

Epitaxial Growth of Double Perovskite Sr_2CrWO_6 Thin Films

J.A. Horn,^{1,*} K.Y. Meng,² and F.Y. Yang²

¹*Northern Michigan University, Marquette, MI 49855 USA*

²*The Ohio State University, Columbus, OH 43210 USA*

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Here, I report on the preparation and characterization of epitaxial thin films of the double-perovskite Sr_2CrWO_6 and $\text{Sr}_2\text{CrReO}_6$ by Ultrahigh Vacuum Magnetron Sputtering. These films were grown on SrTiO_3 and LSAT substrates as an efficient way to tune the growth conditions of these film. In the case of Sr_2CrWO_6 , a sputtering powder was synthesized for use as sputtering target. Purity of target powder was confirmed via Powder X-Ray Diffraction and phase purity of epitaxially grown thin films were confirmed by Thin Film X-Ray Diffraction.

INTRODUCTION AND BACKGROUND

This project is part of a larger-scale ongoing investigation of the possible room temperature magnetic Skyrmion behavior in Double perovskite thin films. Skyrmions are topological magnetic textures that show promise for future electronics and spintronics applications if they can be harnessed and manipulated [1, 2]. At the intersection of two layers of different materials, interfacial DM interactions can give rise to skyrmion activity. This happens because breaking inversion symmetry becomes more energetically favorable as shown by the cross-product in the DMI Hamiltonian:

$$H = -J(S_i * S_j) + D_{ij} * (S_i \times S_j) \quad (1)$$

This project aims to address a modern problem regarding skyrmions: the need for a host for room temperature skyrmion activity. This comes as a concern in the interest and developments of spintronic technology, as skyrmions offer a stable domain for spin storage. However, this technology would not be very useful without a host for room temperature skyrmions. We expect that at interfaces of known double perovskite materials that interfacial DM interaction will occur, and given their characteristic high T_c , they are a likely candidate for room temperature skyrmion activity.

Double-perovskites of the form $\text{A}_2\text{BB}'\text{O}_6$ are interesting materials both due to their rich physics and their interesting properties with respect to applications in spin electronics. They are half metallic, i.e., have a spin-dependent bandgap that leads to high degree of spin-polarization at the Fermi Level [3], and some compounds have a high Curie temperature (T_C), e.g., $T_C = 635$ K in $\text{Sr}_2\text{CrReO}_6$ [4]. Large low field magnetoresistance effects are found in $\text{Sr}_2\text{FeMoO}_6$ [3, 5], $\text{Sr}_2\text{FeReO}_6$ [6], and Sr_2CrWO_6 [7]. Thin films of the well studied system $\text{Sr}_2\text{FeMoO}_6$ have been fabricated by pulsed-laser deposition (PLD) at relatively high temperatures of about 900 °C [8]. However, the epitaxial growth of these films have been found to be complicated and difficult to control. It has also been noted that there is an association between high structural quality of films grown on SrTiO_3

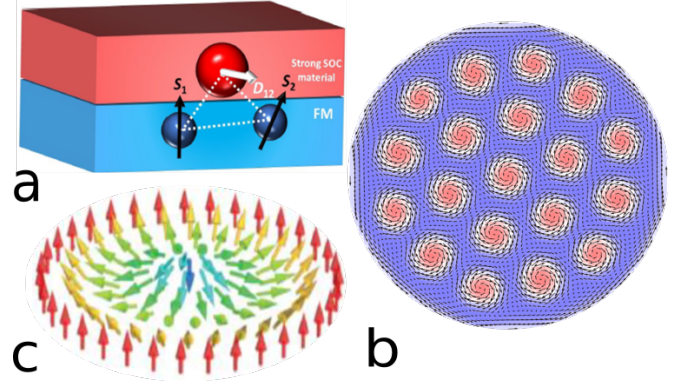


FIG. 1: Illustrations to depict skyrmion and DM interaction outline a) interacting magnetic moments at the interface of a ferromagnetic layer resulting in an energetically favorable configuration according to the second term in equation 1 (b) vector plot of magnetic moments of a single Neel-type (moments rotate in-out of plane) skyrmion and (c) a skyrmion lattice of Bloch-type (moments rotate in plane) skyrmion.

and semiconducting behavior [9, 10].

