Growth, structural, and magnetic characterization of epitaxial Co_2MnSi films deposited on MgO and Cr seed layers

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We report detailed structural characterization and magneto-optical Kerr magnetometry measurements at room temperature in epitaxial Co_2MnSi thin films grown on MgO(001) and Cr(001) buffered MgO single crystals prepared by sputtering. While $\text{Co}_2\text{MnSi}/\text{Cr}/\text{MgO}(001)$ films display the expected cubic anisotropy, the magnetization curves obtained for $\text{Co}_2\text{MnSi}/\text{MgO}(001)$ samples exhibit a superimposed in-plane uniaxial magnetic anisotropy. The evolution of magnetization with film thickness points to a relevant interfacial Co_2MnSi -buffer layer (Cr or MgO) contribution which competes with magnetic properties of bulk Co_2MnSi , resulting in a drastic change in the magnetism of the whole sample. The origin of this interfacial magnetic anisotropy is discussed and correlated with our structural studies. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4789801>]

I. INTRODUCTION

Spintronics is an area in magnetism whose aim is to combine the charges and the spin of electrons to develop innovative devices in which the electronic transport is spin dependent. The development of such devices necessitates the optimization of ferro- and ferrimagnetic materials exhibiting peculiar magnetic properties (high polarization, high saturation magnetization, low coercive field...). In this framework, full-Heusler alloys have received great attention due to their high Curie temperature ($T_C \sim 1000$ K) and because several of them have been predicted to be half-metals and exhibit a low Gilbert damping coefficient, which made them suitable as electrodes in magnetic tunnel junctions (MTJs) and for RF applications at room temperature. Full-Heusler alloys can be described by the formula X_2YZ , where X and Y are 3d transition metal atoms (i.e., Co, Mn, Ni) with a lower atomic number for Y than for X, and Z is a main group *sp*-atom (Al, Si, Ge...). These alloys crystallize in the L_{21} structure, which can be thought of as a simple cubic lattice for X atoms with the Y and Z atoms arranged at alternate body centered positions.

Among the full-Heusler alloys, Co_2MnSi (CMS) has attracted interest because theoretically calculations have predicted its marked half-metallic nature with a relatively large energy band-gap of 0.81 eV for its minority-spin band.¹ The half-metallic behavior has been confirmed experimentally by magnetotransport measurements in $\text{Co}_{75}\text{Fe}_{25}/\text{AlO}_x/\text{CMS}$ (Ref. 2) and $\text{CMS}/\text{AlO}_x/\text{CMS}$ MTJs,³ where tunneling magnetoresistance (*TMR*) ratios of 159% and 570% were obtained at low temperatures, respectively. Those values correspond to a spin polarization (*P*) of $P = 0.89$ according to Julliere's formula, which is the largest value to date reported for a Heusler compound, and is much larger than those of

conventional bulk ferromagnetic materials such as CoFe ($P \approx 0.5$).

The substitution of the AlO_x amorphous barrier by a MgO(001) textured or single crystal MgO(001) barrier plays an important role in the magnetotransport properties of MTJs. MgO(001)-oriented barriers originate electron band symmetry filtering effect and lead to *TMR* enhancement.⁴ The epitaxial growth of CMS films on MgO has been reported elsewhere.⁵ The CMS layer follows an epitaxial relation relative to the MgO buffer layer of $(001)[110]_{\text{CMS}} \parallel (001)[100]_{\text{MgO}}$ (45° in-plane rotated) despite the relatively large mismatch (*f*), defined as $f = [a_s - a_0]/a_0$ (a_s and a_0 being the underlayer and the film plane spacings, respectively), of 5.1%. Similarly, CMS can be epitaxially grown on Cr buffer deposited on MgO with the epitaxial relationship of $(001)[110]_{\text{CMS}} \parallel (001)[110]_{\text{Cr}} \parallel (001)[100]_{\text{MgO}}$ and a misfit between the CMS and Cr cubic structures of $f = 1.8\%$. Recently, Ishikawa *et al.*⁶ fabricated fully epitaxial CMS/MgO/CMS MTJs obtaining high *TMR* ratios of 179% at room temperature (RT) and 683% at 4.2 K, and demonstrated the feasibility of utilizing single-crystalline CMS films with a combination of a single-crystalline barrier as ferromagnetic electrodes in spintronic devices. However, further optimization should be done in order to avoid large decrease of *TMR* with increasing temperature. Both the presence of interface states⁷ and the local atomic disorder⁸ weaken the spin polarization of CMS and reduce the *TMR* ratio.

