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Tuning bad metal and non-Fermi liquid behavior in a Mott material: Rare-earth nickelate thin films

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Resistances that exceed the Mott-Ioffe-Regel limit (known as bad metal behavior) and non-Fermi liquid behavior are ubiquitous features of the normal state of many strongly correlated materials. We establish the conditions that lead to bad metal and non-Fermi liquid phases in NdNiO$_3$, which exhibits a prototype bandwidth-controlled metal-insulator transition. We show that resistance saturation is determined by the magnitude of Ni $e_g$ orbital splitting, which can be tuned by strain in epitaxial films, causing the appearance of bad metal behavior under certain conditions. The results shed light on the nature of a crossover to a non-Fermi liquid metal phase and provide a predictive criterion for Anderson localization. They elucidate a seemingly complex phase behavior as a function of film strain and confinement and provide guidelines for orbital engineering and novel devices.

INTRODUCTION

Mott metal-insulator transitions (MITs) are key to some of the most fascinating topics in materials physics, such as the pathways from a doped Mott insulator to a high-temperature superconductor (1) and the feasibility of electronic devices that utilize switchable MITs. Rare-earth nickelates (RNiO$_3$, where $R$ = trivalent rare-earth ion) exhibit a prototype bandwidth-controlled MIT (2, 3). RNiO$_3$ films have recently attracted renewed interest resulting from a prediction that orbital engineering can promote a Fermi surface that resembles that of the cuprate high-temperature superconductors (4, 5). Furthermore, recent discoveries in RNiO$_3$ films point to strikingly similar physics as found in unconventional superconductors. Non-Fermi liquid (NFL) behavior (6) and pseudogap phases (7) indicate a continuous bandwidth-driven quantum phase transition between a paramagnetic metal and an antiferromagnetic insulator. The RNiO$_3$ family has also been discussed as a new class of “bad metals” (8) in the sense that their resistivity escalates above the semiclassical Mott-Ioffe-Regel limit and does not saturate at high temperatures (9). Bad metal behavior and NFLs are essential yet poorly understood features of the phase diagrams of unconventional superconductors (10). However, neither bad metals nor their counterparts (metals that exhibit resistance saturation) are understood; in both cases, materials enter a regime where classical Boltzmann theory should no longer apply (11–14).

In this work, we show that rare-earth nickelates are not bad metals: accounting for resistivity saturation is key to correctly describing their electrical transport behavior. The resistivity saturation limit is, however, highly sensitive to the degree of $e_g$ orbital polarization, leading to resistances that exceed the semiclassical Mott-Ioffe-Regel limit. Furthermore, accounting for saturation clarifies many aspects of the strain–film thickness phase behavior and the quantum critical point in the RNiO$_3$ system. In particular, an abrupt crossover between classical Landau Fermi liquid (LFL) and NFL metallic regimes occurs with the suppression of temperature-driven MIT. The metallic phase is an LFL in all cases where a robust MIT is present. We also clarify the conditions leading to Anderson localization in this system, namely, a second disorder-driven MIT: it appears when the resistivity at 0 K approaches the saturation resistance. We develop phase diagrams that can serve as practical guidelines for stabilizing robust MITs in ultrathin nickelate films and that identify new opportunities for control of MITs in general.

RESULTS

NdNiO$_3$ thin films with thicknesses ranging between 4 and 15 unit cells (u.c.) were grown on substrates chosen to obtain a wide range of epitaxial strains (in parentheses): YAlO$_3$ (−3.58%), LaAlO$_3$ (−1.20%), NdGaO$_3$ (+0.86%), (LaAlO$_3$)$_{0.3}$(Sr$_2$AlTaO$_6$)$_{0.7}$ (+0.93%), SrTiO$_3$ (+1.72%), and DyScO$_3$ (+2.96%). Figure 1 shows their electrical resistivities as a function of temperature. Several different types of behavior can be discerned, including temperature-driven MITs and films that are metallic or insulating at all temperatures.

