Strain dependence of antiferromagnetic interface coupling in $La_{0.7}Sr_{0.3}MnO_3$/$SrRuO_3$ superlattices

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We have investigated the magnetic response of $La_{0.7}Sr_{0.3}MnO_3$/$SrRuO_3$ superlattices to biaxial in-plane strain applied *in-situ*. Superlattices grown on piezoelectric substrates of $0.72PbMg_{1/3}Nb_{2/3}O_3$-$0.28PbTiO_3$(001) (PMN-PT) show strong antiferromagnetic coupling of the two ferromagnetic components. The coupling field of $\mu_0 H_{AF}$ = 2.8 T is found to increase by $\mu_0 \Delta H_{AF}/\Delta\varepsilon$ ~ -520 mT $\%^{-1}$ under reversible biaxial strain ($\Delta\varepsilon$) at 80 K in a $[La_{0.7}Sr_{0.3}MnO_3(22$ Å$)/SrRuO_3(55$ Å$)]_{15}$ superlattice. This reveals a significant strain effect on interfacial coupling. The applied in-plane compression enhances the ferromagnetic order in the manganite layers which are under as-grown tensile strain. It is thus difficult to disentangle the contributions from strain-dependent antiferromagnetic Mn-O-Ru interface coupling and Mn-O-Mn ferromagnetic double exchange near the interface, since the enhanced magnetic order of Mn spins leads to a larger net coupling of $SrRuO_3$ layers at the interface. Strain-dependent orbital occupation in a single-ion picture cannot explain the sign of the observed strain dependence, whereas the enhanced Mn order at the interface is qualitatively in line with it.

I. Introduction

Magnetic order and coupling at coherent interfaces between oxides of perovskite type have received increasing interest during the last decade. This includes the search for phenomena already known from metal films, e. g. the exchange bias effect between a ferro- and an antiferromagnetic layer[1] and the interlayer coupling through non-magnetic spacer layers responsible for giant magnetoresistance in Co/Cu/Co[2,3]. Additionally, new phenomena have been discovered reminding of the two-dimensional electronic states at semiconductor interfaces, but adding the magnetic degree of freedom to electronic interface states[4]. The most prominent example is the conducting electron gas at the interface between the insulators $LaAlO_3$ and $SrTiO_3$[5]. The interface of ferromagnetic $SrRuO_3$ (SRO) with ferromagnetic manganites such as $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) is in a focus of interest, because it shows an antiferromagnetic coupling with thus far unparalleled coupling strength in oxides[6]. The antiferromagnetic exchange coupling at the interface leads to antiparallel orientation of the magnetizations of thin adjacent SRO and LSMO layers which can be sustained in a magnetic field of several Tesla[6,7,8]. The origin of the coupling is the Mn-O-Ru superexchange interaction across the coherent interface[6,9]. Subsequent work showed the complexity of magnetic order arising from combination of the antiferromagnetic interface coupling with magnetic anisotropies of the components which are perpendicular to the film plane and strong for SRO and in-plane and weak for LSMO on $SrTiO_3$(001) substrates, respectively. An inhomogeneous magnetization depth profile with in-plane Ru spins near the interface and perpendicular Ru spins inside the SRO layer has been detected by neutron reflectivity measurements[10]. The magnetic order at low temperatures depends heavily on the cooling history of samples.[11] One reason for this is the alignment of Ru spins during cooling through $T_C^{SRO} \sim 150$ K according to the more dominant energy of either (i) the exchange coupling to ordered Mn spins ($T_C^{LSMO} \geq 250$ K) at the interface, or (ii) the magnetic anisotropy energy of SRO, or (iii) the Zeeman energy in an applied magnetic field[11]. At low temperatures, the magnetic anisotropy of SRO is so large that full alignment of Ru spins is hard to achieve in applied magnetic fields of a few Tesla. Hence, the arrangement of Ru spins during cooling is (partially) "frozen in".

