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Strains in Fe/Cr trilayers and multilayers epitaxied on MgO and STO\MgO

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Abstract:

High Cr ferritic martensitic steels are good candidates as structural or cladding materials for the next generation of nuclear power plants (generation IV and fusion reactors). The addition of Cr prevents corrosion and the FeCr ferritic form is only weakly sensitive to the swelling phenomenon and minimizes atomic segregation under high-energy neutron irradiation. Diffusion processes and magnetic properties need to be fully understood to optimize these structural materials for high temperature and high flux use. Here, Fe/Cr trilayers and multilayers are prepared as model systems designed to furnish simple data comparable with calculation results of diffusion or magnetic properties. Their structure (epitaxy, residual strains and dislocations) is characterized in detail. Superlattices with high crystalline quality are prepared, with Fe and Cr in coherent epitaxy. In-plane residual strain in Fe is +0.40(5)% on MgO substrates and varies from 1.70(9)% to 0.47(2)% depending on the total thickness of the trilayers on STO\MgO substrates. Thanks to these strains, the contrast between Fe and Cr is enhanced, which opens the way to future kinetics studies using XRD in this system, which is far more efficient (non-destructive and rapid) than HRSTEM-EELS or APT for obtaining data at different times and annealing temperatures.

1) Introduction

High Cr ferritic martensitic steels are good candidates as structural or cladding materials for the next generation of nuclear power plants (generation IV and fusion reactors) [1-3]. The addition of Cr prevents corrosion and the FeCr ferritic form is only weakly sensitive to the swelling phenomenon and minimizes atomic segregation under high-energy neutron irradiation. Many teams have studied the electronic structure of these alloys [4-8] in order to understand the origin of this unusual behaviour that has been often attributed to chemical and magnetic short-range order effects [9-10]. The knowledge of the Fe magnetic behaviour in the Fe-Cr system [11,12] is difficult to elucidate correctly through calculations due to the itinerant nature of iron magnetism. Experiments providing some basic values are needed to compare with theoretical results [13-16]. The proximity effects at the interface between an Fe layer and a Cr layer have also been calculated using sophisticated models [17], but are not easily obtained experimentally because ideal interfaces have to be constructed. In this paper, we describe the preparation of Fe/Cr trilayers and multilayers with flat interfaces and limited defects and strains, in order to make some kinetic and magnetic measurements in a second step. The multilayers are model systems designed to furnish simple data comparable with calculation results.

Fe/Cr superlattices generated scientific interest at the end of the last century because of the magnetoresistance behaviour discovered by Peter Grünberg [18] and Albert Fert [19]. Since this discovery, many researchers have tried to improve the magnetoresistance effect for potential applications in magnetic sensors or magnetic data storage systems [20]. Among other studies, the dependences of magnetoresistance on epitaxial quality [21] and on level/direction of stress [22] were considered. To our knowledge, the stress effects were

only studied in Co/Cu systems, focusing on the magnetoresistance and not on the magnetization properties. Moreover, to date, no study has been published describing epitaxial Fe/Cr(100) on MgO buffer layers on SrTiO₃(100) substrates, despite this system having a smaller lattice mismatch with Cr and Fe than the widely used substrate MgO(100) [19,21,23,24]. STO is also an easier substrate to deal with for the preparation of transmission electron microscopy (TEM) cross-sections and atom probe tomography (APT) tips.

This paper reports the preparation of Fe/Cr trilayers and multilayers on MgO and STO substrates and the detailed study of their structural properties. We show that high-quality superlattices can be prepared and that, thanks to the strains, the contrast between Fe and Cr is enhanced, which opens the way to kinetics studies using XRD in this system.

2) Experimental techniques

Fe-Cr tri-layers and multilayers were deposited by molecular beam epitaxy in a high-vacuum chamber with a base pressure of about 10^{-10} mbar equipped with two electron guns. High-purity targets: 6N Fe and 6N Cr, and 5N Au targets, were used for deposition, at slow speed (0.02 nm/s) for all samples. Reflection High-Energy Electron Diffraction (RHEED) was used at some steps of the growth to get information on the surface quality. The thicknesses of Fe and Cr layers were monitored using a quartz balance placed in the flux, whereas MgO and Au thicknesses were monitored via deposition time (to spare the quartz balance as the atomic weight of Au is high). The surface roughness was also observed using atomic force microscopy (AFM) at some growth steps on test samples.

The surface roughness at different stages of the growth was studied using a Bruker Dimension Icon atomic force microscope (AFM) in tapping mode.

Both TEM and APT samples were prepared using focused ion beam (FIB) on an FEI Helios dual beam Nanolab 650 equipped with a Ga liquid metal ion source. A layer of platinum was deposited on the surface of samples before any ion observation to preserve the surface layers. TEM studies were made on cross-sectional samples. For electron energy loss spectroscopy (EELS), as a very thin sample is necessary to avoid multiple scattering, so some additional Ar-ion milling using a PIPS II (precision ion polishing system from Gatan) was used with low-angle and low-energy.

APT analyses were performed using a CAMECA LEAP 4000XHR at a set-point temperature of 50K in laser-pulsing mode at a wavelength of 382 nm, 200 kHz pulse repetition rate, and 40 pJ pulse energy. For 3D atom reconstruction, visualization and data treatments, the IVAS software by CAMECA was employed. Reconstructions of the volumes were performed using spatial distribution maps on low index crystallographic poles [25].

For conventional TEM experiments, we used a JEOL 2100F microscope, operated at 200kV, equipped with an LaB₆ electron source (point to point resolution: 0.2 nm). STEM imaging in bright and dark field mode was carried out on an FEI TITAN3 transmission electron microscope equipped with a probe-corrected condenser and operating at 300 kV. The resolution in STEM is 70pm.

High resolution STEM in HAADF mode [26,27] and EELS analyses [28] were performed in a Nion Ultrastem 200 cold FEG microscope operating at 100kV. The Cs-corrected probe attains 75pm in HAADF imaging and around 0.5nm in EELS elemental maps in our conditions. The electron spectrometer has a Merlin MEDIPX3 direct electron detector and the operating conditions gave an energy resolution of around 1eV. EELS spectrum-images were recorded using a beam current of around 50pA and a pixel dwell time of 10ms. We used the L_{23} edges for Cr (575 eV) and Fe (710 eV) to quantify the concentration of each element. The quantification was performed using Hyperspy [29] and is based on the use of internal standards generated via independent components analysis, rather than using calculated inelastic scattering cross sections [30]. This enables a more confident measurement of the absolute concentrations. Oxygen (532 eV) is detected in very small quantities on the multilayers, consistent with some surface oxidation of the FIB lamellae.