We first focus on describing the metallic states. Nickelates are often understood to be NFLs (6, 15–19) for which the temperature dependence ($T$) of resistivity follows a power law with an exponent $n < 2$

$$\rho_{NFL}(T) = \rho_0 + A T^n$$

(1)

where $\rho_0$ is the residual resistivity and $A$ is a measure of the strength of electron-electron scattering. For a LFL, $n = 2$. However, Eq. 1 often describes experimental data only in a limited temperature range (20). As will be shown here, the temperature dependence of resistivity in the metallic phase is completely described when we account for resistivity saturation ($\rho_{SAT}$)

$$\rho^{-1}(T) = \rho_{NFL}^{-1}(T) + \rho_{SAT}^{-1}$$

(2)

Equation 2, in which $\rho_{SAT}$ acts as a parallel resistor, is known to apply to a wide range of materials that show resistance saturation, including elemental metals, alloys, and heavy fermion compounds (10, 21–26). The origins of resistivity saturation and the parallel resistor formula remain subjects of significant debate (10–12, 21, 27–31). $\rho_{SAT}$ is often linked to high resistances that approach the Mott-Ioffe-Regel limit ($\rho_{MIT}$) (28),

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which is the semiclassical upper bound for coherent transport in a metal when the carrier mean free path $l$ approaches the interatomic spacing $a$

$$\rho_{\text{MIR}} = \frac{3\pi^2 h}{q^2 k_F^2 a}$$

where $h$ is the reduced Planck constant, $q$ is the elementary charge, and $k_F$ is the Fermi wave vector. For many materials, $\rho_{\text{MIR}}$ estimated from Eq. 3 is similar to the observed $\rho_{\text{SAT}}$ (10). Suggestions as to why $\rho_{\text{SAT}}$ acts as a parallel resistor according to Eq. 2 include semiclassical pictures that invoke the existence of a minimum scattering time (10, 28) as well as interband scattering (29) that acts as a new conducting channel (12, 29). As has also been pointed out in the literature (11, 12, 32), it is questionable to use semiclassical arguments in a regime where $l$ is not a well-defined quantity anymore. Quantum Monte Carlo and dynamical mean field theory calculations indicate that resistances can easily exceed the $\rho_{\text{MIR}}$ value calculated from Eq. 3 for certain materials (13, 14). In particular, certain materials (including some unconventional superconductors) (27) are characterized by saturation at values much higher than the $\rho_{\text{MIR}}$ value calculated from Eq. 3, or even a nonsaturating $\rho$, and these materials have become known as bad metals (9).

For NdNiO$_3$ films, Eq. 2 is remarkably successful in describing the metallic state. This can be seen from the dashed lines in Fig. 1, which are fits to Eq. 2. The saturation behavior is also directly seen in the experimental data when plotted as $d\rho/dT$ as a function of $T$ (Fig. 2A). The key feature is the downturn in $d\rho/dT$ at high $T$ (23, 26). This translates to a sublinear scaling of $\rho$ with $T$, which is due to the increasing contribution from $\rho_{\text{SAT}}$, as expected from Eq. 2. Equation 2 describes the entire metallic state with a single $T$-independent exponent $n$ (inset to Fig. 2A). Additional examples, including those for LaNiO$_3$ films, are shown in figs. S1 to S3.

When the exponent $n$ was an adjustable parameter in the fits, only two values were obtained across the entire sample set (Fig. 2B): for all films displaying a robust temperature-driven MIT, $n = 2$, indicating a classical LFL. For films that are metallic at all temperatures (MIT completely suppressed) and for some films with a weak MIT at very low $T$, $n \approx \frac{5}{3}$, indicating an NFL regime. For example, on YAlO$_3$, all but the 4-u.c. film are metallic at all temperatures and NFLs. The 4-u.c. film shows a MIT and recovers LFL behavior. Noninclusion of $\rho_{\text{SAT}}$ (6, 15–18) would have resulted in the interpretation of the metallic state as an NFL even in the case of an LFL film, with apparent changes in the exponent $n$ for different temperature ranges (Fig. 2A). For example, $n = 1$ is assumed and $\rho_{\text{SAT}}$ is neglected for the dashed orange line in Fig. 2A. For the final fits shown in Fig. 1, the exponent $n$ was fixed at 2 or $\frac{5}{3}$ (by the closest value). The changes in the results were minimal, but a fixed exponent allows for more reliable comparisons across the series for the slope $A$ (fig. S4).

Figure 2C shows $\rho_{\text{SAT}}$ and $\rho(0)$ (metallic resistivity extrapolated to $T = 0$ K, as extracted from the fits) as a function of film thickness. Following Eq. 2

$$\rho^{-1}(0) = \rho_{\text{NFL}}^{-1}(0) + \rho_{\text{SAT}}^{-1} = \rho_0^{-1} + \rho_{\text{SAT}}^{-1}.$$  

For most films, $\rho_0 \ll \rho_{\text{SAT}}$; thus, $\rho(0) \sim \rho_0$. At low thicknesses, $\rho_0$ sharply increases, which is the ubiquitously observed rise of resistivity

