Direct observation of pinned/biased moments in magnetic superlattices

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Abstract

We report the pinned/biased moment in the superlattices consisting of ferromagnetic (FM) SrRuO$_3$ and antiferromagnetic (AFM) SrMnO$_3$. This superlattice system shows anisotropy and oriented pinning/biasing in the field-cooled (FC) hysteresis loop. The in-plane cooling-field provides antiferromagnetic orientations while out-of-plane cooling-field provides ferromagnetic orientations to the pinned/biased moments. The spacer layer thickness, strength and orientation of magnetic field, cooling field, and driving current influence the pinning strength. We propose that the magnetic structure is a repetition of $AFM/\text{Pin}/FM\langle\text{Free}\rangle/\text{Pin}$ unit below a critical field to explain its magnetic and transport properties. The transport behavior is discussed using the spin-dependent conduction.

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A biased magnetic field has been observed on cooling the FM-AFM system below the Curie temperature ($T_C$) of the $FM$ through the Neel temperature ($T_N$) of the $AFM$ in presence of a magnetic field$^{1-5}$. It is believed that the biased field is responsible for the shift of the hysteresis loop along the field axis which has been observed in a wide variety of $FM$ – $AFM$ systems, many of which do not exhibit a simple spin structure at the interface to the $FM$ -AFM layers or materials. In general, a biased field is established through field-cooling in the film plane where the magnetic easy axis of soft ferromagnetic materials normally lies in plane. A shift in hysteresis loop along the magnetization axis in addition to the shift along the field axis is also observed$^6$. The authors have explained the shift in hysteresis loop along the magnetization axis by the pinned uncompensated spin at the interfaces. Recently Maat et al.$^7$ have shown that exchange bias can also be observed for the magnetization perpendicular to the film plane in $Co/Pt$ multilayers biased by $CoO$. They investigated the biasing in various directions and found substantially more within the sample plane, which they related to the anisotropy of the single-$q$ spin structure of the $CoO$. Several theoretical models have been proposed to explain the origin of the biased field. Indeed, most of the theoretical models assume a single domain state of the ferromagnetic layer and focus on the domain structure of the $AFM$ layer for different types of interfaces. However, despite the enormous research done in this field, this effect is poorly understood.

Here, we report the direct observation of pinned/biased moments of the $SrRuO_3$ ($SRO$) layers by the $SrMnO_3$ ($SMO$) layers in the $SRO/SMO$ superlattices grown on (001)-$SrTiO_3$ ($STO$) substrates. To the best of our knowledge, this observation has not been reported so far. The presence of pinned/biased effect can be realized in the magnetic hysteresis loop with field range below certain critical magnetic field ($H_P$). Various factors such as the $SMO$ layer thickness, strength and orientation of the external magnetic field and cooling field influenced the strength of the pinned/biased moments of $SRO$, thus providing a way to control it. Consequently, this presents the tantalizing possibility of controlling the pinning of a $FM$ layer by the $AFM$ layer in an oxide multilayer, which is a necessary step towards a better understanding and improvement of modern magnetic devices.
The fabrication with optimized growth conditions and structural characterizations of the superlattices have been reported elsewhere. The superlattice structures were synthesized by repeating 15 times the bilayer comprising of 20-(unit cell, u.c.) SRO and n-(u.c.) SMO, with n taking integer values from 1 to 20. In all superlattices, SRO is the bottom layer and the modulated structure was covered with 20 u.c. SRO to keep the structure of the top SMO layer stable. The samples were characterized by resistivity (\(\rho\)) and magnetization (\(M\)) measurements, in addition to x-ray diffraction and transmission electron microscopy. Transport and magnetization measurements were performed at 10 K with magnetic field along the [100] and [001] directions of STO. The samples were cooled to a desired temperature (\(T\)) from room temperature in the absence of electric and magnetic field to perform zero-field-cooled (ZFC) measurements. The field-cooled (FC) measurements were always performed with the same orientation of cooling field.