Texture, mosaicity, epitaxy quality, and residual stress were determined using a four-axis RIGAKU x-ray diffractometer operated at 45kV and 200mA equipped with a rotating Cu anode and a Ge(220) crystal monochromator to select $Cu-K\alpha_1$. The beam divergence was 0.03° . $\theta-2\theta$ scans at wide angles were performed to determine the preferential orientation of the film. Rocking curves (ω -scans) give information on the mosaicity (a FWHM below 1° is satisfactory for metals). The epitaxial quality of the layers was determined by measuring pole figures. The average in-plane and out-of-plane residual strains of Fe and Cr were measured using the $\sin^2(\psi)$ method on (200), (110), (220), (130), (310), (222), and (211) reflections. The in-plane stress and the swelling were deduced.

3) Preparation

(100)-oriented SrTiO₃ (STO) and MgO were chosen as substrates because of the small lattice mismatch between these materials and Cr/Fe, favouring a coherent epitaxy. The iron lattice parameter ($a_{\text{Fe}}^{\text{bulk}} = 0.28684 = 0.4057/\sqrt{2}$ nm) with a 45° rotation has a -3.8% mismatch on MgO [31] ($a_{\text{MgO}} = 0.42112$ nm) and a +3.7% mismatch on STO ($a_{\text{STO}} = 0.3905$ nm). The chromium lattice parameter ($a_{\text{Cr}}^{\text{bulk}} = 0.2884 = 0.4079/\sqrt{2}$ nm) is very close (+0.66% mismatch with Fe), and the epitaxy conditions are equivalent. MgO(100) and STO(100) substrates were mounted on the same metallic plate to have the same conditions of deposition. Before the growth, the substrates were heated to 723K for 15 hours and to 873K for 20 minutes. A 20 nm-thick MgO buffer layer was deposited at 873K on both types of substrate to improve the surface quality, as verified by RHEED patterns (not shown). Figure 1 presents some AFM images on degassed substrates and on the MgO buffer layer. The surface is indeed improved by the buffer layer deposition, which smooths the steps on STO substrates and buries the islands present after annealing on MgO substrates (carbon aggregates on scratches or on steps) [32].

A series of trilayers was studied. The first layer was iron deposited at room temperature (RT) and annealed for 2h at 773K to improve flatness and crystallinity [33]. After cooling down, the Cr\Fe bilayer (where A\B means B deposited on A) and the covering gold layer were deposited at RT. Figure A1 shows the RHEED images of the first Fe layer deposition before and after annealing. The smoothness is indeed improved by the heat treatment. The chromium deposition and the second layer of iron are slightly rougher but the surface quality is still good. We call **TXFY** the trilayer XO\MgO_z\Fe_Y\Cr_Y\Fe_Y\Au_y, with Y = 2, 5, 10, 15, 20, 25, 30 nm (XO = MgO or STO; in the names X = M for MgO and S for STO). For

comparison, the buffer layers **BXFY** = $XO \setminus MgO_z \setminus Fe_Y \setminus Au_y$ with $Y = 5, 10, 20$ nm were prepared. For APT, EELS-STEM and future interdiffusion studies, multilayers **MXF10** = $XO \setminus MgO_z \setminus Fe_Y \setminus (Cr_Y \setminus Fe_Y)_5 \setminus Au_y$ with $Y = 10$ nm were also prepared on both substrates. For all layers, z is around 20nm and y around 5nm.

4) Architecture and chemistry of the layers

As an example, **Figure 2a** shows the nanometer scale arrangement of the Fe-Cr layers in MSF10. The thickness of both Fe and Cr layers is about 9.5 nm with a fluctuation of 0.3 nm. The layers appear very flat. Multilayers present diffraction contrast inside the layers. We attributed this contrast to dislocations that cross all the layers. The misfit dislocations generated at the $STO \setminus MgO$ interface and at the $MgO \setminus Fe$ interface are studied in **§7**.

APT was performed to determine the chemical composition within Fe-Cr multilayers. **Figures 2b and 2c** show APT results obtained on MSF10. The layers are homogeneous in composition, suggesting that defects observed by TEM do not affect their chemical composition. The atomic composition of each layer has been measured in the middle of layers (averaged over 2.5 nm) to prevent any APT artefact at interfaces due to the difference of evaporation field between Fe and Cr which may produce some local compression or tension of the ion trajectories **[34]**. This artefact also induces an apparent thickness difference that is not real. Results show a purity level higher than 99.7% for all the Fe and Cr layers. The oxygen distribution was also analysed due to its high reactivity with Cr. Oxygen levels remain extremely low throughout the sample. The oxygen is mainly localized at interfaces with a local maximum composition at 2%. The location of every detected oxygen

atom is shown on the top of Fig. 2c. The Fe layers are almost free of O and the Fe\Cr interfaces (note the growth direction from the right towards the left) are the most polluted by O, as if Fe layers rejected the O atoms and/or the Cr target trapped the oxygen atoms of the vacuum chamber during Fe deposition. Concerning the C and N distributions, levels remain extremely low and never exceed 0.1% for the whole volume. Figure 2d shows the chemical maps obtained by EELS which confirm (with lower precision) the APT findings (> 98% purity for the layers).

Reflectometry measurements give information on the thickness and roughness for the layers. The Fe and Cr layers have similar electronic densities and cannot be separated using reflectometry with a Cu anode, thus only the total thicknesses and the roughness above the last 3d-metal layer are determined. The total Fe,Cr thickness e_R is 5% smaller than the nominal value (in agreement with STEM observations). In TSYF, we found a roughness of 0.2-0.3 nm at the STO surface and of 0.05-0.25 nm at the MgO buffer surface. In TMFY, the roughness at the MgO buffer surface is 0.2-0.95 nm. At the Fe\Au interface, the roughness is 0.2-0.5 nm in TMFY, and 0.1-0.2 in TSYF. Some examples of curves and fits are shown in Figure 3.

