurs between 283 K and 313 K, approximately. It is characterized by a striped self-organized pattern of α (ferromagnetic) and β (paramagnetic) phases [7], oriented parallel to the GaAs(1 0 0)-axis, or equivalently to the MnAs *c*-axis. Actually, MnAs/GaAs(1 0 0) structures have been proposed as templates of very thin magnetic layers. The former studies on Fe/MnAs bilayers [8–10] focused especially on the MnAs phase coexistence region. They revealed that the Fe layer couples antiferromagnetically to the MnAs buffer in the temperature range where the α/β coexistence occurs. The origin of this coupling was ascribed to the finite size of the α-MnAs stripes.

In this paper, we analyze the magnetic anisotropy of the Fe film, the interlayer coupling between both layers, and the correlation with the chemical composition of the Fe–MnAs interface.

The samples were prepared by molecular beam epitaxy (MBE) on GaAs(0 0 1) epi-ready substrates [9]. In situ RHEED and ex situ transmission electron microscopy (TEM) showed that Fe epitaxially grows on MnAs, with $[2\ -1\ 1]_{Fe} \parallel [1\ -1\ 0\ 0]_{MnAs} \parallel [0\ 0\ 1]_{GaAs}$ and $[1\ 1\ -1]_{Fe} \parallel [0\ 0\ 0\ 1]_{MnAs} \parallel [1\ -1\ 0]_{GaAs}$. The lattice misfit along the $[1\ 1\ -1]_{Fe}$ direction for this stacking is around 15%, and the misfit along the [0 1 1] direction is 8%. Due to the large lattice mismatch between Fe and MnAs lattices, the crystal structure of the 5 nm Fe layer may be strained and distorted, or relaxed by the introduction of defects [11]. On the other hand, the existence of Fe–Mn–As alloys at the Fe/MnAs interface cannot be excluded either, due to the high chemical reactivity of the components [12–14].

The FMR measurements were performed in an ESP-300 Bruker spectrometer at a microwave frequency $\omega \simeq 9.4$ GHz (X-band). We measured the angular dependence of the spectrum with the static magnetic field (H) rotating in the plane of the films (in-plane), or within a plane perpendicular to the surface of the samples (out-of-plane), and the temperature variation was done between 230 K and 350 K. The chemical composition of the sample was analyzed by performing a depth-profile analysis using XPS combined with an Ar beam (3 keV). The XPS spectra were acquired using $Al-K_\alpha$ radiation ($h\nu = 1486.6$ eV) while keeping the sample at room temperature. The $2P_{3/2}$ photoemission peaks of Zn, Fe, Mn and As were recorded after each sputtering step.

The in-plane FMR spectra of the Fe/MnAs bilayers show two resonance absorption lines, between 0 and 10 kOe. A thin line at low magnetic fields (hundreds of oersteds) is univocally associated with the Fe layer [15]. The second absorption, broader and much less intense, is centered close to 3 kOe. This corresponds to a gyromagnetic factor $g \simeq 2$ and so could be attributed to the resonance of

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paramagnetic ions or, alternatively, to the microwave absorption of an interlayer Mn–As–Fe compound. Actually, TEM measurements performed on similar samples put in evidence the existence of an interfacial alloy between Fe and MnAs [16]. As we discuss below, our XPS results reinforce this hypothesis.

Fig. 1a shows the in-plane angular variation of the resonance field (H_{res}) of the Fe layer at temperatures that are representative of the α-MnAs and β-MnAs phases. There is a dominant uniaxial term that superimposes to a subtle fourfold, cubic component. The minima of the curve at $\Phi_H = 0^\circ, 180^\circ$ signal the easy axis of magnetization [17]. The experimental data exhibits the same symmetry at both temperatures, revealing that the anisotropy terms (shape, magnetocrystalline, substrate-induced, etc.) of the Fe layer have approximately the same weight in the α and β phases of MnAs. In Fig. 1b, we show an out-of-plane variation of H_{res} of a Fe/MnAs bilayer at $T = 230$ K, i. e. when MnAs is in the FM, α phase.

The experimental points follow two behaviors: while the bottom curve is related to the resonance of the Fe layer, the upper curve corresponds to the absorption of the MnAs layer. The fact of being able to separate the data into two well different behaviors associated with the resonances of two single, independent layers, suggests that the films are very weakly coupled through a direct exchange mechanism, or not coupled at all when the MnAs layer is in the FM phase. The easy axis of magnetization of the Fe layer is parallel to the MnAs stripe pattern (i.e. to the hard axis of magnetization of MnAs), as deduced from additional data (not shown). In Ref. [9], the easy axis of the Fe films was found instead to be perpendicular to the MnAs c-axis. These results are related to the close dependence of the magnetic anisotropy of Fe deposited on MnAs thin films, on the growth procedure. Preliminary results obtained from numerical simulations support this hypothesis.

