



Correlated Insulating Behavior in Infinite-Layer Nickelates

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Unlike their cuprate counterparts, the undoped nickelates are weak insulators without long-range antiferromagnetic order. Identifying the origin of this insulating behavior, found on both sides of the superconducting dome, is potentially a crucial step in the development of a coherent understanding of nickelate superconductivity. In this work, we study the normal-state resistivity of infinite-layer nickelates using high magnetic fields to suppress the superconductivity and examine the impact of disorder and doping on its overall temperature (7) dependence. In superconducting samples, the resistivity of Nd- and La-based nickelates continues to exhibit weakly insulating behavior with a magnitude and functional form similar to that found in underdoped electron-doped cuprates. We find a systematic evolution of the insulating behavior as a function of nominal hole doping across different rare-earth families, suggesting a pivotal role for strong electron interactions, and uncover a correlation between the suppression of the resistivity upturn and the robustness of the superconductivity. By contrast, we find very little correlation between the level of disorder and the magnitude and onset temperature of the resistivity upturn. Combining these experimental observations with previous Hall effect measurements on these two nickelate families, we consider various possible origins for this correlated insulator behavior and its evolution across their respective phase diagrams.

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INTRODUCTION

The recent discovery of superconductivity in the infinite-layer nickelates (ILN) [1–4] represents the culmination of a three-decade-long search to successfully dope the $3d^9$ (Ni¹⁺) configuration in a square planar geometry as a means of replicating the structural and orbital motif found in high- T_c cuprates. Unlike the cuprates, whose parent ground state is a Mott insulator with long-range antiferromagnetic (AFM) order, the undoped ILN were found to be metallic at elevated temperatures with a crossover to a weakly insulating state below approximately 100 K, at which the resistivity starts to develop a moderate upturn. While static AFM order has thus far remained undetected in the nickelates, recent resonant x-ray scattering [5] and nuclear magnetic resonance (NMR) experiments [6, 7] reported signatures consistent with fluctuating AFM paramagnon excitations. Other NMR studies, however, claimed an absence of magnetic order in the nickelates [8]. The occurrence of weakly insulating behavior at a high hole doping level, beyond the range within which superconductivity is realized, further contrasts with the

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correlated but nonetheless metallic ground state found in highly overdoped cuprates [9]. Numerous theoretical calculations [10–22] have indeed pointed out that the 5*d* (and possibly 4*f*) band of the rare-earth (*RE*) elements contributes a finite density of states at the Fermi level, highlighting a fundamental difference between the two $3d^9$ oxides. The sizeable negative Hall coefficient [1, 3] and the finite spectral weight at the Fermi level [23] experimentally found in undoped nickelates appear to corroborate this picture.

Despite the recent progress in understanding the low-energy electronic structure of superconducting nickelates, an understanding of the anomalous insulating behavior that is ubiquitously found in ILN is lacking. Here, we present a systematic study of the normalstate transport of two doped families of ILN-the Nd- and La-based systems-by employing high magnetic fields up to 35 T to fully suppress the superconductivity. The effect of varying the rare-earth (RE) element on the functional form of the insulating resistivity, as well as the impact of (hole) doping and disorder level on the transport characteristics are also investigated. By taking into account the evolution of the Hall coefficient in both systems, we arrive at a number of salient points with regards to the origin of the insulating behavior: 1) The resistive upturns at low doping are likely to be due to a partial gapping of the states derived from the RE ions. 2) Hole doping x is much more effective in suppressing the resistivity upturn than a decrease in disorder (as inferred from the residual resistivity ratio). 3) The upturns, though notably weaker in the superconducting samples, nevertheless persist into the superconducting regime, and show a different functional form depending on the choice of RE. 4) In this region of the phase diagram, the insulating behavior is more likely to be associated with the correlated 3d states on the Ni. 5) The field dependence of the magnetoresistance in superconducting samples appears to rule out localization or the Kondo effect as the origin of the resistive upturns. 6) The RE dependence on the functional form of the low-Tresistivity, as well as its overall magnitude, are more reminiscent of that seen in electron-doped cuprates than in hole-doped cuprates. 7) Finally, we find that T_c in the nickelates is sensitive to the level of disorder, suggesting that superconductivity in the ILN is unconventional in nature.

MATERIALS AND METHODS

La_{1-x}Sr_xNiO₂ and PrNiO₂ thin films were grown by pulsed laser technique described in [3, 24], respectively. Electrical resistivity was measured with a four-point configuration using the ac lock-in technique, with an alternating current $I = 10 \,\mu\text{A}$ applied within the *ab*-plane at a frequency between 13 and 30 Hz. Static magnetic fields up to 35 T, applied parallel to the crystalline *c*-axis, were generated using a Bitter magnet at the High Field Magnet Laboratory in Nijmegen, the Netherlands.