5) Crystalline quality

Figure 4 shows the global high-angle $\theta-2\theta$ scans of BXYF buffer layers, TXYF tri-layers for the different Y, and MXF10 multilayers. Besides the (h00) reflections of the substrates, of the MgO buffer layer in BSYF, TSYF and MSF10, and of the Au coverage, the (200) reflections of Fe and Cr layers are present, indicating a good (100) texture of Fe and Cr films along the

[100] direction of the substrate. Due to the used wavelength, (400) reflections of Fe and Cr are not accessible. There is a single (200) peak in this range for the buffer layers and the thinnest trilayers (for thin layers, the width of the peaks is large, preventing any fine structure from appearing). For $Y > 10$ nm, there are two well-separated peaks. In the multilayers, satellites due to the alternating Cr and Fe layers are present. Note the Laue fringes around the 200 Au Bragg peak at 44.4° in $\theta-2\theta$ scans of TXFY samples with a period corresponding to the Au thickness (5nm or 9 nm depending on the sample). Their presence indicates a very good flatness of the layer. They are negligible around the other peaks due to the small density contrast at the other interfaces.

The rocking curve (Fig. A3a) full widths are smaller than 0.5° for $Y \geq 10$ nm, indicating a very good crystalline quality (mosaicity smaller than 0.5°) and a large in-plane coherence length (larger than $L_{//} = 2\pi/\Delta Q_{//} \approx 20$ nm with $Q_{//} = \Delta\omega Q$ and $Q = 4\pi \sin(\theta)/\lambda$). The thicker the layers, the narrower the rocking curve width; hence the mosaicity and/or lateral coherence length improve during the growth.

To determine the epitaxial relationship, pole figures were measured for the {110} plane family of Fe and Cr. Because the substrates have a four-fold symmetry, quarter pole figures were sufficient. For all layers, we observe a single intense and narrow (110) spot around the angles $\phi=45^\circ$ (from the 020 peak of the substrate) and $\chi=45^\circ$ (Figure A2) corresponding to the epitaxy relationship¹ $M(100)[011] // XO(100)[010]$ for $M=Fe$ or Cr and $XO=MgO$ or STO . The same epitaxial relationship was obtained by Fullerton *et al.* [24,35,36] for Fe/Cr multilayers grown on $MgO(100)$ by magnetron sputtering, but with a poorer crystalline quality. All the pole figures were measured in the same conditions so that we can compare

¹ This relationship indicates that the in-plane [011] direction of the (100) plane (film surface) is aligned on the [010] direction of the (100) substrate surface plane.

their intensities. On Fig. A2c, the intensity of the (110) peak increases more rapidly than the thickness, indicating that the global epitaxy improves with Y .

High resolution images were acquired from different cross-sectional samples with the electron-beam direction along $[001]_{\text{STO}}$ and $[001]_{\text{MgO}}$. The (100) planes of Fe, MgO and STO are observed, indicating a good epitaxy of Fe and MgO on STO (Fig. 5a, Figs. 9 in §7). Electron diffraction patterns (Fig. 5b) show the expected spots for Fe, MgO and STO in both $[010]_{\text{STO}} = [010]_{\text{MgO}} = [011]_{\text{Fe}}$ (in-plane) and $[100]_{\text{STO}} = [100]_{\text{MgO}} = [100]_{\text{Fe}}$ (out-of-plane) directions, confirming the good epitaxy of the MgO and Fe layers on the substrate as well as the epitaxy relationships.

6) Strains

6.1 Out-of-plane strains

In a first approximation, the coherence length of the (200) periodicity can be estimated from the widths of the peaks fitted as Gaussians. In the buffer layers, the widths of the peaks correspond to a coherence length similar to the nominal thickness. In the thickest trilayers (Figures A3b), the two main peaks are separated enough to provide individual coherences. The coherence length corresponds to the whole thickness in the two thinnest trilayers, whereas in TMF15 and TMF30 trilayers, the three layers diffract incoherently ($L_{\perp} = e_R$). In the other trilayers, the coherence length is larger than Y , showing that the interferences between the different diffracted beams have to be taken into account to simulate the curves. The trilayers have thus to be considered as multilayers.

Figure 6 shows zooms on the (200) Fe,Cr reflections. Due to the interference effects, the scans are not a superposition of the individual peaks of Fe and Cr layers. They can be simulated with a finite number of parameters: N_1 , N_2 , N_3 : the numbers of atomic layers in individual layers (N_3 Fe planes on N_2 Cr planes on N_1 Fe planes), $a_{\text{Fe}\perp}$ and $a_{\text{Cr}\perp}$, the lattice parameters in the growth direction in the Fe and Cr layers. The set of parameters has been adjusted by a least square method on the experimental curves. In the simulations, we assume absolutely abrupt and planar interfaces (interplanar distance at the interface: $(a_{\text{Fe}} + a_{\text{Cr}})/2$). The numbers of atoms per unit surface were assumed equal for Fe and Cr (indeed, we show in §6.2 that the in-plane lattice parameters are equal). The interference scattering simulations take into account the anomalous scattering coefficients from [37]. A convolution was applied with a 0.23° -wide Gaussian adjusted on Fe-buffer layers to take into account the global (due to the diffractometer and the sample) experimental resolution.

A first fit with the constraint $N_3=N_1$ was performed and gave quite satisfactory results. The Fe thickness (Figure A3-c,d) is found to be less than a third of the thickness deduced from reflectometry (this effect is the largest in TMF25 and TSF25), showing the presence of an initial Fe layer with many defects, not perfect enough to be in coherence with the rest of the trilayer (in agreement with the STEM observations of §6). In a second step, the constraint was released allowing $N_1 < N_3$, a lack of coherence in the first few Fe planes near the MgO\Fe interface diminishing N_1 . The χ^2 of the fit decreased by about 20% for TXFY with $10 \leq Y \leq 25$. The thin red lines in Figure 6 show the result of this fit for those trilayers and of the simpler fit for TXF5 and TXF30 trilayers.

