Infinite-layer Nd_{1-x}Sr_{x}NiO_{2} thin films with Sr doping level $x$ from 0.08 to 0.3 were synthesized and investigated. We found a superconducting dome to be between $0.12 < x < 0.235$ which is accompanied by a weakly insulating behaviour in both underdoped and overdoped regimes. The dome is akin to that in the electron-doped 214-type and infinite-layer cuprate superconductors. For $x \geq 0.18$, the normal state Hall coefficient ($R_{H}$) changes the sign from negative to positive as the temperature decreases. The temperature of the sign changes monotonically decreases with decreasing $x$ from the overdoped side and approaches the superconducting dome at the mid-point, suggesting a reconstruction of the Fermi surface as the dopant concentration changes across the center of the dome.
The recent discovery of superconductivity in doped infinite-layer nickelate Nd$_{0.8}$Sr$_{0.2}$NiO$_2$ thin films has hailed another milestone in the field of high-temperature superconductivity [1]. In contrast to the antiferromagnetic (AF) order in the cuprates, the parent compound NdNiO$_2$ shows no sign of magnetic order down to a temperature of 1.7 K [2], despite its similarities in the crystalline and electronic structures to the high-$T_c$ cuprates. This severely challenges the scenario of AF spin fluctuation as the superconducting pairing mechanism which is prevalent in the cuprates [3-5]. This discovery may represent a new type of superconductivity and has motivated numerous theoretical work to explore the similarities and differences to the cuprates [6-21]. For example, it is proposed that the low-density Nd 5$d$ conduction electrons couple with the localized Ni-3$d$ electron, suppressing the long-range AF order and forming Kondo spin singlets, leading to the self-doped Mott insulator in the parent compound [15]. In Nd$_{1-x}$Sr$_x$NiO$_2$, the doped holes reside in the Ni ions in contrast to the cuprates in which the holes are located at the oxygen site due to the strong hybridization of Cu 3$d_{x^2-y^2}$ and O 2$p$ orbitals [11, 14]. Electronic-structure calculations showed that RNiO$_2$ ($R =$ La, Pr, Nd) are promising as cuprate analogs with a similarly large value of the long-range hopping $t'$ and the $e_g$ energy splitting [6]. Moreover, the role of Nd 4$f$ state in NdNiO$_2$, which shows no substantial effect in cuprates, has also been investigated [7, 9]. The $t$-$J$ model study proposed a $d$-wave symmetry of paring, analogous to cuprates [12, 13]. Experimentally, X-ray spectroscopy revealed a weakly interacting three-dimensional 5$d$ metallic state in the rare-earth spacer layer, hybridizing with the strongly correlated 3$d_{x^2-y^2}$ symmetric state in the NiO$_2$ layer, differing from the cuprates [22]. Spin fluctuations in bulk NdNiO$_2$ have been explored by Raman spectroscopy and showed the AF exchange of $J = 25$ mV [23]. Nd$_{1-x}$Sr$_x$NiO$_2$ ($x =$ 0, 0.2, 0.4) prepared in bulk form shows the absence of superconductivity even under 50.2 GPa pressure, probably due to the creation of nickel deficiency during the reduction process [24]. In addition, attempting to grow the infinite-layer films at low oxygen partial pressure without chemical reduction resulted in insulating thin films [25]. It is clear that the synthesis of the superconducting nickelate is challenging and critical.
One important feature in most unconventional superconductors is that the superconductivity is caused by charge doping in the parent compound, and there is superconducting dome embedded within other competing ordered phases such as insulator and metal [26, 27]. For the isostructural cuprates, increasing chemical doping causes the transition from Mott insulators to high-$T_c$ superconductors and then Fermi-liquid metals [26]. Moreover, charge doping suppresses the long-range AF order forming spin fluctuations in the CuO$_2$ planes, which is considered as the pairing mechanism in the cuprates. The type of the dopant atom and the dopant level in the firstly synthesized nickelate superconductor, Nd$_{0.8}$Sr$_{0.2}$NiO$_2$, were initially inspired by the need to achieve a maximum conductivity in the La$_{1-x}$Sr$_x$NiO$_2$ ($x$=0.2) through substituting La with a smaller ionic radius atom Nd and thus increasing the electronic bandwidth [1]. In addition, the normal-state Hall coefficient of Nd$_{0.8}$Sr$_{0.2}$NiO$_2$ showed a negative sign at room temperature, in contrast to the expectation of hole doping by Sr. Whether the superconductivity and normal state transport properties are doping dependent in Nd$_{1-x}$Sr$_x$NiO$_2$ is not clear [28]. Here, we synthesized infinite-layer Nd$_{1-x}$Sr$_x$NiO$_2$ with different Sr doping levels and found a superconducting dome ranging between $0.12 < x < 0.235$ with weak insulator regions in both underdoped and overdoped regimes beside the dome. The overdoped regime behaves differently from the high-$T_c$ cuprates, wherein Fermi liquid metal behaviour is seen.