Meaningful investigation of magnetic coupling at oxide interfaces has been enabled by the advance of experimental tools such as RHEED-assisted layer-wise growth under high oxygen pressure[12] and scanning transmission electron microscopy (STEM). The latter allows for semi-quantitative evaluation of chemical intermixing at interfaces by applying the high angle annular dark field technique (HAADF). Thermal diffuse electron scattering at high angles (>70 mrad) is recorded with the intensity of the localized, incoherent scattering processes proportional to Z^2 (Z denotes the atomic number). Thus the position of atom columns or individual atoms is imaged with a brightness related to their atomic number, usually referred as Z-contrast. This technique has been employed to characterize LSMO/SRO interfaces.[18]

Biaxial epitaxial strain is crucial for magnetic exchange interactions because it systematically alters bond angles and lengths.[May10] It has been shown to strongly affect and even reverse the sign of Mn-O-Ru interface coupling in ultrathin $SrRuO_3$/$AMnO_3$/$SrRuO_3$ (A = Ca or Pr) trilayers as observed by X-ray magnetic circular dichroism.[13] That experiment revealed the impact of strain on the magnetic coupling by comparing trilayers grown coherently on $SrTiO_3$(001) and $LaAlO_3$(001) substrates. Superlattices (SL) of LSMO/SRO could not be grown coherently on different substrates thus far, but rather all published work concentrates on SLs grown on TiO_2-terminated $SrTiO_3$(001). Therefore, it seems useful to attempt *in-situ* strain control on such SLs using piezoelectric $0.72PbMg_{1/3}Nb_{2/3}O_3$-$0.28PbTiO_3$(001) (PMN-PT) substrates.[14,15] The strain dependence of magnetic order in SRO and LSMO single films has been investigated earlier using *in-situ* strain.[16,17] Those results for bulk-like films with thicknesses beyond 50 unit cells (20 nm) can help to understand the properties of ultrathin layers in SLs, but must be considered with care because interfaces don´t matter for the magnetization of bulk-like films. We investigate the strain dependence of the antiferromagnetic coupling in LSMO/SRO superlattices grown on piezoelectric PMN-PT substrates and find a large response to reversible biaxial strain. We consider the different contributions of Mn-O-Ru superexchange interactions across the interface in a single-ion picture, but find no agreement in the sign of the effect. Rather, the strain-dependent order of Mn spins at the interface is suggested to contribute to the strain-induced change of the apparent antiferromagnetic coupling.

II. Experiments

[22 Å $La_{0.7}Sr_{0.3}MnO_3$ (LSMO)/ 55 Å $SrRuO_3$ (SRO)]$_{15}$ superlattices (SLs) have been grown by Pulsed Laser Deposition (PLD) with a KrF laser (wavelength 248 nm) on (100)-oriented $SrTiO_3$ (STO) and $0.72Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.28PbTiO_3$ (PMN-PT) substrates using stoichiometric targets of LSMO and SRO. The laser energy density during deposition was 3 J/cm^2 and the frequency 3 Hz. The SLs are grown in 0.1 mbar of pure oxygen at 700°C substrate temperature. After deposition, in-situ annealing is done at 600 mbar O_2 at 700°C for 45 mins. The deposition started with a LSMO layer and ended with a SRO layer.

The SLs have been structurally characterized by X-ray diffraction in a Bruker D8 Discover diffractometer. The microstructure of the SLs has been investigated by high-angle annular dark field (HAADF) imaging in a TITAN 80-300 (FEI) scanning transmission electron microscope (STEM). The chemical interdiffusion or intermixing at interfaces was probed by an energy dispersive X-ray spectrometer (EDX) attached to the TITAN and operating in the STEM mode. The magnetization of the SLs has been measured in a SQUID (Superconducting Quantum Interference Device) magnetometer. The magnetization is expressed in Bohr magnetons per total number of pseudocubic unit cells. The piezoelectric PMN-PT substrates are used to carry out strain-dependent measurements.[14,16] An electrical voltage is applied along the substrate normal between the top of the SL serving as top electrode and a NiCr/Au

back electrode of the substrate. The piezoelectric strain of the substrate is transferred to the SL layers in spite of the large total thickness.[15,19]

III. Results and Discussion

A. Structural characterization

Fig. 1(a) shows the θ-2θ XRD scans around the (002) reflection of the SL grown on PMN-PT and STO, respectively. A strong main peak and sharp satellite peaks of the SL are observed, indicating good structural quality with sharp interfaces. The differences in peak positions are related to the slightly different in-plane strain of SLs on STO and PMN-PT, respectively. In order to determine the average in-plane (a) and the out-of-plane (c) lattice parameters of the superlattices, reciprocal space maps around the pseudocubic (103) reflections were recorded. The determined lattice parameters of the SL are weighted averages over the components. According to our XRD measurements, SLs grown on STO are strained coherently to the substrate lattice with an in-plane parameter a_{STO} = 3.905 Å. Thus, the LSMO layers in the coherently grown SL are under tensile strain, while the SRO layers experience compressive strain, referring to the bulk lattice parameters of 3.87 Å and 3.93 Å for LSMO and SRO, respectively.