The Cr thickness (Fig. A3c,d) is close to the value deduced from reflectometry ($e_R/3$), except for TXF25, indicating that in these trilayers the crystallographic coherence length is smaller

than the layer thickness. The thickness of the thicker Fe layer is slightly smaller than $e_R/3$ and that of the thinner is 20% smaller. The lattice parameters deduced from the fits (Fig. A3e,f) are not sensitive to the constraint $N_3=N_1$. The Cr lattice parameter is very close to the bulk value. The Fe is lattice parameter smaller, in agreement with the out-of-plane strain in compression expected for an in-plane strain in tension. The strain is more or less constant on the MgO substrate and larger for the thinnest layers on the STO substrate.

The $\theta-2\theta$ scans on the MMF10 and MSF10 multilayers (Fig. 6c,d) present many satellites (the orders -4, 4 and 6 are clearly visible in MSF10 where the signal/noise ratio was the best). This confirms that the multilayers are superlattices. There is a small shift of the peaks in MSF10 compared to MMF10, indicating a difference in average out-of-plane strain in the Fe/Cr multilayer of 0.19(1)%. The distances of the peaks are the same because the periods are equal in agreement with the STEM observations. The simulations have been optimized within the same frame as in the trilayers: N_1 Fe layers covered by 5 ($N_2\text{Cr}+N_3\text{Fe}$) bilayers. The optimum is obtained for $N_1=46$, $N_2=70$, $N_3=64$ in both samples, $a_{\text{Fe}}=0.28452\text{nm}$, $a_{\text{Cr}}=0.28857\text{nm}$ in MMF10 and $a_{\text{Fe}}=0.28397\text{nm}$, $a_{\text{Cr}}=0.28847\text{nm}$ in MSF10. These lattice parameters are close to those obtained in TMF10 and TSF10 respectively (Fig. A3e,f). The fit is satisfactory considering the simple description of the multilayers (integer numbers of atomic layers in the layers, abrupt interfaces). The bilayer thickness is 19.2 nm (9.1 nm of Fe and 10.1 nm of Cr) in both samples. Note that the difference in strain is responsible for the contrast between Fe and Cr layers in the superlattices. As a matter of fact, figure A4 shows that with diffraction contrast alone, the satellites would have very small intensities, preventing any precise study.

6.2 In-plane strains

For all samples, intensity maps in reciprocal space were measured around several non-specular peaks (\mathbf{Q} makes an angle α with \mathbf{u}_1 the unit vector along the normal to the sample) to estimate the in-plane strains (Fig. A5a,d). Non-specular peaks can be measured as $\theta/2\theta$ scans, varying either $\omega = \alpha + \theta/2$ (rotation of the normal of the sample inside the diffraction plane; limited by the condition $\omega > 0$, i.e. reflection mode; the explored plane in reciprocal space contains \mathbf{Q} and \mathbf{u}_1) or ψ (rotation of the sample normal out of the diffraction plane; no limitation but poorer resolution; the explored plane in reciprocal space contains $\mathbf{Q} \times \mathbf{u}_1$ and \mathbf{Q}). The interference satellites are clearly separated from the main peak only in the first method. For multilayers, satellites are observed for 200, 211 and 220 (Fig. A5a,b). The satellite peaks are not resolved for 310, 222 and 130 peaks measured when varying ψ (Fig. A5c,d). For trilayers, the small oscillations observed in $\theta/2\theta$ (Fig. 6) are not resolved on non-specular peaks. A single Fe/Cr/Fe peak is observed for all trilayers for 310, 222 and 130 and for thin trilayers (TMF2, TMF5, and TSF2) for all hkl . Two peaks (one for Cr and one for Fe) are resolved in the thick trilayers for 200, 211 and 220. The 2θ values of the peaks are deduced from least square fits with Gaussians in $2D$.

Fig. 7 shows $\sin^2(\psi)$ plots in BXFY, TXY and MXF10 (X=M and S). The dashed-dotted horizontal lines show the Fe and Cr lattice parameters in the bulk. When the Cr and Fe peaks can be separated, two slopes are observed. As the low- α peak is observed in the Fe buffer layers and as $a_{\text{Cr_bulk}} > a_{\text{Fe_bulk}}$, we attribute the higher a values to the Cr layer and the lower to the Fe layer. The points are fitted with a straight line to deduce the in-plane ($\sin^2(\psi)=1$) and out-of-plane ($\sin^2(\psi)=0$) lattice parameters. It is noticeable that both in-plane lattice parameters are equal (within the error bar) for the lines attributed to Fe and Cr, meaning

that Fe and Cr layers grow coherently. Fe is stressed in tension in-plane whereas Cr is almost relaxed in-plane in thick samples on both substrates.

In the BXFY buffer layers, we observe a positive slope with an in-plane tension strain as expected on an MgO buffer layer with an in-plane lattice parameter 0.5-1% larger than in bulk. A compressive out-of-plane strain is induced through Poisson's effect. Assuming no swelling (the swelling expected for a 0.3% O concentration in Fe layers is 0.02% with the calculated size factor² of O in *bcc* Fe: $SF=0.176$ [38]; we will neglect it as it is smaller than the experimental error bars), the deduced Poisson's coefficients ν are 0.378(96), 0.350(81), 0.397(31) for BMFY and 0.309(23), 0.388(28) and 0.401(25) for BSFY ($Y=5, 10, 20$ respectively). Except in BSF5, these values are in good agreement with the values calculated by D. Sander [39] from elastic constants: $\nu=0.37$. A Poisson's coefficient ν equal to 0.37 means $\varepsilon_{//} / \varepsilon_{\perp} = - (1-\nu)/2\nu = -0.85$ and that the swelling can be read on the curves around $\sin^2(\psi)=0.55$, which is indeed the value where all lines cross the bulk Fe line, except BSF5. We can thus deduce a small positive swelling in BSF5.

The in-plane strains $\varepsilon_{//}$ of Fe in all layers are shown in Figure 8. It is constant on MgO substrate and decreases with Y on STO.

The non-specular peaks of the MgO buffer layers are very close to those of Au and the in-plane lattice parameter could not be measured directly. We deduced it from the out-of-plane lattice parameter with a Poisson's coefficient of $\nu(\text{MgO}) = 0.178(9)$ [40] ($\varepsilon_{//} / \varepsilon_{\perp} = - (1-\nu)/2\nu = -2.31(14)$). The MgO layer is found to be in compression in plane with a strain around -0.4%. The in-plane strains in MgO buffer layers are plotted in Figure 8 for comparison.