Deposition conditions play a crucial role in the microstructure of thin films. Use of single crystalline substrates⁹ and seed layers¹⁰ has extensively demonstrated the feasibility of improving crystal quality. In this article, we will present a detailed structural study of CMS thin film grown on two different buffer layers with large lattice mismatch: MgO(001) and Cr(001) deposited on MgO(001) substrates.

The microstructure of the films will be correlated with magnetic properties at room temperature.

II. EXPERIMENT

CMS films were grown on MgO(001) single crystals in a Plassys MPU 600 S ultrahigh vacuum (UHV) chamber with base pressure better than $2 \cdot 10^{-8}$ Torr. Substrates were heated up to 750 °C to degas for 2 h and covered with a thin layer of MgO (~ 20 nm) grown by RF sputtering in order to obtain carbon free surface of MgO with low roughness.¹¹ CMS was deposited at 600 °C by non-reactive RF magnetron sputtering of two facing targets and subsequently annealed *in situ* up to 800 °C for 1 h. Finally, a $t_{\text{MgO}} \approx 10$ nm thick MgO capping layer was deposited to prevent CMS films from oxidation. The thickness of CMS layer was varied in the range $t_{\text{CMS}} = 6\text{--}100$ nm. A second set of samples with Cr buffer grown at 400 °C with nominal thickness $t_{\text{Cr}} = 17$ nm was prepared prior to CMS deposition following the same procedure. The chamber is also equipped with a reflection high-energy electron diffraction (RHEED) system, which allows for *in situ* monitoring of film quality. Both MgO(10 nm)/CMS(t_{CMS})/MgO(001) and MgO(10 nm)/CMS(t_{CMS})/Cr(17 nm)/MgO(001) films have been characterized by X-ray diffraction (XRD) ($\theta/2\theta$, ω scans), X-ray reflectivity (XRR), high resolution transmission electron microscopy (HRTEM), and longitudinal and transversal¹² magneto-optical Kerr effect (L-MOKE and T-MOKE, respectively) magnetometry.

Cross section transmission electron microscopy (TEM) specimens were prepared by mechanical thinning and in a commercial precision ion polishing system. HRTEM images were obtained in a FEI Titan³ operated at 300 kV, equipped with spherical aberration corrector (CEOS). In addition, XRD and XRR were measured to characterize the macroscopic structure of the films. A Seifert XRD 3000 TT diffractometer with Cu $K\alpha_1$ ($\lambda_{K\alpha_1} = 1.54$ Å) was used for XRD measurements, while rocking curves and XRR data were collected using a PANalytical X'Pert PRO MRD diffractometer with Co $K\alpha_1$ ($\lambda_{K\alpha_1} = 1.79$ Å) equipped with a Ge(002) monochromator and a X'Celerator detector.

Finally, the magnetization measurements at room temperature were carried out in two different home-made MOKE magnetometers with the magnetic field (H) applied in the plane of the films at ONERA (L-MOKE geometry), Toulouse, and at IFIMUP-University of Porto (T-MOKE geometry).

III. RESULTS

Thickness (t) and roughness (rms) of MgO, Cr, and CMS layers were determined from the numerical fits of the XRR data (see Fig. 1). Obtained Cr buffer thickness is $t_{\text{Cr}} \approx 17$ nm, while MgO capping layer is ~ 10 nm thick. A nominal MgO buffer thickness of $t_{\text{MgO}} \approx 20$ nm is considered due to the fact that there is no difference between indexes of refraction for the MgO buffer layer and the MgO substrate, and taking into account that the deposition time for the MgO buffer layer is double that of subsequently deposited MgO capping layer. In order to compare the experimental results

FIG. 1. X-ray reflectivity measurements (black spots) from (a) a 38.14 CMS film deposited on MgO, and (b) a 38.21 nm CMS film deposited on Cr buffered MgO substrate (denoted by S). Both samples were capped with MgO. The red lines correspond to the numerical fits. The results of the fitting are shown in the figure, and the values of the best-fit parameters for all studied samples are summarized in Table I.

for both sets of series, MgO and Cr buffered samples with close t_{CMS} values (relative difference, Δ , between t_{CMS} values on MgO films and counterparts deposited on Cr is less than 10%) have been prepared. t_{CMS} and Δ values are listed in Table I.

Symmetrical $\theta/2\theta$ scans (Fig. 2) in the 2θ range of 20°–70° show a strong reflection from the MgO substrate at $2\theta = 42.9^\circ$. Additional peak at $2\theta \approx 64.5^\circ$ was found for CMS buffered samples, corresponding to the 002 Cr reflection. The appearance of the 002 and 004 reflections from the CMS layer indicates that CMS exhibits (001) preferred orientation both on the MgO and Cr seed layers. This confirms the expected (001)[110]_{CMS}|| (001)[100]_{MgO} and (001)[110]_{CMS}|| (001)[110]_{Cr}|| (001)[100]_{MgO} epitaxial relationships, respectively.