RESULTS AND DISCUSSION

Figure 1 shows the *T*-dependent in-plane resistivity $\rho_{ab}(T)$ of a set of undoped *RENiO*₂ films (*RE* = La, Pr, Nd). Several key

features of its normal-state resistivity are revealed in these plots. Firstly, for all films, $\rho_{ab}(T)$ undergoes a resistivity minimum (ρ_{\min}) at $T = T_{\min}$ that delineates the metallic regime from the insulating-like regime at lower temperatures. Secondly, the absolute values of ρ_{ab} show significant variation between samples, with the newer generation exhibiting lower absolute resistivities as well as a reduced level of disorder, as inferred from the higher ρ_{300K}/ρ_{min} ratios. As $\rho_{300\text{K}}/\rho_{\text{min}}$ increases, T_{min} shifts to lower values, suggesting that disorder plays some role in the insulating behavior, at least in the parent compound(s). (The resistivity of LaNiO₂ from an early report [25] was found to be an exceptionally low yet its $ho_{300\text{K}}/
ho_{\min}$ ratio is the lowest among all samples investigated, the origin of which is yet unclear). Thirdly, in the high-*T* metallic regime for PrNiO₂ and NdNiO₂, the slope $d\rho_{ab}/dT$ is found to be very similar despite a large variation in their absolute values. This suggests that the excess disorder, while increasing the impurity scattering rate (and the magnitude of ρ_{ab}), does not significantly affect the intrinsic metallic resistivity. Fourthly, the functional form of the resistive upturn over the accessible temperature range depends on the choice of RE. In LaNiO₂, for example, $\rho_{ab}(T)$ initially follows a log(1/*T*) behavior for $T < T_{min}$ but then tends towards a constant value below 10 K. In contrast, $\rho_{ab}(T)$ in PrNiO₂ and NdNiO₂ $\rho_{ab}(T) \propto \log(1/T)$ down to the lowest measured temperatures. Whether or not $\rho_{ab}(T)$ in (Pr, Nd) NiO₂ saturates below \approx 2 K, however, remains to be seen.

An emerging picture for the electronic structure of undoped ILN, based on recent spectroscopic studies and realistic theoretical calculations with electron interaction taken into account [15, 16, 23, 26, 27], indicates that its Fermi surface comprises a small electron pocket with a dominant character of the *RE* 5*d* band (which hybridizes with the Ni $3d_{z^2}$ band). The Ni $3d_{x^2-y^2}$ band, on the other hand, is split into the upper and lower Hubbard bands and thus does not directly contribute to the Fermi level. The Hall coefficient $R_{\rm H}(T)$ in both LaNiO₂ and $NdNiO_2$ is found to be negative [3, 28], consistent with the notion that the 3d states on the Ni sites are Mott localized and that the longitudinal and Hall conductivities are dominated by the electron pocket derived from the RE 5d states. Hence, it is these states that must be responsible for the resistive upturns in the parent compounds. Secondly, in both systems, T_{\min} is found to mark the onset of a marked increase in $R_{\rm H}(T)$, possibly indicating some form of gap opening below T_{\min} . Thirdly, the fact that $\rho_{ab}(T)$ appears to saturate eventually, at least in LaNiO₂ (and possibly in PrNiO₂ too), implies that this gapping is only partial and that a finite density of states remains on the electron pocket(s) whose low-T ground state is ultimately metallic.

According to the conventional Drude transport model, the electrical conductivity σ is given by

$$\sigma = \sum_{i} n_{i} e \mu_{i} = \sum_{i} \frac{n_{i} e^{2} \tau_{i}}{m_{i}^{\prime}}, \qquad (1)$$

where *i* denotes the distinct channel of conducting carriers, *e* is the elementary charge, and (μ, τ, m^*) denote the associated



FIGURE 1 In-plane resistivity versus temperature $\rho_{ab}(T)$ for undoped infinite-layer nickelates with selected rare earth elements. (A) Data from a previous generation of samples of PrNiO₂ [2], NdNiO₂ [28], and LaNiO₂ [25] are shown in solid points; data from a new generation of samples of LaNiO₂ [3] and PrNiO₂ (this work) in open points. (B) Normalized resistivity $\rho(T)/\rho_{300 \text{ K}}$ in linear-log scale with the same color code as in (A). Vertical bars indicate T_{min} , the temperature at which ρ_{ab} shows a minimum.