The ceramic targets were prepared by sintering the mixed powders of Nd$_2$O$_3$, SrCO$_3$ and NiO (weighted according to the chemical formula of Nd$_{1-x}$Sr$_x$NiO$_3$) in air for 15 h at 1200, 1220 and 1250 °C, respectively, with regrinding before each sintering. On the final sintering, the powder was pressed into a disk-shaped pellet. Thin films with a thickness of 35 nm were grown on TiO$_2$-terminated (001) SrTiO$_3$ (STO) substrates by a pulsed laser deposition (PLD) system with a 248-nm KrF excimer laser. No STO capping layer is deposited for all samples in this study. The deposition temperature $T$ and oxygen partial pressure $P_{O_2}$ for all samples were 600 °C and 150 mTorr, respectively. The laser energy intensity on the target surface is set to be 2 Jcm$^{-2}$. After
deposition, the samples were cooled to room \( T \) in the deposition pressure at a rate of 8 °C/min. In previous reports, the infinite layer nickelates were obtained by chemical reduction in a sealed vacuum Pyrex glass tube [1, 2, 29-31]. This is crucial since the process requires reduced ambient and the \( \text{H}_2 \) gas produced by the heating of \( \text{CaH}_2 \) to arouse the gas-phase reaction with the sample. Here, we realized the chemical reduction in a vacuum chamber. After deposition, the sample was cut into several pieces with a size of 1.3 × 2.5 mm\(^2\). The piece was then embedded with about 0.1 g of \( \text{CaH}_2 \) powder and wrapped in aluminium foil and then placed into a vacuum chamber. The wrapped sample was heated to 340-360 °C at a rate of 25 °C/min and kept for 80-120 mins and then cooled down to room \( T \) at a rate of 25 °C/min. During the heating process, the chamber was sealed and the pressure changed from the background of 1×10\(^{-5}\) Torr to around 1×10\(^{-1}\) Torr, and then stabilized at around 1×10\(^{-1}\) Torr during the whole reduction process. The increase of pressure may be caused by the release of \( \text{H}_2 \) gas from \( \text{CaH}_2 \) upon heating or/and \( \text{H}_2\text{O} \) gas due to the chemical reaction between the sample and \( \text{CaH}_2 \) [32]. The X-ray diffraction (XRD) measurement was done in the X-ray Diffraction and Development (XDD) beamline at Singapore Synchrotron Light Source (SSLS) with a wavelength of \( \lambda = 1.5404 \) Å. The wire connection for electrical transport measurement was done by Al ultrasonic wire bonding. The transport measurements were performed using a Quantum Design Physical Property Measurement System (PPMS).

