**TITLE AND SUBTITLE**
Low-dimensional Mott material: Transport in ultrathin epitaxial LaNiO3 films

**PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)**
University of California, Santa Barbara, CA 93106, USA

**SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)**
Army Research Office, Durham, NC 27703

**ABSTRACT**
Electrical resistivity and magnetotransport are explored for thin 3–30 nm epitaxial LaNiO3 films. Films were grown on three different substrates to obtain LaNiO3 films that are coherently strained, with different signs and magnitude of film strain. It is shown that d-band transport is inhibited as the layers progress from compression to tension. The Hall coefficient is "holelike." Increasing tensile strain causes the film resistivity to increase, causing strong localization to appear below a critical thickness.
Electron transport in two-dimensional systems has been extensively studied in normal metals and semiconductors. Recently, quantum confined films of strongly electron correlated materials have attracted renewed interest. For example, theory predicts dramatic changes in the magnetic and electronic properties for the confined "Mott material" LaNiO$_3$, including the possibility of high-temperature superconductivity. Bulk rare-earth nickelates (RNiO$_3$ with $R$=rare earth cation), to which LaNiO$_3$ belongs, have been extensively researched because they exhibit a metal-insulator transition (MIT) that is a function of the radius of the rare-earth ion. The transition is believed to be bandwidth-controlled and is associated with an expansion (contraction) of the unit cell. More recent investigations have shown that charge ordering also plays a role in the MIT of the nickelates. Unlike the other nickelates in the series, LaNiO$_3$ remains a paramagnetic metal at all temperatures, albeit a strongly correlated one. In this letter, we show that strong localization can be driven in LaNiO$_3$ films by a combination of strain and reduction in dimensionality.

Epitaxial LaNiO$_3$ films were grown by rf magnetron sputtering. Optimized growth parameters (100 mTorr total pressure, 600 °C substrate temperature) resulted in the smallest film lattice parameter and lowest resistivity. Films were grown on (001) surfaces of cubic (LaAlO$_3$)$_{0.3}$Sr$_2$AlTaO$_6$ (LSAT), orthorhombic DyScO$_3$, and rhombohedral LaAlO$_3$. LaNiO$_3$ films [$(a=0.384 \text{ nm}$ (Ref. 7))] on LSAT and DyScO$_3$ were under tensile strain and on LSAT and LaAlO$_3$ up to 10 nm on DyScO$_3$. Figure 1(a) shows radial high-resolution x-ray diffraction scans through the 002 symmetric reflections of films and substrates. Thickness fringes indicate smooth and coherent films. The epitaxial orientation relationship. All films are continuous. Films were grown on LaAlO$_3$ by rf magnetron sputtering. Optimized growth parameters resulted in films show metallic behavior with a resistivity of 100 mTorr total pressure, 600 °C substrate temperature) resulted in the smallest film lattice parameter and lowest resistivity. Films were grown on (001) surfaces of cubic (LaAlO$_3$)$_{0.3}$Sr$_2$AlTaO$_6$ (LSAT), orthorhombic DyScO$_3$, and rhombohedral LaAlO$_3$. LaNiO$_3$ films [$(a=0.384 \text{ nm}$ (Ref. 7))] on LSAT and DyScO$_3$ were under tensile strain and on LSAT and LaAlO$_3$ up to 10 nm on DyScO$_3$. Figure 1(a) shows radial high-resolution x-ray diffraction scans through the 002 symmetric reflections of films and substrates. Thickness fringes indicate smooth and coherent films. The epitaxial orientation relationship. All films are continuous.

Figure 1(b) shows HAADF images of a 3 nm thick LaNiO$_3$ film on LSAT, which confirmed the cube-on-cube epitaxial orientation relationship. All films are continuous.

Figure 2(a) shows the temperature-dependent resistivity of LaNiO$_3$ films on LSAT as a function of thickness. Thick (10−30 nm) films show metallic behavior with a resistivity of $\sim 150 \ \mu\Omega \ \text{cm}$ at room temperature, independent of film thickness. This value is comparable to the lowest reported values, indicating good oxygen stoichiometry. Below $\sim 10 \ \text{nm}$, the room temperature resistivity increases. The 4 nm films show a resistivity minimum at $\sim 40 \ \text{K}$ [see inset in Fig. 2(a)] below which the resistivity scaled with $\log(T)$...
see inset in Fig. 3. Further decrease in film thickness to 3 nm causes the films to become strongly localized (insulating), showing an increase in resistivity with decreasing temperature over the entire temperature range. For films on DyScO₃, which are under a larger tensile strain than those on LSAT, the transition to strong localization occurs at a larger thickness [Fig. 2b]. Independent of substrate, the critical thickness for strong localization of the tensile-strained films corresponds to a film resistance of \(10^{-4}\) \(\Omega\) cm, the Mott minimum metallic conductivity as calculated from the Ioffe–Regel limit, where strong localization should appear. Compressive-strained LaNiO₃ films on LaAlO₃ show lower resistivities than films on LSAT and DyScO₃ and no MIT down to 2.5 nm. However, the thinnest (2.5 nm) film on LaAlO₃ also shows a resistivity minimum.