A XRD reciprocal space map of the SL on PMN-PT is shown in Fig. 1 (b). The SL is not coherently strained to the PMN-PT substrate because of the larger in-plane parameter of a_{PMN-PT} ~ 4.02 Å (which depends on ferroelectric poling). Strain relaxation occurred at the substrate-SL interface, and the SL assumed a lattice parameter of a = 3.92 Å. In-situ recording of the in-plane parameter by tracking the distance of RHEED diffraction streaks during growth has been used to check for strain relaxation during growth. No strain relaxation has been found, pointing to a coherent growth of the SL (further checked by HR-TEM below). The in-plane lattice parameter of the SL on PMN-PT is slightly larger than that on STO. Hence, LSMO layers are under slightly stronger tensile strain than in the SL grown on STO, while the SRO layers are under weak compressive strain. No conclusion on the lattice symmetry of the SL components (SRO) can be made from the XRD measurements. While in single layers of LSMO or SRO on STO(001) substrates the film structure is expected to be tetragonal (LSMO) or orthorhombic with small monoclinic distortion (SRO), respectively, the symmetry of the layers in the SL might be different. For example, it has been shown that ultrathin SRO layers in SLs with PCMO layers are tetragonal.[20]

High-resolution STEM images of the SL on PMN-PT confirm the absence of dislocations and other crystal defects breaking the coherence of the lattice inside the SL (Fig. 2(a)). Probably due to the less well-defined surface of the PMN-PT substrate (and the lattice mismatch of the components), the SRO layers don´t grow in fully flat way, but show thickness fluctuations of 2-3 unit cells. The intermixing at the interfaces has been probed by

tracking the EDX composition along a line across the interfaces using the Ru-Kα and the Mn-Kα X-ray intensities (Fig.2(b)). From this figure, intermixing of the elements Ru and Mn can be deduced to range over a distance of about 1 unit cell for both interfaces LSMO/SRO and SRO/LSMO. Interestingly, intermixing is very small at the interfaces in spite of the non-ideal flatness of the layers. This indicates the absence of a chemical driving force for intermixing under the applied growth conditions. No clear difference between the interfaces of LSMO/SRO and SRO/LSMO (in the sequence of growth) has been found, contrary to the expectation for a well-defined termination of sharp interfaces between layers of complete perovskite unit cells. This may result from a random termination on the PMN-PT surface or be a consequence of the intermixing. An inspection by STEM of a SL on SrTiO$_3$ substrate revealed fully coherent growth of flat layers comparable to earlier published work by Ziese et al.[6] A similar magnitude of intermixing at the interfaces has been found as for the SL on PMN-PT.

We first discuss magnetization measurements of a representative SL on PMN-PT. Temperature-dependent magnetization curves recorded in a magnetic field of $\mu_0 H$ = 0.1 T after field-cooling in 2T give evidence for the antiferromagnetic coupling of SRO and LSMO layers.[6] The Curie temperatures of the components, T_C^{SRO} = 156 K and T_C^{LSMO} = 263 K, are close to the bulk value for SRO and strongly reduced (because of the tensile strain of $\sim$ 1.3% and the low layer thickness) for the LSMO layers. Magnetic hysteresis curves $M(H)$ have been measured at temperatures between 10 K and 100 K both, in the film plane along a pseudocubic 100 direction and along the film normal, the 001 direction. For T = 80 K (and in the range of 60 – 100 K), $M(H)$ reveals hard-axis behavior with nearly reversible magnetization rotation for the normal direction (Fig.3). This result indicates spontaneous in-plane magnetization for both layers. In-plane M(H) loops measured along a 110 diagonal direction show smaller M(4 T) and smaller remanent magnetization, both indicating {100} easy axes. (In stating that, we assume biaxial in-plane symmetry not to be broken.)