Figure 1 shows the XRD \( \theta-2\theta \) patterns of \( \text{Nd}_{1-x}\text{Sr}_x\text{NiO}_3 \) thin films with Sr doping level \( x \) from 0.08 to 0.3 after chemical reduction. The structural characterization of as-grown \( \text{Nd}_{1-x}\text{Sr}_x\text{NiO}_3 \) thin films can be found in Fig. S1 [33]. Only (00\(l\)) infinite layer peaks were observed, where \( l \) is an integer, confirming the transformation from \( \text{Nd}_{1-x}\text{Sr}_x\text{NiO}_3 \) perovskite to corresponding \( \text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2 \) infinite-layer structures after reduction. As shown in Fig. 1, the thin film peaks slightly shift to lower angles as \( x \) increases, indicating an expansion of the \( c \)-axis. The \( c \)-axis lattice constants \( d \) as a function of \( x \) are plotted in the inset of Fig. 1. It is observed that \( d \) increases with the increase of
Figure 2 shows the resistivity versus temperature (\( \rho - T \)) curves of \( \text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2 \) thin films for \( x \) from 0.08 to 0.3. The \( \rho - T \) curves of the as-grown \( \text{Nd}_{1-x}\text{Sr}_x\text{NiO}_3 \) thin films can be found in Fig. S2 and other multiple \( \text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2 \) samples for \( x \) from 0.12 to 0.3 can be found in Fig. S4 [33]. For \( 0.08 \leq x \leq 0.12 \), the samples show metallic behaviour at high \( T \), with resistivity upturns at low \( T \) between 27-35 K, which is similar to the observation in underdoped cuprates. For \( 0.135 \leq x \leq 0.22 \), the samples also behave like a metal down to the \( T \) near superconducting transition and then evolve into superconducting states. The zoomed-in \( \rho - T \) curves near transition are shown in Fig. 2(b). One example of magnetic field-dependent \( \rho - T \) curves for \( x = 0.2 \) is shown in Fig. S3 [33], the \( T_c \) is suppressed with increasing field, further confirming the occurrence of superconductivity. For higher doping level \( x \geq 0.235 \), the resistive behaviour is similar to underdoped ones but shows lower resistivity upturn \( T \) of 12 - 18 K.

Figure 3(a) shows the temperature dependence of the normal-state Hall coefficient \( R_H \) for \( \text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2 \) thin films. At room \( T \), the \( R_H \) of all samples shows a negative sign and the magnitudes (\( |R_H| \)) show a general decrease with increasing \( x \) and a sign of saturation at \( x > 0.2 \) (Fig. 3(b)), indicating that the charge carrier is an electron and the electron density is increased with \( x \) (considering the carrier density \( n = 1/(e \cdot R_H) \), where \( e \) is the electron charge). This is inconsistent with the expectation of single-band hole doping as a result of Sr substitution for Nd. The \( R_H \) is negative below 300 K for samples with \( x \leq 0.15 \), while it undergoes a smooth transition from negative to positive sign at low-temperature \( T_H \) for samples with \( x \geq 0.18 \). For the samples with \( x \leq 0.135 \), the \( |R_H| \) increases moderately with decreasing \( T \) to 200 - 100 K depending on \( x \) and then decreases with further decreasing \( T \). This behaviour is similar to the observation in both electron-doped \( \text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4 \) and hole-doped \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) cuprates [34-36]. For samples with \( x = 0.15 \), the \( |R_H| \) decreases monotonically as the \( T \) is reduced. For samples with \( x \geq 0.18 \), the \( |R_H| \) decreases with
decreasing $T$ at the negative side and increases at the positive side. This indicates that for those samples, the cancellation of electron and hole contributions occurs as the $T$ decreases. Even though the electron density is higher (lower $|R_{\text{H}}|$) at a higher Sr doping level, the hole contribution is much more obvious, leading to higher $T_{\text{H}}$ at higher $x$ (Fig. 4).

Even though the room-temperature electron density generally increases with $x$, the $\rho$ does not follow a similar trend with increasingly resistive at the overdoped regime for $x \geq 0.235$ (Fig. 2). This counters the trend in over-doped cuprates and could be attributed to the disorder or defect induced in the reduction process. Indeed, previous results in LaNiO$_2$ showed that the resistivity is different in samples reduced by different reduction agents, or at different $T$, even though the films show the similar infinite-layer peaks in XRD [32]. More recently, nickel-deficient bulk Nd$_{1-x}$Sr$_x$NiO$_2$ ($x = 0, 0.2, 0.4$) showed insulating behaviour, also suggesting the influence of composition on resistivity [24]. Even for the cuprate Nd$_{1.85}$Ce$_{0.15}$CuO$_{4+\delta}$ in which reduction is required to obtain superconductivity, the normal state resistivity could be affected when over-reduction occurs [37]. Moreover, we might speculate that during the chemical reduction process, the STO substrates were reduced to generate oxygen vacancies and make STO itself metallic, since the defect levels of oxygen vacancies in STO are shallow (~ 4 - 25 meV) [25, 38, 39]. To rule out this possibility, we treated the bare STO substrate in the same reduction process and found that it is highly resistive beyond the measurement limit, confirming the negligible effect of STO on the electrical transport of the films.

