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Spintronic devices have been playing an important role in magnetic storage and memory applications for the last 20 years. For such a trend to continue, it is critical to develop new magnetic materials, in particular, in the context of antiferromagnetic spintronics, materials with high Curie temperatures, large spin polarizations, and low saturation magnetizations. One of the most unique magnetic elements is manganese. Elemental Mn is not ferromagnetic, despite having a d band that is more than half full. Of all the elements, the unit cell of the stable cubic α-Mn phase is the largest and most intricate. The unit cell of α-Mn is made up of 48 atoms distributed over four inequivalent sites, each site resulting in different magnetic moments ranging between 0.5 µB and 2.8 µB. Below 90 K, Mn orders in various antiferromagnetic and ferromagnetic structures [1, 2]. However, when combined in binary or ternary compounds, it leads to the emergence of a vast range of magnetic properties.

Large exchange bias induced by polycrystalline Mn₃Ga antiferromagnetic films with controlled layer thickness

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Abstract
Polycrystalline Mn₃Ga layers with thickness in the range from 6–20 nm were deposited at room temperature by a high target utilisation sputtering. To investigate the onset of exchange-bias, a ferromagnetic Co₀.₆Fe₀.₄ layer (3.3–9 nm thick) capped with 5 nm Ta, were subsequently deposited. X-ray diffraction measurements confirm the presence of Mn₃Ga (0002) and (0004) peaks characteristic of the D0₁₉ antiferromagnetic structure. The 6 nm thick Mn₃Ga film shows the largest exchange bias of 430 Oe at 120 K with a blocking temperature of 225 K. The blocking temperature is found to decrease with increasing Mn₃Ga thickness. These results in combination with x-ray reflectivity measurements confirm that the quality of the Mn₃Ga/Co₀.₆Fe₀.₄ interface controls the exchange bias, with the sharp interface with the 6-nm-thick Mn₃Ga inducing the largest exchange bias. The magneto-crystalline anisotropy for 6 nm thick Mn₃Ga thin film sample is calculated to be 9 × 10⁴ J m⁻³. Such a binary antiferromagnetic Heusler alloy is compatible with the current memory fabrication process and hence has a great potential for antiferromagnetic spintronics.

Keywords: Heusler alloys, antiferromagnet, exchange bias

(Some figures may appear in colour only in the online journal)
of magnetic properties, including room temperature magnetism, such as the ferromagnetic \( \text{Cu}_2\text{MnAl} \) Heusler alloy with \( T_c = 603 \text{ K} \), discovered by Heusler in 1903 \cite{3, 4}, long before the development of quantum mechanics.

As a binary Heusler alloy, many studies have been carried out experimentally and theoretically that address the magnetic properties of \( \text{Mn}_3\text{Ga} \) alloys. It is possible to stabilise the bulk \cite{5, 6} and thin films of \( \text{Mn}_3\text{Ga} \) (in an experimental context) as \( \varepsilon\text{-Mn}_3\text{Ga} \). This phase is antiferromagnetic with a hexagonal \( D0_{19} \) crystal structure which has a noncollinear triangular magnetic structure with antiferromagnetic behaviour \cite{5, 7}. This is comparable to the triangular magnetic state of a conventional \( \text{IrMn}_3 \) antiferromagnet \cite{8}. In the triangular antiferromagnetic structure three magnetic moments point in three different directions which causes the net magnetization to be zero \cite{9}. \( \varepsilon\text{-Mn}_3\text{Ga} \) has been reported to have a high Néel temperature of 470 K \cite{5}. \( \text{Mn}_3\text{Ga} \) also forms a tetragonal phase of \( \tau\text{-Mn}_3\text{Ga} \) with the \( D0_{22} \) structure \cite{10}. This ferrimagnetic \( \text{Mn}_3\text{Ga} \) is reported to possess a large uniaxial anisotropy of \( 1 \times 10^6 \text{ J m}^{-3} \) \cite{12} and a high Curie temperature of around 770K \cite{10}. Bulk- and thin-film forms of the hexagonal material can be annealed to realise the \( \tau\text{-Mn}_3\text{Ga} \) phase \cite{11}. The \( \text{Mn}_3\text{Ga} \) tetragonal phase has been grown epitaxially on different substrates \cite{13–15}. Both magnetisation and anisotropy are reported to be dependent on the \( \text{Mn}_{2.8}\text{Ga}_{1.2} \) alloy stoichiometry and the growth conditions of the \( \text{Mn}_3\text{Ga} \) films \cite{13, 15}. In this paper, we report on the thickness dependence of the antiferromagnetic/compensated ferrimagnetic behaviour in polycrystalline \( \text{Mn}_3\text{Ga} \) films. \( \text{Mn}_3\text{Ga} \) is reported to have the above properties thus it is important and promising for spintronics application.