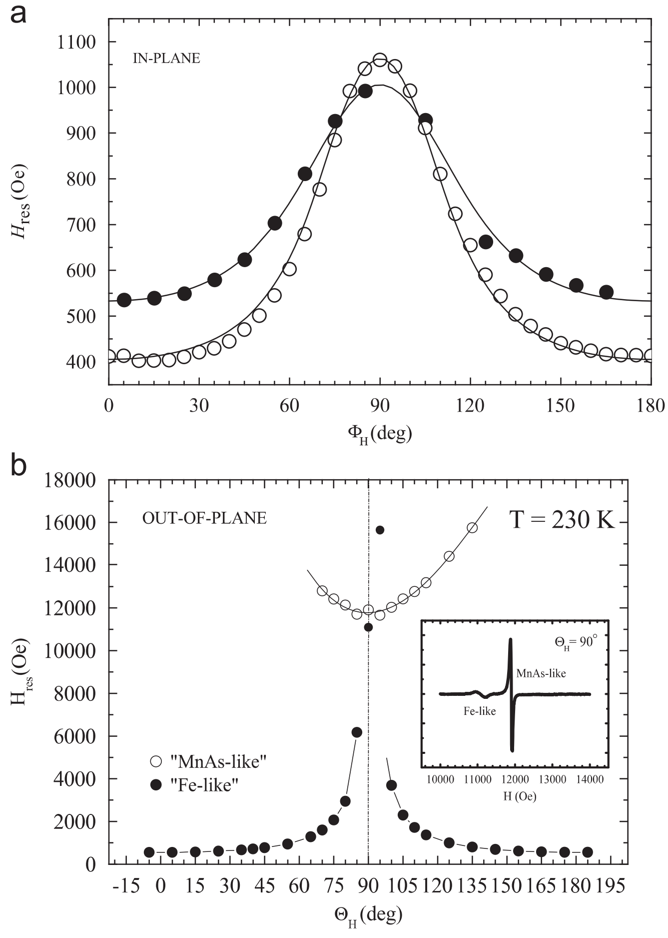

Fig. 1. (a) In-plane angular variation of H_{res} at $T = 230$ K (○) and 336 K (•). The solid lines are best fitting curves of the experimental data. (b) Out-of-plane angular variation of H_{res} at $T = 230$ K. The lines are guides to the eye. Inset: Detailed spectrum measured at the normal position $\Theta_H = 90^\circ$.

We propose a phenomenological model for the resonance of the Fe layer when MnAs is in the pure α phase

$$\begin{aligned} E = & -\mathbf{M}\cdot\mathbf{H} + \frac{\mu_o}{2} M^2 \sin^2\Theta \\ & + \frac{1}{12} K_4 [3\sin^4\Theta - 4\sqrt{2}\sin^3\Theta\cos\Theta\cos 2\Phi \\ & + 6\sin^2\Theta\cos^2\Theta\sin^2\Phi \\ & + 12\sqrt{2}\cos\Phi\sin^2\Phi\sin\Theta\cos^3\Theta \\ & + \cos^4\Theta(4\cos^4\Phi + 3\sin^4\Phi)] \\ & + K_u\cos^2\Theta\sin^2(\Phi - \Phi_u) - K_n\sin^2\Theta, \end{aligned} \tag{1}$$

where E is the energy density, $\mathbf{M}$ is the magnetization vector and Θ, Φ are the corresponding polar and azimuthal angles that give its orientation. The first and second terms are the Zeeman and the demagnetizing energy, respectively, and the third term accounts for the fourfold cubic magnetocrystalline (MC) anisotropy characterized by K_4. The x-axis has been chosen parallel to the c-MnAs and the $[1\ 1\ -1]$ Fe directions. The last two terms take into account interfacial anisotropies. The in-plane uniaxial anisotropy energy is determined by the orientation of the easy axis as given by Φ_u, and the parameter K_u. The out-of-plane term associated with K_n arises from the Fe/MnAs interface.