\(SrRuO_3\) is known as a metallic FM, with a Curie temperature (\(T_C\)) \(\sim\) 160 K in its bulk form. Similar transport and magnetic behaviors are observed in (80 nm)SRO/STO with easy axis along [001] direction of STO, consistent with Ref.10. The saturation field (\(H_S\)), coercive field (\(H_C\)) and saturation magnetization (\(M_s\)) along the easy axis of this film are 0.4 tesla, 0.17 tesla and 1.46 \(\mu_B/\text{Ru}\), respectively. Its ZFC and FC magnetic hysteresis loop (\(M − H\)) remain the same at 10 K under 0.1 tesla cooling field (\(H_{FC}\)). The current-in-plane magnetoresistance (\(MR\)) of this sample with magnetic field along [100] and [001] directions of the STO is negative although it is hysteretic and higher when \(H \perp I\). In contrast, \(SrMnO_3\) is an AFM with a Neel temperature close to 260 K\(^{11}\) and crystallizes in a cubic structure when sandwiched between perovskite layers inside a superlattice\(^8\).

Fig. 1 shows the zero-field-cooled (ZFC) magnetization at 10 K at various magnetic fields oriented along the in-plane and out-of-plane directions of the substrate for the sample with 3 u.c. thick SMO layer. The easy axis of SRO remains same in the superlattices. The in-plane magnetization of the superlattice gradually increases as the magnetic field increases and becomes larger than the calculated value (1.6 \(\mu_B/\text{Ru}\)), based on the only contribution from SRO layer. This larger value of the in-plane magnetization indicates that the SMO layer
contribute to the net magnetization of the superlattice at higher magnetic field. However, the out-of-plane hysteresis loop shows a clear $M_S$ and $H_S$ with enhanced $H_C$. In order to understand the strong anisotropic nature of the ferromagnetic layer in the superlattice and their magnetotransport behavior below $H_C$, we have measured the minor hysteresis loops of this superlattice in the field range between the saturation field of SRO and the out-of-plane $H_C$ of the superlattice with $n = 3$. The minor ZFC hysteresis loops in the field range of ±1 tesla are symmetric with respect to the origin (Fig. 2a and 2b) for magnetic field along the [100] and [001] directions of STO. The gradual increase in magnetization with the increase in magnetic field even above the $H_S$ (0.4 tesla) of SRO indicates that the spin-orbit coupling is modified in SRO layer and that the $MnO_6$ octahedra at the interface influences the magnetic state of the $RuO_6$ octahedra\(^{12,13}\). The $H_C$ of SRO layer (0.02 tesla along [100] and 0.17 tesla along [001]) is reduced in the superlattices (0.001 tesla and 0.0027 tesla respectively). The magnetization of SRO layer (1.46 $\mu_B$/Ru) is decreased to $\approx 0.6$ $\mu_B$/Ru in the superlattices. This large suppression of the FM state of the SRO layer in the superlattice suggests that it is strongly influenced by the G-type AFM state of the SMO layer\(^{11}\). In the case of G-type spin ordering, the (00$l$) planes show the staggered pattern of spin arrangement, which is the source of spin frustration at the compensated SRO – SMO interfaces as well as the spin canting in the SRO layer in the vicinity of the interfaces\(^{14}\). Thus, due to the presence of SMO layer, the spin canting/frustration in the SRO layer is reducing the effective FM layer thickness of the SRO layer in the SRO/SMO superlattice. In others words, the effective ferromagnetic SRO layer thickness is decreasing by the presence of a canted/frustrated spin in the SRO layer close to the interface, which will be detailed hereafter (Fig.5b).

