Study of Strain and Temperature Induced Changes in the Band Structure of Ultra-thin Mangainte Films

Trevor A. Tyson[1], Qing Qian[1], and Yong-Qiang Cai (蔡永強)[2]

New Jersey Institute of Technology, Newark, New Jersey, USA
National Synchrotron Radiation Research Center, Hsinchu, Taiwan

The self-doped system, La_xMnO_3, is a simple manganite with properties analogous to the classic $La_{1-x}Ca_xMnO_3$ system but with fewer chemical components. Bulk magnetization measurements conducted by SQUID magnetrometry on 6, 30, and 160 nm thick films showed a reduction of T_c and magnetic saturation moment with a reduction in thickness. Films of the entire thickness range exhibit metal-like conductivity at low temperature.

Epitaxial $La_{1-x}MnO_{3-\delta}$ ($x \sim 0.2$) films were grown on (012) $LaAlO_3$ substrates (referred to as $LaAlO_3$ (001) by the pseudocubic designation) by metal-organic chemical vapor deposition. Mn K_β high -resolution x-ray emission and x-ray absorption measurements indicate that all films have the same average Mn valence.

X-ray diffraction measurements reveal 1% strain (out-of-plane lattice parameter change) in the 9nm film compared with the 412 nm film. By SQUID magnetometry, the 412 nm film was found to have a T_C of ~270K with a saturation magnetization of 3.7 μ_B/Mn (25 K) while the corresponding T_C and saturation magnetization values for the 9 nm film are ~100 K and 1.4 μ_B/Mn (25 K), respectively.

RIXS measurements were conducted at Spring-8 beamline 12XU. The undulator beamline utilized a Si(111) double crystal monochromator. The measurements on the inelastic x-ray spectrometer were conducted in a closed cycle cryostat. A single 4” Si(220) analyzer with a resolution of ~0. 5 eV was used. The resolution of the complete system was 1eV. In the RIXS scans the incident energy as set at 6556 eV near the peak of the Mn K-edge absorption spectrum.

Significant changes are seen in the 9 nm film (1% strained). The A, B and C features are shifted to higher energy compared to the thick film- occurring at ~6, 9 and above 12 eV respectively. (The change in the width of the elastic line “lifts up” the 300K spectrum but does not produce any significant energy shifts.) The large shift found is due to the effect of strain on the electronic structure. In theoretical simulations, we have found the same upward shifts in the unoccupied bands with increasing pressure (volume reduction). Hence we associate this shift with the substrate strain. In addition to the shifts, the other major difference is that the thin film exhibits a large narrow B peak (oxygen 2p to Mn 3d) at room temperature while the at low temperature both the shape and position of the B region model those of the thick film. Hence is appears that the 3d band of the thin film is transformed from a highly localized band at high temperature to a broad band at low temperature.

The broadening of the 3d band a low temperature can possibly result from structural changes or can be purely electronic –resulting from correlation effects. A detailed study of this narrowing of the bands is essential to understanding this effect in thin films and connecting it with the loss of magnetic moment.

A manuscript is in preparation for submission to the journal Applied Physics Letters.

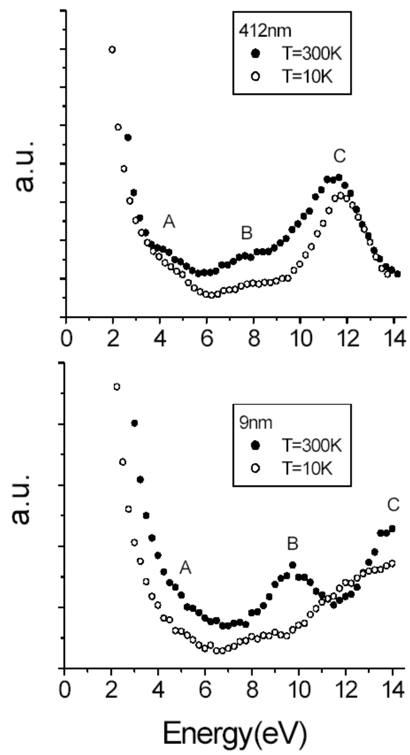

Figure 1. The RIXS (energy loss) spectra for the 412 nm film (top) and 9nm film measured at 10 and 300 K. Note the band broadening (B feature) at low temperature in the 9nm film.