The samples for this study were grown on Si(001) substrates at room temperature with a Ta (5 nm) and Pt (35 nm) seed layer and were capped with a Ta layer (5 nm) using a PlasmaQuest high target utilisation sputtering (HiTUS) system with a base pressure of \( 5 \times 10^{-5} \text{ Pa} \). The plasma was generated by a radio frequency (RF) field of 13.56 MHz in an Ar atmosphere of \( 3 \times 10^{-1} \text{ Pa} \). Platinum was used as a seed layer due to its good lattice match to \( \text{Mn}_3\text{Ga} \) \cite{8, 19}. The (0 0 0 1) plane of \( \text{Mn}_3\text{Ga} \) aligns with the (1 1 1) plane of Pt with a small lattice mismatch of 2%, as shown in figure 1. In order to confirm the film composition, we carried out energy dispersive x-ray (EDX) measurements on a sample area of \( 5 \times 5 \text{ nm}^2 \) and our results indicate the stoichiometry of our films to be \( \text{Mn}_{2.8}\text{Ga}_{1.2} \), which is close to the nominal composition of antiferromagnetic \( \text{Mn}_3\text{Ga} \). The thickness of \( \text{Mn}_3\text{Ga} \) is varied in the range from 3 to 20 nm \cite{16}. A ferromagnetic \( \text{Co}_{0.8}\text{Fe}_{0.2} \)
layer (3.3–9 nm) was deposited onto the Mn$_3$Ga layer in order to study the onset of exchange bias induced at the Mn$_3$Ga/Co$_0.6$Fe$_{0.4}$ interfaces induced by the antiferromagnetic Mn$_3$Ga.

The samples are first characterised by x-ray diffraction (XRD) in order to confirm the crystal structure of the Mn$_3$Ga layers. A Rigaku SmartLab x-ray diffractometer was used in this study. A Cu target was used as the x-ray source. The samples are measured in air. $\theta-2\theta$ XRD scans indicate the variation in the degree of the crystallisation with respect to different thicknesses of the Mn$_3$Ga layers from 3 nm to 20 nm as shown in figure 2. The XRD spectra show that, as the thickness of Mn$_3$Ga layer increases, the (0 0 0 2) and (0 0 0 4) peaks become more intense and sharper. This is because thicker Mn$_3$Ga layer provide a stronger signal. The 20 nm thick Mn$_3$Ga film shows a clear (0 0 0 2) peak at 41.3° as shown in figure 2. When the Mn$_3$Ga layer thickness is reduced to 6 nm the signal is too weak to distinguish the (0 0 0 2) peak from the Pt (1 1 1) peak. A pole figure scan is required for this purpose, shown in figure 2(c) for the 20 nm-thick Mn$_3$Ga film. From the pole figure scan for the 6 nm-thick Mn$_3$Ga film, the central peak confirms the presence of Mn$_3$Ga (0 0 0 2) peak at 41.3°, corresponding to the hexagonal $D_0_{19}$ phase. According to the distance from the outer ring to the centre spot, the in-plane rotation angle alpha is found to be 19.0° which indicates that the outer ring arises from polycrystalline Pt (1 1 1). It should be noted that the central Mn$_3$Ga (0 0 0 2) peak is well separated from that of the Pt (1 1 1). The Mn$_3$Ga layer is reported to be stabilised in a hexagonal $D_0_{19}$ crystal structure $\varepsilon$-Mn$_3$Ga with antiferromagnetic behaviour. The lattice parameters of the Mn$_3$Ga films in the $D_0_{19}$ and $D_0_{22}$ phases are listed in table 1. From the XRD scan, it is proved that the sample has purely $D_0_{19}$ antiferromagnetic phase.

