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Controlling magnetism in epitaxial SrRuO₃ thin films through strain orientation

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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 SrRuO₃. By varying the type and orientation of the substrate as well as film thickness, we find that SrRuO₃ films exhibit saturation magnetization values of over 3 μ_B per formula unit, exceeding the observed bulk values of 1.2-1.6 μ_B and the maximum 2 μ_B spin-only moment value of Ru⁴⁺ in the low-spin configuration.[1-4]

Films were grown on (001) and (110) oriented SrTiO₃, (LaAlO₃)_{0.3}(SrTiO₃)_{0.7} (LSAT), and LaAlO₃ as well as (111) oriented SrTiO₃ using pulsed laser deposition (PLD). These substrate choices introduce compressive strain with lattice mismatches of 0.64% in SrTiO₃, 1.53% in LSAT and 3.56% in LaAlO₃. The different substrate orientations create tetragonal, monoclinic, and trigonal distortions in the SrRuO₃ 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 SrRuO₃ films deposited by PLD.[5-6] The XRD spectra showed only SrRuO₃ 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 SrRuO₃ films.

Using SQUID magnetometry, we found that saturated magnetic moments of SrRuO₃ 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 SrRuO₃ films on (001), (110), and (111) SrTiO₃ as well as (001) LSAT were fully strained to thicknesses of 120nm, films on (110) LSAT as well as (001) and (110) LaAlO₃ 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 SrTiO₃, 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. SrRuO₃ films grown on LaAlO₃ 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 SrRuO₃.

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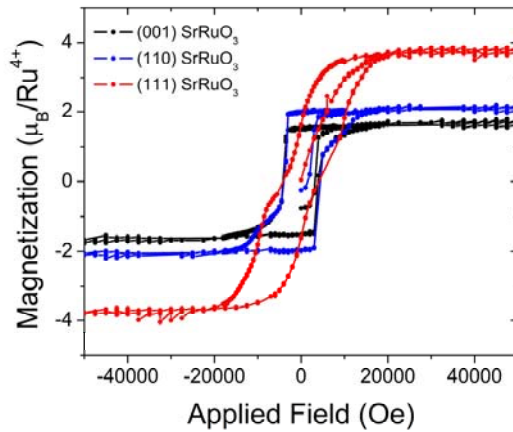


Figure 1 - Hysteresis loops of 24nm thick SrRuO₃ films deposited on (001), (110), and (111) oriented SrTiO₃

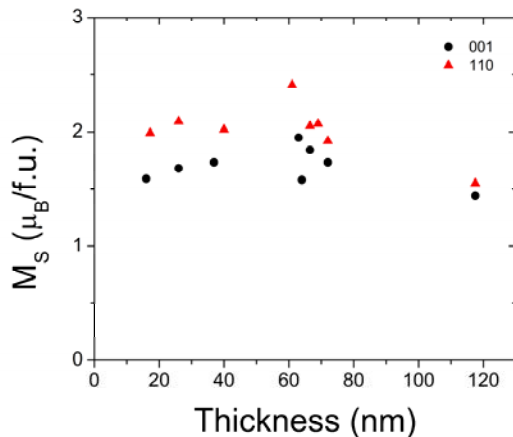


Figure 2 - Saturated magnetic moment (M_S) vs. thickness for SrRuO₃ films grown on (001) and (110) oriented SrTiO₃. M_S is largely insensitive to thickness in the range of 20-80nm.