In this study, we report on the epitaxial growth of Sr_2CrWO_6 . Due to the good lattice match, epitaxial films of this material can be grown on SrTiO_3 substrates in a molecular layer-by-layer growth mode resulting in high crystalline quality.

EXPERIMENTAL DETAILS

Epitaxial Sr_2CrWO_6 and $\text{Sr}_2\text{CrReO}_6$ films are grown on SrTiO_3 (001) and LSMO (001) substrates in an ultrahigh vacuum magnetron sputtering system (depicted in Fig.) with a base pressure on the order of 10^{-7} Torr.

Stoichiometric Sr_2CrWO_6 targets were synthesized via solid state chemical reaction by sintering in forming gas (5% H_2 in N_2) using SrCO_3 , Cr_2O_3 , and WO_3 as reagent powders. Xray Diffraction was used to verify that the Sr_2CrWO_6 targets are pure Double Perovskite phase using a Bruker D8 Advance diffractometer in a Bragg-Brentano geometry. Atomic Force Microscopy (AFM) was used to verify the surface quality of the epitaxial

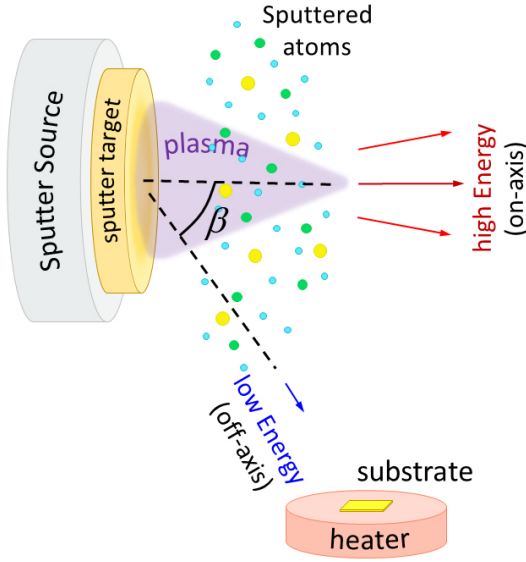


FIG. 2: Magnetron sputtering uses high voltage to produce an Argon-ion plasma to strip off atoms from a target material and deposit them onto a substrate surface. Magnetic fields are used to steer the ions in the plasma for a controlled deposition. Off-axis geometry is used to avoid bombardment of atoms with the surface of the film which would potentially hinder the desired kinetics of epitaxial growth.

thin films.

Sr_2CrWO_6 films were deposited using direct-current (DC) magnetron sputtering with a constant current of 30 mA in the presence of low O_2 partial pressure and total pressure of $p \approx 12$ mTorr. Oxygen concentration was varied between 0.2% and 5% in Ar to find the optimal inert/oxidizing environment for epitaxial growth. The substrate temperatures were varied between 650°C and 780°C .

$\text{Sr}_2\text{CrReO}_6$ films were deposited using Radio-Frequency (RF) magnetron sputtering constant power of 50 W in the presence of low O_2 partial pressure and total pressure of $p \approx 12.5$ mTorr. Oxygen concentration was varied between 0.2% and 1.0% in Ar to find the optimal inert/oxidizing environment for epitaxial growth. The substrate temperatures were varied between 650°C and 700°C .

Standard 90° off-axis geometry was used for with the substrates positioned at a horizontal distance of 6.5 cm from the target and 9.0 cm below the center of the target for all films grown [11]. Conventional on-axis sputtering geometry was also used to deposit Sr_2CrWO_6 films for comparison.