² The size factor SF is the limit of relative volume increase when adding 1% of impurity atoms for large atom numbers in the crystal: $\Delta V/V=3 \Delta \ell/\ell = SF \cdot c$ with c in percent.

For trilayers, assuming a Poisson's ratio of 0.37, the Fe lines are compatible with a zero swelling (except in MSF2 and MSF5). The Cr lines have very small slopes, so the Cr layers are almost relaxed. In the multilayers, the average peak and first superlattice satellites have been reported. The lines converge towards a unique in-plane lattice parameter. The corresponding in-plane strains are reported in [Figure 8](#). The strains of all layers on the MgO substrate are around the same value with an average of $\langle \varepsilon_{//}(\text{Fe})_{\text{MgO}} \rangle = 0.43\% \pm 0.09\%$ in Fe (not considering MMF2). On the STO substrate, there is a variation of the strain in Fe, increasing at small values of Y . Note that the points are on a single $\langle \varepsilon_{//}(\text{Fe})_{\text{MgO}} \rangle + A/Y$ curve (blue curve on Fig. 8) for the buffer layers, the trilayers and the multilayers on STO, which shows that the important parameter to describe this supplementary strain, compared to the layers prepared on MgO substrates, is the thickness of individual layers and not the total thickness. We deduce that the first Fe layer is different on STO.

7) Dislocations

At the STO\MgO interface, if the strain was totally relaxed, the misfit $m=7.8\%$ would give rise to one missing plane in MgO every 13 STO atomic planes ($1/m=12.75$), i. e. dislocations with separation $d=2.685$ nm. At the MgO\Fe interface, the misfit $m=-3.7\%$ corresponds to one half plane added in Fe every 27 MgO atomic layers ($1/m=27.1$), or $d=5.52$ nm. These epitaxy dislocations give rise to extended defects, threading dislocations, corresponding to the ends of these half-planes that are not infinite. These defects are responsible for the contrast observed in [Figure 2a](#). In the samples deposited on STO, some dislocations created at the STO\MgO interface may be annihilated at the MgO\Fe interface. When the samples are annealed, the presence of extended defects will have a strong influence on the diffusion and

so it is important to characterize the epitaxy dislocations in detail. Moreover, XRD detected some residual strain in MgO buffer and in Fe layers. It is interesting to compare them to the strain deduced from the dislocation numbers at the two interfaces.

We therefore acquired high resolution STEM-HAADF images from MSF10. [Figure 9a](#) shows the interface between the STO substrate and the MgO buffer layer. Seven images were Fourier transformed, filtered applying a mask and inverse Fourier transformed to image the atomic planes and evidence the dislocations. Using a mask selecting the (020) peaks of STO and MgO ([Fig. 9c](#)), we reveal the epitaxy dislocations ([Fig. 9b](#)). They are marked by a 'T' (additional half plane below) or a '⊥' (additional half plane above). Some dislocations are in close pairs with opposite Burger's vectors; they are probably artefacts because they depend on the applied mask diameter. We marked those in yellow. The true epitaxial dislocations are marked in red. They are regularly distributed at the STO\MgO interface. The position of the interface is deduced from the disappearance of the STO periodicity in [Fig. 9a](#). Most dislocations are located at the interface and only a few artefacts are present. The true dislocations were counted and their numbers were confirmed by counting the planes between the red arrows in STO and MgO. The average distance between dislocations was measured in the 7 different images. The distribution is shown as narrow blue bars in [Figure 10a](#). The distances between the dislocations and the interface were measured and the corresponding distribution is shown as narrow blue bars in [Figure 10c](#). Many dislocations have migrated into the STO substrate.

The same analysis was performed at the MgO\Fe interface on 5 images. The location of the interface is deduced from the strong dark/bright contrast present in the image ([Figure 9d](#)). The misfit dislocations were imaged with a mask on the $(011)_{\text{Fe}} \approx (020)_{\text{MgO}}$ peaks (green in

Fig. 9g). The dislocations are not regularly distributed (Figure 9e) and there are many parasitic dislocations. Many epitaxy dislocations are located inside the Fe layer. The interface is thus not the preferred location for dislocations. This is in agreement with the observation that the first Fe layer is only partly in coherence with the rest of the trilayers or multilayers, as deduced from the X-ray diffraction simulations of $\theta/2\theta$ scans. Moreover, the distance between two dislocations is larger and fluctuates a lot (thick beige bars in Fig. 10a). The error bar on the deduced residual strain (Figure 10b) is so large that even its sign cannot be deduced from these images. The average distance of the dislocations from the MgO\Fe interface (thick beige bars in Fig. 10c) is around 1 nm. Note that the image displays only 5 nm of Fe. As the dislocations are dispersed in the Fe layer, this view field may not be sufficient to observe all the dislocations, which would explain the discrepancy between TEM and XRD results in terms of in-plane strains. We did not detect any dislocation at the Fe\Cr interface, which is expected because the in-plane lattice parameters of Fe and Cr are equal as deduced from asymmetrical x-ray diffraction.

In *bcc* metals, two types of dislocations are stable with Burger's vector of $\frac{1}{2}\langle 111 \rangle$ -type and $\langle 001 \rangle$ -type respectively [41]. Both Burger's vectors have the same $\frac{1}{2}[011]$ projection along the interface line in Fig. 9d. To distinguish between these two possibilities, we imaged the dislocations with the Burger's vector perpendicular to the interface with a mask on the $(200)_{\text{Fe,MgO}}$ peaks (magenta in Fig. 9g) to have an insight into the perpendicular component of the dislocation Burger's vectors. We observe on Fig. 9f that the misfit dislocations detected in Fig. 9e do not have such a component. We deduce that the misfit dislocations have a $b=\langle 001 \rangle$ -type Burger's vector within the Fe/MgO interface.

8) Discussion

The multilayers have an excellent crystalline quality on both substrates. In this paragraph, we will discuss the strain results from both X-ray diffraction and HRSTEM.

