Interfacial coupling effect of Cr$_2$O$_3$ on the magnetic properties of Fe$_{72}$Ga$_{28}$ thin films

I. Hontecillas$^1$, M. Maicas$^2$, J. P. Andrés$^3$ & R. Ranchal$^{1,4,*}$

Here it is investigated the effect of the antiferromagnet Cr$_2$O$_3$ on the magnetic properties of ferromagnetic Fe$_{72}$Ga$_{28}$ thin films. Sputtered Fe$_{72}$Ga$_{28}$ layers have their magnetization in the sample plane with a magnetic fluctuation that gives rise to magnetic ripple. In order to turn its magnetization into the out of plane (OOP) direction, it has been magnetically coupled with Cr$_2$O$_3$ that has magnetic moments along the c-axis, that is the perpendicular direction when properly aligned. Cr$_2$O$_3$ has been obtained from Cr oxidation, whereas Fe$_{72}$Ga$_{28}$ has been deposited on top of it by sputtering in the ballistic regime. Although a uniaxial in-plane magnetic anisotropy is expected for Fe$_{72}$Ga$_{28}$ thickness above 100 nm, the interfacial coupling with Cr$_2$O$_3$ prevents this anisotropy. The formation of stripe domains in Fe$_{72}$Ga$_{28}$ above a critical thickness reveals the enhancement of the out of plane component of the Fe$_{72}$Ga$_{28}$ magnetization with respect to uncoupled layers. Due to the interface coupling, the Fe$_{72}$Ga$_{28}$ magnetization turns into the out-of-plane direction as its thickness is gradually reduced, and a perpendicular magnetic anisotropy of 3·10$^6$ erg·cm$^{-3}$ is inferred from experimental results. Eventually, the coupling between Cr$_2$O$_3$ and Fe$_{72}$Ga$_{28}$ promotes an exchange-bias effect that has been well fitted by means of the random field model.

Control of the magnetization is an important issue for the development of pioneering magnetic devices. Typically, thin films have the magnetization in the sample plane in order to reduce the energy of the system$^1$. However, in many applications as for example: high density magnetic storage, spintronic devices, non-volatile random access memories, logic devices, skyrmions or sensors, materials with OOP magnetization are desirable$^2$–$^6$.

Systems with large perpendicular magnetic anisotropy (PMA) such as L$_1_0$ FePt and CoPt thin films are extensively investigated but, their large coercivity and switching fields can represent a drawback for their integration in devices$^7$–$^9$. Therefore, it is of interest the investigation of other materials with PMA, or the possibility of turning the magnetization into the OOP direction. Stripe domains appear above a critical thickness when a moderate PMA is present$^9$. In permalloy films, stripes have been observed because of columnar growth$^{10}$, but they have also been promoted when coupled with NdCo$^{11}$. Recently, the magnetization direction has been tuned in Fe–N layers by ion implantation and heat treatment conditions$^8$, and by annealing in Fe$_5$Si$_2$B$_1$.$^{12}$ Even more notably, the control of the magnetic anisotropy by means of voltage has been observed at magnetic transition metal/oxide interfaces$^8$.

FeGa alloys are extensively studied because of their large magnetostriction constant and low coercivity.$^{13}$–$^{16}$ Also interesting is the possibility in sputtered Fe$_{72}$Ga$_{28}$ layers of controlling the in-plane magnetic anisotropy by growth conditions$^{17}$–$^{19}$ or by thermal treatments combined with mechanical stress$^{20}$. Molecular beam epitaxy (MBE) FeGa can exhibit domain stripes due to a low PMA$^{21}$–$^{23}$, but the general behavior observed by magnetic force microscopy (MFM) in FeGa deposited either by electro deposition$^{19}$, sputtering$^{20}$ or even MBE$^{21}$ is a magnetic ripple due to magnetic fluctuations in layers with magnetization in the sample plane.

In this work, we have explored the possibility of turning the Fe$_{72}$Ga$_{28}$ magnetization into the OOP direction when appropriately coupled with Cr$_2$O$_3$, an antiferromagnet that has its magnetic moments along the hexagonal c-axis being possible to have moments in the perpendicular direction when properly aligned$^{27}$–$^{29}$. Although Cr$_2$O$_3$ has already been coupled with ferromagnetic metals,$^{30}$–$^{32}$ the interaction with Fe$_{72}$Ga$_{28}$ seems not a trivial problem. Fe$_{72}$Ga$_{28}$ has already been coupled with TbFe in [Fe$_{72}$Ga$_{28}$/TbFe$_1$] multilayers to turn its magnetization into the OOP direction$^{33}$–$^{34}$. However, only a tilt of the magnetization of around 25° with respect to the sample...
Experimental section

