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ABSTRACT

The ability to tune magnetic ordering in complex oxide based correlated antiferromagnetic insulators, due to the coupling between the charge, spin, lattice, and orbital degrees of freedom, opens a vast playground in spintronics. Here, we study a tensile strain induced coexistence of a wide range of magnetic ordering, as established from the temperature dependence of the spin Hall magnetoresistance (SMR) and spin Seebeck effect (SSE) studies and complemented by structural and bulk magnetization measurements. The temperature dependence of the SMR, SSE, and bulk magnetization studies fingerprints the competition between different magnetic domains across the manganite film thickness. Our work demonstrates that strain induced spatial variation of magnetization in such nominal antiferromagnetic manganite, SrMnO_3 , can be tuned by orbital ordering and opens research opportunities in antiferromagnetic spintronics.

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In recent years, spin Hall magnetoresistance (SMR) and Spin Seebeck Effect (SSE), relying on the spin-charge inter-conversion by Spin (Inverse) Hall effect (SHE and ISHE), have emerged as efficient electrical means to probe magnetic ordering in a wide range of magnetic systems such as ferromagnets, ferrimagnets,^{1–4} and antiferromagnets.^{5–9} The versatility of this approach in providing additional information, on the surface magnetization^{10,11} or on magnetic anisotropy,^{9,12} that otherwise need dedicated microscopic techniques or synchrotron based facilities, makes it a useful all-electrical scheme for the study of complex magnetic ordering in thin films of magnetic materials. In this context, strongly correlated rare earth manganites, exhibiting competing magnetic ground states in their phase diagram,¹³ provide an important platform for the study and identification of magnetic ordering, essential for developing antiferromagnetic oxide based spintronics, but is largely unexplored.

SrMnO_3 (SMO) is one of the end members of the $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ family. In bulk, it exhibits three different polymorph phases, namely, cubic, 4H, and 6H hexagonal phases, stabilized at different growth temperatures.¹⁴ All the polymorphs exhibit fully compensated G-type antiferromagnetic-insulating behavior with Neel temperature (T_N) varying between 260 and 278 K.^{14,15} The antiferromagnetic exchange in bulk SMO is governed by the superexchange interaction between the Mn^{4+} ions that preserve the degeneracy of the, e.g., orbitals of Mn^{4+} . Thin films of SMO, on the other hand, can be tailored to exhibit a wide range of magnetic ordering. For example, by tuning

epitaxial strain, the antiferromagnetic (AFM) ordering in thin SMO films can be tuned from G-type to C/A-type.^{16,17} In addition, controlled tuning of different growth conditions can also lead to coexistence of different magnetically ordered phases for this end member of the manganite family, typical among them being a ferromagnetic insulating phase brought about by the structural distortion of the oxygen octahedra and leading to an oxygen deficient $\text{SrMnO}_{3-\delta}$ phase.¹⁸ Moreover, first principles calculations have predicted that tensile or compressive strain can drive a series of magnetic phase transitions that couples with the ferroelectric order in SMO and were corroborated by experimental studies.^{17,19–22}

In this work, we study the temperature dependence of magnetic phases that coexist in thin films of SrMnO_3 (SMO) using complementary techniques, (i) bulk magnetization studies using Superconducting Quantum interference device (SQUID), (ii) Surface sensitive SMR response, and (iii) bulk sensitive SSE response. Two different film types, deposited by adopting different growth strategies and of varying thicknesses (S1 and S2) are studied in this work. The different approaches allow for the study of the correlated interplay between tensile strain and oxygen vacancies across $\text{SrMnO}_3/\text{SrTiO}_3$ interfaces. This work demonstrates the inevitability of coexisting magnetically ordered states, intrinsic to such tensile strained SMO thin films that are nominally antiferromagnetic, in spite of adopting careful growth strategies, as revealed by spin transport studies.

