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Strain-dependent transport properties of the ultra-thin correlated metal, LaNiO$_3$

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Abstract. We explore the electrical transport and magneto-conductance (MC) in quasi-two-dimensional strongly correlated ultra-thin films of LaNiO$_3$ (LNO) to investigate the effect of hetero-epitaxial strain on electron–electron and electron–lattice interactions from the low to intermediate temperature range (2–170 K). The fully epitaxial 10 unit cell thick films spanning tensile strain up to $\sim 4\%$ are used to investigate the effects of enhanced carrier localization driven by a combination of weak localization (WL) and electron–electron interactions at low temperatures. The MC data show the importance of the increased contribution of WL to low-temperature quantum corrections. The obtained results demonstrate that with increasing tensile strain and reduced temperature, the quantum-confined LNO system gradually evolves from the Mott into the Mott–Anderson regime.

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1. Introduction

Low-dimensional strongly correlated electron systems composed of artificial layers of transition metal oxides (TMO) have attracted a great deal of attention and have been an active area of research for many years due to their intriguing electronic and magnetic properties \([1–3]\). Despite extensive research efforts, limited experimental information is available on the behavior of correlated carriers in reduced dimensions and their response to the magnitude and sign of a strain state. For example, in TMO thin films, since strain due to the lattice mismatch between epitaxial film and substrate can modify the electronic bandwidth, the balance between Coulomb repulsion energy \(U\) and electron hopping \(t\) can be used to control the metal-to-insulator transition (MIT). Bulk rare-earth nickelates (RENiO\(_3\), RE = La, Pr, Nd, . . . , Lu) represent an ideal candidate system, where the one-electron bandwidth derived from the Ni–O chemical bond length and the Ni–O–Ni bond angle primarily controls the electronic ground state \([4]\). In the ultra-thin limit, this is further amplified by the intrinsic tendency of correlated metals to localize in two dimensions (2D) \([5]\). Recently, extensive transport measurements have been reported on films of LaNiO\(_3\) (LNO) to address the role of reduced dimensionality and externally applied electric fields on conductivity \([6, 7]\). A few studies have focused on the effects of disorder in strongly correlated electronic systems, such as the resistivity minima at low temperature \(T_{\text{min}}\), which reveals the presence of quantum corrections to the conductivity (QCC). This quantum effect has been reported for SrRuO\(_3\) (SRO). SRO, another important candidate for the electrical conductivity of metallic perovskites, has a comparable Ioffe–Regel limit \((k_B l \sim 1, k_B\) is the Boltzmann constant and \(l\) is the mean free path\) to LNO \([8]\). The upturn in the electronic resistivity of ferromagnetic SRO is driven by renormalization of electron–electron interactions (REEI) due to the strong internal field \([9]\), whereas in LNO weak localization (WL) is significant \([7, 10]\). For LNO ultra-thin films (few nanometers), limited systematic information on the contributions of REEI and WL in terms of strain is available.

In this paper, we present results on the systematic investigation of dc and magneto-transport properties of 10 unit cell (u.c.) thick (~3.83 nm) high-quality epitaxial LNO films as a function of tensile strain. Unlike the bulk, at low temperatures the dc transport measurements show that, depending on the magnitude of the strain, QCC arise from two distinct sources: a dominant electron–electron interaction (EEI) and WL. Additionally, magneto-conductance (MC) as a function of magnetic field corroborates the dominant role of WL induced by electron scattering at the reduced dimensional regime. Therefore, here we investigate the importance of epitaxial strain and its sign on metallicity as the material reaches the quasi-2D limit in LNO.
2. Experimental procedures