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**Fig. 1. MITs and temperature dependence of resistivity.** The graphs show the resistivity as a function of temperature for NdNiO$_3$ (NNO) films with thicknesses ranging between 4 and 15 u.c. on six different substrates [YAO, YAlO$_3$, LAO, NdGaO$_3$, LSAT, (LaAlO$_3$)$_{0.3}$(Sr$_2$AlTaO$_6$)$_{0.7}$; STO, SrTiO$_3$; DSO, DyScO$_3$]. Each panel corresponds to a different substrate, with the corresponding epitaxial strain noted in parentheses. Solid lines are experimental data and dashed lines are fits using Eqs. 1 and 2. The horizontal solid line is the Mott-Ioffe-Regel limit according to Eq. 3.
in ultrathin nickelates (33–36). In contrast, $\rho_{\text{SAT}}$ is essentially thickness-independent. $\rho_{\text{SAT}}$ does depend, however, on the magnitude of epitaxial strain, as determined by the substrate. This is further illustrated in Fig. 2D, where $\rho_{\text{SAT}}$, $\rho_0$, and $\rho(0)$ are shown as a function of the in-plane strain $e_{xx}$ for 15-u.c. films.

Discussion

Tunable bad metal

A key result is that NdNiO$_3$ clearly exhibits resistance saturation in the high-temperature limit. It is thus a bad metal only in the sense that the saturation resistance exceeds the resistance predicted by the Mott-Ioffe-Regel...
criterion (Eq. 3), but not in the sense that its resistance escalates without saturation (as in the cuprates). In the following, we discuss the origins of this behavior and its correlation to the electronic structure of this system.

Several theoretical studies have pointed to the importance of orbital degeneracy and specific scattering mechanisms in determining $\rho_{\text{SAT}}$ (30, 32). Transport calculations that include interband scattering show that this produces a new conducting channel whose magnitude is proportional within the first order to the interband spacing $\Delta$ (29, 30). In this theory, interband currents act as a parallel conducting channel that reduces resistance, leading to a saturating resistance as described by Eq. 2, with $\rho_{\text{SAT}} \sim |\Delta|$. The behavior of $\rho_{\text{SAT}}$ as a function of strain in NdNiO$_3$ thin films can then be rationalized as a consequence of $e_g$ band splitting. In the rare-earth nickelates, two Ni $e_g$ bands cross the Fermi level. These are derived from orbitals having $x^2 - y^2$ and $3z^2 - r^2$ symmetry, which are degenerate in unstrained bulk material. Experiments have shown that epitaxial strain lifts degeneracy and causes orbital polarization, with tensile strains lowering the energy $E$ of $x^2 - y^2$ orbitals and with compressive strains lowering the energy $E$ of $3z^2 - r^2$ orbitals (37–39). Figure 5 shows the magnitude of the orbital splitting $|\Delta| = E(3z^2 - r^2) - E(x^2 - y^2)$ for NdNiO$_3$ as a function of strain, as estimated by density functional theory. With increasing epitaxial strain, orbital polarization increases, in keeping with prior experimental and theoretical findings (37–39), as does $\rho_{\text{SAT}}$. Moreover, electron-electron scattering strength ($A$ in Eq. 1) follows a similar trend with strain as $\rho_{\text{SAT}}$ (Fig. S4). This trend may also be an indication of the increasing importance of interband scattering, which can lead to resistance from electron-electron scattering, in addition to Umklapp processes (40).

Calculations for cuprates predict that these compounds will also saturate [which appears to be confirmed in the experiment (10)] but that $\rho_{\text{SAT}}$ will be very large because of the fact that only a single $x^2 - y^2$ orbital band crosses the Fermi level (32, 41) (in contrast to the nickelates studied here) and because of strong electron correlations. A large $\rho_{\text{SAT}}$ makes the second term in Eq. 2 small and causes a nonsaturating resistance. The experimental results confirm the importance of orbital degeneracy: as we lift degeneracy toward a more cuprate-like Fermi surface with a single band, $\rho_{\text{SAT}}$ increases.

Our results also show that although $\rho_{\text{SAT}}$ is sensitive to the degree of orbital polarization, it is relatively insensitive to disorder. This can be seen from the very different behaviors of $\rho_{\text{SAT}}$ and $\rho_0$ with decreasing film thickness: $\rho_0$ sharply increases presumably because of increased scattering by the surface, whereas $\rho_{\text{SAT}}$ remains approximately constant (Fig. 2C).