In situ RHEED experiments are a preliminary indication of the epitaxial quality of the studied samples. In Fig. 3, we

TABLE I. Nominal CMS layer thickness (t_{CMS}) and full width at half maximum (FWHM) of the rocking curves for MgO and Cr buffered samples. The shift field (H_S) is given by the weighted sum of the fields where magnetic subloops are symmetric around in MgO/CMS/MgO(001) bilayers. Absolute value of the difference between coercive fields for a field applied along the two easy axes (δH_C) in MgO/CMS/Cr/MgO(001) trilayers, and relative difference between t_{CMS} values in MgO buffered films and counterparts deposited on Cr (Δ).

Buffer MgO			Buffer Cr			
t_{CMS} (nm)	FWHM (°)	H_S (Oe)	t_{CMS} (nm)	FWHM (°)	δH_C (Oe)	Δ (%)
7.07	1.612	5.0	6.37	1.272	4.5	9.9
15.17	1.610	3.2	14.80	1.186	3.4	2.4
38.14	1.237	1.7	38.21	0.970	~ 0	0.2
66.76	0.878	0.5	69.50	0.818	~ 0	4.1
92.72	0.735	~ 0	96.30	0.805	~ 0	3.9

FIG. 2. θ - 2θ scans of (a) the bilayer MgO(10.57 nm)/CMS(38.14 nm) and (b) the trilayer MgO(11.24 nm)/CMS(38.21 nm)/Cr(17.46 nm), both grown on MgO(001) substrates. The not-indexed peaks arise from spectral contamination in diffraction patterns (see Ref. 34).

show RHEED patterns measured along the $\langle 100 \rangle$ and $\langle 110 \rangle$ azimuths at subsequent deposition stages. The RHEED image for the MgO showing characteristic narrow and intense streaks as well as Kikuchi lines indicates high quality starting surface. Cr layer also exhibit sharp streaks, confirming the expected cube-on-cube relationship with MgO with smooth surface. Attending to the RHEED images, CMS growth does not differ significantly for MgO and Cr buffered samples. The former case exhibits the presence of spotty diffraction stripes (see Fig. 3(a)) while the latest shows chevron features (Fig. 3(b)). These results notice the faceted structure of the CMS surface on both MgO and Cr buffered samples. Afterwards a MgO capping layer was deposited in order to protect the CMS film. The epitaxial growth of MgO on half-metal^{5,6,13,14} and metal^{15,16} thin films requires smooth underlayer with low roughness. In case of our non-perfectly CMS flat surfaces, a non-epitaxial MgO layer is observed with the appearance of rings (polycrystalline) and diffuse background (amorphous) in our RHEED patterns. It should be noted that MgO epitaxial growth on CMS films has been reported only in samples prepared by electron beam deposition method under high vacuum conditions,^{5,6,13,14,17} where no additional oxygen gas is needed¹⁸ contrary to our sputtering setup. Plasma oxidation can induce Mn/Si segregation and residual disorder¹⁹ at the upper CMS interface.

FIG. 3. Collected RHEED images during subsequent growth steps along the directions [100] and [110] of the MgO single crystal substrate of the (a) MgO(10.57 nm)/CMS(38.14 nm) bilayer and (b) MgO(11.24 nm)/CMS(38.21 nm)/Cr(17.46 nm) trilayer.

FIG. 4. (a) Low magnification and (b) high-resolution cross sectional TEM images of the bilayer MgO(10.64 nm)/CMS(7.07 nm)/MgO(001). Two lattice dislocations are circled in the latter. (c) Indexed electron diffraction measurement with subscripts denoting crystallographic planes of MgO and CMS.