30 nm-thick Cr₂O₃ was synthesized from the evaporation of Cr on glass substrates that was subsequently oxidized in oxygen atmosphere at 750 °C during 3 h. Fe₇₂Ga₂₈ layers with a thickness ranging from 240 to 20 nm were grown by the DC magnetron sputtering technique in the ballistic regime at room temperature on top of the Cr₂O₃. The sputtering deposition was carried out in oblique incidence with an angle between the vapor beam and the perpendicular to the sample of about 25° and a distance of 9 cm between target and substrate. This direction of the vapor beam within the sample plane is taken as the reference direction to control the direction of the in-plane uniaxial anisotropy axis when created. Fe₇₂Ga₂₈ films were deposited from a target with a composition of Fe₇₂Ga₂₈ with a diameter of 5 cm and a thickness of 2 mm using an Ar pressure of 3·10⁻³ mbar and a growth power of 90 W in all cases. A 20-nm thick Mo layer was used as a capping layer to avoid FeGa oxidation. Mo was also deposited with a power of 90 W and with an Ar pressure of 3·10⁻³ mbar. Therefore, the structure of the studied samples is: glass/ Cr₂O₃/Fe₇₂Ga₂₈/Mo. For further comparisons, we have also deposited single Fe₇₂Ga₂₈ layers with thickness of 20 and 150 nm between Mo buffer and capping layers to avoid oxidation.

We have used X-ray diffractometry (XRD) in the Bragg–Brentano configuration to study the structural properties. Measurements were performed in a Philips X’Pert MPD using the Cu Kα wavelength (1.54056 Å). A Digital Instruments Nanoscope IIIa instrument was used to obtain MFM images. We monitored the cantilever’s phase of oscillation while the magnetic tip was scanning the sample surface to work in the phase detection mode. The distance between sample and surface was 40 nm on average (lift mode). The MFM measurements were performed without magnetic field after an OOP magnetic field of 10 kOe was applied. The topography of the samples has also been obtained in this microscope working as an Atomic Force Microscope (AFM).

In-plane and OOP hysteresis loops were performed in a vibrating sample magnetometer (VSM) from LakeShore 7304 at room temperature. In the sample plane, we measured loops at different angles between the applied magnetic field and the in-plane reference direction. As a reference direction, we considered as 0° the beam incidence direction in the sample plane. Hysteresis loops at 5 K were measured in a SQUID EverCool MPMS SQUID magnetometer from Quantum Design after field-cooling (FC) at 1 kOe from 360 K and zero-field-cooling (ZFC) at 0 Koe at 360 K. To reduce any hypothetical systematic error from the SQUID, a field step of 10 Oe was also deposited with a power of 90 W and with an Ar pressure of 3·10⁻³ mbar. Therefore, the structure of the studied samples is: glass/ Cr₂O₃/Fe₇₂Ga₂₈/Mo. For further comparisons, we have also deposited single Fe₇₂Ga₂₈ layers with thickness of 20 and 150 nm between Mo buffer and capping layers to avoid oxidation.

Results and discussion

In the XRD measurements we have only observed diffractions peaks related to either Cr₂O₃, FeGa or Mo (Fig. 1a). We have analyzed the possibility of Fe₇₂Ga₂₈ oxidation because of its growth on top of Cr₂O₃, but XRD measurements do not show evidences of oxidation within the resolution technique. The diffraction peaks related to FeGa are similar to what we have previously reported about sputtered Fe₇₂Ga₂₈ thin films being the (110) the main diffraction peak. For Fe₇₂Ga₂₈ we can determine the lattice parameter (a) thanks to Bragg’s law:

\[ n = 2d_{hkl} \sin \theta \]

where \( d_{hkl} \) is the family of planes, \( \theta \) the diffraction angle, and \( \lambda \) the radiation wavelength (Cu Kα in this case). In the cubic system, we can obtain \( a \) from \( d_{hkl} \) since:

\[ a = d_{hkl}(h^2 + k^2 + l^2) \]

where \( h, k, \) and \( l \) are the Miller indexes of the family of planes. From experimental results for the (110) peak we have determined a lattice parameter of 2.90 Å, in agreement with previous works.