X-ray reflectivity (XRR) measurements carried out for the 6 nm-thick-Mn$_3$Ga film confirm the presence of a sharp interface between the Mn$_3$Ga and FeCo layers as shown in
Using the GenX software to fit the data, the estimated thicknesses are: Ta (6.0 ± 0.4 nm)/Pt (34 ± 2 nm)/Mn$_3$Ga (5.8 ± 0.9 nm)/CoFe (4 ± 1 nm)/Ta (4 ± 1 nm).

The magnetic characteristics of the samples are measured using a vibrating sample magnetometer (VSM). The 20 nm-thick Mn$_3$Ga film without the CoFe ferromagnetic layer shows no magnetization. This result suggests that the film is antiferromagnetic. The magnetization curves for the polycrystalline Mn$_3$Ga/Co$_{0.6}$Fe$_{0.4}$ samples measured at 120 K after cooling under an applied magnetic field of 20 kOe are shown in figure 4. As can be seen in figure 4, the magnetization curves for films with varying Mn$_3$Ga thicknesses exhibit different behaviors. The CoFe layer exhibits a significant exchange bias, which increases with increasing Mn$_3$Ga thickness.

Figure 4. (a) Magnetization curves for the polycrystalline Mn$_3$Ga/Co$_{0.6}$Fe$_{0.4}$ (3.3 nm) films versus Mn$_3$Ga thicknesses of 6 (blue line), 10 (red line) and 20 nm (black line) measured at 120 K. (b) Magnetization curves for the polycrystalline Mn$_3$Ga (6 nm)/Co$_{0.6}$Fe$_{0.4}$ films CoFe thicknesses of 3.3 (black line), 6 (red line) and 9 nm (blue line) measured at 120 K.

Figure 5. Magnetization curves for the 6 nm-thick Mn$_3$Ga/Co$_{0.6}$Fe$_{0.4}$ film for different activation temperatures between 100 K and 350 K.

Figure 6. Temperature dependence of exchange bias determined for the Mn$_3$Ga/Co$_{0.6}$Fe$_{0.4}$ films with the Mn$_3$Ga thickness of 6, 10 and 20 nm.

Figure 7. (a) Plan-view TEM image for grain size analysis for 6 nm thick Mn$_3$Ga thin film sample. (b) Grain size distribution for 6 nm thick Mn$_3$Ga thin film sample.
be seen, all $M$-$H$ curves are shifted horizontally, which demonstrates the presence of exchange-bias in the system. The 6 nm-thick Mn$_3$Ga film shows the largest exchange bias of 430 Oe at 120 K. The saturation magnetization of the Mn$_3$Ga/Co$_{0.6}$Fe$_{0.4}$ sample, $1300 \pm 100$ emu cm$^{-3}$, arises from the 3.3 nm-thick ferromagnetic Co$_{0.6}$Fe$_{0.4}$ layer, since the individual Mn$_3$Ga layer shows no magnetization between 100 and 300 K. Hence the observed magnetization can be attributed to that of the Co$_{0.6}$Fe$_{0.4}$ layer, which is very close to the bulk value for Co$_{0.6}$Fe$_{0.4}$ (1450 emu cm$^{-3}$) [21]. For the 10 and 20 nm-thick films, the exchange bias is measured to be 299 and 270 Oe, respectively. By increasing the Mn$_3$Ga thickness, the exchange bias is found to decrease. Such effect can be due to a combination of factors: on the one hand, at small thicknesses, one may expect a larger epitaxial strain, which may lead to an increased magnetocrystalline anisotropy in the Mn$_3$Ga; at larger thicknesses, strain relaxation sets in, leading to rougher interfaces and a possible reduction in the anisotropy, hence to a smaller exchange bias. For a fixed Mn$_3$Ga thickness (6 nm), we find that, as the thickness of CoFe increases from 3.3 nm to 9 nm, the exchange bias decreases significantly from 430 Oe to 130 Oe, as seen in figure 4(b). This is in agreement with other studies, where a thin ferromagnetic layer attached to an AF layer with good crystalline ordering maximises the interfacial exchange coupling [22].