FIGURE 2 | Hall mobility $\mu_{\rm H}$ of undoped LaNiO₂ and NdNiO₂. $\mu_{\rm H}$ is estimated using $\mu_{\rm H} = R_{\rm H}/\rho$ with the Hall coefficient data $R_{\rm H}$ reported in [3, 28] and ρ as shown in **Figure 1**. The locations of $T_{\rm min}$ are marked by vertical arrows.

mobility, relaxation time, and effective mass, respectively. Consequently, $\rho = 1/(ne\mu) = R_H/\mu$ for a single-band metal, where $R_H = \frac{1}{ne}$ is the Hall coefficient. From **Eq. 1**, it can be seen that a reduction of conductivity (i.e. a metal-insulator transition) can be caused by a reduction of *n* (loss of carrier) or τ (increased scattering rate), an increase in m^* (effective mass enhancement), or a combination of these factors. For simplicity, here we estimate the carrier mobility using the measured R_H , known as the Hall mobility $\mu_H = R_H/\rho$ for undoped LaNiO₂ and NdNiO₂, for which the single-band picture is most likely to apply. As shown in **Figure 2**, both LaNiO₂ and NdNiO₂ show a relatively unchanged μ_H above T_{min} , below which μ_H increases moderately. Crucially, the increase in μ_H below T_{min} indicates that the increase in ρ below T_{\min} is not related to a reduction of mobility (i.e. a change in τ/m^*), but is most likely caused by a reduction in n. The minimization of $R_{\rm H}$ at low T as x approaches 0.20, at which T_{\min} and $\rho_0 - \rho_{\min}$ are most suppressed (see **Figure 4**) further supports this scenario. Possible candidates responsible for the loss of carriers below T_{\min} include the emergence of a secondary order parameter (e.g. magnetic, charge, or stripe order), the opening of a pseudogap that partially depletes the density of states at the Fermi level [29], or a transfer of spectral weight to higher energy [30, 31].

With hole-doping, the situation evolves in a systematic fashion. In $Nd_{1-x}Sr_xNiO_2$, we revealed previously by destroying superconductivity with a large magnetic field, that the resistivity upturn, though persisting throughout the doping range of superconductivity, is progressively suppressed, essentially vanishing as x approaches the edge of the superconducting dome $x_c \approx 0.225$ [32]. Here, we examine the evolution of the low-T resistivity in $La_{1-x}Sr_xNiO_2$ in the fieldinduced normal state for $0.15 \le x \le 0.20$, i.e. across much of the superconducting doping range [3]. A contrasting behavior manifests in the functional form of $\rho_{ab}(T)$ below T_c , as shown in **Figure 3**. Similar to the undoped compound, $\rho_{ab}(T)$ in the field-induced normal state of superconducting La1-xSrxNiO2 exhibits an initial $\log(1/T)$ -behavior followed by a leveling off as $T \rightarrow 0$. The magnitude of the resistive upturn decreases as x increases from 0.15 to 0.18, after which it again increases at x = 0.20.

The overall magnitude of the resistive upturn, on the order of 10% between 0.5 K and T_{min} , is considerably smaller than that observed in the underdoped hole-doped cuprates (\geq 100%) [33, 34, 35] but is comparable with that reported in the electron-doped cuprates $RE_{2-x}Ce_xCuO_4$ below optimal doping [36–38]. A direct comparison of the low-*T* resistivities in the ILN and $RE_{2-x}Ce_xCuO_4$ is shown in **Figure 3C**. Intriguingly, the functional form of the low-*T* resistivity in $RE_{2-x}Ce_xCuO_4$ also



FIGURE 3 | Normal-state resistivity of superconducting nickelates at low temperatures. (A) $\rho_{ab}(T)$ of La_{1-x}Sr_xNiO₂ and (B) Nd_{1-x}Sr_xNiO₂ [32] thin films with 0.15 $\leq x \leq 0.20$ measured at zero applied magnetic field (lines) and at 35 T (solid points). Magnetic field is applied along the crystalline *c*-axis. Right axis shows the estimates of (k_F)⁻¹ assuming a two-dimensional free electron model (see main text for details). (C) Comparison of resistivity normalized by its 10 K value in the field-induced normal state, $\rho/\rho_{10 \text{ K}}$, for selected hole-doped nickelates (x = 0.15) and electron-doped cuprates below optimal dopings as specified. LCCO: representative rho_{ab}(T) La_{2-x}Ce_xCuO₄ measured at $\mu_0 H = 10$ T [38]. NCCO: representative rho_{ab}(T) Nd_{2-x}Ce_xCuO₄ measured at $\mu_0 H = 14$ T [37]. Note that the temperature axes are shown in log-scale and a vertical shift is applied to (C) for clarity. For LCCO, a rescaling factor of 0.50 is applied to the change in $\rho/\rho_{10 \text{ K}}$, which does not affect the functional form of $\rho(T)$.

