Tailoring magnetization and anisotropy of tetragonal Mn₃Ga thin films by strain-induced growth and spin orbit coupling

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ABSTRACT

Tetragonal Mn₃Ga thin films were epitaxially grown with and without strain on Cr and Mo crystalline buffer layers, respectively, using rf-magnetron sputtering. Epilayers grown on Cr with a lattice mismatch of 4.16%, exhibit a high magnetization of 220 kAm⁻¹ and high perpendicular magnetic anisotropy. These characteristics are attributed to interfacial strain. Additionally, a soft ferromagnetic component is observed in these films but not in relaxed layers grown on Mo, where $\Delta a/a$ is -0.1% . These latest films exhibit a low magnetization of 80 kAm⁻¹ and both perpendicular and in-plane magnetic anisotropies. We propose that high spin orbit coupling of Mo-5s¹4d⁵ orbitals from the buffer layer and strong hybridization with Mn³⁺-3d⁴ orbitals from the magnetic layer are at the origin of in-plane anisotropy at the interface, while Mn₃Ga magnetocrystalline anisotropy leads to perpendicular anisotropy on the rest of the film.

1. Introduction

One of the most successful applications of materials sciences in spintronics is the construction of magnetic random access memories (MRAM), whose principal feature is its non-volatile capacity to store information [1–3]. The core of a MRAM is a magnetic tunnel junction (MTJ) where the writing process consists on switching the magnetic orientation of the free layer [4–6]. A practical method for this purpose, which simplifies the architecture of the MRAM [7], is the use of the spin-transfer torque (STT) phenomenon consisting on the magnetization or magnetization reversal of the free layer by the injection of a spin polarized pulse [8,9].

A material that has been proposed to fabricate the recording media on MTJs, which magnetization reversal can be achieved by the STT phenomenon, is the tetragonal phase of Mn₃Ga (D0₂₂) due to its high coercivity and high crystal anisotropy (without needing the presence of noble metals, e.g., FePt [10]). These are highly suitable magnetic properties to ensure the magnetic stability of single patterned bits at the sub-10 nm scale [11]. Additionally, its low magnetization allows faster magnetic switching with lower current pulses. On the other hand, tetragonal Mn₃Ga Heusler alloy is ferrimagnetic (FiM), with two Mn sublattices oppositely oriented along the *c* axis with magnetic moments (*m*) of 1.6 μ_B/Mn_X and $-2.8 \mu_B/\text{Mn}_Y$. Mn_X and Mn_Y atoms are

antiferromagnetically coupled while atoms of the same (001) plane (same Mn type atoms) are ferromagnetically coupled [12]. The growth of Mn₃Ga thin films is highly influenced by the substrate temperature (*T_s*) and the selected substrate or buffer layer. The effect of strain at the interface plays an important role in modulating the magnetic properties, e.g., magnetization and perpendicular magnetic anisotropy (PMA) [13]. Mn₃Ga thin films epitaxially grown on MgO(001)/Pt buffer layers, with a low lattice mismatch ($\Delta a/a$) of 0.4% exhibit a magnetization of 110 kAm⁻¹ whereas strained films grown on Cr-buffered substrates exhibit an increase up to 140 kAm⁻¹. In these samples, the uniaxial anisotropy constant (*K_u*) varies from 0.89 MJm⁻³ to 0.40 MJm⁻³, and the coercivity, $\mu_0 H_c$, remains scarcely unchanged: 1.7 T and 1.8 T, respectively [12,14]. The demanding size reduction of spintronic devices needs magnetic layers with high PMA. PMA has been observed at the interface between magnetic and heavy-non-magnetic layers as a consequence of strong hybridization between 3d orbitals from magnetic atoms and 4d or 5d orbitals from heavy metals [15]. The combination of spin-orbit coupling and the strong hybridization of *d* orbitals with heavy atoms as Pt, Pd, Au, Mo and W, produces a charge transfer thus leading to PMA [16].

In this paper we report the magnetization and anisotropy dependence on the crystalline quality of Mn₃Ga epilayers. Strained films grown on Cr buffer layers exhibit larger magnetization and PMA than

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relaxed layers grown on Mo buffer layers; this feature is attributed to interfacial strain. The Mn_3Ga layers grown on Mo exhibit also in-plane anisotropy. This issue is explained in terms of orbital hybridization of Mn^{3+} - $3d^4$ orbitals and Mo- $5s^14d^5$ orbitals with high spin-orbit coupling.