In order to determine the blocking temperature ($T_B$), i.e. the point at which the exchange bias vanishes, the activation temperature must be considered. The determination of $T_B$ is usually carried out by raising the activation temperature until the loop shift becomes zero. A single grain has a unique $T_B$ in polycrystalline systems. Figure 5 illustrates $T_B$ measured using the York protocol [17]. In order to ensure that no magnetic history affects the measurements, the sample temperature is set to $T_{SET} = 500$ K (lower than $T_N$) for 90 min under an external field of 20 kOe, then cooled to $T_{NA} = 100$ K. The external field is then reversed to $-20$ kOe, and the samples are then thermally activated at a temperature between 100 K and 350 K for 30 min. The magnetisation curves are then taken at 100 K. The result of a sequence of such measurements for the 6 nm-thick Mn$_3$Ga sample is shown in figure 5. The data clearly shows the evolution of the loop shift from a negative magnetic field shift of $-420$ Oe to $+500$ Oe by increasing the activation temperature from 100 K to 350 K. The loop shift is caused by the reorientation of the magnetisation of the Mn$_3$Ga/CoFe with the individual Mn$_3$Ga grains due to the thermal activation in a negative field. By increasing the activation temperature, the magnetisation in smaller grains can be reversed [19]. Therefore, the temperature where the exchange bias becomes zero represents the equilibrium state between the total volume of the grains aligned along the initial magnetic field and that of the grains reversed by the thermal energy induced by the increase in temperature. This is the definition of the median blocking temperature of an antiferromagnetic material. For the 6 nm-thick Mn$_3$Ga sample, $\langle T_B \rangle$ is estimated to be 225 K.

Figure 6 shows that the exchange bias varied monotonically as a function of the thermally activating temperature. Interestingly, the blocking temperature is found to decrease when the Mn$_3$Ga thickness is increased, from 235 K for the 10 nm-thick film to 175 K for the 20 nm-thick film. This is attributed to the larger epitaxial strain to the Pt buffer layer and a smoother interface at smaller Mn$_3$Ga thicknesses. These results indicate that the quality of the Mn$_3$Ga/Co$_{0.6}$Fe$_{0.4}$ interface controls the exchange. For the 6 nm-thick Mn$_3$Ga, we find a reduction in the blocking temperature, which we attribute to a reduction in the total magnetic anisotropy of the Mn$_3$Ga due to the reduced thickness [18]. Nevertheless, the 6 nm-thick Mn$_3$Ga induces the largest exchange bias of 430 Oe at 120 K.

It is worth noting that there is a large difference between the Néel temperature and blocking temperature of Mn$_3$Ga, which might be related to the weak anisotropy and a small grain size. To ascertain the role of the grain size, we carried out transmission electron microscopy (TEM) grain size analyses (JEOL JEM-2011 TEM). More than 500 individual grain
particle was measured which follows a lognormal distribution shown in figure 7. The mean grain size was obtained to be 13.2 nm with 0.4 standard deviation.

For the case when the sample is thermally stable at the temperature of measurement and has been fully set, the magneto-crystalline anisotropy can estimated using the expression:

\[ K_{AF} \left( \langle T_B \rangle \right) = \frac{\ln \left( 1800 f_0 \right) kT_B}{\langle V \rangle} \]  

(1)

where \( K_{AF} \) indicates the magneto-crystalline anisotropy, \( T_B \) is the median blocking temperature, \( f_0 \) is an attempt frequency generally taken to be \( 10^9 \) s\(^{-1} \) and \( V \) is the median grain volume. In our report the anisotropy value was calculated to be \( 9 \times 10^4 \) J m\(^{-3} \). For comparing, the value for IrMn is more than 2 orders of magnitude higher. This leads to a lower blocking temperature for the Mn\(_3\)Ga sample.