In general, the magnetic interactions across the interface between the FM and AFM are known as exchange coupling (EC), with phenomenological features such as an enhancement and an unidirectional anisotropy of $H_C$.\(^{1-5}\) To study the exchange coupling at the FM – AFM interfaces, we have measured the FC hysteresis loop of this sample. The FC hysteresis loop of the superlattice with $n = 3$ for in-plane and out-of-plane orientations of the
magnetic field are shown in Fig. 2a and 2b respectively. It shows several interesting features. First, the center of in-plane as well as out-of-plane FC hysteresis loop is shifted along the magnetization axis. Second, the FC hysteresis loops show a negligibly small change in the values of $H_C$ compared to the ZFC hysteresis loop. Third, the values of the in-plane magnetization in the FC hysteresis loop is lower (Fig. 2a) while the out-of-plane magnetization in the FC hysteresis loop is higher (Fig. 2b) than its corresponding magnetization in the ZFC hysteresis loop. These features indicate that the spin configuration that was realized in the ZFC state is modified in presence of cooling magnetic field. From the observed ZFC and FC hysteresis loop one can conclude that the canted/frustrated spins are aligned antiferromagnetically in the presence of in-plane cooling-field and are aligned ferromagnetically for out-of-plane cooling-field. So we define the oriented interfacial canted/frustrated spins as the pinned/biased moments at the interfaces. The in-plane pinned/biased moment can be defined as $M_{\parallel P} = M_S(0) - M_S(H_{FC})$. Taking into account the weak diamagnetic response of the substrate, the $M_S$ has been extracted by extrapolating the linear part of the $(M - H)$ curve to $H = 0$. The value of $M_{\parallel P}$ when $H$ is antiparallel to $H_{FC}$ is larger by $\approx 0.302 \times 10^{-4}$ emu (a factor of 0.3) compared to the value of $M_{\parallel P}$ when $H$ parallel to $H_{FC}$. This indicates the presence of moments at the interfaces which do not flip $180^\circ$ with the flipping of the magnetic field. So the canted/frustrated layer, partially close to the $SRO - SMO$ interface, is pinned/biased along the direction of the cooling magnetic field. In other words, this is a signature of uniaxial pinning/biasing of moments at the interfaces. The value of $M_{\parallel P}$ changes significantly at cooling fields below $\pm 0.03$ tesla and remains constant for higher values of $H_{FC}$. Similarly, the out-of-plane pinned/biased moment $M_{\perp P}$ can be defined by analogy to the bias field as $M_{\perp P} = M_{R}^{+} + M_{R}^{-}$, where $M_{R}^{+}$ and $M_{R}^{-}$ are field-increasing and field-decreasing remanent magnetization respectively. The same sign of the field for increasing and decreasing $M_{R}$ (Fig. 2b) indicates the uniaxial pinning/biasing of moments. The value of $M_{\perp P}$ increases with $H_{FC}$ and changes negligibly when $H_{FC} > 0.1$ tesla. $M_{\perp P}$ also depends on the magnetic field that is applied and becomes zero when a magnetic field larger than 1.5 tesla is applied. We have also measured the $M_{\perp P}$ for various superlattices and the results are
given in Fig. 3. It decreases as the SMO layer thickness increases above 1 u.c., and remains the same for \( n > 7 \). Since \( M^\perp \) varies with SMO layer thickness, this indicates that the EC at the interfaces is a combination of the exchange coupling \( (J_{exch}) \) between SRO layer and SMO layer and the interlayer exchange coupling \( (J_{int}) \) between the SRO layers. Note that for SRO/SMO superlattice, the Neel temperature of SMO layer is higher than the Curie temperature of SRO layer. Since the exchange coupling also depends on the thermal energy, the physical processes responsible for the effective exchange coupling \( (J_{eff} \approx J_{Exch} + J_{int}) \) is expected to be different from the AFM/FM system where \( T_C > T_N \).

From the ZFC and FC magnetization measurements of the SRO/SMO superlattices, we have observed a strong anisotropy and pinned/biased moments. To understand the effects of these magnetic behavior we have also studied their electronic transport in presence of magnetic field below \( H_P \). The ZFC and FC current-in-plane magnetoresistance for various magnetic fields \((MR - H)\) in the range of magnetic field \((\pm 2 \text{ tesla})\) below \( H_P \) of the sample with \( n = 3 \) for field along [100] and [001] directions of STO are shown in Fig. 4. The ZFC out-of-plane MR (Fig. 4b) is negative as well as positive with hysteretic and asymmetric nature. As the field sweep starts, the MR increases and shows a sharp change from positive to negative value at \( + H_{flip} \). On reverse sweep of \( H \) to zero from \( + 2 \text{ tesla} \), the MR decreases with a lower value than the MR in the field-increasing branch. As \( H \) increases in the negative direction, the MR becomes positive until the field is smaller than \(- H_{flip} \) and at \(- H_{flip} \), the MR becomes negative. The negative field decreasing branch is similar but opposite to the reverse positive field sweep branch. In presence of \( H_{FC} \) the out-of-plane MR (Fig. 4a and 4c) is negative as well as positive, less hysteretic, more asymmetric and higher in magnitude compared to the ZFC MR. The ZFC in-plane MR (Fig. 4e) is negative, non-hysteretic and symmetric with respect to origin. In presence of \( H_{FC} \), the \((MR - H)\) curve becomes asymmetric (Fig. 4d and 4f). For a field applied along the direction of \( H_{FC} \) the in-plane MR is larger than the opposite direction field. Furthermore, the origin of the FC \((MR - H)\) shows a small shift towards the field antiparallel to the \( H_{FC} \). These phenomena are not the cumulative effect of the interfaces because of the shortening of the
top conducting layer. We attribute this asymmetric nature of the field-cooled \((MR - H)\) loop to the uniaxial pinning/biasing of moments observed in the \(FC\) magnetic hysteresis loop.