RESULTS AND DISCUSSION

Unfortunately, after working towards a Sr_2CrWO_6 powder target with high purity, we were unable to grow

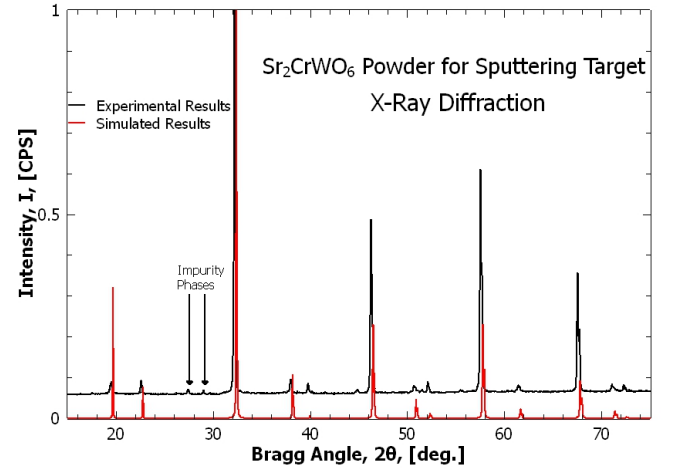


FIG. 3: Results from powder X-Ray Diffraction on the final powder sample of Sr_2CrWO_6 show small impurity peaks from presence of oxidized phases: SrCrO_3 and SrWO_3 . Simulated results show clearly that these two impurity phase peaks do not belong to any Sr_2CrWO_6 phases. Simulation was done using VESTA structural analysis software.

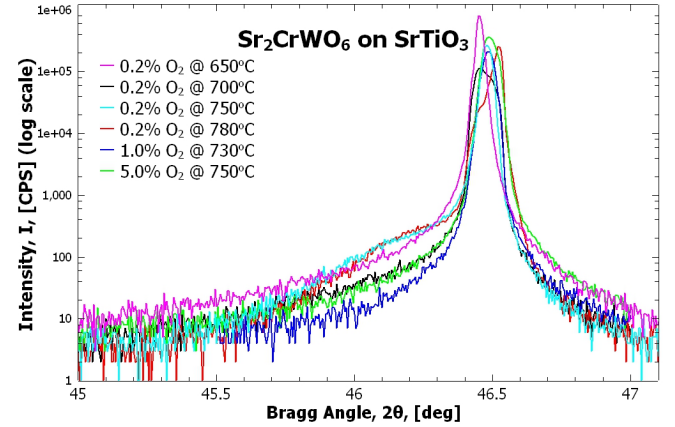


FIG. 4: Results from single crystal X-Ray Diffraction on Sr_2CrWO_6 deposited on SrTiO_3 substrate suggests that Sr_2CrWO_6 has not been able to deposit epitaxially at reasonable parameters (see e.g. [7]).

epitaxial Sr_2CrWO_6 films. Lack of distinct peak in X-Ray diffraction pattern near the SrTiO_3 substrate peak indicates that we do not have a good epitaxial thin film for any of the films grown at reasonable parameters. Reasonable parameters come from past successful growth of epitaxial Sr_2CrWO_6 films on SrTiO_3 substrates (unpublished results). Temperature and oxygen partial pressure were tuned around those used in the past results but as shown, none of these parameters were successful. These lack of epitaxial films is likely due to impurities in the target powder, as verified by the powder X-Ray diffraction results. On the the hand, we were able to produce epitaxial films of $\text{Sr}_2\text{CrReO}_6$ on both SrTiO_3 and LSAT substrates. Results from single crystal X-Ray