2. Experimental

Thin film growth has been performed in a magnetron-sputtering system with a base pressure of 5×10^{-8} Torr. The growth chamber is equipped with a reflection high-energy electron diffraction (RHEED) system, from STAIB operating at 30 kV, used to control the film crystalline quality and the lattice deformation. Mn and Ga atomic flux is obtained from a homemade Mn-Ga target fabricated according to the desired composition of the films [17]. A radio-frequency (rf) power source was used to erode the target at 25 W with a corresponding deposition rate of 0.6 nm/min. The growth of the Mn_3Ga layers was performed at $T_s = 340^\circ\text{C}$. The surface morphology was observed using atomic force microscopy with a Veeco SPM MultiMode equipment. The hysteresis magnetic loops were obtained in a superconducting quantum interference device (SQUID), from Quantum Design, at room temperature (300 K) and with an applied magnetic field up to 5 T. The diamagnetic contribution of the substrate has been removed in all the loops by subtracting the magnetic moment of a MgO substrate with the contribution of only the buffer layers.

3. Results and discussion

The selection of transition metals Cr ($a = 2.88 \text{ \AA}$) and Mo ($a = 3.15 \text{ \AA}$), both with cubic bcc crystalline structure, to elaborate the buffer layers on MgO(001) is based on the lattice mismatch between the buffer layers and the in-plane lattice of Mn_3Ga ($a = 3.91 \text{ \AA}$ and $c = 7.1 \text{ \AA}$) and hence on the effect of deformation of the Mn_3Ga layers. The growth of Mn_3Ga on the Cr buffer layer is achieved by a rotation of 45° of the Mn_3Ga cell regarding the [100] direction of Cr, i.e. a 1×1 cell of Mn_3Ga matches a $(\sqrt{2} \times \sqrt{2})R45^\circ$ cell of Cr within a lattice mismatch $\Delta a/a = 4.16\%$. The positive sign of $\Delta a/a$ implies that tensile deformation is induced on the film according to the expression $\Delta a/a = (a_{\text{substrate}} - a_{\text{Mn}_3\text{Ga}})/a_{\text{Mn}_3\text{Ga}}$, where a are lattice parameters. The growth on Mo is achieved after the Mn_3Ga cell rotates 30° regarding the [100] direction of the Mo buffer. Under this condition $\Delta a/a$ is reduced to -0.1% .

The epitaxial growth of Cr and Mo buffer layers on MgO(001) substrates has been performed at 600°C and 850°C , respectively, choosing the most convenient temperature, according to the literature [18,19]. The thickness of the buffer layers are 50 nm for Cr and 10 nm for Mo whereas all Mn_3Ga layers are of 50 nm. The RHEED patterns of the Cr and Mo buffer layers taken along the [100] and [110] directions are shown in the first row in Fig. 1(a) and (b), respectively. All buffer layers exhibit sharp streaky-RHEED patterns indicating that the films

are perfectly flat and monocrystalline along the complete thickness of the layer. Furthermore, on the RHEED pattern of the Mo surface along the [110] direction one can observe $\frac{1}{2}$ order streaks corresponding to a 2×1 reconstruction of the surface, confirming the high crystal quality of the film.

The second row shows the streaky RHEED patterns of the initial growth stage of Mn_3Ga on the Cr and Mo buffer layers indicating that a wetting layer is successfully formed. At the end of the growth (third row) 3-dimensional (3D) RHEED patterns consist on vertically elongated spots indicating that the films are rough at the surface but still monocrystalline as no rings are observed on the RHEED patterns, which would correspond to polycrystalline layers. The fact that the RHEED patterns consist on 3-dimensional spots and not on sharp streaks is due to the fact that electron diffraction takes place in transmission through Mn_3Ga bumps originated from the roughness at the surface. The layer-by-layer growth, at the initial stages, followed by 3D growth corresponds to an epitaxial Stranski-Krastanov mode.