The \(SRO/SMO\) superlattices exhibit anisotropy with the orientation of magnetic field to the sample in \((MR - H)\) as well as \((M - H)\) measurements. The major contributions to this anisotropy behavior is from the strong anisotropy of the \(SMO\) layers and the additional periodicity of the magnetic layer along the out-of-plane direction of the sample. The \(ZFC\) hysteresis loop measured with \(H > H_P\) for both orientations of \(H\) indicates that the hard axis of \(SMO\) is along [001] direction of \(STO\) (Fig. 1). At a field much below 4 \(tesla\) but larger than the \(H_S\) (0.4 \(tesla\)) of \(SRO\), both \(H_C\) and magnetization (at 1 \(tesla\)) of the superlattice is lower compared to the thin film of \(SRO\) on \(STO\) by \(\sim 95\%\) and \(\sim 32\%\) in-the-film-plane and \(\sim 84\%\) and \(\sim 52\%\) out-of-plane respectively. This suggests that the ideal \(SRO/SMO\) magnetic structure (Fig. 5a) is lost as the sample is cooled down to 10 \(K\) due to the strong anisotropy of \(SMO\) layer, crystallographic and/or magnetic reconstructions and relaxation at the interfaces\(^{12-14}\). We attribute the suppression of \(H_C\) and magnetization to the pinning/biasing of \(SRO\) layer by the \(SMO\) layer due to the strong exchange coupling between them (at a field below \(H_P\)). At \(H < H_P\) the magnetization results partially from the part of the \(SRO\) layer which rotates coherently with the magnetic field. This part of the \(SRO\) layer is identified as the free layer. Using this picture, we can model the ideal structure as a repetition of \(AFM/(pin)/FM(Free)/(pin)\) unit (Fig. 5b). In the \(ZFC\) state, the net magnetization of the pin layer is negligible, i.e., antiferromagnetic orientation of the spin in the pin/bias layers. But in the \(FC\) state, the net magnetization of the pinned/biased layer is lower by the same value as \(M_P^{\parallel}\) for in-plane \(H_{FC}\), while for out-of-plane \(H_{FC}\) it is increasing to a finite value equal to \(M_P^\perp\). Since \(M_P^\perp\) is much larger than the \(\frac{M_P^+ - M_P^-}{2}\) on both \(ZFC\) and \(FC\) states, we conclude that the volume of the free layer is smaller than the volume of pinned/biased layer. Thus, the effective volume of the free layer depends on the \(SMO\) layer thickness, magnetic field and cooling field. Since the FC hysteresis loop of the superlattices shifts along the magnetization axis, this effect in \(SRO/SMO\) superlattices are seen at 0.1 \(tesla\) field low enough not to saturate the \(FM\) magnetization of \(SRO\) \((H_S =\)

\[7\]
0.4 tesla), these processes must occur in the SRO layer. This is in contradiction with the shifts in FC hysteresis loop along the magnetic field axis, in a magnetic field high enough to saturate the FM magnetization - where the irreversible process occurs at the interfaces and in the AFM\textsuperscript{1–5}. In the range of 1 tesla (< $H_P$) magnetic field, the orientation of spin in SMO layer is along the film-plane, for field along [100] and [001] directions of STO. Since the anisotropy axis of SMO layer is fixed, the magnetic field along the easy axis of SMO layer, decreases the angle between the magnetization of SRO layer and the easy axis of SMO layer, while their angular separation increases as the magnetic field is rotated 90°. So, the in-plane $H_{FC}$ may induce bilinear coupling of the spins of SMO and SRO at the interfaces, while the out-of-plane $H_{FC}$ induces biquadratic coupling.

Transport processes in magnetic structures as spin-dependent tunneling\textsuperscript{15} and scattering of spin-polarized carriers\textsuperscript{16} are influenced by the spin-orientations of the pinned/biased layers and free layers of SRO. The FC magnetic field dependent $MR$ in this structure can be explained by using the spin dependent scattering\textsuperscript{17} and the uniaxially pin/bias spin. When the net moment in the pin and free layers are parallel, the in-plane $MR$ is higher and out-of-plane $MR$ is negative; while the antiparallel alignment of the net moments in the bias/pin and free layers results in a lower in-plane $MR$ and a positive out-of-plane $MR$. This correlation between the FC out-of-plane magnetization and $MR$ with the change in magnetic field is sketched in Fig. 5(c).