The films, S1 (SMO 2 nm/STO) and S2 (SMO 17 nm/STO), were grown using Pulsed Laser Deposition (PLD) techniques. The deposition conditions and structural characterization by $2\theta - \omega$ scans along (001) direction and reciprocal space mapping (RSM) are discussed in the [supplementary material](#). Whereas the ultrathin sample (S1) does not show x-ray diffraction (XRD) peaks in the $2\theta - \omega$ scan, the analysis of $2\theta - \omega$ scan and reciprocal space mapping (RSM) in S2 yields a tensile strain of 2.9%. Temperature and field-dependent magnetization studies of both SMO films were done using SQUID magnetometer (Quantum Design). [Figure 1\(a\)](#) shows the temperature dependent in-plane hysteresis loops for samples S1 and S2, after subtracting the linear diamagnetic contribution arising from the STO substrates. For sample S1, hysteresis loops with finite coercivity and large saturation magnetization are observed at different temperatures bearing signatures of a predominant ferromagnetic behavior. In bulk, SMO is an antiferromagnetic insulator with a G-type ordering.^{14,23} However, in thin films, the effect of tensile strain, oxygen vacancies, and different growth environment promotes competing magnetic states with ferromagnetic, ferrimagnetic, and antiferromagnetic ordering.^{17–21} In our case, the thinner sample S1 shows a strong ferromagnetic behavior that arises from the mixed valence states in Mn, prevalent in such competing magnetic phases, due to tensile strain and oxygen vacancies.^{17–20} Owing to its ultrathin nature in sample S1, no structural characterization and quantification of the strain were possible.

On the other hand, the thicker sample S2 shows a saturation magnetization of 200 emu/cc at 10 K and requires larger magnetic field to reach saturation as shown in the [supplementary material](#) (Fig. S2). The structural characterization in sample S2 (Fig. S1 in the [supplementary material](#)) reveals a tensile strain of 2.9% and tetragonality ratio similar to previous reports.^{16,20} Besides, both S1 and S2 exhibit distinct magnetic phase transitions ($T_N = 290$ K for S1, $T_N > 350$ K for S2), as shown from the bifurcation points of zero field cooled (ZFC) and field cooled (FC) curves in [Fig. 1\(b\)](#) (blue arrow).

FIG. 1. Temperature dependence of magnetic hysteresis loops of (a) sample S1: SMO (2 nm)/STO (top panel), sample S2: SMO (17 nm)/STO (bottom panel). (b) Temperature dependent magnetization plots of S1 and S2, recorded in both zero-field cooled and field cooled modes ($H_{FC} = 70$ kOe with a running field of 500 Oe). The black arrows near 40 K indicate the magnetic phase transitions due to secondary phases, and the blue arrow indicates the bifurcation of the ZFC and FC curves at 300 K indicating the magnetic phase transition of SMO film in sample S1.

Magnetization vs Temperature (M-T) plots show an additional magnetic ordering around 40 K for both S1 and S2 as seen from the cusp like feature (black arrow) in the ZFC curve similar to previous reports.¹⁸ Secondary phases of MnO_x with mixed oxidation states of Mn (Mn^{2+} , Mn^{3+} , and Mn^{4+}) that could segregate during the deposition of $ReMnO_3$ are well documented in the literature. The competing magnetic phases are reported to be ferrimagnetic Mn_3O_4 ^{24–27} or $\gamma - Mn_2O_3$ ²⁸ with transition temperature around 40–46 K and antiferromagnetic $MnO/x - Mn_2O_3$ with transition temperatures around 80–100 K.²⁹ Further, computationally both the cubic and hexagonal structures are shown to exhibit several magnetically ordered structures, depending on the interactions between the neighboring B site cations and with an AF ground state.¹⁴