Epitaxial LNO ultra-thin films on (LaAlO$_3$)$_{0.3}$(Sr$_2$AlTaO$_6$)$_{0.7}$ (LSAT), SrTiO$_3$ (STO), TbScO$_3$ (TSO) and GdScO$_3$ (GSO) substrates were grown by pulsed laser deposition (PLD) with in situ monitoring by a recently developed high-pressure reflection high-energy electron diffraction (HP-RHEED) system, which operates in an O$_2$ background pressure of up to 400 mTorr. The HP-RHEED image of LNO/STO is shown in figure 1(a). Details regarding the 2D layer by layer growth PLD for LNO films are given elsewhere [11]. After the completion of deposition, the films were annealed in 1 atm of ultra-pure oxygen to minimize possible oxygen deficiency that adversely affects the conductivity [12, 13]. AFM imaging revealed smooth surface morphology with a surface roughness of $\sim 70$ pm or better (see figure 1(b)). Photoemission spectra were recorded at room temperature in a spectrometer equipped with a VG-Scienta R3000 electron energy analyzer and a Vacuum Generators twin crystal monochromatized Al-Ka ($h\nu = 1486.6$ eV) source. As shown in figure 1(c), the band crossing of the Fermi level is clearly observed, which affirms the metallicity of 10 u.c. LNO films. Quantitative modeling of the spectrum is, however, a complicated issue due to the correlated nature of Ni 3$d$ electrons. The problem is even more intractable because the O 2$p$ to Ni 3$d$ charge transfer energy in LNO is small and perhaps even negative [14], leading to strong charge fluctuations involving many different electronic configurations [15]. However, recently for an LNO film extensive local density approximation (LDA)+U calculations were reported and the results of the calculations are in excellent agreement with the presented XPS data [16].

3. Results and discussion

We begin with a discussion of the temperature-dependent dc transport of LNO ultra-thin films. At finite temperatures, resistivity as a function of temperature can be conventionally defined as $\rho(T) = \rho_0 + AT^\alpha$, where $\rho_0$ is the temperature-independent residual resistivity. $A$ is a constant, and the power exponent $\alpha$ depends on details of the scattering mechanism.
Figure 2. Resistivity as a function of temperature for a LNO 10 u.c. film grown on (a) an LSAT substrate, (b) an STO substrate, (c) a TSO substrate and (b) a GSO substrate with each fitting curve. Insets: corresponding temperature dependence of the deviation from the fittings in the 3D limit.

framework of Fermi liquid theory, Coulomb interaction yields $\alpha = 2$ (or $T^2$ dependence) [17, 18]. However in complex oxides at lower temperatures, the carriers can localize, and a metal–insulator transition may occur in these marginal metals. The nature of the transition is then conventionally explained by the emergence of QCC. The quantum corrections are derived from two important mechanisms: WL, a self-interference effect, and EEIs [19].

Figure 2 shows the resistivity of LNO films on the four different substrates along with the fitting curves. The temperature-dependent resistivity was measured from 300 K to 2 K in van der Pauw geometry with a commercial physical properties measurement system (PPMS, Quantum Design). The resistivity upturn is clearly visible at low temperatures. The upturn can be described by considering both the EEI and WL contributions. Based on the localization-interaction model for a disordered metallic (e.g. ‘marginal’ metal) system in the 3D limit, the temperature dependence of resistivity is given by [20–22]

$$\rho(T) = \frac{1}{\sigma_0 + a_1 T^{p/2} + a_2 T^{1/2}} + b T^\alpha,$$

where $\sigma_0$ implies the classical temperature-independent Drude conductivity, $a_1$ takes into account the 3D WL contribution, and the last term, $a_2$, introduces the EEI in transport. The variable $p$, the second term in the denominator of the first term, is an exponent that describes localization effects. It is well known that $p = 2$ implies the dominance of EEIs, while $p = 3$ is
attributed to electron–phonon scattering [20]. As seen in figure 2, for each LNO film, the value of $p = 2(0.003)$ results from the fitting to equation (1). Given the ultra-thin nature of epitaxial LNO films, an attempt to fit the experimental data in the 2D limit (i.e. including the $\ln T$ term instead of $a_1 T^{\alpha_1} + a_2 T^{\alpha_2}$ in equation (1) [20] was also made, but the worse $\chi^2$ (1–2 order of magnitude of difference) confirmed that the 3D fit is more appropriate. The MC data, discussed later in the paper, corroborate the 3D nature of electronic transport. Thus, the best fit to the data for the 10 u.c. LNO films testifies to the 3D nature of the localization.

