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Conductivity enhancement of ultrathin LaNiO_3 films in superlattices

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The resistance of superlattices composed of bilayers of ultrathin (~ 4 unit cells) of LaNiO_3 and ~ 3 unit cells of insulating SrTiO_3 is explored as a function of temperature and the number of bilayers. All superlattices with more than one bilayer are metallic, whereas a single bilayer is insulating. Two possible interpretations of the electrical characteristics of the superlattices are discussed. The first model involves conduction in parallel-connected layers, whereas the second model assumes coupling of layers, each of which is near the percolation threshold for a metal-insulator transition. © 2010 American Institute of Physics. [doi:10.1063/1.3511738]

Oxide heterostructures have attracted significant attention as they may exhibit novel properties at interfaces that are not present in either of the bulk phases of the materials that make up the heterojunction.¹ In contrast to perovskite heterostructures with manganites, titanates, or cuprates, which have been studied extensively, nickelate thin films remain relatively unexplored, despite the fact that they may exhibit many interesting properties arising from strong electron correlations.² Bulk LaNiO_3 shows correlated Fermi liquid behavior and enhanced Pauli paramagnetism at all temperatures^{3,4} and, unlike the other rare-earth nickelates, does not undergo a metal-to-insulator transition.⁵ Recent band structure calculations predict dramatic changes in the magnetic and electronic properties of confined LaNiO_3 films including the possibility of high-temperature superconductivity.^{6,7} Experimental studies have shown that ultrathin (few unit cells) LaNiO_3 films exhibit strongly localized (insulating) behavior below a critical thickness, whereas thicker films remain metallic.^{8,9} The goal of this letter is to report on the transport properties of ultrathin LaNiO_3 films that are embedded in superlattices. It is shown that LaNiO_3 films in the superlattices show an enhanced conductivity compared to single LaNiO_3 layers of the same thickness.

Epitaxial $[\text{SrTiO}_3(3 \text{ u.c.})/\text{LaNiO}_3(4 \text{ u.c.})]_n$ superlattices, where n is the number of bilayers ($n=2,3,5,10$) and u.c. stands for the (pseudo)cubic unit cell ($a_{\text{LaNiO}_3}=0.384 \text{ nm}$), were grown on (001) $(\text{LaAlO}_3)_{0.3}(\text{Sr}_2\text{AlTaO}_6)_{0.7}$ (LSAT) substrate surfaces by rf magnetron sputtering. All superlattices were coherently strained to the LSAT substrate. The growth parameters for the LaNiO_3 films and their electrical properties as a function of thickness and lattice mismatch strain have been reported previously.⁸ The resistivity of thick ($>10 \text{ nm}$) metallic LaNiO_3 films is comparable to the lowest values reported in literature,^{10,11} indicating good oxygen stoichiometry. To preserve oxygen stoichiometry of the LaNiO_3 films during superlattice deposition, SrTiO_3 films were grown with the same high pressure used for LaNiO_3 growth, which resulted in slightly off-stoichiometric, but insulating, SrTiO_3 layers. The surfaces of the superlattice samples showed clear step-and-terrace structure in atomic force microscopy. High-angle an-

nular dark-field (Z-contrast) imaging was performed using a field emission scanning transmission electron microscope (STEM) (FEI Titan 80-300) operated at 300 kV. Electrical transport measurements were carried out down to 2 K using a physical property measurement system (PPMS, Quantum Design) using mesa Hall bar structures with Ni/Au Ohmic contacts.⁸ To confirm the insulating properties of the ultrathin SrTiO_3 layers, the sheet resistance of 3 u.c. SrTiO_3 layers was measured on LSAT substrates and confirmed to be greater than $1 \text{ M}\Omega/\square$.

Figure 1(a) shows radial high-resolution x-ray diffraction scans through the 002 symmetric reflections of a $[\text{SrTiO}_3/\text{LaNiO}_3]_{10}$ superlattice (the pseudocubic notation is

FIG. 1. (Color online) (a) High-resolution x-ray diffraction radial scans through the 002 reflections of a $[\text{SrTiO}_3(3 \text{ u.c.})/\text{LaNiO}_3(4 \text{ u.c.})]_{10}$ superlattice on LSAT. Note the satellite peaks around the 002 Bragg reflection. (b) Atomic resolution cross-section high-angle annular dark-field image of a superlattice.