In order to gain more insights into the temperature dependence of magnetic ordering in SMO thin films, we have performed simultaneous measurements of spin Hall magnetoresistance (SMR) and Spin Seebeck Effect (SSE) using spin Hall effect based spin transport. UV-lithography followed by electron beam evaporation were used to pattern and fabricate a 7 nm thick-Pt Hall bar device (dimensions: width $w = 50 \mu m$, length $l = 3000 \mu m$) on SMO thin films as shown in [Fig. 3\(a\)](#). Simultaneous angle dependent spin Hall magnetoresistance (SMR) and Spin Seebeck effect (SSE) measurements were performed on a physical property measurement (PPMS) system (Quantum Design) in the temperature range of 5–300 K and the magnetic field range of 0–7 T to get insights about the magnetic order of SMO thin films. For field-dependent SMR and SSE measurements, a sinusoidal alternating current of amplitude $I_0 = 1$ mA ($J_0 = 2.8 \times 10^9$ A/m²) and frequency (ω) = 7.77 Hz were driven across the Pt Hall bar in the y-direction (see the [supplementary material](#)). Four Lock-in amplifiers were simultaneously used to measure the first harmonic ($V_{yy}^{1\omega}$ and $V_{xy}^{1\omega}$ at phase = 0°) and second harmonic ($V_{yy}^{2\omega}$ and $V_{xy}^{2\omega}$ at phase = -90°) voltages with a time constant of 3 s by rotating the sample along the x-y (α), x-z (β), and y-z (γ) planes, respectively.

[Figure 2](#) shows the angular dependent magnetoresistance (ADMR) responses for both S1 and S2, along the plane of transport (x-y plane). The details of the ADMR responses in the longitudinal contacts are discussed in the [supplementary material](#). For simplicity, we focus on the transverse measurements, as any background resistances are minimized in the x-y plane owing to its measurement geometry. On application of the charge current J_{ac} (J_e marked in [Fig. 2](#)) along the Pt strip (y-direction), a spin current (J_s) in the Pt layer due to spin Hall effect is induced (SHE)³⁰ that exerts a torque on the magnetization (M) vector of the underlying SMO layer. Depending on the relative orientation of M in SMO and μ the spin accumulation, reflection or absorption of spin current occurs at the Pt/FM interface, which in turn causes the modulation of spin Hall magnetoresistance (SMR)^{30,31} in an applied magnetic field.

A typical transverse SMR contribution arising at the interface of Pt/SMO is described as follows:³¹

$$\frac{\Delta R_{xy}}{R_o} = \frac{\Delta \rho_{xy}}{\rho_o} \frac{w}{l} \langle m_x m_y \rangle \propto \sin(2\alpha), \quad (1)$$

where ΔR_{xy} is the change in the SMR response measured in the longitudinal and transverse configurations, respectively. $\Delta R_{xy} = R_{yy}(\alpha = 45^\circ) - R_{yy}(\alpha = 135^\circ)$. The transverse SMR variations in the x-y plane (α -direction) shown for S1 and S2 samples ([Fig. 2](#)) with a 7 T field, follows a $\sin(2\alpha)$ variation, and consistent with Eq. (1). The spin resistance

FIG. 2. The angular dependence of transverse SMR signals shown for three representative temperatures obtained at an applied magnetic field (B) = 7 T along α rotation plane for both S1 and S2 samples. Here, J_e and μ represent the corresponding electron flow and spin accumulation directions, respectively.

displays a maximum (absorbed) and minimum (reflected) at $\alpha = 45^\circ$ and $\alpha = 135^\circ$ ($B \perp J_e$) configurations, respectively. This is similar to SMR signals observed on Pt/YIG and other ferrimagnetic spinel systems, commonly attributed to positive SMR,³² and occurs when the applied magnetic field exceeds the anisotropy fields in Pt/FM heterostructures. In addition, the transverse SMR signals display a $\sin(2\alpha)$ behavior according to Eq. (1) [Figs. 2(a) and 2(b)]. The different contributions to the magnetoresponse, which can simultaneously occur across Pt/insulator hybrid systems, are discussed in the [supplementary material](#). By systematic cross-check and analysis, we ascribe the observed signals, shown in Fig. 2, to originate due to SMR.