The high-resolution TEM cross-section micrographs yield further insights into the microstructure of the films. In Fig. 4(b), we show the HRTEM image of the thinnest sample ($t_{\text{CMS}} = 7.07$ nm) deposited on MgO(001) substrate. As can be seen in Fig. 4(c), electron diffraction measurements confirm the epitaxial growth of the films with the expected out-of-plane (001) orientation with a 45° rotation of the in-plane axes: $(001)[100]_{\text{MgO}} \parallel (001)[110]_{\text{CMS}}$. The diffraction image indicates the expected cubic structures of both the MgO buffer layer and the CMS film. The diffraction reflections for CMS are clearly seen at intermediate positions with respect to the MgO spots due to larger lattice parameter in CMS, e.g., the interplanar spacings (d) for planes with Miller indices $\{002\}$ are $d_{[002]\text{-CMS}} = 2.835$ Å and $d_{[002]\text{-MgO}} = 2.107$ Å for CMS and MgO, respectively. The observed 111 CMS spots evidence the presence of the L_{21} structure^{5,20} in the MgO/CMS/MgO films. Additionally, preliminary results of high angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) (data not shown here) indicate the L_{21} structure and that the partially disordered B2 structure also exists. CMS/MgO(001) bottom interface is smooth and flat. The accommodation of the large-lattice mismatch between MgO [100] and CMS [220] ($f = 5.1\%$) results

FIG. 5. (a) Low magnification cross-sectional TEM image of the trilayer MgO(11.64 nm)/CMS(6.37 nm)/Cr(16.31 nm)/MgO(001). (b) and (c) are magnifications of (a), with MgO/Cr and Cr/CMS interface regions, respectively. (d) Selected area electron diffraction pattern, revealing an orientation relationship $(001)[100]_{\text{MgO}} \parallel (001)[110]_{\text{Cr}} \parallel (001)[110]_{\text{CMS}}$.

in a strained layer relaxed by misfit dislocations formed at the CMS-MgO interface (see Fig. 4(b)).

On the other hand, HRTEM cross-section image of the thinnest Cr buffered sample with $t_{\text{CMS}} = 6.37$ nm is shown in Fig. 5. HRTEM micrographs and indexed electron diffraction measurements (Fig. 5(d)) confirm the expected 45° rotation of the Cr(001) and MgO(001) surfaces, and a cube-on-cube relationship $(001)[110]_{\text{Cr}} \parallel (001)[110]_{\text{CMS}}$. The nearly 2:1 relation between the lattice parameters ($a_{\text{Cr}} = 2.89$ Å, $a_{\text{CMS}} = 5.67$ Å) leads to the misfit of $f = 1.8\%$ between $[110]_{\text{Cr}}$ and $[220]_{\text{CMS}}$. In this case, however, weak 002 and 111 CMS reflections are not distinguishable on the diffraction pattern because of the low CMS content and possibly reduced by the presence of an intermixed Cr-CMS layer (not observable in the TEM images). We should note that these 002 and 111 CMS reflections are clearly seen in thicker CMS films (not shown). A set of misfit dislocations along the MgO-Cr interface are observable in the micrographs as a consequence of the stress due to mismatched lattice spacing between MgO $[100]$ and Cr $[110]$, $f = 3.2\%$. The Cr-CMS interface is barely distinguishable due to low Z contrast.

Once the structural quality was confirmed, another topic of intensive study is the determination of magnetic anisotropies in these CMS films. In Fig. 6, the longitudinal MOKE measurements at room temperature for thinnest and thickest CMS films grown on MgO (Figs. 6(a) and 6(b)) and Cr seed layer (Figs. 6(c) and 6(d)) are shown. The magnetization features vary with t_{CMS} thickness and with the presence or not of the buffer as well. $M(H)$ curves for films directly

FIG. 6. Longitudinal MOKE signal at room temperature for films with nominal structure (a) MgO(10.64 nm)/CMS(7.07 nm)/MgO(001); (b) MgO(11.14 nm)/CMS(92.72 nm)/MgO(001); (c) MgO(11.64 nm)/CMS(6.37 nm)/Cr(16.31 nm)/MgO(001); and (d) MgO(10.80 nm)/CMS(96.30 nm)/Cr(16.20 nm)/MgO(001). The magnetic field H is applied along the $[110]$ (red line), $[1-10]$ (black line; left panel), and $[100]$ (black line; right panel) directions. The shift field where subloop is symmetric around at positive fields (H_{S1}) is remarked within the figure.