Figure 4 shows the phase diagram of Nd$_{1-x}$Sr$_x$NiO$_2$. The onset $T_c$ is defined to be the temperature at which the resistivity drops to 90% of the value at 15 K. A superconducting (SC) dome ranging $0.12 < x < 0.235$ is seen, which is comparable to that in a recent report [28]. Li et. al. found a small dip at $x = 0.2$ in the dome, which is also visible in our results while it is at $x = 0.18$ [28]. A similar dip has also been seen in La$_{2-x}M_x$CuO$_4$ ($M =$ Sr and Ba) at $x =$1/8 which is ascribed to the charge stripe order [40, 41]. However, the dip in nickelate needs to be further explored since there exists
a variation of the dip position. The SC dome of Nd$_{1-x}$Sr$_x$NiO$_2$ is much narrower compared to the canonical hole-doped cuprate La$_{2-x}$Sr$_x$CuO$_4$ [40]. Note that even for 214-type cuprates with similar crystal structures, the dome range of electron-doped R$_{2-x}$Ce$_x$CuO$_4$ ($R =$ Nd and Pr, $0.10 < x < 0.24$) is narrower than that of La$_{2-x}$Sr$_x$CuO$_4$ ($0.6 < x < 0.25$) [40, 42, 43], suggesting the asymmetric SC dome at asymmetric charge doping. The dome range of Nd$_{1-x}$Sr$_x$NiO$_2$ is much more similar to that of Pr$_{2-x}$Ce$_x$CuO$_4$. More interestingly, the $R_{II}$ of Pr$_{2-x}$Ce$_x$CuO$_4$ is negative for $x \leq 0.15$ and changes sign at low $T$ for $x \geq 0.16$, which is similar to that of Nd$_{1-x}$Sr$_x$NiO$_2$ observed here [34]. Moreover, the comparable dome ($0.05 < x < 0.15$) was also observed in the electron-doped infinite layer cuprate Sr$_{1-x}$La$_x$CuO$_2$ [44]. Therefore, the SC dome of nickelate Nd$_{1-x}$Sr$_x$NiO$_2$ behaves more like the one observed in electron-doped cuprates. The temperature $T_{II}$ where the $R_{II}$ changes from negative to positive sign for the sample with $x \geq 0.18$ is also plotted in Fig. 4. As $x$ decreases from the overdoped side, $T_{II}$ monotonically decreases. A simplified two-band model with both electron- and hole-like Fermi surfaces is schematically showed in the inset of Fig. 4. The $T_{II} \sim x$ curve separates the normal state regime into two panels, with the right panel dominated by hole-like band and left panel by electron-like band. With increasing Sr doping and temperature, the band which dominates the normal state electrical transport, evolves from electron- to hole-like bands. Therefore, the $R_{II}$ changes sign from negative to positive. It is worth noting that the extension of the $T_{II} \sim x$ curve reaches the middle of the superconducting dome, suggesting a reconstruction of the Fermi surface as the doping changes across the middle point of the dome.

In summary, we have synthesized the infinite-layer Nd$_{1-x}$Sr$_x$NiO$_2$ thin films with doping level $x$ from 0.08 to 0.3 and found a SC dome ranging $0.12 < x < 0.235$. In both underdoped and overdoped regimes adjacent to the dome, weakly insulating behaviour is observed, which is different from the high-$T_c$ cuprates in which Fermi liquid metal is seen in the overdoped regime. The dome range is comparable to that in the electron-doped infinite-layer Sr$_{1-x}$La$_x$CuO$_2$ and 214-type Pr$_{2-x}$Ce$_x$CuO$_4$. Moreover, in the current study, the synthesis of infinite-layer nickelate thin films via chemically
topotactic reduction was realized in a sealed vacuum chamber, facilitating future studies of \textit{in-situ} reaction by-product monitoring, and thus control of the reduction process.