The induced deformation (ϵ) on the films was evaluated with $\epsilon = (a_{\text{film}} - a_{\text{bulk}})/a_{\text{bulk}}$ where a_{bulk} stands for the theoretical lattice parameter of Mn_3Ga and a_{film} for that measured by RHEED at the surface of the films at different thicknesses. For the growth on Cr, the relaxation of the films occurs progressively as the thickness increases. Hence, at the interface the film matches perfectly the lattice parameter of the buffer layer with $\epsilon = 4.16 \pm 0.3\%$; as the thickness increases to 20 nm, the induced deformation decreases to $\epsilon = 1.07 \pm 0.3\%$, and for a thickness of 50 nm the film is completely relaxed, showing no deformation. Meanwhile, in the case of Mo, the negligible value of the lattice mismatch, $\Delta a/a = -0.1\%$, allows the growth of a relaxed film along all the thickness of the film (within the accuracy of the measurement).

Fig. 2(a) and (b) show $2 \mu\text{m} \times 2 \mu\text{m}$ atomic force microscopy images of the surface of 50 nm-thick Mn_3Ga films grown on Cr and Mo buffer layers, respectively. The film grown on Cr exhibit higher root-mean-square roughness (rms = 11 nm) and z-scale ($z = 80$ nm) than the film grown on Mo (6.3 and 57 nm, respectively). The high rms of the former is attributed to different growth rate at different regions of the surface. Strained regions are not energetically favorable as adsorption sites for adatoms, thus the growth is favored at relaxed regions (i.e., between dislocations, which may be the relaxation mechanism). Mn and Ga atoms arise uniformly to the surface of the film but surface diffusion promotes the migration of adatoms towards unstrained regions.

The lower rms and z scale of the films grown on Mo agrees well with the higher surface free energy of Mo (3 J/m^2) compared to that of Cr (2.3 J/m^2) [20]. The color contrast in the images corresponds to terraces composed of several atomic layers. The island-like growth agrees well with the 3D-RHEED patterns of 50 nm-thick films where electron diffraction occurs in transmission within Mn_3Ga bumps.

Fig. 3 shows the hysteresis loops, parallel ($\parallel$) and perpendicular ($\perp$) to the film, measured at room temperature for the Cr/ Mn_3Ga (a) and

Fig. 1. The first row shows the RHEED patterns of the surface of (a) Cr and (b) Mo buffer layers along [100] and [110] directions. Second and third rows are the RHEED patterns of Mn_3Ga films at two different thicknesses: 1 nm and 50 nm.

Fig. 2. Atomic force microscopy images of the surface of 50 nm-thick Mn_3Ga layers grown on (a) Cr and (b) Mo buffer layers.

Fig. 3. SQUID $M^{\perp}(H)$ and $M^{\parallel}(H)$ of Mn_3Ga layers grown on (a) Cr and (b) Mo buffer layers at 340 °C. Decomposition of the FM and FiM components for both samples are shown in the insets.

$\text{Mo}/\text{Mn}_3\text{Ga}$ (b) bilayers. First of all, it is noteworthy that all the measurements, except the parallel one of the $\text{Cr}/\text{Mn}_3\text{Ga}$ bilayer (see Fig. 3(a)), exhibit a characteristic “hummingbird-like” shape, with a low-field step in the upper branch hysteresis loop. This shape is typically found when there is coexistence of two magnetic phases [21,22]. In our case, it seems therefore that a soft ferromagnetic (FM) phase appears in addition of the hard FiM one.

The contribution of this soft FM phase can be fitted, for example, using the Brillouin function $B(J, a')$ and then subtract it to the experimental data to evaluate the pure FiM component of the film [17]:

$$\frac{M}{M_s} = \frac{2J+1}{2J} \coth\left(\frac{2J+1}{2J}a'\right) - \frac{1}{2J} \coth\frac{a'}{2J} \quad (1)$$