depends on the RE elements in a similar manner to what is seen in the ILN. For $La_{2-x}Ce_{x}CuO_{4}$ (LCCO, x = 0.08) $\rho_{ab}(T)$ appears to saturate below 4 K, while for $Nd_{2-x}Ce_xCuO_4$ (NCCO, x = 0.14), $\rho_{ab}(T) \propto \log(1/T)$ down to the lowest measured temperature. This close alignment to the experimental situation in the n-doped cuprates is curious, but may simply be a consequence of the way in which carriers are doped into each system. In the cuprates, doped holes sit preferentially on the O sites while doped electrons reside on the Cu sites [39]. In the ILN, it is thought that the carriers are also introduced directly into the $3d_{x^2-y^2}$ orbital on the Ni sites [40]. At the same time, the similarities found in the low-T $\rho_{ab}(T)$ behavior of the ILN (for which no long-range AFM order exists at half-filling) and the *n*-doped cuprates suggests that the resistive upturns in the latter are not necessarily caused by shortrange spin correlations, as is believed to be the case for the pdoped cuprates.

The evolution of $R_{\rm H}(T)$ with doping in both ILN families is qualitatively the same, with a gradual reduction in the overall magnitude of $R_{\rm H}$ culminating in a crossover from negative to positive $R_{\rm H}(0)$ (the Hall coefficient in the low-*T* limit) around optimal doping [1, 3]. In any two-band metallic system, the sign of $R_{\rm H}(0)$ reflects the sign of the most mobile carriers. Hence, the observed sign change signals a delocalization of the 3*d* hole states on the Ni sites with hole-doping until eventually, they become the most mobile carriers in each system. Nevertheless, the fact that $\rho_{ab}(T)$ continues to exhibit a logarithmic divergence (at least in Nd_{1-x}Sr_xNiO₂) implies that these carriers are also prone to some form of localization, however weak. (Note that $R_{\rm H}(T)$ exhibits no upturns within the superconducting doping range, and so it is unlikely that the resistive upturns here are due to partial gapping).

It was noted early on that the insulator-to-metal crossover in the cuprates occurs at a threshold value of $k_F \ell > 10$ for both the

hole- [35] and electron-doped [36] compounds, far higher than the usual criterion $k_{\rm F}\ell \approx 1$. Assuming that the suppression of the resistive upturn in Nd_{0.775}Sr_{0.225}NiO₂ reflects a metallic ground state and using the two-dimensional free electron model [35]:

$$\rho_{ab}/d = h/\left(e^2 k_{\rm F}\ell\right),\tag{2}$$

where *d* is the *c*-axis lattice spacing, $k_{\rm F}$ is the Fermi wavevector, and *l* is the electronic mean free path, we find a threshold $k_{\rm F}l \approx 2 - 10$ for low-*T* metallicity in the ILN. We note, however, that the assumption of a single-band, 2D Fermi surface is likely not valid for the entire series of hole-doped nickelates (due to the expected presence of a 3D electron pocket derived from the 5*d* band of *RE* elements); therefore the estimates of $k_{\rm F}l$ here should be interpreted with caution.

Several proposals have been put forward to explain the anomalous upturn in the normal-state resistivity in the nickelates [31, 41, 42]. Two well-known mechanisms to produce a logarithmically diverging resistivity at low *T* are weak localization due to disorder [43, 44] and Kondo scatterings due to magnetic impurities [14, 45]. In both circumstances, however, a strong negative magnetic-field dependence of the insulating resistivity is expected, which is not observed in the nickelates. Moreover, a monotonic suppression of the insulating behavior with decreasing residual resistivity, expected for a localization-driven origin, is not seen (**Figure 4**) while the re-entrant insulating behavior found at high dopings also cannot be naturally explained by a Kondo-like mechanism.