In-plane $M(H)$ loops (Fig.3) show a two-step switching process in the field. Firstly, the LSMO layers align along the field. This is not immediately obvious, because strong antiferromagnetic interlayer coupling may lead to different switching sequences depending on the magnetic moments of both layers.[6] Next to the Zeeman energy in the applied field and the magnetic anisotropy energy of the respective layer, the interface coupling governs the switching and may lead to different loop shapes / switching sequences.[22,?] Based on layer thicknesses and magnetization values of 3.7 μ_B/Mn for LSMO and 1.1 μ_B/Ru one expects the magnetic moment of LSMO layers to be larger than that of SRO layers. This would mean the first switching step is related to LSMO alignment (Fig.3), whereas the second is the SRO alignment with the applied field. Confirmation for this assumption is found below in the strain response. We assign the midpoint of the SRO transition (defined as the point where 50% of the SRO magnetization has been switched) as the coupling field H_{AF}. H_{AF} increases from 1.4 T to 2.8 T when the sample is cooled from 100 K and 10 K. The magnitude and temperature dependence of H_{AF} approximately agree with earlier work on SLs on

SrTiO$_3$(001) substrates.[7,8] In more detail, H_{AF} depends on several parameters including SRO layer thickness, in-plane strain and interdiffusion at the interfaces:[6] H_{AF} is proportional to the inverse SRO thickness,[23] and decreases with increased level of interface roughness / interdiffusion. There is no information on the impact of biaxial in-plane strain on the coupling strength available thus far. The observed strong AFM coupling in the SL on PMN-PT indicates good structural interface quality in agreement with the chemically sharp interfaces found by STEM. The fluctuations in SRO layer thickness surely have the effect of broadening the switching transition. We note that other samples prepared under less favourable growth conditions did not show strong (or even any) coupling; deposition parameters are vital to obtain strongly coupled samples on PMN-PT.

At 10 K where the anisotropy of SRO is very large, the out-of-plane magnetization is more hysteretic and reveals some remanent magnetization (Fig.4a). This indicates that some SRO spins are canted out-of-plane at 10 K. A canted or vertical easy axis may be present in an inner section of the SRO layers[10] at low temperatures. Therefore, strain-dependent measurements have been restricted to T ≥ 60 K where M essentially lies in the film plane.

For inspecting the effect of biaxial strain, Fig.5 gives a comparison of the $M(H, T = 80$ K) loops in the as-grown and a biaxially compressed ($\Delta\varepsilon \sim$ -0.07%) state. The change between the two loops is reversible and controlled by the piezoelectric substrate strain. Similar loops have been measured between 60 K and 100 K. The immediately obvious impact of the compression is an enlargement of the saturated magnetization (at $\mu_0 H$ = 4 T) which roughly agrees with the enlargement seen after the first switching step (at $\mu_0 H$ = 1 T) (Fig.5). Ferromagnetic order in LSMO is known to be very sensitive to tensile strain, reflected in strong strain-induced shifts of T_C for thicker LSMO films.[16] Ultrathin films like those in the present SL sample show some magnetic disorder at the interfaces which substantially contributes to the total magnetization (which takes, as an absolute value, only about half of the fully ordered value). The latter fact makes the LSMO magnetization strain-dependent through the influence of strain on the ferromagnetic double exchange interaction. The applied reversible compression releases a small part of the as-grown tensile strain of ~1.3% in the LSMO layers. This has a profound effect on LSMO magnetization at $T << T_C^{LSMO}$ which increases by 6.3% (at 60 K), 5.5% (80 K) or 4.4% (100 K), respectively. Unfortunately, this also reveals a general problem in assessing the interlayer exchange coupling as an independent parameter of interest. Stronger apparent AFM coupling of the SRO layer at the interface as detected by strain-dependent magnetization measurements may result from both, (i) stronger Mn-O-Ru exchange interaction and (ii) higher ordered Mn moment at the interface. (We note that the extreme case of randomly oriented Mn moments would offer no net coupling to ferromagnetically aligned Ru moments.) The issue is further discussed below.