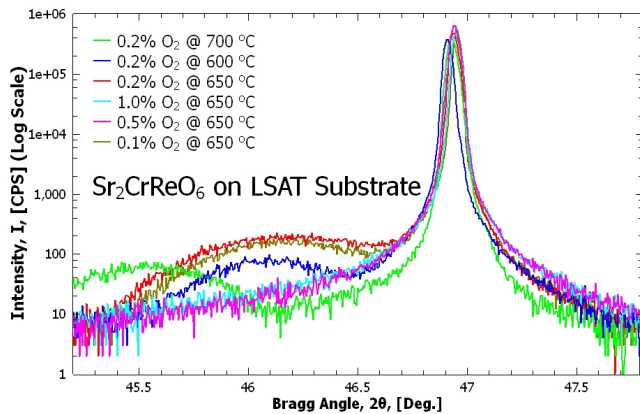


FIG. 5: Results from single crystal X-Ray Diffraction on $\text{Sr}_2\text{CrReO}_6$ deposited on LSAT (001) substrate suggests that $\text{Sr}_2\text{CrReO}_6$ will deposit epitaxially in highest quality (of the parameters varied) at 650 °C and low oxygen partial pressure (0.2% O_2 in inert mixture).

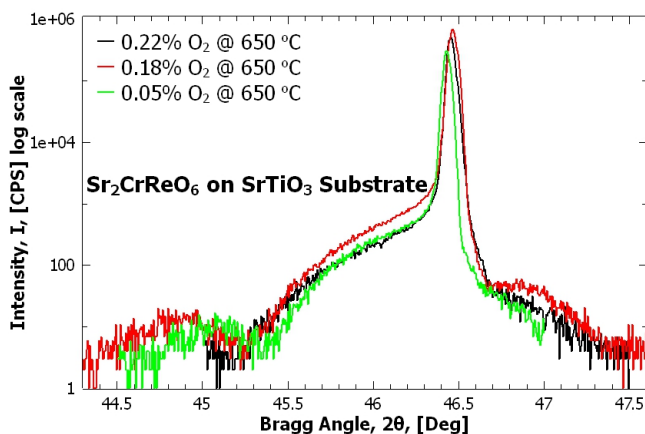


FIG. 6: Results from single crystal X-Ray Diffraction on $\text{Sr}_2\text{CrReO}_6$ deposited on SrTiO_3 (001) substrate suggests that $\text{Sr}_2\text{CrReO}_6$ epitaxial growth on this substrate is not very oxygen sensitive in low oxygen partial pressures ($0.05\% \leq \text{O}_2 \leq 0.22\%$) at 650°C.

diffraction show clear evidence of epitaxial $\text{Sr}_2\text{CrReO}_6$ from the presence of a $\text{Sr}_2\text{CrReO}_6$ film peak next to the SrTiO_3 substrate peak, accompanied by Laue oscillations

which is an indication of good epitaxial growth quality.

CONCLUSION

As of right now in this project: We know that X-ray diffraction revealed impurities in Sr_2CrWO_6 sputtering target which may have hindered the growth of these films. However, $\text{Sr}_2\text{CrReO}_6$ thin films have been able to be grown in good quality as verified by single crystal X-Ray diffraction. Moving forward, next logical steps would include growing a strong spin-orbit coupling material on $\text{Sr}_2\text{CrReO}_6$ to induce interfacial skyrmion activity. Work to develop Sr_2CrWO_6 powder in high purity is currently under way and will hopefully give rise to promising results in the future.

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* Corresponding author: jahorn@nmu.edu

- [1] A. Fert et al., Nature Reviews Materials **2** 17031 (2017)
- [2] A. Rosch et al., Nature Nanotechnology **12** 103-104 (2017)
- [3] K.-I. Kobayashi et al., Nature (London) **395**, 677 (1998).
- [4] H. Kato et al., Appl. Phys. Lett. **81**, 328 (2002).
- [5] M. Garca-Hernandez et al., Phys. Rev. Lett. **86**, 2443 (2001).
- [6] K.I. Kobayashi et al., Phys. Rev. B **59**, 11159 (1999).
- [7] J. B. Philipp et al., Appl. Phys. Lett. **79**, 3654 (2001).
- [8] T. Manako et al., Appl. Phys. Lett. **74**, 2215 (1999).
- [9] H. Asano et al., Appl. Phys. Lett. **74**, 3696 (1999).
- [10] W. Westerburg et al, Phys. Rev. B **62**, R767 (2000).
- [11] C. B. Eom et al., Appl. Phys. Lett. **55**, 595 (1989).