The temperature range for fitting to equation (1) was extended up to intermediate temperatures (~170 K), because the upturn minima $T_{\text{min}}$ of the four films vary between ~7 and ~50 K. To investigate the effect of quantum corrections over the same temperature range and consider the same factors for all four films, the fit range includes both the rise in the resistivity $\Delta \rho / \rho \sim T^{5/2}$ at low temperature and the intermediate metallic phase. In addition, to demonstrate the validity of the fitting parameters, the deviation plot defined as $\Delta \rho / \rho = ((\rho_{\text{obs}}(T) - \rho_{\text{fit}}(T))/\rho_{\text{obs}}(T)) \times 100$ is shown in figures 2(a)–(d), and the fitting parameters for the different substrates that correspond to varying the level of the epitaxial biaxial tensile strain on LNO are displayed in table 1.

Information on the nature of localization regardless of quantum corrections can be obtained from MC measurements [20, 21, 23]. Specifically for a 3D system with dominant WL, the field-dependent correction to the conductivity $\Delta \sigma(\mu B, T)$ takes on a simple power law in limiting fields [20, 24]. For small applied fields, the MC is $\propto B^{1/2}$ ($g \mu_B B < k_B T$), and at the limit of high magnetic field the MC is $\propto B^{1/2}$ ($g \mu_B B > k_B T$) [21]. We performed MC measurements on the LNO sample grown on the GSO substrate (+ 4% tensile strain) in an applied magnetic field of up to 7 T parallel to a substrate. As seen in figure 3, the MC data taken at 2 K follow the anticipated power-law behavior at both limits. The upper limit for the low-field dependence corresponds to $B = 1.3$ T at 2 K. The MC measurement provides additional information on the modulation of the low T localization by the magnitude of strain, which is largely attributed to WL. Again, observation of this field-dependent MC supports the presence of WL of charge carriers in metallic films. The strain alters the distortion of the NiO$_6$ octahedra and varies the $d$-band transport along with other factors that may contribute to interaction mechanisms.

We now focus on the second power exponent term, $\sim T^\alpha$, of equation (1) to obtain the contribution to the low $T$ quantum corrections from the metallic phase up to the intermediate

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Table 1. Results of fits to equation (1) over the temperature range from 2 to 170 K for LNO films with thickness 10 u.c. grown on various substrates. The substrate parameters compared with LNO are also displayed.

<table>
<thead>
<tr>
<th>Substrate</th>
<th>Lattice parameter (Å)</th>
<th>Strain with LNO (%)</th>
<th>$a_0$ ($\Omega^{-1} \text{cm}^{-1}$)</th>
<th>$a_1$ ($\Omega^{-1} \text{cm}^{-1} \text{K}^{-1}$)</th>
<th>$a_2$ ($\Omega^{-1} \text{cm}^{-1} \text{K}^{-1/2}$)</th>
<th>$b$ ($\Omega \text{cm K}^{-\alpha}$)</th>
<th>$\alpha$</th>
</tr>
</thead>
<tbody>
<tr>
<td>LSAT</td>
<td>3.87</td>
<td>0.78</td>
<td>2795.8</td>
<td>6.6553</td>
<td>76.113</td>
<td>2.1135 x 10^{-7}</td>
<td>1.3099</td>
</tr>
<tr>
<td>STO</td>
<td>3.901</td>
<td>1.915</td>
<td>3050.4</td>
<td>4.9923</td>
<td>68.258</td>
<td>1.8915 x 10^{-7}</td>
<td>1.1999</td>
</tr>
<tr>
<td>TSO</td>
<td>3.954</td>
<td>3.224</td>
<td>3729.5</td>
<td>6.3772</td>
<td>62.489</td>
<td>3.6222 x 10^{-7}</td>
<td>1.2558</td>
</tr>
<tr>
<td>GSO</td>
<td>3.969</td>
<td>3.612</td>
<td>4509.8</td>
<td>10.065</td>
<td>30.387</td>
<td>2.6661 x 10^{-7}</td>
<td>1.2915</td>
</tr>
</tbody>
</table>

* Based on the ratio of $a_1$ and $a_2$ as a function of strain, it is plausible to conclude that the LNO ultra-thin film grown on GSO substrate is the best candidate for WL.

New Journal of Physics 13 (2011) 073037 (http://www.njp.org/)
Figure 3. MC varies in field with its different power factor at the same system. The dotted lines are fits to $B^2$ in the low-field and to $B^{1/2}$ in the high-field regime.