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\textbf{References}

[1] D. Li, K. Lee, B. Y. Wang, M. Osada, S. Crossley, H. R. Lee, Y. Cui, Y. Hikita, and H. Y. Hwang, \textit{Superconductivity in an infinite-layer nickelate}, Nature \textbf{572}, 624 (2019).
[2] M. A. Hayward and M. J. Rosseinsky, \textit{Synthesis of the infinite layer Ni(I) phase NdNiO\textsubscript{2+x} by low temperature reduction of NdNiO\textsubscript{3} with sodium hydride}, Solid State Sci. \textbf{5}, 839 (2003).
[3] P. W. Anderson, \textit{The Resonating Valence Bond State in La\textsubscript{2}CuO\textsubscript{4} and Superconductivity}, Science \textbf{235}, 1196 (1987).
[4] G. A. Sawatzky, \textit{Superconductivity seen in a non-magnetic nickel oxide}, Nature \textbf{572}, 592 (2019).
[5] R. M. Wilson, \textit{Superconductivity is found in a nickel oxide}, Phys. Today \textbf{72}, 19 (2019).
[6] A. S. Botana and M. R. Norman, \textit{Similarities and Differences between LaNiO\textsubscript{2} and CaCuO\textsubscript{2} and Implications for Superconductivity}, Phys. Rev. X \textbf{10}, 011024 (2020).
[7] P. Jiang, L. Si, Z. Liao, and Z. Zhong, \textit{Electronic structure of rare-earth infinite-layer RNiO\textsubscript{2} (R=La, Nd)}, Phys. Rev. B \textbf{100}, 201106(R) (2019).
[8] Y. Nomura, M. Hirayama, T. Tadano, Y. Yoshimoto, K. Nakamura, and R. Arita, \textit{Formation of a two-dimensional single-component correlated electron system and band engineering in the nickelate superconductor NdNiO\textsubscript{2}}, Phys. Rev. B \textbf{100}, 205138 (2019).
[9] M. Y. Choi, K. W. Lee, and W. E. Pickett, Role of 4f states in infinite-layer NdNiO$_2$, Phys. Rev. B 101, 020503(R) (2020).
[10] F. Lechermann, Late transition metal oxides with infinite-layer structure: Nickelates versus cuprates, Phys. Rev. B 101, 081110(R) (2020).
[11] M. B. Mi Jiang, and George A. Sawatzky, The critical nature of the Ni spin state in doped NdNiO$_2$, https://arxiv.org/abs/1909.02557
[12] X. Wu, D. Di Sante, T. Schwemmer, W. Hanke, H. Y. Hwang, S. Raghu, and R. Thomale, Robust $dx^2-\gamma^2$-wave superconductivity of infinite-layer nickelates, Phys. Rev. B 101, 060504(R) (2020).
[13] Y. H. Zhang and A. Vishwanath, Type II $t$–$J$ model in superconducting nickelate Nd$_{1-x}$Sr$_x$NiO$_2$, https://arxiv.org/abs/1909.12865
[14] H. Zhang, L. Jin, S. Wang, B. Xi, X. Shi, F. Ye, and J. W. Mei, Effective Hamiltonian for nickelate oxides Nd$_{1-x}$Sr$_x$NiO$_2$, Phys. Rev. Res. 2, 013214 (2020).
[15] G.-M. Zhang, Y.-f. Yang, and F.-C. Zhang, Self-doped Mott insulator for parent compounds of nickelate superconductors, Phys. Rev. B, 101, 020501(R) (2020).
[16] J. E. Hirsch and F. Marsiglio, Hole superconductivity in infinite-layer nickelates, Physica C 566 135354 (2019).
[17] H. Sakakibara, H. Usui, K. Suzuki, T. Kotani, H. Aoki, and K. Kuroki, Model construction and a possibility of cuprate-like pairing in a new d$^9$ nickelate superconductor (Nd,Sr)NiO$_2$, https://arxiv.org/abs/1909.00060
[18] S. Ryee, H. Yoon, T. J. Kim, M. Y. Jeong, and M. J. Han, Induced magnetic two-dimensionality by hole doping in the superconducting infinite-layer nickelate Nd$_{1-x}$Sr$_x$NiO$_2$, Phys. Rev. B 101, 064513 (2020).
[19] L.-H. Hu and C. Wu, Two-band model for magnetism and superconductivity in nickelates, Phys. Rev. Res. 1, 032046(R) (2019).
[20] F. Bernardini, V. Olevano, and A. Cano, Magnetic penetration depth and $T_c$ in superconducting nickelates, Phys. Rev. Res. 2, 013219 (2020).
[21] Y. Gu, S. Zhu, X. Wang, J. Hu, and H. Chen, Hybridization and correlation effects in the electronic structure of infinite-layer nickelates, https://arxiv.org/abs/1911.00814
[22] M. Hepting, D. Li, C. J. Jia, H. Lu, E. Paris, Y. Tseng, X. Feng, M. Osada, E. Been, Y. Hikita, Y. D. Chuang, Z. Hussain, K. J. Zhou, A. Nag, M. Garcia-Fernandez, M. Rossi, H. Y. Huang, D. J. Huang, Z. X. Shen, T. Schmitt, H. Y. Hwang, B. Moritz, J. Zaanen, T. P. Devereaux, and W. S. Lee, Electronic structure of the parent compound of superconducting infinite-layer nickelates, Nat. Mater. 19, 381 (2020).
[23] Y. Fu, L. Wang, H. Cheng, S. Pei, X. Zhou, J. Chen, S. Wang, R. Zhao, W. Jiang, C. Liu, M. Huang, X. Wang, Y. Zhao, D. Yu, F. Ye, S. Wang, and J. Mei, Core-level X-ray photoemission and Raman spectroscopy studies on electronic structures in Mott-Hubbard type nickelate oxide NdNiO$_2$, https://arxiv.org/abs/1911.03177
[24] Q. Li, C. He, J. Si, X. Zhu, Y. Zhang and H. H. Wen, Absence of superconductivity in bulk Nd$_{1-x}$Sr$_x$NiO$_2$, Comms. Mater. 1, 16 (2020).
[25] X.-R. Zhou, Z.-X. Feng, P.-X. Qin, H. Yan, S. Hu, H.-X. Guo, X.-N. Wang, H.-J. Wu, X. Zhang, H.-Y. Chen, X.-P. Qiu, and Z.-Q. Liu, Absence of Superconductivity in Nd$_{0.8}$Sr$_{0.2}$NiO, Thin Films without Chemical Reduction, Rera Metal 39, 368 (2020).
[26] P. A. Lee, N. Nagaosa, and X. G. Wen, Doping a Mott insulator: Physics of high-temperature superconductivity, Rev. Mod. Phys. 78, 17 (2006).
[27] H. Hosono and K. Kuroki, Iron-based superconductors: Current status of materials and pairing mechanism, Physica C 514, 399 (2015).

