Nanoskins on Layered Manganites

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Introduction

What happens to ferromagnetism at the surface of a complex magnetic oxide? With the competition between charge, spin, and orbital degrees of freedom, it is not surprising that the surface behavior may be profoundly different than that of the bulk. Using a powerful combination of two surface probes-tunneling and polarized x-ray interactions-this work reveals the intrinsic electronic and magnetic surface states of a layered manganite, $La_{2-2x}Sr_{1+2x}Mn_2O_7$, that is ferromagnetic and conducting in the bulk [1]. The results of these combined probes present clear evidence for an intrinsic insulating non-ferromagnetic surface layer atop adjacent subsurface layers that display the full bulk spin polarization. This abrupt intrinsic magnetic interface is attributed to the weak inter-bilayer coupling native to these quasi-two-dimensional materials. The stratified magnetic structure persists up to the Curie temperature, T_C, and is thus in marked contrast to the non-layered manganite system ($La_{1-x}Sr_xMnO_3$), whose spin polarization in the near surface region degrades at temperatures significantly below T_C

Methods and Materials

To study this problem in detail we chose a well-defined single-crystal system, which cleaves readily, providing an atomically flat surface with terraces on the order of 1 μm. To determine the electronic and magnetic states at the surface, we used a combination of two surface probes-tunneling and polarized x-ray interactions-to study the intrinsic electronic and magnetic surface. These probes present clear evidence for an intrinsic insulating non-ferromagnetic surface layer atop adjacent ferromagnetic subsurface layers.

An element resolved polarized x-ray probe is an ideal tool because the magnetism is localized on the Mn site. Using surface senstitive x-ray magnetic circular dichroism measured via total electron yield at the Mn L_3 edge, we can determine the average magnetic properties measured over the escape depth of the electron (1-2 nm). This signal shows a reduced value compared to that expected for Mn possessing a large (3-4 μ_B) magnetic moment. Such a measurement is an average though and does not provide any spatial information, which might give insight into the mechanism behind the reduction. For this we turn to magnetic scattering which is ideal for determining the magnetic profile in the surface region. By utilizing the large magnetic contrast at the Mn L edge, angle and energy dependent x-ray resonant magnetic scattering (XRMS) can provide a direct means to determine the magnetic profile (see Fig. 1). The clear change in sign with angle for a bulk crystal can only result from the magnetic and electronic surfaces being out of coincidence.

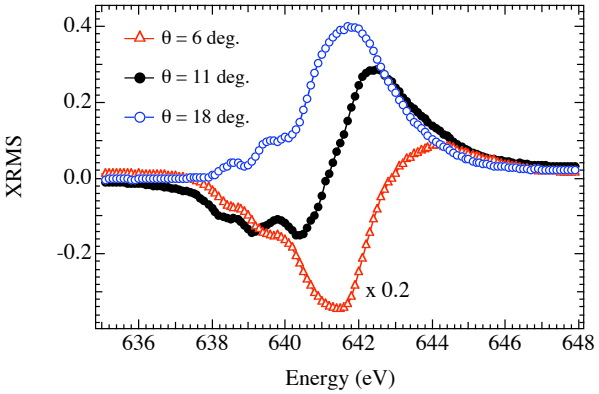

Fig. 1. Angle dependent x-ray resonant magnetic scattering (XRMS) showing clearly the sign change with angle, which results from the chemical and magnetic interfaces being out of registry.

Results

To quantify the details of the magnetic profile requires modeling the XRMS data. Since x-rays at the Mn L edge have a wavelength of ~2 nm, the scattering from the structure is well described by the magneto-optics formalism. By assembling a layered structure matching the unit cell, we used the dielectric tensor (determined from the polarization dependent absorption) to model the scattering as a function of the magnetic profile near the surface. A comparison to simulations of magnetic profiles ranging from 0 to 2 non-magnetic bilayers (see Fig. 2) clearly demonstrates that a single bilayer alone is non-ferromagnetic. From this we estimate that the spin polarization in the second bilayer at 35 K is the same as the bulk value, with an uncertainty < 20%. The XRMS data thus reveal the presence of an extremely thin (~1 nm) non-ferromagnetic layer below which subsequent bilayers are fully magnetized.

While this layer shows no signs of ferromagnetism, competing energies can result in an anti-ferromagnetic state. Subsequent measurements in magnetic fields up to 7 T have shown a linear magnetic response from the surface bilayer, which is consistent with canting of spins in an anti-ferromagnet. From this we conclude the surface bilayer is not ordered ferromagnetically like the bulk, but has stabilized in an anti-ferromagnetic state.

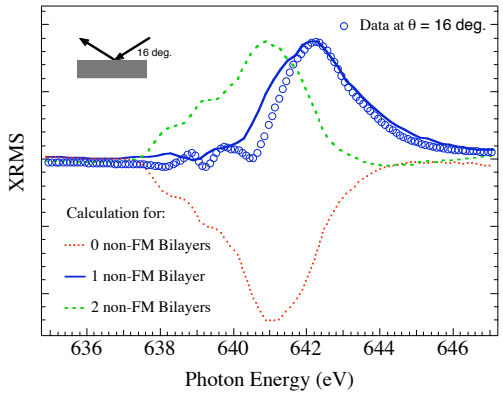

Fig. 2. Calculated XRMS energy dependence at θ = 16 degrees for the case of 0, 1, and 2 non-ferromagnetic (non-FM) bilayers. From the profound lineshape change we can pinpoint that the data are consistent only with the case of a single non-FM bilayer.

Since ferromagnetism in this system is mediated by the double-exchange mechanism, there is a symbiotic relationship between conductivity and ferromagnetism. Lack of ferromagnetism in this model would imply an insulating groundstate. To determine if the surface is insulating we performed point contact tunneling measurements at low temperature. The results are well described by a high-quality square tunnel barrier with a thickness in agreement with the non-ferromagnetic surface bilayer.

An exciting aspect of this result is that nature has provided us with a self-assembled tunnel barrier on top of a high-spin polarization ferromagnet. Due to the high quality of the single crystal, this barrier is nearly perfect.

Acknowledgements

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[1] J.W. Freeland, K.E. Gray, L. Ozyuzer, P. Berghuis, Elvira Badica, J. Kavich, H. Zheng, and J.F. Mitchell, Nat. Mater. **4**, 62 (2005).