deposited on MgO reveal an in-plane magnetic anisotropy with the easy axis oriented along $[110]_{\text{MgO}}$, i.e., along the $[100]$ axis of the Heusler alloy. The expected four fold anisotropy due to epitaxial growth is found to be superimposed to an additional uniaxial magnetic anisotropy with the easy-axis along the MgO $[1-10]$ in-plane direction. Indeed, when the external magnetic field is applied along the $[110]$ axis, the longitudinal magnetization switches in two defined Barkhausen jumps separated by an intermediate plateau (see Fig. 6(a): left column, red line), while when the magnetic field is applied along $[1-10]$ (Fig. 6(a): left column, black line), the hysteresis loop exhibits a “classical” square shape with only two stable magnetic configuration. This clearly demonstrates the different magnetization reversal processes for two crystallographically equivalent $\langle 110 \rangle$ axis. Similar plateaus at low fields were reported earlier measuring hysteresis loops on epitaxial CMS⁸ and Co₂MnGe (Ref. 21) Heusler films grown on GaAs(001) substrates, and even before on epitaxial Fe thin films prepared onto MgO(001),^{22,23} GaAs(001),²⁴ Ag(001),²⁵ and W(001)²⁶ surfaces. We observed that these plateaus coincide to an absolute maximum T-MOKE signal in the hysteresis loops^{22,25} (see Fig. 7). Both L- and T-MOKE loops indicate that when the external field is applied along $[110]_{\text{MgO}}$, the magnetization is first rotating perpendicular to the applied field before completely switching in the direction parallel to it. The origin of this superimposed uniaxial anisotropy is unclear. Various explanations have been proposed considering surface anisotropy contribution induced by the strain of the system, dangling bonds at the interface, or even related to technological deposition conditions (see Ref. 23 and references therein). In the latter case, the oblique-deposition geometry has been pointed out as a source of additional magnetic anisotropies in magnetic thin films provided by an oriented growth²⁷ or by lattice distortion.²⁸ We should mention that both MgO/CMS//MgO(001) and MgO/CMS/Cr//MgO(001) set of samples were deposited at normal incidence. A possible explanation for the additional magnetic anisotropy we observed could come from the presence of terraces on the surface of MgO single crystals.²⁹ Recently, it has indeed been demonstrated

FIG. 7. Longitudinal (black line; top panel) and transversal (red line; bottom panel) magneto-optical signal for different azimuths measured in a MgO(8.18 nm)/CMS(15.17 nm)/MgO(001) bilayer.

that nanometer-scale steps introduce an in-plane magnetic easy direction perpendicular to the step edge in $\text{Fe}_3\text{O}_4//\text{MgO}(001)^{29}$ half metal. Similar results were reported earlier in $\text{Fe}/\text{W}(001)^{26}$, $\text{Fe}/\text{Ag}(001)^{30}$, and $\text{Fe}/\text{Au}(001)^{30}$ thin films. McGuigan *et al.*²⁹ suggest that the origin of this magnetic anisotropy can arise from the preferential alignment of structural defects with step edges. In a similar manner, the work by Leeb *et al.*³⁰ pointed out that the magnitude of the magnetic anisotropy is determined by the large lattice vertical mismatch which gives rise to strong strain in the vicinity of monoatomic steps. Such a contribution should vanish with increasing film thickness because the local strain relaxation is confined to the interface. The shift field is defined as $H_S = (H_{S1} - H_{S2})/2$, where H_{S1} and H_{S2} denote the fields where sub-loops separated by the plateaus are symmetric around at positive and negative fields along $[110]$ direction (see Figure 6(a), left column, for clarity), respectively. For the $\text{MgO}/\text{CMS}/\text{Cr}/\text{MgO}(001)$ film with $t_{\text{CMS}} = 92.72$ nm, sub-loops are barely distinguishable with our MOKE setup and magnetic curves with H applied along $[110]$ and $[1-10]$ axes superimpose into a square loop with coercive field (H_C) value of ~ 6 Oe, as can be seen in Figure 6(b) (left). On the contrary, Table I shows that H_S value increases as t_{CMS} decreases for samples with $t_{\text{CMS}} \leq 66.76$ nm. Although we only have four values for the latter range, the t_{CMS} dependence of H_S can be approximated by the expression $H_S = A + B/t_{\text{CMS}}$ with $A = 0.5 \pm 0.5$ Oe and $B = 34 \pm 6$ Oe·nm. A linear dependence of H_S with $1/t_{\text{CMS}}$ was found previously in $\text{Co}_2\text{MnSi}/\text{GaAs}(001)$ thin films.⁸ Unfortunately, the obtained values for the numerical fit determined by Wang *et al.*⁸ are not given and we are not able to compare our results quantitatively with theirs.