X-ray diffraction showed:

- identical in-plane lattice parameter for Fe and Cr,
- residual strain in Fe of +0.40(5)% on MgO substrate,
- residual strain in MgO of -0.40(9)% on STO substrate,
- dependence of strain in STO-substrate trilayers on Y: decreasing from 1.70(9)% at Y=2 nm down to 0.47(2)% at Y=30 nm;
- in-plane strain of 0.79(2)% in MSF10 and 0.59(2)% in MMF10.

HRSTEM results in the multilayers on STO\MgO can be summarized as:

- the dislocations are regularly distributed at the STO\MgO interface with an average separation of 2.8(8) nm and with an average distance of -0.2(6) nm from the interface,
- at the MgO\Fe interface, the dislocation average separation is 5.3(2.7) nm and the dislocations are distributed inside the Fe layer with an average distance of 1.0(8) nm from the interface.
- no dislocations were observed at the Fe\Cr or Cr\Fe interfaces.

8.1 Strains in MgO

In both MgO and Fe layers, we get a strain smaller than 1%, which is in agreement with the maximum values expected in metals [42,43] and oxides (dislocations are formed when the elastic energy stored in the strained layer is large enough to overcome the activation energy of formation of a new dislocation). Note that at the STO\MgO interface the dislocations are formed during the growth of MgO at 873K. The expansion coefficients of MgO and STO are not equal so that the strain at the end of the preparation was different from the strain measured at room temperature. De Ligny and Richet [44] showed that the thermal linear expansion in SrTiO₃ has a constant value $\alpha_L = 1.077(7) \cdot 10^{-5} \text{ K}^{-1}$ up to 1800K. Data reported by Leroy [45] for MgO can be interpolated as $\alpha_L = (6.73 \cdot 10^{-6} + 1.49 \cdot 10^{-8} T - .402 \cdot 10^{-12} T^2) \text{ K}^{-1}$ between 300 and 900K and, for Fe, data reported by [46] give $\alpha_L = (8.37 \cdot 10^{-6} + 7.25 \cdot 10^{-9} T) \text{ K}^{-1}$ between 300 and 1100K.

The expansion coefficient of MgO is larger than that of STO above 300K (Fig. A6a). For the MgO buffer layer, the compression strain thus decreases when decreasing the temperature after the growth (Fig. A6b). We can thus assume that the number of epitaxial dislocations does not vary during cooling. The residual strain of -0.37% at 300K corresponds to a strain of -0.54% at 874K. Dislocations were formed in the MgO buffer layer until the strain was around 0.5% as expected in oxides. Indeed, the residual strains deduced from x-ray diffraction and HRSTEM are equal for MgO.

The relaxation due to the dislocations observed through HRSTEM has been calculated considering that in MgO, an *fcc* system, the stable Burgers vector is of type $\frac{1}{2}\langle 011 \rangle$ [41].

Calling the interface plane (100) and the beam direction [010], the gliding plane is (100) and the edge contribution of the Burgers vector in the image plane is $\frac{1}{2}[001]$. Writing the coincidence between n planes of MgO and $(n+1)$ planes of STO between two dislocations separated by d , we get:

$$d = (n+1) \times a_{\text{STO}}/2 = n \times a_{\text{MgO}} \times (1+\varepsilon_{\text{MgO}}) / 2 \rightarrow n = a_{\text{STO}} / (a_{\text{MgO}} \times (1+\varepsilon_{\text{MgO}}) - a_{\text{STO}})$$

$$\rightarrow d = a_{\text{MgO}} \times (1+\varepsilon_{\text{MgO}}) \times a_{\text{STO}} / (a_{\text{MgO}} \times (1+\varepsilon_{\text{MgO}}) - a_{\text{STO}})/2$$

$$d = 2.685 \text{ nm for } \varepsilon_{\text{MgO}} = 0 \text{ and } d = 2.83(4) \text{ nm for } \varepsilon_{\text{MgO}} = -0.40(9)\% \text{ (from XRD).}$$

The dislocation separation distance observed with HRSTEM is in very good agreement with the value deduced from XRD results.

8.2 Strains in first layer of Fe

The same calculation is done for Fe, taking into account the 45° rotation of the Fe lattice with respect to the MgO lattice. In Fe, a *bcc* system, the stable Burgers vectors are of $\frac{1}{2}\langle 111 \rangle$ or $\langle 100 \rangle$ type, with a small energy difference [41]. If we call the interface plane (100) and the beam direction [0-11], the gliding plane is (100) and the edge contribution of the Burgers vector $\frac{1}{2}[011]$ in the image plane for the $\langle 100 \rangle$ -type dislocations. Equivalent equations can be written:

$$d = a_{\text{MgO}} \times (1+\varepsilon_{\text{MgO}}) \times a_{\text{Fe}} \times (1+\varepsilon_{\text{Fe}}) \times \text{sqrt}(2) / (a_{\text{MgO}} \times (1+\varepsilon_{\text{MgO}}) - a_{\text{Fe}} \times (1+\varepsilon_{\text{Fe}}) \times \text{sqrt}(2)) / 2$$

$$d = 5.36 \text{ nm for } \varepsilon_{\text{MgO}} = \varepsilon_{\text{Fe}} = 0 \text{ and } 7.79(32) \text{ nm for } \varepsilon_{\text{MgO}} \text{ and } \varepsilon_{\text{Fe}} \text{ measured by XRD.}$$

In the first Fe layer, the average separation of dislocations (5.3 ± 2.7 nm) is slightly smaller (Fig. 10a) than the value expected considering the average residual strain in the Fe layers (7.8(3) nm) in multilayers. It is nevertheless within the error bars.

Fe is more ductile than MgO. This explains why the dislocations are formed after some Fe planes are grown on MgO. As in a Stranski-Krastanov growth mode, the first Fe layers grow coherently. When the elastic energy gets too high, dislocations are formed and slide towards the interface. This effect is much more rapid in oxides like MgO, which is why the dislocations are exactly at the STO\MgO interface.

We did not observe any dislocation with Burger's vector perpendicular to the interface as observed in [47]. The deposition of a 20nm-thick MgO buffer layer to get rid of the surface pollution and steps appears thus to be a better method than that used by the authors of [47]: etching in 85% o-phosphoric acid and electron cyclotron resonance microwave plasma in the vacuum chamber. The high vacuum clean surface and the deposition of Fe at room temperature followed by an annealing, as we used in this work, indeed improves the interface and avoids the formation of such dislocations that do not contribute to the lattice misfit relaxation at the interface.