Strain-induced changes of H_{AF} have been determined as the difference of H_{AF} values in two investigated strain states. The values are $\Delta H_{AF}/\Delta\varepsilon$ = 650 mT %$^{-1}$, 520 mT %$^{-1}$, and 410 mT %$^{-1}$ (with an error of ~20%) at the temperatures of 60K, 80 K, and 100 K. (Lower temperatures have not been investigated because the spontaneous magnetization shows

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some reorientation out of the film plane as discussed above.) Fig.6 provides a direct view on the change of H_{AF} induced by the piezo-compression in the following way: the $\varepsilon = 0$ loop has been shifted vertically by a constant value to match the loop under strain at saturation (4 T). In this way, the strain-enhanced LSMO magnetization is compensated. One notes the shift of H_{AF} at the 50% level of the transition. Further, there is a lower slope dM/dH of LSMO around 1 T in the strained case. The latter results from better ferromagnetic order of the LSMO layers in small fields after partial release of tensile strain.

The magnetic behavior of the reference SL sample grown on STO substrate is useful to compare because of its smaller in-plane lattice parameter. The Curie temperatures of the components are T_C^{SRO} = 143 K and T_C^{LSMO} = 305 K. T_C^{SRO} is not so far from the SRO bulk value, but smaller than that of the SL on PMN-PT, in qualitative agreement with the increase of T_C^{SRO} between a = 3.905 Å and 3.92 Å.[17] T_C^{LSMO} is about 40 K higher on STO, an expectable shift for the 0.4% weaker tensile strain of the LSMO layers. The magnetic anisotropy of both SLs is quite different (Fig.4): curiously, the in-plane and out-of-plane $M(H)$ loops for both cases appear nearly like interchanged at 10 K. Weak hysteresis and rotation of magnetization in the field occurs for the in-plane (100$_{pc}$) direction on STO, whereas the out-of-plane M shows a distinct transition at an antiferromagnetic coupling field of H_{AF} = 2.8 T. Hence, both layers of LSMO and SRO in the SL on STO have a spontaneous perpendicular (or canted) magnetization which is antiferromagnetically coupled. This coupling is of similar strength like the in-plane coupling for the SL on PMN-PT. This change of the magnetic anisotropy is consistent with the known influence of epitaxial strain on the anisotropy in single SRO layers, where compressed films on STO(001) substrate show tilted perpendicular anisotropy.[24]

Regarding the origin of strain-dependent antiferromagnetic coupling, Seo et al.[13] have proposed a strain-dependent orbital occupation of Ru^{4+} ions at the interface as briefly summarized here. The four 4d electrons of the Ru^{4+} ion occupy t_{2g} states as three majority-spin electrons ($t_{2g}^{\uparrow}$) and one minority-spin electron ($t_{2g}^{\downarrow}$). The $t_{2g}^{\uparrow}$ electrons half-fill all t_{2g} orbitals, providing antiferromagnetic coupling with the half-filled $t_{2g}^{\uparrow}$ Mn 3d orbitals according to the Goodenough-Kanamori rules. The in-plane (x-y) orbitals don´t contribute to the Mn-O-Ru coupling in cubic symmetry (and little in tetragonal symmetry with a small distortion of the lattice as present in our SL). The $t_{2g}^{\downarrow}$ electron of Ru^{4+} may reside in the xy in-plane orbital or in the xz or yz out-of-plane orbitals, in the second case contributing a ferromagnetic component to Mn-O-Ru coupling. In-plane strain may alter the relative occupation of in-plane and out-of-plane orbitals, with compression favouring the out-of-plane ones. Hence, the smaller the in-plane lattice parameter, the larger would be the ferromagnetic contribution of this fourth Ru electron. The absence of orbital degeneracy for the other three electrons makes their contribution insensitive to strain.

