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Tailoring magnetization and anisotropy of tetragonal Mn_3Ga thin films by strain-induced growth and spin orbit coupling

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ABSTRACT

Tetragonal Mn_3Ga 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-$5s^1 4d^5$ orbitals from the buffer layer and strong hybridization with Mn^{3+}-$3d^4$ orbitals from the magnetic layer are at the origin of in-plane anisotropy at the interface, while Mn_3Ga 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 Mn_3Ga ($D0_{22}$) 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 Mn_3Ga 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 μ_B/Mn_Y. Mn_X and Mn_Y atoms are antiferromagnetically coupled while atoms of the same (00*l*) plane (same Mn type atoms) are ferromagnetically coupled [12]. The growth of Mn_3Ga 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]. Mn_3Ga 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 3*d* orbitals from magnetic atoms and 4*d* or 5*d* 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 Mn_3Ga 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-$5s^1 4d^5$ 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 $T_s = 340$ °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 ($a = 2.88$ Å) and Mo ($a = 3.15$ Å), 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 ($a = 3.91$ Å and $c = 7.1$ Å) 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 $(\sqrt{2} \times \sqrt{2})R45°$ 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 $\Delta a/a = (a_{substrate} - a_{Mn3Ga})/a_{Mn3Ga}$, 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 ½ 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 $\varepsilon = (a_{film} - a_{bulk})/a_{bulk}$ 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 $\varepsilon = 4.16 \pm 0.3\%$; as the thickness increases to 20 nm, the induced deformation decreases to $\varepsilon = 1.07 \pm 0.3\%$, 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 μm × 2 μm 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 (∥) and perpendicular (⊥) to the film, measured at room temperature for the Cr/Mn_3Ga (a) and

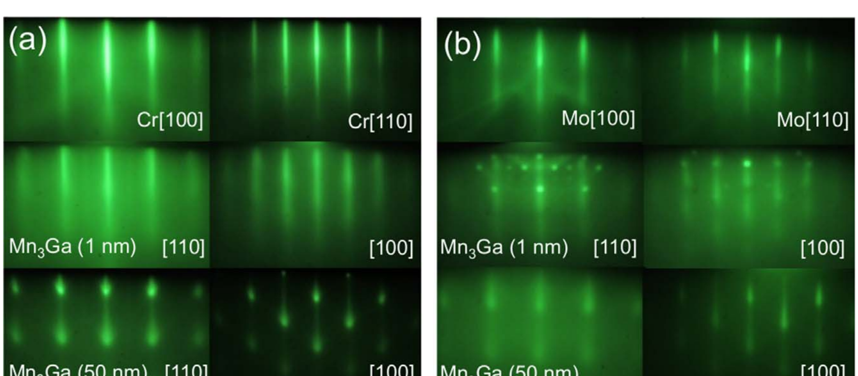

Fig. 1. The first row shows the RHEED patterns of the surface of (a) Cr and (b) Mo buffer layers along [100] and [110] directions. Second and third rows are the RHEED patterns of Mn_3Ga films at two different thicknesses: 1 nm and 50 nm.