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Authors
Grutter, A.
Wong, F.
Arenholz, E.
et al.

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Controlling magnetism in epitaxial SrRuO$_3$ thin films through strain orientation

A. Grutter$^{1,2}$; F. Wong$^1$; E. Arenholz$^3$; M. Liberati$^2$; A. Vailionis$^4$; Y. Suzuki$^{1,2}$

1. Materials Science and Engineering, UC Berkeley, Berkeley, CA, USA.
2. Materials Science Division, Lawrence Berkeley National Laboratory, Berkeley, CA, USA.
3. Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA, USA.
4. Geballe Laboratory for Advanced Materials, Stanford University, Stanford, CA, USA.

Oxide heteroepitaxy has enabled us to isolate ground states of transition-metal oxide thin films not accessible in their bulk counterparts. Through thin film epitaxy, we have been able to enhance the magnetization of the itinerant 4d ferromagnet SrRuO$_3$. By varying the type and orientation of the substrate as well as film thickness, we find that SrRuO$_3$ films exhibit saturation magnetization values of over 3 $\mu_B$ per formula unit, exceeding the observed bulk values of 1.2-1.6$\mu_B$ and the maximum 2 $\mu_B$ spin-only moment value of Ru$^{5+}$ in the low-spin configuration.[1-4] Films were grown on (001) and (110) oriented SrTiO$_3$, (LaAlO$_3$)$_{0.3}$(SrTiO$_3$)$_{0.7}$ (LSAT), and LaAlO$_3$ as well as (111) oriented SrTiO$_3$ using pulsed laser deposition (PLD). These substrate choices introduce compressive strain with lattice mismatches of 0.64% in SrTiO$_3$, 1.53% in LSAT and 3.56% in LaAlO$_3$. The different substrate orientations create tetragonal, monoclinic, and trigonal distortions in the SrRuO$_3$ unit cell, respectively.

We have performed structural, magnetic and transport characterization, including electron transport, X-ray diffraction, X-ray absorption (XA) spectroscopy, X-ray magnetic circular dichroism (XMCD), and Rutherford backscattering spectroscopy (RBS). Transport measurements showed resistance vs. temperature curves with residual resistivity ratios similar to those of other SrRuO$_3$ films deposited by PLD.[5-6] The XRD spectra showed only SrRuO$_3$ film peaks with the same orientation as the underlying substrates. XA spectroscopy was used to rule out the possible presence of magnetic impurities while XMCD confirmed that the magnetism originated in Ru ions in an octahedral environment.[7] RBS measurements confirmed stoichiometry and determined the thickness of the SrRuO$_3$ films.

Using SQUID magnetometry, we found that saturated magnetic moments of SrRuO$_3$ films grown on (110) and (111) substrates are consistently higher than those on (001) substrates, c.f. Fig. 1. The enhancement of (110)-oriented films is consistent in thicknesses from 15-120nm, c.f. Fig. 2, while (111)-oriented samples exhibit a thickness dependent enhancement.

X-ray diffraction revealed that the strain state of the films is crucial in determining the magnetic properties. Using reciprocal space maps, we were able to probe the film strain and show a correlation between lattice deformation and saturated magnetization. While most SrRuO$_3$ films on (001), (110), and (111) SrTiO$_3$ as well as (001) LSAT were fully strained to thicknesses of 120nm, films on (110) LSAT as well as (001) and (110) LaAlO$_3$ were relaxed to bulk lattice parameters. In all cases, unstrained films were found to have lower saturated moments than their strained counterparts. This trend holds even in the case of (111)-oriented films on SrTiO$_3$, where unstrained films demonstrate significantly lower moments than strained films of comparable thickness.

Magnetic transition temperatures followed similar trends, with the $T_c$'s of (111) and (110) samples (> 145K) greater than those of (001) films ($T_c$ ~ 130K). The saturation magnetization values decreased with increasing thickness and approached bulk values for films on the order of 120nm. SrRuO$_3$ films grown on LaAlO$_3$ and LSAT substrates also displayed lower saturation magnetization values.

Our results suggest that the significant factor in this magnetic enhancement is, in fact, the strain state of the samples. This demonstrates not only the importance of coherent epitaxial strain, but also the effects of the symmetry of lattice deformation for different substrate orientations on the magnetism in SrRuO$_3$. 
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Figure 1 - Hysteresis loops of 24nm thick SrRuO$_3$ films deposited on (001), (110), and (111) oriented SrTiO$_3$

Figure 2 - Saturated magnetic moment ($M_s$) vs. thickness for SrRuO$_3$ films grown on (001) and (110) oriented SrTiO$_3$. $M_s$ is largely insensitive to thickness in the range of 20-80nm.