On the other hand, the diffraction peaks of Cr₂O₃ show no variations upon the deposition of Fe₇₂Ga₂₈ on top of it as it can be inferred from the comparison with a single layer of Cr₂O₃ deposited in the same growth conditions (Fig. 1a). Experimental diffraction peaks for Cr₂O₃ appear at the same diffraction angles than for the file used for identification (01-084-0312) and therefore, it has a rhombohedral structure with lattice parameters \( a = b = 4.95 \) Å and \( c = 13.56 \) Å. Cr₂O₃ layers are not fully \( c \)-oriented since (012), (104) and (116) diffraction peaks for Cr₂O₃ have been detected by XRD. However, it is expected a magnetic contribution along the perpendicular direction due to these family of planes. The Néel temperature \( T_\text{N} \) of the Cr₂O₃ has been obtained from the measurement of the magnetization as a function temperature (Fig. 1b). The experimental value of \( T_\text{N} = 297 \) K is just 10 K below that of bulk Cr₂O₃, \( T_\text{N} = 307 \) K. Finally, we have also analysed the morphology of the samples by AFM (Fig. 2) being obtained a root mean square (rms) roughness of around 2 nm for the final stack.

Sputtered Fe₇₂Ga₂₈ layers deposited in the ballistic regime develop in-plane magnetic anisotropy above 100 nm. Nevertheless, the coupling with Cr₂O₃ completely eliminate this in-plane anisotropy even for thicknesses well above 100 nm (Fig. 3a). This can be understood considering that a sample will show PMA if two conditions are fulfilled: i) it is magnetically isotropic in the sample plane, and ii) the OOP direction is an easy axis in comparison to any direction in the sample plane. Therefore, the absence of in-plane magnetic anisotropy is a necessary condition for the PMA to be developed. We have ruled out that magnetostriction and strain have any effect on this modification of the magnetic anisotropy since we have not observed differences of the Fe₇₂Ga₂₈...
Figure 1. (a) XRD diffraction patterns of the samples studied in this work. The measurement for a 30 nm-thick Cr$_2$O$_3$ layer on glass has been included for further comparisons. Curves are shifted for clarity. (b) Magnetic moment versus temperature for the Cr$_2$O$_3$/Fe$_{72}$Ga$_{28}$(20 nm) sample. Inset: Detail of the measurement.

Figure 2. Morphology of the sample Cr$_2$O$_3$(30 nm)/Fe$_{72}$Ga$_{28}$(20 nm) measured by AFM in an area of 5 µm × 5 µm.
lattice parameter with respect to uncoupled single layers. In fact, when strain has been used to manipulate the magnetic anisotropy in Ga-rich FeGa thin films, only the direction within the sample plane was modified20. The VSM hysteresis loops reveal the change of the magnetization direction from in-plane to OOP as the Fe72Ga28 thickness is reduced (Fig. 3b). We can quantitively monitor this evolution by means of the OOP squareness ($\frac{M_r}{M_{\text{max}}}$) defined as the ratio between the remanence ($M_r$) and the maximum magnetization ($M_{\text{max}}$) obtained in the perpendicular hysteresis loops. In Table 1 we can observe how the squareness increases as the Fe72Ga28 thickness in the Cr2O3/Fe72Ga28 system, and for an uncoupled single Fe72Ga28 layer. Perpendicular magnetic anisotropy ($K_{\text{Fm}}$) for the Cr2O3/Fe72Ga28 system in those samples in which the OOP direction is a clear easy axis.

Table 1. OOP squareness ($\frac{M_r}{M_{\text{max}}}$) calculated from the perpendicular hysteresis loops as a function of the Fe72Ga28 thickness in the Cr2O3/Fe72Ga28 system, and for an uncoupled single Fe72Ga28 layer. Perpendicular magnetic anisotropy ($K_{\text{Fm}}$) for the Cr2O3/Fe72Ga28 system in those samples in which the OOP direction is a clear easy axis.

| Fe72Ga28 thickness (nm) | Cr2O3/Fe72Ga28 system FeGa thickness (nm) | Single Fe72Ga28 thickness (nm) |
|------------------------|------------------------------------------|-------------------------------|
|                        | 20 | 40 | 80 | 160 | 240 | 20 |
| $\frac{M_r}{M_{\text{max}}}$ (OOP direction) | 0.5 | 0.3 | 0.2 | 0.2 | 0.2 | 0.2 |
| Perpendicular $K_{\text{Fm}}$ (erg/cm³) | 2.8·10⁴ | 3.3·10⁴ | – | – | – | – |