We further used Spin Seebeck studies to understand the bulk magnetic ordering in samples S1 and S2. In Figs. 3(a) and 3(b), the SSE signals are detected in the longitudinal and transverse configurations, respectively, at 5 K by sweeping the in-plane 7 T field (α -scan) for sample S2. These responses are detected as second harmonic resistances ($R_{yy}^{2\omega}$ and $R_{xy}^{2\omega}$) in the lock-in amplifier. An applied current bias ($I_0 = 1.5$ mA in this case) induces a thermal gradient (∇T) creating a magnon spin accumulation (μ_m) in the bulk SMO layer, which in turn diffuses a spin current (I_s) in the Pt layer, that is detected as charge resistance due to inverse spin Hall effect (ISHE).^{33,34} From Figs. 3(a) and 3(b), we observe a sinusoidal variation of the SSE signal. The SSE signals show a peak at 0° and 180° for the longitudinal contacts [Fig. 3(a)] when the applied magnetic field is parallel to the magnon spin accumulation, leading to a detectable charge resistance in Pt due to ISHE. Similar signals are observed for sample S1, which are shown in the [supplementary material](#). These obtained signals scale quadratically with current bias (see the [supplementary material](#)) indicating a current induced SSE.²⁻⁴ On the other hand, we observe a phase shift of 90° in the observed response for the transverse configuration [Fig. 3(b)].

In order to further investigate the observed SSE response, we performed angular dependent measurements (see the [supplementary material](#)) in the out-of-plane direction to rule out the contribution of thermal response arising from Anomalous Nernst Effect in Pt, in close proximity with a magnetic insulator.^{35,36} We have performed field dependent measurements of the SSE signals, at angles, where the SSE response showed a maximum. This is shown in the inset of Figs. 3(a) and 3(b) for both longitudinal and transverse configurations. The field dependent SSE measurements display a non-linear dependence, attributed to either a weak-ferromagnetic behavior³⁷ or to an easy-plane antiferromagnetic ordering,^{7,8} as inferred from the absence of any hysteretic behavior in the trace and retrace of the magnetic field, as reported for other ferri/ferromagnetic materials.^{2,4}

We now discuss our findings, in light of the different probing techniques used in this work, to understand the role of the coupling between the coexisting magnetic domains in the tensile strained S1 and S2 films. From the bulk magnetization studies, we find the coexistence of different magnetically ordered phases and their coupling with temperature as shown in Fig. 2. We now take into account the temperature dependent SMR and SSE amplitudes for samples S1 and S2 at an applied in-plane magnetic field of 7 T, as shown in Figs. 4(a) and 4(b). S1 shows a temperature dependence atypical of SMR response in Pt/YIG hybrid systems;^{38,39} and the variation of S2 is unique. In S2, it saturates beyond a critical temperature, T_{crit} but for $T < T_{crit}$ a steep decrease is observed. This temperature range corresponds to the temperature at which the ZFC curves, in Fig. 2(b), exhibit a cusp in SQUID studies, and is ascribed to the presence of antiferromagnetic ordering in the bulk. As discussed earlier, our tensile strained films exhibit ferromagnetic ordering, evinced in the M-H loops in Fig. 2(a) due to a possible $\text{SrMnO}_{3-\delta}$ phase (low oxygen pressure during growth). To understand the temperature variation of the SMR response in S2, we recall that the additional annealing step induces a competing magnetically frustrated ordering. Additionally, the absence of any hysteresis in the field dependent SSE measurements shown for sample S2, [insets of Figs. 3(a) and 3(b)] indicate the presence of a bulk antiferromagnetic ground state, where the antiferromagnetic sublattices gradually cant in the direction of the magnetic field.⁸ Moreover, the SSE signals, which indicate the thermal excitation of the magnons in SMO films, decrease with increasing temperature and are not detected beyond 40 K as shown in Fig. 4(b). From a collective inference drawn from the bulk magnetization, SMR, and SSE studies, we consider the thin film to comprise of competing magnetic ordering at the surface (S-SMO) and in the bulk (B-SMO), whose strength is tuned by temperature. Thus, below $T < T_{crit}$, enhancement in the antiferromagnetic ordering of the magnetic domains near the surface leads to a reduction in the SMR response similar to that observed in Pt/YIG hybrids with surface treatment in YIG³⁸ or by insertion of antiferromagnetic NiO layer.⁴⁰ Above $T > T_{crit}$, surface ferromagnetic ordering dominates the SMR response. Additionally, no SSE signals are detected above T_{crit} for both S1 and S2, indicating a lack of exchange stiffness and long range magnetic ordering above T_{crit} .