Using the data presented in Fig. 2C, we can make predictions for the occurrence of Anderson localization in this system. In particular, films become insulating at all temperatures when $\rho(0) = \rho_{\text{SAT}}$; in other

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**Fig. 3.** Strain-thickness phase diagram. (A) Prototypes for the four basic behaviors seen in the $p$-$T$ curves shown in Fig. 1. (B) Phase diagram for $e_{xx}$ versus $t_{\text{NNO}}$. The boundaries are drawn between the four basic behaviors shown in (A). Each point indicates a transport curve in Fig. 1. Black diamonds represent predictions for MIT based on $\rho(0) = \rho_{\text{SAT}}$.

**Fig. 4.** Strain-temperature phase diagram. Each panel corresponds to a different NdNiO$_3$ (NNO) thickness. Symbols indicate MIT temperatures measured for films under different strains. The colors of the regions correspond to those in Fig. 3, YAO, YAlO$_3$; LAO, LaAlO$_3$; NGO, NdGaO$_3$; LSAT, (LaAlO)$_3$Sr$_2$AlTaO$_6$; STO, SrTiO$_3$; DSO, DyScO$_3$.
words, \( \rho_0 \) becomes so large that, essentially at all temperatures, the first term in Eq. 2 is smaller than the second term and the resistance is dominated by \( \rho_{\text{SAT}} \). Such a material clearly cannot be a metal anymore, and films become insulating at all temperatures. The black diamonds in the phase diagram shown in Fig. 3B are predictions for the critical thickness for this transition, obtained by extrapolating \( \rho(0) \) to the point where it intersects with \( \rho_{\text{SAT}} \). They agree with the experimental transition within one unit cell. Because \( \rho_0 \) contains the effects of disorder, RNiO\(_3\) films that are insulating at all temperatures are strongly localized because of disorder, as has also been suggested in the literature (33, 34, 42). We loosely term this the Anderson insulator, although correlations presumably play a role and the insulator may be magnetic (43). The criterion \( [\rho(0) \approx \rho_{\text{SAT}}] \) established here has an interesting implication, namely, that the Anderson insulating state is tunable in a similar fashion as \( \rho_{\text{SAT}} \). Specifically, materials with a large \( \rho_{\text{SAT}} \) will require larger \( \rho_0 \) values to become insulating and can be considered as more disorder-tolerant, all other things being equal. Unlike \( A \) and \( \rho_{\text{SAT}} \), however, \( \rho_0 \) does depend on the sign of the strain and not only on its magnitude. This is attributable to \( \rho_0 \) being a function of the size of the Fermi surface (44), which changes with strain (38). Tensile strained films, with their larger \( \rho_0 \), fulfill \( \rho(0) \approx \rho_{\text{SAT}} \) at larger thicknesses than compressively strained films (see data on SrTiO\(_3\) in Fig. 2C).

**NFL behavior**

The phase diagrams shown in Figs. 3 and 4 illustrate the two distinct types of MITs: the quantum phase transition at 0 K between an NFL and an antiferromagnetic insulator (blue-yellow crossover) and the disorder-driven transition to the Anderson insulator (yellow-red crossover), when \( \rho(0) \approx \rho_{\text{SAT}} \). The quantum phase transition as a function of strain has been documented in relatively thick RNiO\(_3\) films (6, 45–47). The nature of this MIT—as being driven by Fermi surface nesting and spin density wave (promoting the insulating state) (38, 43, 48, 49) or in terms of bandwidth and charge-transfer energy (6, 45, 50)—has been extensively discussed in the literature. The insulating phase at low thicknesses has also been linked to the stabilization of the spin density wave order, acting similarly to tensile strain (37, 38). This work establishes that the location of the quantum phase transition is highly sensitive to both strain and confinement. This is reflected in the curved phase boundaries in Fig. 3 and, equivalently, in the continuous shift of the entire phase diagram toward compressive strain at low thickness in Fig. 4. Moreover, at high tensile strains, the Mott MIT and the disorder-driven Anderson transition are brought in increasingly close proximity to each other. The dual nature of the driving mechanisms promoting the insulating transition should be an important consideration in interpreting phenomena such as magnetism (34). The two transitions can be decoupled using strain (for example, as in NdNiO\(_3\) grown on LaAlO\(_3\)).