The Smit–Beljers [18] equation (2) was solved in a self-consistent way to obtain a numerical simulation for the behavior of H_{res}, and an estimation of the optimum set of the constants K_4, K_u and K_n for each temperature. It reads as

$$\left(\frac{\omega_{\mathrm{res}}}{\gamma}\right)^2 = \frac{1}{M^2\cos^2\Theta_{eq}}\left[\frac{\partial^2 E}{\partial\Theta^2}\frac{\partial^2 E}{\partial\Phi^2} - \left(\frac{\partial^2 E}{\partial\Theta\partial\Phi}\right)^2\right], \tag{2}$$

where ω_{res} is the resonance frequency, γ is the gyromagnetic ratio, and M is the magnetization of the Fe layer. The fits were done by taking [19] $M = 1723$ emu/cm^3 for $T = 230$ K, $M = 1692$ emu/cm^3 for $T = 336$ K, and by setting $\Phi_u = 0$ for the orientation of the easy axis.

The results are shown in Table 1. The out-of-plane constant K_n is of the same order of magnitude that previously reported data of surface and step anisotropies measured on Fe layers [20]. K_n is nearly temperature independent, and so cannot be associated with the striped domain structure of the underlying MnAs. Breitwieser et al. [12] recently showed that the surface morphology of the MnAs films exhibits elongated structures of 2 nm height and around 40 nm width, arranged periodically along the c-axis and highly disordered along the perpendicular direction. We associate the appearance of K_n to the array of troughs at the MnAs surface. The MC anisotropy constant K_4 is largely depressed as compared to the bulk iron value (48 kJ/m^3 at room temperature) [19]. The lack of fourfold symmetry, in addition to the appearance of a strong in-plane uniaxial anisotropy, strengthen the picture of a Fe layer grown with highly distorted structure, or the formation of a Fe–Mn–As alloyed layer, that has a crystalline structure of twofold symmetry [13,14]. The FeAs–MnAs magnetic

Table 1
Magnetic anisotropy constants of the iron layer, as determined from FMR data.

T (K)	K_4 (kJ/m^3)	K_u (kJ/m^3)	K_n (kJ/m^3)
230	9.0 (0.3)	30.0 (1)	−580 (10)
336	6.5 (0.3)	20.5 (1)	−630 (10)

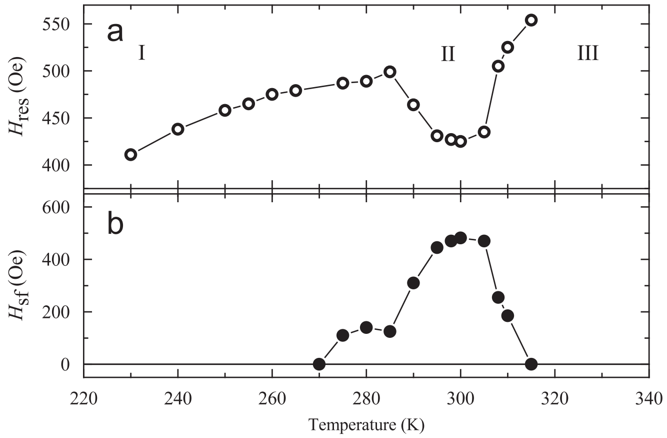

Fig. 2. (a) H_{res} vs. temperature. (b) Simulated H_{sf} coupling.

phase diagram [13] shows that only Mn-rich compounds exhibit ferromagnetism at room temperature, so we may discard the existence of a 5 nm fully-alloyed layer. However, we will show below that a thinner interfacial layer is consistent with our XPS results.

The T-dependence of H_{res} is shown in Fig. 2a, as measured in the in-plane geometry with **H** oriented parallel to the Fe easy-axis. A drastic decrease of H_{res} is observed in $280\ K \lesssim T \lesssim 315$ K. This behavior should be ascribed to the existence of a strong interaction between the MnAs magnetic structure and the Fe layer.

In Ref. [9], it has been analyzed the relevance of stray fields induced by the α-MnAs stripes onto the Fe film, and suggested that this effect gives rise to the magnetic coupling between MnAs and Fe in the MnAs phase coexistence regime. There are two main competing processes that govern the stray-coupling. On the one hand, as the temperature is increased the FM α stripes become narrower, the stray field becomes stronger, and the strength of the coupling increases. On the other hand, the magnitude of the stray-field depends also on the magnetization of MnAs: when the temperature increases the magnetization diminishes and the coupling field becomes weaker. To take into account this effect we fitted the FMR data as a function of temperature. The free energy density of the Fe layer was modified from Eq. (1) to consider the Fe/MnAs coupling. The stray fields are modeled by the introduction of an effective field, H_{sf} as follows: $E^{coupled} = E - H_{sf} \cdot M_{Fe}$. The H_{sf} direction is opposite to that of the MnAs magnetization. The anisotropy constants in the T-range $280\ K \lesssim T \lesssim 315$ K have been estimated by interpolating the previously calculated values. K_4 and K_u were made to vary linearly with temperature, while K_n was kept fixed. This assumption is based on previously reported data obtained from Fe/GaAs(1 0 0) films [21]. In Fig. 2b, we show the calculated stray field vs. temperature. It starts to grow at $T \simeq 275$ K and the maximum values are attained in the range $297\ K \simeq T \simeq 305$ K. Indeed, the maximum coupling of 470 Oe agrees very well with the ~ 450 Oe shift of the Fe layer hysteresis loop observed in XRMS measurements [9].