The VSM hysteresis loops reveal the change of the magnetization direction from in-plane to OOP as the Fe72Ga28 thickness is reduced (Fig. 3b). We can quantitively monitor this evolution by means of the OOP squareness ($\frac{M_r}{M_{\text{max}}}$) defined as the ratio between the remanence ($M_r$) and the maximum magnetization ($M_{\text{max}}$) obtained in the perpendicular hysteresis loops. In Table 1 we can observe how the squareness increases as the
Fe$_72$Ga$_{28}$ thickness is reduced, mostly for a thickness below 80 nm. The OOP squareness and hysteresis loops of coupled layers can be compared with those of a Fe$_72$Ga$_{28}$ thin film grown in similar conditions (Table 1; Fig. 3c). The OOP squareness of a single layer is much lower with respect to that of a Fe$_72$Ga$_{28}$ with the same thickness but coupled to Cr$_2$O$_3$. Finally, in Fig. 3d it is presented the comparison between in-plane and OOP hysteresis loops for the Cr$_2$O$_3$/Fe$_72$Ga$_{28}$(20 nm) sample to show the magnetization direction has been turned into the perpendicular direction becoming the OOP direction an easy axis.

The perpendicular magnetic anisotropy, $K_{FM}$, can be experimentally inferred taking into account that:

$$K_{FM} = \frac{M_{FM} \cdot H_K}{2}$$

being $M_{FM}$ the saturation magnetization of the Fe$_72$Ga$_{28}$, and $H_K$ the anisotropy field in the hard direction. For the samples with Fe$_72$Ga$_{28}$ thickness of 20 nm and 40 nm the in-plane is a hard axis, and we have obtained an average value of $K_{FM}$ of $3 \times 10^6$ erg cm$^{-3}$ (Table 1) for a $M_{FM}$ = 1100 emu cm$^{-3}$. For higher Fe$_72$Ga$_{28}$ thickness, the OOP direction is not a clear easy axis due to the progressive turning of the magnetization into the perpendicular direction as the FeGa thickness is reduced.

MFM images can also be used to monitor the influence of the Cr$_2$O$_3$ on the Fe$_72$Ga$_{28}$ magnetic behavior (Fig. 4). When Fe$_72$Ga$_{28}$ is no coupled (Fig. 4a), it is observed the magnetic contrast known as ripple in agreement with previous works$^{24-26}$. However, due to the interfacial coupling with Cr$_2$O$_3$, the OOP component of the magnetization of Fe$_72$Ga$_{28}$ is enhanced, and stripe domains start to be visible in the MFM images (Fig. 4b–d).

In ferromagnetic materials with PMA, the quality factor $Q$ is defined as$^{39}$:

$$Q = \frac{K_{FM}}{2\pi M_{FM}^2}$$

For materials with moderate or low PMA, $Q < 1$, stripe domains appear above a critical thickness ($t_{cr}$):

$$t_{cr} = \frac{2\pi \sqrt{A_{ex}}}{K_{FM}}$$
where $A_{ex}$ is the exchange energy per unit length. When $Q > 0.1$, stripe domains are wider than the layer thickness, whereas they exhibit a periodicity equals to the layer thickness if $Q < 0.1$.

Taking into account the experimental value inferred for the perpendicular magnetic anisotropy, $K_{FM} = 3 \times 10^6$ erg/cm$^3$, and $M_{FM} = 1100$ emu cm$^{-3}$, it is inferred a $Q$ of 0.3 in our samples and therefore, stripes are expected above a critical thickness. Considering $A_{ex} = 1.7 \times 10^{-6}$ erg cm$^{-1}$ from the literature, and using Eq. (5), it is obtained an experimental $t_{cr}$ of 44 nm. This is in agreement with our experimental findings in which stripes have only been observed for Fe$_{72}$Ga$_{28}$ thickness $\geq$ 40 nm. In fact, if we take this experimental value as $t_{cr}$, a $K_{FM}$ of $4 \times 10^6$ erg cm$^{-3}$ is calculated, close to the experimental inferred value from experimental hysteresis loops, $3 \times 10^6$ erg cm$^{-3}$. Finally, from the MFM images we have obtained a stripe period between 110 and 125 nm by means of the power spectral density (supplementary information). This stripe periodicity higher than the layer thickness is consistent with the quality factor $Q$ higher than 0.1 calculated in our samples.

In addition to the rotation of the Fe$_{72}$Ga$_{28}$ direction magnetization towards the perpendicular direction, we have observed an exchange-bias effect related to the Cr$_2$O$_3$/Fe$_{72}$Ga$_{28}$ interfacial coupling as indicated by the shift of the OOP hysteresis loop in the horizontal axis ($H_E$) at 5 K after a FC process at 1 kOe (Fig. 5). This exchange-bias phenomenon in the perpendicular direction is related to the exchange-coupling between the antiferromagnetic Cr$_2$O$_3$, and the ferromagnetic Fe$_{72}$Ga$_{28}$.