Different features were obtained in $\text{MgO}/\text{CMS}(t_{\text{CMS}})/\text{Cr}/\text{MgO}(001)$ films (Figs. 6(c) and 6(d)). For these samples, the longitudinal L-MOKE signal presents typical square hysteresis loops with 100% remanence for magnetic fields applied along both $[110]_{\text{MgO}}$ and $[1-10]_{\text{MgO}}$ axes, while the magnetization reversal along the $[100]_{\text{MgO}}$ axis is a typical hard magnetic axis loop showing a continuous rotation of the magnetization, followed by an abrupt switch. Nevertheless, the two coercive fields in the two easy $\langle 110 \rangle_{\text{MgO}}$ directions are not equal in the sample with $t_{\text{CMS}} = 6.37$ nm (see Figure 6(c)). This difference is given by $\delta H_C = |H_{C1} - H_{C2}|$, being H_{C1} and H_{C2} the coercive fields along $[110]_{\text{MgO}}$ and $[1-10]_{\text{MgO}}$ axes. We observed that δH_C diminishes as t_{CMS} increases, reaching 0 Oe for $t_{\text{CMS}} \geq 38.21$ nm, as can be seen from values taken of Table I. L-MOKE curves along the principal axes superimpose for samples with larger t_{CMS} values, being $H_{C1} \approx H_{C2} \approx 11$ Oe.

Despite the established presumption of the influence of the miscut on the growth of thin films deposited on MgO single crystals, direct evidences for induced strain are lacking. We then performed local strain measurements on the HRTEM images of the $\text{MgO}/\text{CMS}/\text{MgO}$ and $\text{MgO}/\text{CMS}/\text{Cr}/\text{MgO}$ stacking using the geometrical phase analysis (GPA) method.³¹ Basically, the GPA method consists on studying any change in the lattice parameters analyzing the changes in contrast periodicity within a HRTEM image and selecting a given area as a “reference.” All contrast

periodicities variations compared to the reference area are associated to lattice deformations, allowing drawing a quantitative 2D map of the relative strain. In our case, we use the substrate lattice contrast as reference area to map the relative deformation tensor ε_{xx} in the strained CMS layer. Figs. 8(b) and 9(b) correspond to the maps of the ε_{xx} in-plane strain relative to the MgO substrate in the $\text{MgO}/\text{CMS}/\text{MgO}(001)$ bilayer and the $\text{MgO}/\text{CMS}/\text{Cr}/\text{MgO}(001)$ trilayer with thinnest t_{CMS} values, respectively. The measured value of the former is $\varepsilon_{xx} = -5.1\% (\pm 0.2\%)$, which corresponds to the misfit between the $\text{MgO}(200)$ ($d_{[200]\text{-MgO}} = 2.107$ Å) planes and the $\text{CMS}(220)$ ones ($d_{[220]\text{-CMS}} = 2.005$ Å) ($f = 5.2\%$). A set of periodic misfit dislocations clearly appears at the CMS-MgO buffer layer interface (see Fig. 8(b)). The mean misfit dislocation periodicity is $l = 3.9 \pm 0.2$ nm for the thinnest CMS film ($t_{\text{CMS}} = 7.07$ nm). This value is in perfect agreement with the theoretical distance between the misfit dislocations, which is given by $l = (d_{[220]\text{-CMS}}/f) = 3.9$ nm.

Performing the same measurement of the CMS deformation relative to the MgO lattice in the trilayer $\text{MgO}/\text{CMS}/\text{Cr}/\text{MgO}(001)$, we get a slightly lower strain value of $\varepsilon_{xx} = -4.5\% (\pm 0.2\%)$ (see Fig. 9(b)). The presence of a Cr spacer also seriously affects the CMS layer morphology. In the case of the $\text{MgO}(11.64\text{ nm})/\text{CMS}(6.37\text{ nm})/\text{Cr}(16.31\text{ nm})/\text{MgO}(001)$ trilayer, the presence of non-periodical dislocations is not reduced at CMS-Cr and Cr-MgO interfaces, but also at critical CMS and Cr layer thicknesses, as it can be seen in Fig. 9(b). The obtained results can be explained in terms of different epitaxial growth modes of the CMS layer on top of MgO and Cr buffered substrates. For low lattice mismatch ($0.2\% < |f| < 5\%$), the thin film growth is often “pseudomorphic” at the very first

FIG. 8. (a) High-resolution cross sectional TEM image of the bilayer $\text{MgO}(10.64\text{ nm})/\text{CMS}(7.07\text{ nm})/\text{MgO}(001)$ and (b) ε_{xx} strain relative to MgO. The inset in (a) shows the fast fourier transform (FFT) of the area of interest.