In order to investigate the interface composition, we studied the samples by XPS. The main results are shown in Fig. 3, where the evolution of the intensity of the $2P_{3/2}$ peaks as a function of the sputtering time is shown. These intensities were obtained from the peak area after a Shirley-type background substraction [22]. The initial raise of the Zn yield corresponds to the elimination of the surface contaminant layer. Instead, the decrease observed at 5 min of sputtering time corresponds to the removal of the ZnSe film deposited on top of the FeMnAs bilayer. Below the Zn layer, the Fe contribution appears and it extends from approximately 2–11 min of sputtering time. The long tail at longer etching times (> 11 min) can be attributed to re-implantation of Fe atoms inside the MnAs film. We also see (arrow in Fig. 3) that the Mn and As yields extend inside the Fe region, which can be related to an intermixing of the Fe/MnAs interface. This is deduced from the fact that the Mn and Fe $2P_{3/2}$ peaks have similar kinetic energies (842 eV and 774 eV, respectively), and therefore the corresponding electron inelastic mean free path, electron-analyzer transmission and photoemission cross section are similar. As a consequence, the Fe/MnAs interface is not abrupt, and the intermixing extends approximately half of the Fe layer. Assuming that the nominal width of this layer is 5 nm, the interface alloy should extend ~ 3 nm.

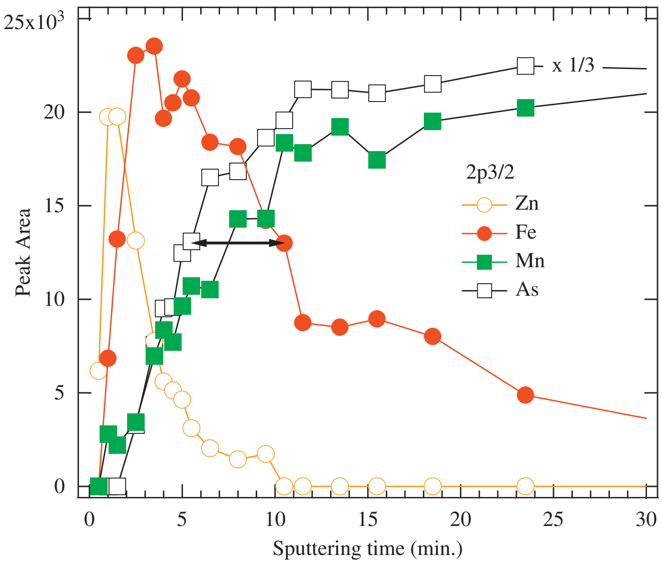

Fig. 3. Intensity of $2P_{3/2}$ peaks as a function of the sputtering time.

In summary, we have performed FMR and XPS experiments on Fe/MnAs bilayers epitaxially grown on GaAs(1 0 0) substrates. We calculated the different contributions to the magnetic anisotropy of the Fe layer as a function of temperature in the whole T-regime. The magnetization of this layer is similar to that of the bulk material, discarding the existence of a whole Fe–MnAs alloyed layer grown onto the MnAs underlayer. Besides, we note a strong reduction in the MC anisotropy and the appearance of an in-plane uniaxial contribution. We deduce that the magnetic anisotropy of the Fe layer do not seem to be particularly influenced by the MnAs α/β phase coexistence regime, but it seems to be mostly affected by the MnAs underlayer surface texture, the existence of the interfacial alloy, and the probable Fe lattice distortion due to the large lattice mismatch between film and underlayer. The out-of-plane FMR results at low temperature suggest that the direct exchange coupling between both layers is very weak in the FM (α) phase of MnAs. This fact is also reasonably explained by the XPS results, that allowed us to probe the existence of a Fe–Mn–As interfacial alloy at the Fe/MnAs interface.

We also showed that FMR spectrum of the Fe layer changes notably in the temperature region where α and β MnAs phases coexist. H_{res} exhibits an abrupt drop that we explain in terms of the MnAs stray field contribution to the free energy density of the Fe film. Finally, we have estimated a maximum value of this Fe/MnAs coupling of around 500 Oe between 297 K and 305 K.

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