The results have further intriguing implications for the nature of NFL-LFL crossover in the metallic phase, which is abrupt and coincides with the suppression of the temperature-driven MIT. The exponent \( n \) is insensitive to disorder, which increases (according to \( \rho_0 \)) with decreasing film thickness (Fig. 2, B and C). This is contrary to the expectations of NFL behavior driven by spin fluctuations, which should yield \( n \) values that are highly sensitive to disorder (51). Furthermore, it is constant across all of the NFL phases observed here. \( n \approx \gamma/3 \) has also been observed for PrNiO\(_3\) and EuNiO\(_3\) under pressure when the temperature-driven MIT is suppressed (52, 53), as well as in overdoped cuprates (54, 55). This points to a common origin that requires further theoretical investigations. Specifically, future studies should address the question of whether a quantum critical point or a distinct NFL phase is the origin of NFL metal. Our results emphasize the need for a quantitative treatment of electronic structure and, in keeping with earlier suggestions (4, 5), that nickelates are a fertile ground to investigate aspects of the normal states of cuprates.

**Applications**

The phase diagram presented in Fig. 3 can be used as a guide in designing future devices based on controlling the MIT of nickelates. The M(NFL)\(\rightarrow\)I pocket at compressive strains and low film thicknesses could be useful for electrostatic control (56–60), which requires thin films to make carrier density modulation feasible while still permitting a sharp MIT with many orders of magnitude of resistance change. Strain control of MIT may also be of interest for low-voltage digital switches (61). On the tensile side of the phase diagram, the sharp M(NFL)\(\rightarrow\)I boundary is noteworthy because a small amount of strain, controlled by a piezoelectric material, can have a large effect on resistivity at temperatures relevant to practical devices. In particular, Fig. 3 shows a strain-tunable transition between the Anderson insulator \( [\rho(0) = \rho_{\text{SAT}}] \) and the Mott MIT.

**MATERIALS AND METHODS**

Films were grown by radio frequency magnetron sputtering in a 95% Ar/5% O\(_2\) mixture, a total pressure of 9 mTorr, and a sputter power of 15 W. The optimization of growth conditions, the structure and chemical composition of films, and the quantification of mismatch strains have been reported elsewhere (47). Methods used to characterize the films included high-resolution x-ray diffraction, scanning transmission electron microscopy, and Rutherford backscattering spectrometry, as described by Hauser et al. (47). Films of a given thickness were simultaneously deposited on different substrates. The magnitude of the in-plane strain was calculated as \( e_{xx} = (a_0 - a_\parallel)/a_0 \) where \( a_0 \) is the measured in-plane lattice constant and \( a_\parallel \) is the unstrained (intrinsic) lattice parameter, which was extracted from x-ray diffraction as described by Hauser et al. (47). Resistivity was determined from measurements in a van der Pauw configuration with Ni\(_{30}\) nm/Au\(_{300}\) nm ohmic contacts.

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**Fig. 5. Saturation resistance and orbital splitting as a function of strain.** (A) Schematic showing the lifting of \( e_g \) orbital degeneracy in NdNiO\(_3\). (B) Magnitude of the calculated orbital splitting \( |\Delta| \) in NdNiO\(_3\) and measured \( \rho_{\text{SAT}} \) as a function of \( \varepsilon_{xx} \). (Inset) Correlation between the two quantities.
Electronic structure calculations were performed using projector-augmented wave formalism (62), as implemented in the QUANTUM ESPRESSO package (63). Electronic wave functions were expanded up to a kinetic energy cutoff of 50 Ry. Brillouin zone integrations were performed on a \(8 \times 8 \times 8\) special \(k\)-point grid and using Methfessel-Paxton smearing (64) of the Fermi-Dirac distribution function with a smearing width of 0.01 Ry. We used the generalized gradient approximation for the exchange correlation functional [Perdew-Burke-Ernzerhof (PBE)] (65). The crystal structure was constrained to a tetragonal unit cell with the \(ab\) plane fixed to the lattice constant of the substrate, whereas the \(c\) lattice parameter was allowed to relax. For the calculation of crystal field splittings, we constructed maximally localized Wannier functions (66) and a real-space Hamiltonian in Wannier function basis. The splitting between the diagonal elements of the real-space Hamiltonian of the \(c\)-like Wannier functions for different strain configurations was interpreted as crystal-field splitting for the \(e_g\) states, defined as \(\Delta = H(z^2-R^2) - H(x^2-y^2, x^2-y^2)\), with \(H\) representing the real-space Hamiltonian in Wannier function basis. Although the PBE functional and the tetragonal unit cell did not reproduce the insulating and \(E\)-type antiferromagnetic ordering of bulk NdNiO\(_3\), they provided the correct qualitative behavior and trends for the dependence of \(\Delta\) as a function of strain. In addition, we expected the metallic solution to provide a better description of the electronic properties of metallic NdNiO\(_3\) films, compared to those of bulk NdNiO\(_3\), within the PBE functional.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/1/15/00797/DC1

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