FIG. 9. (a) High-resolution TEM image of the trilayer MgO(11.64 nm)/CMS(6.37 nm)/Cr(16.31 nm)/MgO(001) and (b) the corresponding ϵ_{xx} strain map relative to MgO. The inset in (a) shows the FFT of the area of interest.

stages,³² and the epilayer could accommodate the misfit through biaxial in-plane and out-of-plane strains. For thicker deposited layer, the elastic energy has to be released by the insertion of misfit dislocations, which accommodate the stress and relax the epilayer. Due to further rearrangements of the surface, gliding dislocations can migrate to deeper layers³³ and end up at the epilayer-underlayer interface (Fig. 9(b)). On the contrary, for larger misfit ($|f| > 5\%$),³¹ the epitaxial stress is directly relaxed through misfit dislocations confined at the epilayer-underlayer interface (Fig. 8(b)), which could underlie the observed magnetic uniaxial anisotropy in CMS films deposited on MgO single crystals. The presence of buried dislocations can affect the subsequent growth of the CMS layer by perturbing the activation energy for diffusion steps in neighboring atoms,³³ giving rise to mounds on the surface of the film, in agreement with results observed in RHEED images in both MgO/CMS//MgO(001) bilayers and MgO/CMS/Cr//MgO(001) trilayers (see Fig. 3).

As it has been mentioned before, formed dislocations have influence on the succeeding steps of thin film growth despite being completely overgrown. As the film thickness increases, the average vertical and lateral lattice spacings

continuously approach the undisturbed bulk values.³³ Rocking curve measurements around the (004) reflection of CMS support this theory. The full width at half maximum (FWHM) of the rocking curves decreases with increasing t_{CMS} for both MgO and Cr buffered samples (see Table I). The ω -scan for a single crystal shows an intrinsic FWHM value, but strain and density of defects cause the broadening of the rocking curve.

IV. CONCLUSIONS

Epitaxial Co₂MnSi films were deposited on MgO(001) and Cr buffered MgO(001) substrates by sputtering. RHEED patterns suggest that the Co₂MnSi growth follows a 3D nucleation on MgO and Cr seed layers. The films exhibit a cubic magnetic anisotropy with an additional superimposed uniaxial magnetic anisotropy in the case of samples deposited on MgO(001). In such a case, the magnetization reversal switches in one Barkhausen jump when the field is applied along [1–10] axis. In contrast to this result, longitudinal Kerr loop along [110] axis exhibits two plateaus at low fields, which correspond with two maxima of the magnetic signal in the in-plane transversal direction. Differently, magnetic measurements performed on Co₂MnSi films deposited on Cr buffer layer show square loops when the field is applied along the easy axis. Nevertheless, coercive field values in [110] and [1–10] axes differ for ultrathin MgO/Co₂MnSi/Cr//MgO films. Both effects uniaxial magnetic anisotropy in MgO/Co₂MnSi/MgO(001) films, and different values of the coercive field in MgO/Co₂MnSi/Cr//MgO(001) films—vanish with increasing nominal Co₂MnSi thickness. This suggests an interfacial origin for the observed phenomena. Contrary to the established explanation which relates the presence of step-terrace topography on single-crystal substrates with magnetic anisotropies in deposited ferromagnetic thin films, the local strain investigations in our films suggest that the presence of misfit dislocations underlie the observed magnetic anisotropy. Determining the origin for this anisotropy is a key factor to optimize promising Co₂MnSi-based nanodevices.

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¹S. Picozzi, A. Continenza, and A. J. Freeman, *Phys. Rev. B* **66**, 094421 (2002).

²Y. Sakuraba, J. Nakata, M. Oogane, H. Kubota, Y. Ando, A. Sakuma, and T. Miyazaki, *Jpn. J. Appl. Phys.* **44**, L1100 (2005).

³Y. Sakuraba, M. Hattori, M. Oogane, Y. Ando, H. Kato, A. Sakuma, T. Miyazaki, and H. Kubota, *Appl. Phys. Lett.* **88**, 192508 (2006).

⁴S. Yuasa, *J. Phys. Soc. Jpn.* **77**, 31001 (2008).

⁵T. M. Nakatani, Y. K. Takahashi, T. Ishikawa, M. Yamamoto, and K. Ono, *J. Magn. Magn. Mater.* **322**, 357 (2010).

⁶T. Ishikawa, S. Hakamata, K. Matsuda, T. Uemura, and M. Yamamoto, *J. Appl. Phys.* **103**, 07A919 (2008).

⁷H. L. Yu and G. W. Yang, *Appl. Phys. Lett.* **98**, 011910 (2011).

⁸W. H. Wang, M. Przybylski, W. Kuch, L. I. Chelaru, J. Wang, Y. F. Lu, J. Barthel, H. L. Meyerheim, and J. Kirschner, *Phys. Rev. B* **71**, 144416 (2005).