Figure 4. (a) $\rho_{\text{normal}}(T)$ versus $T^\alpha$ ($\alpha = 1.3$ or $4/3$ as shown in table 1 in the range of $2 \sim 170 \text{ K}$) of LNO 10 u.c. films grown on different substrates to introduce various strain mismatches. The curvy arrows (gray color) across the four curves of resistivity show an evolution of the shape of upturn as a function of strain. The black-colored arrows under the curves indicate the temperature in which the resistivity is minimum (the position of the upturn). (b) Film biaxial strain versus $\sigma_0^{-1/2}$ (left axis) and $a_2$ (right axis). (c) Coefficient $a_2$ and $\sigma_0^{-1/2}$ as a function of strain mismatch between an LNO film and a substrate (see table 1).

temperature range. Figure 4(a) shows the $\rho_{\text{normal}}$ temperature dependence for LNO under increasing tensile strain from 2 to 170 K; here the value of $\rho_{\text{normal}}$ is defined as $\rho_{\text{normal}} = \rho(T) - \rho_{\text{con}} = \rho_{\text{QCC}} + T^\alpha$, where $\rho_{\text{con}}$ is a constant to rescale each resistivity curve to cross zero resistivity at 0 K. As seen in figure 2 and table 1, the power $\alpha$ yields $1.29(0.03)$ ($\sim 4/3$).

Upon approaching the low-temperature range, $dT^{4/3}/d\rho_{\text{normal}}(T)$ gradually changes with strain. As shown in figure 4, the region within the yellow-filled enclosed box exhibits significant changes of $dT^{4/3}/d\rho_{\text{normal}}(T)$ in terms of strain. Furthermore, the minimum of resistivity $T_{\text{min}}$
decreases with strain as illustrated by the black arrows under the resistivity curves in figure 4(a). This resistivity upturn (or MIT) may result from several plausible causes. For instance, it is well known that after reaching the Ioffe–Regel limit, in which the mean free path equals the interatomic spacing, the film may undergo a transition from the metallic to insulating ground state [25]. The thickness of the LNO sample (few nanometers around the critical thickness) will also enhance the propensity towards localized behavior [7]. Here we note, however, that the minimum conductivity from the Mott–Ioffe–Regel limit (MIRL) [26] of bulk LNO is \( \sim 300 \text{ S cm} \) [27], whereas the conductivity of each strained LNO ultra-thin film from LSAT to GSO (up to the tensile strain of 4%) is well above this MIRL. Given these results, the localization observed in the quality of 10 u.c. films is not primarily driven by the intrinsic disorder and/or the reduced layer thickness (size effect). Instead, based on the strong correlation between the low \( T \) localization and the magnitude of strain, the lattice mismatch that modifies the one-electron bandwidth is the prime source for such response. To further quantify this, we consider the relative contribution of the fitting parameters of equation (1) \( (a_1, a_2, \sigma_0) \) (see table 1) versus strain by means of the empirically derived relation reported in [10, 28]. As shown in figures 4(b) and (c), parameter \( a_2 \) decreases with increasing tensile strain, indicating that EEI is suppressed with tensile strain, while the WL contribution \( (a_1 \) parameter) is clearly enhanced with strain. The relative contribution of localization and Coulomb interactions in the transport properties of the film might be controlled by strain. Furthermore, localized states in the presence of Coulomb interactions in strongly correlated materials lead to the formation of a soft gap, which can be determined by the scaling function of \( \rho(T) = \exp(T_0 / T)^{1/2} \) at low temperature, suggested by the Efros–Shklovskii model [29–31]. The resistivity curves are fitted well below \( \sim 15 \text{ K} \), and this result can be expected from the fitting parameters above. These trends of the fitting and scaling parameters may indicate that LNO ultra-thin films are closer to Mott–Anderson materials, which are defined by the presence of a soft gap at the Fermi level near the MIT, than Mott materials. Indeed, the evolution of the transition driven by biaxial strain indicates that the MIT is tuned as a function of strain. The synergetic contribution of these two effects is perhaps responsible for the peculiar low-temperature dynamics with strain.

4. Summary

We have found the QCC for charge carriers in LNO ultra-thin films and modeled the temperature-dependent resistance in terms of 3D WL and EEIs in a disordered metallic system. The synergetic contribution of these two effects is perhaps responsible for the peculiar low-temperature dynamics with strain.

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