For a Fe$_{72}$Ga$_{28}$ thickness of 20 nm, $H_E$ is $\sim$ 21 Oe, and $\sim$ 11 Oe for a thickness of 40 nm. For the lowest FeGa thickness, exchange-bias effect has been observed at least up to 200 K.

Since the perpendicular magnetic anisotropy inferred in this work for Fe$_{72}$Ga$_{28}$ ($K_{FM} = 3 \times 10^6$ erg cm$^{-3}$) is higher than the theoretical value of Cr$_2$O$_3$ ($K_{AF} = 2 \times 10^5$ erg cm$^{-3}$), and some chemical roughness is expected at the Fe$_{72}$Ga$_{28}$/Cr$_2$O$_3$ interface, we have used the random field model proposed by Malozemoff$^{35-37}$ to calculate the theoretical $H_E$ values. In this random field model, the AF layer breaks into domains, and the exchange-bias field is obtained by means of the expression:

$$H_E = \frac{2z\sqrt{A_{AF}K_{AF}}}{\pi^2M_{FM}^2}$$

where $z$ is generally taken as the unity, and $A_{AF}$ is the exchange stiffness of the antiferromagnet that takes a value of $4 \times 10^{-7}$ erg cm$^{-1}$.$^{42}$ With this expression (6) we obtain $H_E$ equals to $\sim$ 26 Oe and $\sim$ 13 Oe for Fe$_{72}$Ga$_{28}$ thickness of 20 and 40 nm, respectively, that are pretty close to the experimental $\sim$ 21 Oe and $\sim$ 11 Oe, respectively. This well agreement confirms the possibility of using this model in Cr$_2$O$_3$-based exchange-biased systems as also previously reported.$^{42}$

Figure 5. OOP hysteresis loops recorded at 5 K after FC at 1 kOe (filled triangle) and ZFC (open circle) from 360 K for samples with a FeGa thickness of (a) 20 nm, and (b) 40 nm.
Finally, from $H_e$ experimental values the interfacial exchange energy $\Delta E$ can be calculated:

$$\Delta E = H_e M_{FM} M_{FM}$$

(7)

For Fe$_{72}$Ga$_{28}$ of 20 nm and 40 nm, it is obtained a $\Delta E$ of 0.046 and 0.048 erg cm$^{-2}$ that is similar than reported in previous works in which Cr$_2$O$_3$ has been coupled with other ferromagnets with values of 0.05 erg cm$^{-2}$ at 5 K \cite{89}. However, it is important to remark that we have obtained that interfacial energy using polycrystalline Cr$_2$O$_3$, not a fully c-oriented Cr$_2$O$_3$ with all the magnetic moments aligned in the perpendicular direction. Nevertheless, our experimental results point out that even in this situation, it is possible to gradually turn the Fe$_{72}$Ga$_{28}$ magnetization into the out of plane direction due to the combination of Cr$_2$O$_3$ with Fe$_{72}$Ga$_{28}$ that has a magnetic fluctuation that promotes magnetic ripple in the uncoupled layers \cite{21}. Therefore, these results show the possibility of using polycrystalline layers for further applications such as tailoring of the magnetization direction.

Conclusions

In summary, we have studied the effect of the interfacial coupling between an antiferromagnet with magnetic moments along the c-axis (Cr$_2$O$_3$) and a ferromagnet (Fe$_{72}$Ga$_{28}$) with the magnetization in the sample plane but with a fluctuation that produces magnetic ripple. First of all, the in-plane magnetic anisotropy of Fe$_{72}$Ga$_{28}$ is completely vanished due to the coupling. Secondly, stripe domains are promoted due to the enhancement of the OOP component of the Fe$_{72}$Ga$_{28}$ magnetization. We have observed that the magnetization direction of Fe$_{72}$Ga$_{28}$ is gradually turned from in-plane to the OOP direction as the Fe$_{72}$Ga$_{28}$ thickness is reduced A perpendicular magnetic anisotropy of 3·10$^6$ erg·cm$^{-3}$ has been inferred from experimental results in the Fe$_{72}$Ga$_{28}$ layers. It has also been observed exchange-bias phenomena in the perpendicular direction fitting the experimental data to the random field model.

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Author contributions

I.H. grew part of the samples and magnetically characterized the samples by VSM and MFM. M.M performed part of the MFM experiments, and helped with VSM measurements. J.P .A. helped with the VSM and performed SQUID experiments. RR. thought the experiments, and grew part of the samples. All authors contributed to the discussion of the results. R.R wrote the main manuscript. All authors reviewed the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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