In summary, the magneto-transport properties of SRO/SMO superlattices deposited on (001) – STO substrates were studied. Our data provide the direct evidence for the manifestation of the uniaxial pinned/biased moments in the FM/AFM superlattice. The pinned/biased moments becomes uniaxial as the superlattice is cooled in presence magnetic field. The in-plane cooling field orients pinned/biased moments antiferromagnetically while they orient ferromagnetically with the out-of-plane cooling field. The electronic transport in these superlattices shows the evidence of spin coupling of the mobile carriers to the interfacial pinned/biased layer. The field dependent in-plane $MR$ is negative while the out-of-plane $MR$ is negative as well as positive. We explain the magnetization and $MR$ by
the spin dependent scattering due to the relative orientation of the net magnetization of
the pinned/biased and free layers. Since progress towards understanding and use of spin-
electronic is growing rapidly, these results should provide fundamentally new advances in
both pure and applied sciences.

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Figures Captions:

Fig. 1: Isothermal (10 K) zero-field-cooled magnetization of the (20 u.c.) SRO/(3 u.c.) SMO superlattice at various fields oriented along the [100] and [001] directions of the substrate, respectively.

Fig.2(a) and (b): Isothermal (10 K) zero-field-cooled and field-cooled magnetization of the (20 u.c.) SRO/(3 u.c.) SMO superlattice at various fields oriented along the [100] and [001] directions of the substrate, respectively.

Fig. 3 Out-of-plane field-cooled biased/pinned moment ($M^\perp_P$) of several superlattices at 10 K.

Fig.4 Current-in-plane zero-field-cooled and field-cooled magnetoresistance $MR (MR = \frac{R(H) - R(H=0)}{R(H)})$ of the (20 u.c.) SRO/(3 u.c.) SMO superlattice at various fields at 10 K. Panels a, b and c show the $-0.1$ tesla FC, ZFC, and $0.1$ tesla FC magnetoresistance respectively at various magnetic fields along the [001] direction of the substrate. Panels d, e and f show the $-0.1$ tesla FC, ZFC, and $0.1$ tesla FC magnetoresistance respectively at various magnetic fields along the [100] direction of the substrate. The arrows indicate the directions of the field sweep with the thicker arrow denoting the commencement.

Fig. 5(a) and (b) Schematic view of the cross section of two interfaces of SRO/SMO multilayer at room temperature and 10 K, respectively. (c) Schematic comparison of FC magnetization and magnetoresistance measured with magnetic field lower than the critical pinning field oriented along the [001] direction of the substrate. In the rectangle box, thick and thin arrows represent the relative orientations of the pinned and free layer net moments, respectively.
Fig. 1 Padhan and Prellier

Magnetization $\mu_B$ per Ru vs Magnetic field (tesla)

- $\bullet$ $\mathbf{H} \parallel [001]$
- $\bigcirc$ $\mathbf{H} \parallel [100]$
Fig. 2 Padhan and Prellier

(a) $n = 3$

$H // [100]$

(b) $n = 3$

$H // [001]$

Magnetization ($\mu_B$ Per Ru)

Magnetic field (tesla)
Fig. 3 Padhan and Prellier

![Graph showing the relationship between SMO thickness (u.c.) and $M_P^\perp$ (μB per Ru). The graph indicates a decrease in $M_P^\perp$ as the SMO thickness increases. The title of the graph is "H // [001]."]
Fig. 4 Padhan and Prellier

(a) $H \parallel [001]$
FC (-0.1 tesla)
n = 3

(b) $H \parallel [001]$
ZFC
n = 3

(c) $H \parallel [001]$
FC (+0.1 tesla)
n = 3

(d) $H \parallel [100]$
FC (-0.1 tesla)
n = 3

(e) $H \parallel [100]$
ZFC
n = 3

(f) $H \parallel [100]$
FC (+0.1 tesla)
n = 3

Magnetoresistance (%) vs. Magnetic field (tesla)
Fig. 5 Padhan and Prellier

(a) IDEAL

(b) ZFC/FC

SMO

SRO

SMO

pinned

free

pinned

(c) $H_{\text{FC}} // [001]$

$M$

$(0, M_p)$

$(0, -M_p)$

$H$

$(0,0)$

MR

H

$(0,0)$