8.3 Y variation of strain

The additional strain in Fe deposited on STO\MgO varies as $1/Y$. Only the first Fe layer was annealed for 2h at 773K after deposition at room temperature. All other sub-layers were deposited at room temperature and not annealed. The thermal strain has no reason to vary

with Fe thickness. In addition, its sign would be negative as the expansion coefficient is smaller in STO than in Fe (Fig. A6).

A possible explanation of these observations would be that some strontium atoms segregate to the MgO surface during the MgO growth at 873K. The diffusion of Sr through 20nm of MgO is possible at 873K as deduced from the data of [48]. EELS spectra acquired in the most favourable conditions to observe Sr in MgO and Fe showed that the strontium concentration in the MgO layer and in the first Fe layer is smaller than the detection limit of about 1at.%.

During Fe growth at room temperature, if an Sr layer either floated at the Fe surface or stayed at the MgO\Fe interface, during the 2h 773K annealing subsequent to Fe growth, these Sr atoms would interdiffuse inside the Fe layer to attain a homogeneous distribution [49] leading to an Sr concentration varying as $1/Y$. The increase of in-plane lattice parameter would thus correspond to a swelling of the first Fe layer due to the presence of Sr. Taking into account the calculated size factor of Sr in *bcc* Fe: $SF=1.473$ [50] we can estimate the amount of Sr from the additional strain in TSY compared to TMFY. We find a third of a monolayer and an average concentration around 0.5%. This concentration is below the detection limit of EELS in our microscope. If Sr is present in the first Fe layer, it would behave as a buffer layer with a lattice parameter varying as $1/Y$ because a constant amount of Sr would be dissolved in a Y -thick film.

The other layers do not contain any Sr as they were grown at room temperature and were not annealed afterwards. They are slightly more strained in tension than if the swelling of the first Fe layer was not present. The swelling of the first Fe layer cannot be observed directly from the out-of-plane lattice parameter because only an average lattice parameter is

measured and because the other Fe layers have a negative perpendicular strain associated with this in-plane positive strain.

9) Conclusion

In this paper, we have shown that using MBE it is possible to grow perfectly coherent Cr/Fe superlattices. The in-plane lattice parameters are equal in the Fe and Cr layers.

The residual in-plane strains are smaller than 0.5% in all trilayers and multilayers deposited on an MgO substrate. On an STO substrate, the residual in-plane strains are larger but remain smaller than 1% in trilayers with individual layers thicker than 5nm, i.e. 35 monolayers. The additional strain has been attributed to a swelling of the first Fe layer attributed to a small contamination by Sr that was present at the MgO surface at the end of the MgO growth and had diffused inside the Fe first layer during its post-growth annealing.

In conclusion, these samples appear to be suitable for diffusion and magnetic measurements in multilayers with very few defects at the interfaces. The strain difference notably enhances the XRD contrast making it possible to study the interdiffusion kinetics in FeCr system on such superlattices. The swelling of the first Fe layer on STO could be used to prepare a buffer layer with finely accorded lattice parameter in future studies.

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Appendix (or Supplementary material)

Some additional and more technical figures are given here to complete the main text.

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Figure captions:

Figure 1: AFM images on the degassed substrate (a,c) and the MgO buffer layer (b,d) on the MgO (top) and STO (bottom) substrates.

Figure 2: MSF10: (a) BF-STEM image. (b) 3D reconstruction from APT around the 4 top Cr layers. (c) top: Maps of Fe,Cr and O from a $7*7*60 \text{ nm}^3$ box extracted close to the (100) pole (black rectangle in (b)). Bottom: the relative atomic composition of Fe, Cr and O along the growth direction (scale on the right for O), (d) HAADF and EELS maps on Ti, O, Fe, and Cr from EELS (Mg and Sr cannot be measured with the used incident energy).

Figure 3: Some examples of reflectometry measurements and fits with the densities ρ (in g/cm^3 : $\rho_{\text{STO}}=5.12$, $\rho_{\text{MgO}}=3.58$, $\rho_{\text{Fe}}=7.8$, $\rho_{\text{Au}}=19.3$) and the following thicknesses (d in nm) and roughnesses (σ in nm); in inset: zoom showing the oscillations due to the Fe/Cr thickness.

BSF5: $d_{\text{MgO}}=19.02$, $d_{\text{Fe}}=4.66$, $d_{\text{Au}}=9.75$, $\sigma_{\text{MgO}}=0.26$, $\sigma_{\text{Fe}}=0.19$, $\sigma_{\text{Au}}=0.22$; TSF30: $d_{\text{MgO}}=19.97$, $d_{\text{Fe/Cr}}=83.61$, $d_{\text{Au}}=4.76$, $\sigma_{\text{STO}}=0.21$, $\sigma_{\text{MgO}}=0.3$, $\sigma_{\text{Fe}}=0.24$, $\sigma_{\text{Au}}=0.21$.

Figure 4 : Global high-angle $\theta-2\theta$ scans of BXFY buffer layers, TXFY tri-layers and MXF10 multilayers for the X=S (STO substrate; left) and M (MgO substrate; right). The sample names are indicated in the margin (thicknesses Y from the bottom: 2, 5, 10, 15, 20, 25, and 30 nm for TXFY, 10 and 20 nm for BMFY, 5, 10, and 20 nm for BSFY; MXF10 are the topmost curves in blue). The curves are shifted for clarity.

Figure 5: HRTEM image and diffraction on BSF20.

Figure 6: XRD normalized intensity (linear scale) of the Fe/Cr 200 peaks ($\theta/2\theta$ scans). The experimental intensities are plotted as circles and the simulations as lines. (a,b) Measured and simulated in trilayers (a: TMFY and b: TSFY). (c,d): Measured and simulated intensities in MMF10 and MSF10 multilayers, amplified by a 1 and (10 or 100) factor. The names of the different satellites are given in (d).

Figure 7: $\sin^2(\psi)$ plots for Fe,Cr in (a) BXFY, (b) TMFY, (c) MXF10, and (d) TSFY.