[28] D. Li, B. Wang, K. Lee, Shannon P. Harvey, M. Osada, B. H. Goodge, L. F. Kourkoutis, and H. Y. Hwang, Superconducting Dome in Nd$_{1-x}$Sr$_x$NiO$_2$ Infinite Layer Films, https://arxiv.org/abs/2003.08506

[29] K. Lee, B. H. Goodge, D. Li, M. Osada, B. Y. Wang, Y. Cui, L. F. Kourkoutis, H. Y. Hwang, Aspects of the synthesis of thin film superconducting infinite-layer nickelates, APL Mater. 8, 041107 (2020).

[30] M. Kawai, S. Inoue, M. Mizumaki, N. Kawamura, N. Ichikawa, and Y. Shimakawa, Reversible changes of epitaxial thin films from perovskite LaNiO$_3$ to infinite-layer structure LaNiO$_2$, Appl. Phys. Lett. 94, 082102 (2009).

[31] M. A. Hayward, M. A. Green, M. J. Rosseinsky, and J. Sloan, Sodium Hydride as a Powerful Reducing Agent for Topotactic Oxide Deintercalation-Synthesis and Characterization of the Nickel(I) Oxide LaNiO$_2$, J. Am. Chem. Soc. 121, 8843 (1999).

[32] A. Ikeda, T. Manabe, and M. Naito, Comparison of reduction agents in the synthesis of infinite-layer LaNiO$_2$ films, Physica C, 506, 83 (2014).

[33] See Supplemental Material for the following: Fig. S1 for XRD $\theta$–2$\theta$ scans of the as-grown Nd$_{1-x}$Sr$_x$NiO$_3$ thin films; Fig. S2 for $\rho$-$T$ curves of as-grown Nd$_{1-x}$Sr$_x$NiO$_3$ thin films; Fig. S3 for $\rho$-$T$ curves of a sample with $x = 0.2$ under various magnetic field; Fig. S4 for $\rho$-$T$ curves of multiple samples for $x$ from 0.12 to 0.3.