In order to gain further insights into the origin of the resistive upturns, we have examined the impact of disorder and doping on the insulating characteristics in the ILN, namely the onset temperature $(T_{\rm min})$ and the size of the resistivity upturn $(\rho(T \rightarrow 0) - \rho(T_{\rm min}))$, denoted as $\rho_0 - \rho_{\rm min})$, as shown in **Figure 4**. We find no clear correlations



between T_{\min} and ρ_{\min} (Figure 4A) nor between $\rho_0 - \rho_{\min}$ and ρ_{\min} (Figure 4B), for both Nd_{1-x}Sr_xNiO₂ and La_{1-x}Sr_xNiO₂. Meanwhile, T_{\min} and $\rho_0 - \rho_{\min}$ both appear to collapse near *x* = 0.20 (Figures 4C,D), though deviations from the overall trends are visible both at zero doping and at the highest dopings. Notably, while T_{\min} and $\rho_0 - \rho_{\min}$ are gradually suppressed with improved sample quality (Figure 1B), varying x is seen as much more effective in suppressing the insulating behavior, suggesting that it is sensitive to carrier screenings and primarily driven by electron correlation effects. A number of non-Fermi-liquid models have been proposed to explain the anomalous insulating behavior seen in underdoped cuprates, including those based on the marginal Fermi liquid [46], the 2D Luttinger liquid [47] and the polaronic Bose liquid [48] model. The relevance of these more exotic models to the ILN, whose distinction from the (hole-doped) cuprates has become increasingly established, remains to be examined.

Lastly, we examine the impact of disorder on the critical temperature of superconducting nickelates. **Figure 5** shows a compilation of Nd_{0.8}Sr_{0.2}NiO₂ resistivity data reported to date [1, 28, 32, 49–53]. A large difference in the absolute values of ρ_{ab} is found, with ρ_{ab} (300 K) ranging from ~ 0.1–6.75 m Ω cm for nominally the same samples. Meanwhile, the agreement in the normalized resistivity $\rho/\rho_{300 \text{ K}}$ is much better across different reports, as shown in **Figure 5B**, with a good overlap found in 6 out of 9 traces. This suggests the

discrepancy in the absolute resistivities arises from geometric uncertainties. Importantly, we find that T_c depends strongly on the residual resistivity ratio, defined as $\rho_{300 \text{ K}}/\rho_{20 \text{ K}}$, with T_c increasing from $\approx 5 \text{ K}$ to over 12.5 K as $\rho_{300 \text{ K}}/\rho_{20 \text{ K}}$ increases. Such a strong dependence of T_c with respect to the level of disorder points to an unconventional nature of the superconductivity in the nickelates, and hints at a possible further increase in T_c with improved sample quality.

CONCLUSION

In summary, by suppressing superconductivity with high magnetic fields, we find the unusual resistivity upturn in the undoped infinite-layer nickelates persists into the superconducting regime and appears to be maximally suppressed near x = 0.20 in both La- and Nd-based systems. The resilience of the resistivity upturn against magnetic fields rules out localization and Kondo effect as its origin, and points to a partial gapping of the states with dominant RE 5d character as its cause at low doping as supported by Hall mobility analysis. In the superconducting doping range, the resistive upturn is found to be highly reminiscent in both its functional form and its overall magnitude to that found in electron-doped cuprates, which suggests the insulating behaviour is associated with the correlated Ni 3d states. While disorder has only a minor impact on the insulating behavior, the robustness of



FIGURE 5 [Impact of disorder on the critical temperature T_c of Nd_{0.8}Sr_{0.2}NiO₂. (**A**) A compliation of representative resistivity data on Nd_{0.8}Sr_{0.2}NiO₂ tims reported in [1, 28, 32, 49–53]. Data reported in [49] is rescaled by a factor of 0.4 for clarity. (**B**) Normalized resistivity $\rho/\rho_{300 \text{ K}}$ of data shown in (**A**). (**C**) T_c versus residual resistivity ratio $\rho_{300 \text{ K}}/\rho_{20 \text{ K}}$. T_c is defined as the midpoint of the resistive superconducting transition and the error bars reflect the 10–90% transition width. The same color code is applied to all panels. Inset: The impact of disorder on T_c of Bi-based cuprates. For both Bi₂(Sr, La)₂CuO_{6+δ} (Bi2201) and Bi₂Sr₂CaCu₂O_{8+δ} (Bi2212), T_c is found to be strongly reduced from the optimized value T_{c0} with an increase in residual resistivity per CuO₂ plane (ρ_0^{2D} , similar to that found in Nd_{0.8}Sr_{0.2}NiO₂. Data reproduced from [54].

superconductivity is strongly affected by the level of disorder, pointing towards the unconventional nature of nickelate superconductivity.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

YTH, HYH and NEH conceived the experiments. MO, BW, SH, KL, and DL grew and prepared the thin-film samples. YTH, MB, CD, and SW performed the resistivity measurements. YTH and NEH analyzed the data and wrote the manuscript with contribution from all authors.

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