Figure 8: Residual in-plane strains from $\sin^2(\psi)$ plots for Fe and MgO in BSFY, TSFY, and TMFY. The weighted averages of in-plane strains in Fe for TMFY and in MgO for TSFY are shown as horizontal bars. Blue line: $(0.43 + 2.85/Y)$: $1/Y$ fit of the departure of $\varepsilon_{//}(\text{Fe})$ on STO from the average $\varepsilon_{//}(\text{Fe})$ on MgO.

Figure 9: Examples of HRSTEM images: raw data (a,d) and data treated to evidence misfit dislocations (b,e) at the STO\MgO (a,b) and Fe\MgO (d,e) interfaces in the MSF10 multilayer. Raw images (a,d) were Fourier transformed (c,g), filtered applying a mask on the 020 (resp. 011) in-plane peak of MgO and STO (resp. Fe) (green on (c,g)), and inverse Fourier transformed to image the atomic planes and evidence the epitaxy dislocations (b,e). The blue lines show the approximate location of the interface. In red the dislocations due to interfacial strain relaxation and in yellow the pairs of opposite dislocations due to artefacts. The dislocations are counted in 5 (resp. 7) images (between the red arrows). We also imaged the component of dislocations with the Burger's vector perpendicular to the interface with a mask on the 200 peaks of Fe and MgO (magenta on (g)). We observe that there is no such component associated to the misfit dislocations. The latter have thus $b=1/2 [002]$ (projected on the image as $1/2[011]$), and not $b=1/2[111]$ (with the same projection).

Figure 10: (a) Distribution of individual dislocation distances at both interfaces (narrow dark blue bars: STO\MgO; large light beige bars: MgO\Fe) and the corresponding averages. The different lines show the expected values in the cases: MgO/STO interface (long lines): Black (left): $\varepsilon_{\text{MgO}} = 0$; Green (right): $\varepsilon_{\text{MgO}} = -0.4\%$. Fe/MgO interface (short lines): Blue (left): $\varepsilon_{\text{MgO}} = \varepsilon_{\text{Fe}} = 0$; Red (center): $\varepsilon_{\text{MgO}} = -0.4\%$, $\varepsilon_{\text{Fe}} = 0$; Magenta (right): $\varepsilon_{\text{MgO}} = -0.4\%$, $\varepsilon_{\text{Fe}} = 0.79\%$ (MSF10). (b) Average strains deduced from STEM in each sample (●: MgO/STO interface; ▼: Fe/MgO interface; the mean values and the standard deviations are indicated as hatched bars)

compared to the strains deduced from XRD on MgO in MSF10 (\square) and on Fe in MMF10 (violet star) and MSF10 (\triangle). (c) Distribution of distances of individual dislocations to the interface: in narrow blue bars at STO\MgO interface, in beige large bars at Fe\MgO interface.

Figure A1: RHEED images of the first-deposited Fe layer before (a) and after annealing (b), of the Cr layer (c) and the second Fe layer (d) deposited at 300K. The substrate is MgO in the left panel (azimuth [100] in (a,b) and [110] in (c,d)) and STO in the right panel (azimuth [100] in (a,b,d) and [110] in (c)).

Figure A2: Pole figures: to determine all the orientations of (hkl) planes present in the sample, the pole figure method consists in fixing the diffraction angle θ to the value corresponding to the d_{hkl} inter-reticular distance and collecting the diffracted intensity at different ϕ ($0^\circ < \phi < 360^\circ$) and χ ($0^\circ < \chi < 90^\circ$) angles, where ϕ is the rotation angle of the sample around its normal and χ the tilt angle of the sample perpendicularly to the diffraction plane. An epitaxial layer gives intense and narrow (hkl) spots at specific ϕ and χ angles corresponding to the symmetry of the crystal. If the growth is only textured (all the crystals have the same growth direction but no common direction in the sample plane), a ring is observed at a specific χ . If the growth is polycrystalline, the intensity is spread on the whole solid angle. (a) Definition of angles. (b) Geometry of the stereographic projection plots. (c) Maximum intensity of the (011) peak as a function of Y . (d) Pole figures in TSF2. (e) Pole figure in TSF25 (the intensity is multiplied by 45 in the two light squares to make the 200 peak of the substrate visible).

Figure A3: Characteristics deduced from the fits of the $\theta/2\theta$ x-ray diffraction scans: (a) Rocking curves full widths; (b) Coherence length deduced from the width of the two main peaks in BXFY and TXY (pink dot-dashed line: expected total thickness; vine dashed line: expected thickness of the bilayer from reflectometry $2e_R/3$; green dot line: expected individual thickness $e_R/3$). (c,d) Individual thicknesses in the Cr and Fe layers in the two kind of fits ($N_3=N_1$; $N_3>N_1$); (e,f) Out-of-plane lattice parameters in the two types of fits ($N_3=N_1$; $N_3>N_1$) in Cr and Fe layers of trilayers and in multilayers.

Figure A4: Simulations of the $\theta/2\theta$ x-ray diffraction scans in MMF10 in \log_{10} plot. Circles: experimental points. Red lines: simulation with diffraction length and perpendicular-strain contrasts. Dotted blue lines: simulation with diffraction length contrast only. The light colors correspond to simulations with a 0.002 background. Architecture of the superlattice: $\text{Fe}_{46}(\text{Cr}_{70}\text{Fe}_{64})_5$ (the subscripts are the N_i 's). Parameters of the simulations: $d_{\text{Fe}}=0.28452$ nm, $d_{\text{Cr}}=0.28857$ nm for the red lines; $d_{\text{Fe}}=d_{\text{Cr}}=0.2866$ nm for the dotted blue lines.

Figure A5: Examples of maps in the reciprocal space in the MMF10 sample: (a) 200 peak ($\psi=0^\circ$), (b) 211 peak ($\psi\approx 34^\circ$), (c) 222 peak ($\psi\approx 55^\circ$), and (d) 130 peak ($\psi\approx 72^\circ$).

Figure A6: (a) Thermal expansion coefficients, α_L , of bulk STO, MgO, Fe, and Cr between 300K and 900K. (b) Lattice parameter normalized to its value at 773K in bulk STO, MgO, Fe, and Cr between 300K and 900K deduced from α_L by integration after interpolating α_L with the second degree polynomial function (the hatched range containing the Néel temperature is discarded).