- ⁹A. García-García, J. A. Pardo, P. Štrichovanec, C. Magén, A. Vovk, J. M. De Teresa, G. N. Kakazei, Y. G. Pogorelov, L. Morellón, P. A. Algarabel, and M. R. Ibarra, *Appl. Phys. Lett.* **98**, 122502 (2011).
- ¹⁰C. Magén, E. Snoeck, U. Lüders, and J. F. Bobo, *J. Appl. Phys.* **104**, 013913 (2008).
- ¹¹M. Rickart, B. F. P. Roos, T. Mewes, J. Jorzick, S. O. Demokritov, and B. Hillebrands, *Surf. Sci.* **495**, 68 (2001).
- ¹²J. M. Teixeira, R. Lusche, J. Ventura, R. Fermento, F. Carpinteiro, J. P. Araujo, J. B. Sousa, S. Cardoso, and P. P. Freitas, *Rev. Sci. Instrum.* **82**, 043902 (2011).
- ¹³T. Ishikawa, T. Marukame, H. Kijima, K.-I. Matsuda, T. Uemura, M. Arita, and M. Yamamoto, *Appl. Phys. Lett.* **89**, 192505 (2006).
- ¹⁴T. Ishikawa, N. Itabashi, T. Taira, K. Matsuda, T. Uemura, and M. Yamamoto, *Appl. Phys. Lett.* **95**, 232512 (2009).
- ¹⁵Y. Park, E. F. Fullerton, and S. D. J. Bader, *Vac. Sci. Technol. A* **13**, 301 (1995).
- ¹⁶S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, *Nature Mater.* **3**, 868 (2004).
- ¹⁷M. Yamamoto, T. Ishikawa, T. Taira, G. Li, K. Matsuda, and T. Uemura, *J. Phys.: Condens. Matter* **22**, 164212 (2010).
- ¹⁸S. Yadavalli, M. H. Yang, and C. P. Flynn, *Phys. Rev. B* **41**, 7961 (1990).
- ¹⁹J. Schmalhorst, S. Kämmerer, M. Sacher, G. Reiss, A. Hütten, and A. Scholl, *Phys. Rev. B* **70**, 024426 (2004).
- ²⁰G. Li, T. Taira, K. Matsuda, M. Arita, T. Uemura, and M. Yamamoto, *Appl. Phys. Lett.* **98**, 262505 (2011).
- ²¹T. Ambrose, J. J. Krebs, and G. A. Prinz, *Appl. Phys. Lett.* **76**, 3280 (2000).
- ²²J. L. Costa-Krämer, J. L. Menéndez, A. Cebollada, F. Briones, D. García, and A. Hernando, *J. Magn. Magn. Mater.* **210**, 341 (2000).
- ²³C. Martínez Boubeta, J. L. Costa-Krämer, and A. Cebollada, *J. Phys.: Condens. Matter* **15**, R1123 (2003).
- ²⁴O. Thomas, Q. Shen, P. Schieffer, N. Tournier, and B. Lépine, *Phys. Rev. Lett.* **90**, 017205 (2003).
- ²⁵R. P. Cowburn, S. J. Gray, J. Ferré, J. A. C. Bland, and J. Miltat, *J. Appl. Phys.* **78**, 7210 (1995).
- ²⁶J. Chen and J. L. Erskine, *Phys. Rev. Lett.* **68**, 1212 (1992).
- ²⁷J. H. Wolfe, R. K. Kawakami, W. L. Ling, Z. Q. Quiu, R. Arias, and D. L. Mills, *J. Magn. Magn. Mater.* **232**, 36 (2001).
- ²⁸O. Durand, J. R. Childress, P. Galtier, R. Bisaro, and A. Schuhl, *J. Magn. Magn. Mater.* **145**, 111 (1995).
- ²⁹L. McGuigan, R. C. Barklie, R. G. S. Sofin, S. K. Arora, and I. V. Shvets, *Phys. Rev. B* **77**, 174424 (2008).
- ³⁰T. Leeb, M. Brockmann, F. Bensch, S. Miethaner, and G. Bayreuther, *J. Appl. Phys.* **85**, 4964 (1999).
- ³¹M. J. Hÿtch, E. Snoeck, and R. Kilaas, *Ultramicroscopy* **74**, 131 (1998).
- ³²M. Ohring, *Materials Science of Thin Films*, 2nd ed. (Academic Press, 2001).
- ³³M. Walther, M. Biehl, and W. Kinzel, *Phys. Status Solidi C* **4**, 3210 (2007).
- ³⁴See: <http://prism.mit.edu/xray/Basics%20of%20X-Ray%20Powder%20Diffraction.pdf> for detailed information about spectral contamination in X-ray diffraction measurements.