[34] Y. Dagan, M. M. Qazilbash, C. P. Hill, V. N. Kulkarni, and R. L. Greene, Evidence for a quantum phase transition in Pr$_2$Ce$_x$CuO$_4$ from transport measurements, Phys. Rev. Lett. 92, 167001 (2004).

[35] Y. Ando, Y. Kurita, S. Komiya, S. Ono and K. Segawa, Evolution of the Hall coefficient and the peculiar electronic structure of the cuprate superconductors, Phys. Rev. Lett. 92, 197001 (2004).

[36] H. Y. Hwang, B. Batlogg, H. Takagi, H. L. Kao, J. Kwo, R. J. Cava, J. J. Krajewski, and W. F. Peck, Jr., Scaling of the temperature dependent Hall effect in La$_{2-x}$Sr$_x$CuO$_4$, Phys. Rev. Lett. 72, 2636 (1994).

[37] W. Jiang, S. N. Mao, X. X. Xi, X. G. Jiang, J. L. Peng, T. Venkatesan, C. J. Lobb, and R. L. Greene, Anomalous transport-properties in superconducting Nd$_{1.85}$Ce$_{0.15}$CuO$_4$, Phys. Rev. Lett. 73, 1291 (1994).

[38] Z. Q. Liu, D. P. Leusink, X. Wang, W. M. Lu, K. Gopinadhan, A. Annadi, Y. L. Zhao, X. H. Huang, S. W. Zeng, Z. Huang, A. Srivastava, S. Dhar, T. Venkatesan, and Ariando, Metal-Insulator Transition in SrTiO$_{3-x}$ Thin Films Induced by Frozen-Out Carriers, Phys. Rev. Lett. 107, 146802 (2011).

[39] S. W. Zeng, X. M. Yin, T.S. Herng, K. Han, Z. Huang, L. C. Zhang, C. J. Li, W. X. Zhou, D. Y. Wan, P. Yang, J. Ding, A. T. S. Wee, J. M. D. Coey, T. Venkatesan, A. Rusydi, and A. Ariando, Oxygen Electromigration and Energy Band Reconstruction Induced by Electrolyte Field Effect at Oxide Interfaces, Phys. Rev. Lett. 121, 146802 (2018).

[40] H. Takagi, T. Ido, S. Ishibashi, M. Uota, S. Uchida, and Y. Tokura, Superconductor-to-nonsuperconductor transition in (La$_{1.3}$Sr)$_2$CuO$_4$ as investigated by transport and magnetic measurements, Phys. Rev. B 40, 2254 (1989).

[41] A. R. Moodenbaugh, Y. Xu, M. Suenaga, T. J. Folkerts, and R. N. Shelton, Superconducting properties of La$_{2-x}$Ba$_x$CuO$_4$, Phys. Rev. B 38, 4596 (1988).
[42] Y. Krockenberger, J. Kurian, A. Winkler, A. Tsukada, M. Naito, and L. Alff, Superconductivity phase diagrams for the electron-doped cuprates $R_{2-x}Ce_xCuO_4$ ($R=La$, Pr, Nd, Sm, and Eu), Phys. Rev. B, 77, 060505(R) (2008).

[43] J. L. Peng, E. Maiser, T. Venkatesan, R. L. Greene, and G. Czjzek, Concentration range for superconductivity in high-quality $Pr_{2-x}Ce_xCuO_4$ thin films, Phys. Rev. B, 55, R6145 (1997).