In addition, x-ray photoemission electron microscopy (XPEEM) measurements were carried out at the SIM beamline at the Paul Scherrer Institut (PSI). In this technique, fully polarised x-ray light illuminates homogeneously the sample, and high resolution images of the local photomitted electron intensity of the sample, proportional to the x-ray absorption, are recorded. Using circularly polarised light and the x-ray magnetic circular dichroic (XMCD) effect and the elemental sensitivity of x-rays, separate magnetic contrast images of different layers can be obtained sequentially on the very same region of the sample. Magnetic contrast images of a Mn\(_3\)Ga/CoFe sample were obtained at the Co and Mn L\(_3\) edges at 150K to probe the CoFe and Mn\(_3\)Ga layers simultaneously. As shown in figure 8, we find a strong magnetic contrast in the CoFe layer, as expected for such a ferromagnetic material. The different black and white regions correspond to areas
with opposite magnetisation, showing the presence of a multi-domain state. When probing the Mn L3 edge, we find the presence of a clear magnetic contrast which correlates exactly to that of the CoFe layer, showing the presence of a spin-polarised Mn3Ga layer at the interface.

A schematic diagram of the interface with spin structures are shown in figure 9. This indicates possible formation of an uncompensated Mn3Ga spin layer in the vicinity of the interface against the CoFe layer in these films since they exhibit a small saturation magnetisation. This also agrees with the large exchange-bias effects observed in this system.

By taking a sequence of images as a function of photon energy, we can obtain the local XAS spectra in a single magnetic domain. Figures 10(a) and (b) shows the XAS spectra taken at room temperature at the Mn and Co edge with right (C+) and left (C−) circular polarisations for the region marked with a blue circle shown in figure 10(c). The subtraction of these two signals provide XMCD spectra shown in figures 10(c) and (d). For the Co spectrum, one can clearly see the domain structure using right-circularly-polarised light at L3 and L2 edges as shown in figure 10(e) for the L3 edge. For the Mn spectrum, the domain structure and corresponding XMCD spectra can also be observed. In the Mn3Ga (6 nm)/CoFe (2 nm)/Al (2 nm) orbital and spin magnetic moments of Mn and Co can therefore be estimated from XMCD spectra using the sum rules [23]. The orbital and spin moments of Co are estimated to be (0.299 ± 0.005) µB and (1.268 ± 0.005) µB, respectively. The total moment of Co is calculated to be (1.57 ± 0.01) µB which agrees with the theoretical value of 1.60 µB [24]. The orbital and spin moments of Mn are also estimated to be (0.270 ± 0.005) µB and (0.320 ± 0.005) µB respectively. The total moment of Mn is calculated to be (0.59 ± 0.01) µB which is within the literature magnetic moment of Mn (0.5–2.8) µB [1]. These estimated values represent those in the vicinity of Mn3Ga/CoFe interface, confirming that the quality of these layers is not affected by their neighbouring layers.

Conclusion

Polycrystalline Mn3Ga films grown by high target utilisation sputtering system are confirmed to crystallise in the DO19 antiferromagnetic hexagonal structure. By coupling to a CoFe layer, we observe the presence of large exchange-bias fields, of up to 430 Oe at 120 K for a 3.3 nm ferromagnetic CoFe layer deposited on top of a 6 nm Mn3Ga layer. The blocking temperature for 6 nm-thick Mn3Ga is found to be 225 K. The blocking temperature of Mn3Ga decreases as the thickness of Mn3Ga layer increases. The value of the exchange bias and the blocking temperature can be further increased by substituting some of the Mn and/or Ga atoms with the other elements as reported by Nayak et al. [20], which warrants the possibility of Mn3Ga being used in future antiferromagnetic spintronic devices.

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