The observed resistivity minima and logarithmic temperature dependence in the intermediate regime can be due to weak localization or electron-electron interactions. Figure 3 shows that negative magnetoresistance in a perpendicular field is observed below the resistivity minimum. This is consistent with weak localization; a magnetic field suppresses the coherent interference needed for weak localization. The Hall coefficient of LaNiO₃ layers on all substrates was positive, “holelike,” while the Seebeck coefficient was negative, similar to bulk LaNiO₃. A Fermi surface with any complexity can give rise to opposite signs. Band structure calculations and photoemission spectroscopy show small electron Fermi surfaces with an enhanced effective mass at the I point and a large hole Fermi surface around the R point. Figure 4 shows that \(R_H\) for films on LSAT and LaAlO₃ exhibits a strong temperature dependence, inconsistent with normal metallic behavior, which indicates temperature-dependent scattering times and/or carrier concentrations/asymmetry. Because two carrier types with different mobilities are present, \(R_H\) cannot be easily converted into carrier concentrations.

At first glance the transport on the three substrates is similar: (i) metallic temperature dependence at room temperature for most thicknesses; (ii) emerging weak and then strong localization as the layer resistance approaches the Mott minimum conductivity, (iii) temperature dependent holelike \(R_H\), and (iv) thickness dependent \(R_H\), beyond the trivial \(1/T\). Behind these similarities, a systematic trend toward localization or inhibited transport appears, which we will discuss next.

In the literature, thickness-dependent conductivity of metallic oxide thin films has been attributed to a “dead layer.” We use the high temperature resistivity to explore thickness dependence and the potential role of dead layers. We use a phenomenological, classical description of the resistivity \(\rho\).
ment does not produce changes in the band structure. For equal to the measured thickness, such as surface/interface scattering. The derivative of Eq. (1) with temperature, at high temperature, is given by

$$\frac{\partial \rho}{\partial T} = \frac{m}{n} \left[ \frac{1}{e^2} \frac{1}{\tau_{\text{ph}}^*} + \frac{1}{\tau_{\text{surface}}} + \frac{1}{\tau_{\text{impurity}}} \right],$$

where $e$ is the electron charge, and the band structure is captured by the term that contains the carrier mass $m$ and density $n$. The scattering rate is the sum of temperature dependent, intrinsic, phonon scattering ($\tau_{\text{ph}}$), and temperature independent terms, such as surface/interface ($\tau_{\text{surface}}$) and impurity ($\tau_{\text{impurity}}$) scattering. The derivative of Eq. (1) with temperature is independent of $t$, if the electrical thickness is equal to the measured thickness (no dead layer) and confinement does not produce changes in the band structure. For thick films on the same substrate, $\partial \rho / \partial T$ at room temperature is similar [see, i.e., Fig. 2(a)]; thus there is no need to invoke position dependent transport such as a “dead layer.” The thick film, low temperature ($2–80$ K), resistivity exhibits quadratic temperature dependence, $\rho = \rho_0 + AT^2$, characteristic of electron-electron interactions. Table I shows the coefficients $A$ and $\rho_0$, and the high temperature $\partial \rho / \partial T$ ($250–300$ K). Both $A$ and $\partial \rho / \partial T$ are measures of changes in intrinsic transport as the biaxial strain evolves from compression to tension. Increasing $\partial \rho / \partial T$ is consistent with a narrowing of the $d$-band with tensile strain. Changes in $d$-band transport drive the films toward a localization transition. As the transport is inhibited the resistance reaches the two-dimensional Iofe–Regel limit and strong localization sets in. Furthermore, although we are reluctant to over-interpret the Hall data, biaxial expansion may also reduce the effective number of carriers.

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7. We use the pseudocubic unit cells for denoting the surface orientations and lattice parameters for all noncubic substrates and the LaNiO$_3$ films.
19. If magnetotransport can be measured in the regime where $\mu_B B > 1$, where $\mu_B$ is the respective carrier mobility, the Hall effect will be a nonlinear function of magnetic field and it may be possible to distinguish different carrier contributions, see Ref. 18. The layers explored here exhibit linear Hall effect and appear to be far from this limit.