In this eq. $a' = mH/kT$, m is the magnetic moment of a single Mn atom and $J = L + S$, where L is the orbital momentum, S is the spin momentum and J is the total angular momentum. In the perpendicular loop of the sample deposited on a Cr buffer, the best fit to the FM component is obtained with $J = 2$ (used in all the fits) and $M_s^{\text{FM}} = 80 \text{ kA m}^{-1}$. The pure FiM and FM components are shown in the inset of Fig. 3(a). The former has $M_s^{\text{FiM}} = 138 \text{ kA m}^{-1}$, $H_c^{\text{FiM}} = 1.4 \text{ T}$, and remanent magnetization, $M_r^{\text{FiM}} = 66\%$. The magnetization saturation of the loop (perpendicular to the film), $M_s^{\perp} = 215 \text{ kA m}^{-1}$, is quite high. A similar value of $M_s^{\perp} = 220 \text{ kA m}^{-1}$ has been reported by Kurt et al. on epitaxial Mn_3Ga films grown on Cr monocrystalline substrates at 250 °C whereas epitaxial Mn_3Ga films grown on a Cr buffer over a MgO substrate presented a fairly lower magnetization (140 kA m^{-1}) [13,14]. The enhancement of M_s in our samples is attributed to the modification of the interatomic distances between Mn_x and Mn_y atoms and between atoms of the same sublattice, by means of induced strain, thus increasing the direct exchange interaction, and modifying, therefore, the type of magnetic coupling: from AFM to FM at the strained regions. The fact that the saturation magnetization of our Cr-buffered sample is equal to the value reported by Kurt et al. [13] grown on Cr substrates can be attributed to the high crystalline quality of the buffer layer as depicted by the sharp streaky RHEED patterns in Fig. 1(a). It is possible that the growth of buffer layers without RHEED control can be rough thus introducing interfacial structural defects leading to low values of M_s .

Meanwhile, the loop measured with the applied field parallel to the film plane is practically anhysteretic and with a $M_s^{\parallel} = 120 \text{ kA m}^{-1}$ (see Fig. 3(a)). The slope of the magnetization at high fields is quite small (i.e., it seems to have reached saturation or almost at 5 T), which was not expected, given the PMA (it would be expected that the magnetization keeps increasing with the field, reaching the same M_s as $M_s^{\perp}$ at a higher magnetic field). The second phase (soft FM one) appearing in the perpendicular loop is most likely also present in this case but it is not discerned as the FiM phase is also anhysteretic and we are therefore not able to see the “hummingbird-like” shape (as there are not two magnetic phases with different coercive fields).

We have calculated K_u from the in-plane measurement using the expression $K_u = \mu_0 H_k M_s / 2$ (according to the Stoner-Wohlfarth model), finding a value of $K_u = 0.32 \text{ MJ m}^{-3}$, which evidences its high PMA.

Fig. 3(b) shows the hysteresis loops (perpendicular and parallel to the film) of the Mn_3Ga layer grown on Mo at $T_s = 340 \text{ °C}$. The saturation magnetizations $M_s^{\perp} = 74.8 \text{ kA m}^{-1}$ and $M_s^{\parallel} = 38.4 \text{ kA m}^{-1}$ are smaller than those of the sample grown on Cr due to the lack of strain. This lower magnetization can be quite relevant for certain applications, as low magnetization allows faster magnetization reversal and with a lower electric demand in magnetic tunnel junctions [19]. The soft FM components of the in-plane and out-of-plane measurements of 5 kA m^{-1} and 3 kA m^{-1} , respectively are shown in the inset of Fig. 3(b). It is important to note that both $M^{\parallel}$ and $M^{\perp}(H)$ loops are hysteretic for this

sample (in contrast to the previous sample). The observation of in-plane anisotropy on the relaxed Mo-buffered films is in agreement with a reduction of the PMA from $K_{u,\text{Cr}/\text{Mn}_3\text{Ga}} = 0.32 \text{ MJm}^{-3}$ to $K_{u,\text{Mo}/\text{Mn}_3\text{Ga}} = 0.15 \text{ MJm}^{-3}$, which agrees well with the value of 0.2 MJm^{-3} found by Kurt et al. [20]. Similar simultaneous IP and OP anisotropies have been observed on cubic Mn_{3-8}Ga films epitaxially grown on GaN [23].