As shown in Fig. 4(c), below $T < T_{crit}$ and for a field of 7 T, the enhanced antiferromagnetic ordering across the S-SMO and B-SMO layers facilitates magnon excitation and propagation that is detected as a finite spin current at the Pt electrodes. However for $T > T_{crit}$, the coupling between the S- and B-SMO layers is weakened, hindering efficient magnon spin transport, leading to a vanishing SSE signal.

FIG. 3. (a) and (b) Angular dependent SSE signals measured in the longitudinal and transverse SSE configurations at 5 K at 7 T in-plane field (α scan) for a current bias of $I_0 = 1.2$ mA for sample S2. The insets show the field dependent SSE signals acquired at different angles.

FIG. 4. (a) and (b) Temperature dependence of SMR (top panel) and SSE (bottom panel) signals at current bias of $I_0 = 1$ mA and 1.5 mA, respectively, and at an applied magnetic field of 7 T. (c) The schematic on the right hand side shows the field dependent exchange coupled ordering of the magnetic moments in the surface (S-SMO) and bulk (B-SMO) layers of the SMO films for the temperature ranges, $T < T_{crit}$ and $T > T_{crit}$.

Overall, our findings reiterate (i) the presence of a magnetic ordering at the surface of the SMO films that is responsible for the observed SSE signals¹¹ and (ii) the surface sensitivity of the SMR in detecting competing magnetic ordering in such tensile strained SMO thin films. Moreover, as reported earlier, enhancement of SSE responses at low temperatures in Pt/YIG,⁴¹ Pt/Cr₂O₃⁸ hybrids, indicate the role of the magnon-phonon interaction due to the thermal induced phonon drag. This is demonstrated by infrared and Raman studies (below T_N) for bulk SMO.⁴² In our case, the temperature dependence of magnetic ordering, evident from the M–T and SSE studies hint at magnon-phonon interaction as a possible reason for the decrease in SSE signals with increasing temperature. However, further studies are needed to establish this. In order to further ascertain that the SSE signals originate in Pt due to SMO, control experiments were performed on Pt/SrTiO₃, where no such modulations are observed in the second harmonic signal with an applied magnetic field of 7 T. This is shown in Fig. S12 (see the [supplementary material](#)).

Our work represents a first comprehensive study of the temperature and field dependence of the magnetic ordering in SMO thin films utilizing complementary all-electrical transport. Furthermore, our study also highlights the predominant contribution of the surface magnetic ordering from the temperature dependence of the SMR and SSE studies, promoted by a spatially varying strain across the films and the choice of growth parameters. The ability to tune the magnetic ordering with strain and oxygen vacancies in thin films of SrMnO₃ is an important step toward using such materials for magnon spin transport studies and their control by electric field in such multiferroics.

See the [supplementary material](#) for discussion on the growth and structural characterization of the SrMnO₃ thin films, magnetization and magnetic phase transition, signals due to Spin Hall Magnetoresistance and Magnetoresistance in Pt and angular Spin Seebeck responses in sample S1.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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