[44] S. Karimoto, K. Ueda, M. Naito, and T. Imai, Single-crystalline superconducting thin films of electron-doped infinite-layer compounds grown by molecular-beam epitaxy, Appl. Phys. Lett. 79, 2767 (2001).
Fig. 1. XRD $\theta$–$2\theta$ scan patterns of Nd$_{1-x}$Sr$_x$NiO$_2$ thin films on STO substrates. The intensity was vertically displaced. The inset shows the room-temperature $c$-axis lattice constants, $d$, as a function of Sr doping level $x$ for Nd$_{1-x}$Sr$_x$NiO$_2$. The red circles represent the data extracted from the $\theta$–$2\theta$ scans in Fig. 1, the black circles represent another set of samples at each doping level for $0.12 \leq x \leq 0.3$. The dashed line in the inset is a guide to the eye. The measurements were done at room temperature.
Fig. 2. (a) The resistivity versus temperature ($\rho$-$T$) curves of Nd$_{1-x}$Sr$_x$NiO$_2$ thin films with Sr doping level $x$ from 0.08 to 0.3. (b) The zoomed-in $\rho$-$T$ curves at temperatures from 50 to 2 K for superconducting Nd$_{1-x}$Sr$_x$NiO$_2$ with $x$ from 0.135 to 0.22.
Fig. 3. (a) The temperature dependence of the Hall coefficient ($R_H$) for Nd$_{1-x}$Sr$_x$NiO$_2$ thin films with Sr doping level $x$ from 0.08 to 0.3. (b) The room-temperature $R_H$ as a function of $x$. The red dash line is a guide to the eye.
Fig. 4. The onset critical-temperature $T_c$ and temperature $T_H$ where the Hall coefficient ($R_H$) changes from negative to positive sign. The $T_c$ is defined to be the temperature at which the resistivity drops to be 90% of the resistivity value at 15 K which is near to the onset of superconductivity. The red circles represent the data extracted from $\rho-T$ curves in Fig. 2, the black triangles and blue inverted triangles represent other sets of samples at each doping level shown in Fig. S4. The $T_H$ is defined to be the temperature at which the $R_H$ curve crosses the axis at zero in Fig. 3. The blue dash line is a guide to the eye. The inset schematically shows the electron band ($e^-$) and hole band ($h^+$) which dominates the charge carriers in the two panels separated by the $T_H-x$ curve. The black dash line in the inset is the Fermi energy ($E_F$).
Supplementary materials for

Phase diagram and superconducting dome of infinite-layer Nd$_{1-x}$Sr$_x$NiO$_2$ thin films

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Fig. S1. (a) XRD $\theta$–$2\theta$ scan patterns of the as-grown Nd$_{1-x}$Sr$_x$NiO$_3$ thin films on STO substrates. The intensity was vertically displaced. Only (00$l$) perovskite peaks are observed, where $l$ is an integer, confirming the $c$-axis oriented epitaxial growth. (b) The room-temperature $c$-axis lattice constants of Nd$_{1-x}$Sr$_x$NiO$_3$ thin films as a function of Sr doping level $x$. The red dot line is a guide to the eye. The $c$-axis lattice constants, in general, increase slightly with $x$, from 3.776 Å for NdNiO$_3$ ($x = 0$) to 3.818 Å at $x = 0.3$. The $c$-axis lattice constant obtained for NdNiO$_3$ film is lower than that of the bulk (3.81 Å for pseudocubic lattice) due to the epitaxial tensile strain imposed by STO substrate.
Fig. S2. (a) The logarithmic-scale resistivity versus temperature ($\rho$-$T$) curves for undoped NdNiO$_3$ and Sr-doped Nd$_{1-x}$Sr$_x$NiO$_3$ thin films with different Sr doping level $x$ from 0.08 to 0.3. (b) The linear-scale $R$-$T$ curves for Nd$_{1-x}$Sr$_x$NiO$_3$ thin films with $x$ from 0.08 to 0.3. NdNiO$_3$ thin film shows a metal-insulator transition with decreasing temperature and the transition is suppressed by Sr doping. All Nd$_{1-x}$Sr$_x$NiO$_3$ thin films show metallic behaviour and a slight resistance upturn at low temperatures.

Fig. S3. The resistivity versus temperature ($\rho$-$T$) curves for sample with $x = 0.2$ under various magnetic field applied perpendicularly to the $a$-$b$ plane.
Fig. S4. The resistivity versus temperature ($\rho$-$T$) curves of multiple samples for $x$ from 0.12 to 0.3. (a) $x$ = 0.12; (b) $x$ = 0.135; (c) $x$ = 0.15; (d) $x$ = 0.18; (e) $x$ = 0.2; (f) $x$ = 0.22; (g) $x$ = 0.235; (h) $x$ = 0.25; (i) $x$ = 0.3. The red curves for specific doping are the ones showed in Fig. 2 in the main text. The curves shown in other colours correspond to samples different from the ones in Fig. 2 in the main text.