Regarding the hummingbird-like loops and the soft FM phase, Jamer et al. [24] on $\text{Mn}_{2.5}\text{Ga}$ polycrystalline layers attributed to secondary phases with different stoichiometry or to the existence of strain in their nanoparticles. Taking into account all the precedent (including the RHEED measurements, pinpointing a high strain close to the Cr buffer), this FM soft phase is most likely originated at the strained regions, near the interface with the Cr buffer layer, as the changes in the interatomic distances affect the magnetic coupling. The presence of interfacial alloying at the interface (due to Cr diffusion into the first atomic layers of the Mn_3Ga layer) as another possible explanation is discarded as Cr is usually used as a diffusion barrier [25]. In the case of the film deposited on Mo, the strain is very low and, hence, this soft FM phase is fairly smaller, almost negligible. The presence of other secondary phases (e.g., $\text{Mn}_{1.6}\text{Ga}$ or $\text{Mn}_{2.5}\text{Ga}$) can be not totally be discarded but seems unlikely given the RHEED characterization and the fact that no reflections corresponding to other phases were found in the x-ray diffraction spectra obtained (not shown). Furthermore, Mo diffusion from the buffer layer towards the film cannot be discarded leading to the formation of secondary phases or simply doping the Mn_3Ga unit cell.

Note that in all the samples $M_s^\perp$ is higher than $M_s^\parallel$. This difference is likely originated from IP tensile deformation and contraction of the cell along the c axis [26]. This suggestion is valid for strained Mn_3Ga layers grown on Cr where $M_s^{\text{Cr}\perp}/M_s^{\text{Cr}\parallel} = 1.8$. However, the ratio of the films grown on Mo $M_s^{\text{Mo}\perp}/M_s^{\text{Mo}\parallel} = 2$ cannot be explained in this manner as the film is completely relaxed. An approach to solve this issue can be the elaboration of thinner layers with thickness $t = 5 \text{ nm}$, to have an insight of the pure interface interaction with the buffer layer and to confirm the fact that IP anisotropy takes place only near to the interface. Similar difference of M_s between IP and OP $M(H)$ loops has been observed on diluted magnetic oxides and on CeO_2 layers [27,28], however, this kind of PMA is not yet understood and deserves further investigation.

To further try to explain this issue in our samples we focus our explanation on the magnetic coupling at the interface between $\text{Mn}^{3+}-3d^4$ and Mo-5s^14d^5 orbitals inducing IP oriented domains. The valence state of Mn^{3+} with d^4 occupancy is found on Mn atoms octahedrally coordinated [29]. For Mo- or Ta-buffered CoFeB ultrathin films $t \sim 1 \text{ nm}$, the hybridization of Co-Fe- $3d$ orbitals with the d orbitals of heavy atoms of the buffer layer possessing high spin orbit coupling produces OP anisotropy [15,16]. Films with barely higher thicknesses exhibit IP anisotropy. Thus, in our $\text{Mo}/\text{Mn}_3\text{Ga}$ system the interface anisotropy produces IP anisotropy over a critical thickness at the interface with the buffer layer, probably larger than 1 nm , while the bulk anisotropy of the film produce PMA for the rest of the film. The geometry of the domains of the Mn_3Ga layer may obey to a flux closure domain pattern [30], which consists on vertical aligned domains (up and down orientations) and horizontal thin domains at the interface and at the surface of the layer completing a closed magnetic circuit. This model reinforces the idea of IP domain alignment at the interface. More research work is on progress to try to clarify these issues.

4. Conclusions

In summary, we have modified the magnetic properties of tetragonal Mn_3Ga thin films grown on Cr and Mo buffer layers at 340°C by magnetron sputtering. Epitaxial growth was achieved as depicted by RHEED patterns. Strained films grown on Cr, exhibit high perpendicular magnetic anisotropy and high magnetization while relaxed films

grown on Mo buffers exhibit low magnetization and both in-plane and out-of-plane anisotropy. On the Cr-buffered layers, additionally to ferrimagnetic ordering, a soft ferromagnetic component arises from strained regions, which magnetization depends on the growth temperature. This component is negligible on unstrained layers grown on Mo buffers. The in-plane anisotropy of $\text{Mo}/\text{Mn}_3\text{Ga}$ layers is attributed to a strong hybridization of $\text{Mn}^{3+}-3d^4$ orbitals with Mo-5s^14d^5 orbitals with high spin orbit coupling.

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