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FIG. 2. (Color online) Sheet resistance as a function of temperature for superlattices with different number of bilayers n . The dotted line shows the sheet resistance of $10 \text{ k}\Omega/\square$, corresponding to the Mott minimum conductivity. All superlattices show a resistivity minimum at 20 K, typical for weak localization. The sheet resistance of a single 4 u.c. LaNiO₃ layer is also shown (open squares).

used for LaNiO₃). The main peak of the superlattice appears near the LSAT substrate peak. Moreover, satellite peaks up to second order are observed. This is in contrast to recent reports of [LaAlO₃/LaNiO₃] superlattices¹² and suggests sharper interfaces in the present study. Thickness fringes are visible around the peaks and indicate smooth interfaces between the SrTiO₃ and the LaNiO₃ layers. From the distance between adjacent satellite peaks, the thickness of one period is estimated to be 2.73 nm, which is consistent with the nominal thickness of 3 u.c. SrTiO₃ and 4 u.c. LaNiO₃. Figure 1(b) shows a cross-section Z-contrast STEM image of the superlattice, which confirms the cube-on-cube epitaxial orientation relationship. Due to the atomic number contrast in these images, the LaNiO₃ layers appear brighter than the SrTiO₃ layers. Although individual layers can be clearly distinguished, the interface roughness is about 1 unit cell. Furthermore, disordered regions were occasionally detected on top of the SrTiO₃ layers, most likely to accommodate nonstoichiometry.

Figure 2 shows the sheet resistance of the superlattices as a function of temperature and the number of bilayers. All superlattices with $n \geq 2$ are metallic, i.e., their resistance decreases with decreasing temperature. In contrast, 4 u.c. LaNiO₃ films and structures with $n=1$, i.e., a single, 4 u.c. LaNiO₃/3 u.c. SrTiO₃ bilayer (also shown in Fig. 2), are all insulating, showing an increase in resistance with decreasing temperature. Thus, the ultrathin LaNiO₃ layers are insulating, independent of whether the film surface is exposed to air or capped. The sheet resistance of superlattices systematically decreases with n , for $n \geq 2$, which indicates that the current passes through all SrTiO₃ barriers, which are sufficiently thin to allow for tunneling,¹³ and the resistance of all LaNiO₃ layers in the superlattice contributes to the measured total sheet resistance, R_S . We next attempt to describe the sheet resistance of the superlattices with conduction through parallel-connected LaNiO₃ layers. A linear relationship between $1/R_S$ and n is then expected, as indeed observed [see Fig. 3(a)]. The nonzero intercept in Fig. 3(a) implies an additional contribution to the measured conductance. Assuming

FIG. 3. (Color online) (a) The reciprocal of the superlattice sheet resistance, $1/R_S$, as a function of the number of bilayers n measured at four different temperatures. The solid lines are linear fits to the data. The inset shows a schematic of the parallel resistor model used to describe $1/R_S$ of the superlattices. (b) Measured sheet resistance of a single LaNiO₃ layer and the extracted sheet resistance of an individual LaNiO₃ layer in the superlattices, using Eq. (1). (c) Resistivity of the superlattices multiplied with $(2^n - 1)/2^n$ as a function of temperature.

parallel-connected conduction in LaNiO₃ layers (sheet resistance R_{LNO}) and additional “interface layers” (sheet resistance R_i), the inverse of the total sheet resistance can be modeled if $2n-2$ interfaces are conducting, i.e.,

$$\frac{1}{R_S} = \frac{1}{R_i}(2n-2) + \frac{1}{R_{\text{LNO}}}n. \quad (1)$$

If all $(2n-1)$ interfaces in the superlattices are assumed to be conductive, a negative value for R_{LNO} is obtained, which is unphysical. The physical picture behind Eq. (1) is then that

only interfaces, which interface with LaNiO_3 from both sides, are conductive. Using Eq. (1), R_{LNO} can be determined as a function of temperature [see Fig. 3(b)]. Comparison of R_{LNO} extracted using Eq. (1) with the measured sheet resistance of the single 4 u.c. LaNiO_3 layer shows that the LaNiO_3 layers in the superlattices are more conductive than a single LaNiO_3 layer. This would indicate that coupling between the LaNiO_3 layers in superlattices through tunneling through the SrTiO_3 layers results in a conductivity enhancement.

An alternative explanation for the differences in conductivity of single layers and superlattices, which does not involve conductive interface layers, can also be proposed. Specifically, if thin LaNiO_3 layers possess some degree of disorder or are near a first order phase transition, both metallic as well as poorly conductive regions may be present, causing the thinnest layers to be just below the percolation threshold for metallic conduction. This would be consistent with slightly thicker layers being metallic.⁸ Adding more layers to the superlattice and allowing for tunneling through the SrTiO_3 will increase the number of conduction pathways. For example, if a single layer consists of a 50:50 mixture of conducting and insulating regions, each additional layer may double the number of conduction pathways, i.e., the resistivity of the superlattice should scale according to $(2^n - 1)/2^n$. The connected fraction of the superlattice moves away from the critical point as more layers are added. Although a more sophisticated description of the superlattice conductivity would involve effective medium theory,¹⁴ Fig. 3(c) shows that the superlattice resistance can be described surprisingly well with this simple model.

The results show that care must be taken in drawing conclusions from the transport properties of ultrathin metal oxide layers in superlattices to those of individual layers.¹⁵ In

particular, in metal oxide systems for which some degree of disorder causes a high residual resistance, individual layers may be insulating, whereas superlattices remain metallic.

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