This scenario applied to our experiment implies reduced effective antiferromagnetic coupling for the smaller in-plane parameter, in contrast to observation. In a step beyond, the contributions of e_g orbitals have been considered. In $SrRuO_3$, the Ru^{4+} e_g orbitals are empty because of the large crystal field splitting. In Mn^{4+}, they are empty, whereas in Mn^{3+} there is

one $e_g^{\uparrow}$ electron. Nominally, LSMO contains 30% of Mn^{4+} and 70% Mn^{3+} ions. Coupling via the e_g $3z^2$-r^2 orbitals of Mn and Ru would thus be antiferromagnetic for Mn^{4+} and ferromagnetic for Mn^{3+} at the interface (according to the Goodenough-Kanamori rules). The e_g $3z^2$-r^2 orbital occupation of Mn^{3+} is expected to increase with in-plane compression, because the single e_g electron gets a higher probability to leave the tensile-strain-stabilized x^2-y^2 orbital. Again, this e_g-orbital-related mechanism reduces the total antiferromagnetic coupling upon in-plane compression and thus disagrees with our result. Possibly, such a single-orbital consideration fails to describe the covalent behavior of $SrRuO_3$.

However, as mentioned above, it is difficult to isolate the Mn-O-Ru interface coupling based on magnetization measurements if the Mn-O-Mn coupling at the interface is changing simultaneously. This is clearly true for our experiment, as is seen in the enhanced saturated magnetization of the LSMO layers upon piezo-compression. Understanding interface coupling means to take into account the magnetic order in both components at the interface, next to the exchange interaction through the interface. Manganite layers are known to show some degree of magnetic disorder at interfaces. In our experiment, this is evident from the lower saturated moment of LSMO as follows (Fig.5). For the as-grown state, the magnetic moment of ~0.6 μ_B per unit cell of the superlattice at 1 T is assumed to represent LSMO layers aligned and SRO layers anti-aligned with the field. The reversal of SRO layers yields a change by ~0.3 μ_B / u.c., leading to an estimated ordered moment of 2.6 μ_B / Mn, in contrast to 3.7 μ_B / Mn for fully ordered Mn spins. Release of tensile strain is known to enhance the ferromagnetic Mn-O-Mn double exchange interaction in LSMO, in line with the observed larger LSMO magnetization upon in-plane compression. Hence, we expect the increased antiferromagnetic coupling of SRO layers to result, at least partially, from better ordered Mn spins at the interfaces.

Summarizing, coherent superlattices of LSMO(22 Å)/SRO(55 Å) on piezoelectric PMN-PT substrates show strong antiferromagnetic interface coupling with profound strain dependence. The coupling field H_{AF} is enhanced by ~50 mT per 0.1% of reversible biaxial compression (for a superlattice in-plane parameter of 3.92 Å). Simultaneously, the magnetic order of the LSMO layers grows. We see the latter effect as an important second influence on H_{AF} besides the strength of the Mn-O-Ru superexchange interaction; it is possibly dominating the observed strain effect. Mechanisms of strain-dependent orbital occupation in a single-ion picture are not in agreement with the experiment, because in-plane compression moves the balance of ferro- and antiferromagnetic exchange couplings towards the ferromagnetic side. Strictly speaking, it is hard to discriminate between the contributions from Mn-O-Ru and Mn-O-Mn couplings at interfaces based on magnetization measurements, i. e. the effective interface coupling also depends on the magnetic order of the components at the interface.

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Figure Captions

Fig 1: (a) *Θ-2Θ* X-ray diffraction scans around the (002) reflection of the superlattices on STO and PMN-PT substrates, respectively. (b) Reciprocal space map around the (103) reflection on PMN-PT.

Fig: 2 (a) HAADF-STEM images of sample SL on PMN-PT, (b) EDX line scans of Ru and Mn, crossing LSMO/SRO layers. The dashed lines indicate an intermixing depth of about 4 Å.

Fig. 3: Field cooled (FC) in-plane (ip) and out-of-plane (op) magnetization loops of the superlattice on PMN-PT .

Fig. 4: Field cooled (FC) in-plane (ip) and out-of-plane (op) magnetization loops at *T*=10K of the superlattices on (a) PMN-PT and (b) STO, respectively.

Fig.5: In-plane magnetization loops of the superlattice on PMN-PT in the as-grown state (ε = 0) and after piezocompression (ε = - 0.07%).

Fig.6: Direct view on the change of antiferromagnetic coupling field (H_{AF}) induced by the piezo-compression at (a) T= 80K and (b) T = 60K. We define H_{AF} as the field where 50% of the SRO magnetization has been switched.