Superconductivity in nickel-based 112 systems

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Graphical abstract

Public summary
- The infinite-layer nickel-based 112 thin films \( R_{1-x}A_x\text{NiO}_2 \) can host superconductivity up to 15 K
- \( R_{1-x}A_x\text{NiO}_2 \) is a multiband system, in which the short-range antiferromagnetic fluctuations can be detected
- \( R_{1-x}A_x\text{NiO}_2 \) has an unconventional superconducting pairing state with a robust \( d \)-wave gap and a full gap without unified understanding
- The nickelate system provides a new platform for researching unconventional superconductivity
Superconductivity has been discovered recently in infinite-layer nickel-based 112 thin films $R_1-x$NiO$_2$ ($R =$ La, Nd, Pr and $A =$ Sr, Ca). They are isostuctural to the infinite-layer cuprate (Ca,Sr)CuO$_2$ and are supposed to have a formal Ni $3d^8$ valence, thus providing a new platform to study the unconventional pairing mechanism of high-temperature superconductors. This important discovery immediately triggers a huge amount of innovative scientific curiosity in the field. In this paper, we try to give an overview of the recent progress on the newly found superconducting nickelate systems, both from experimental and theoretical aspects. We mainly focus on the electronic structures, magnetic excitations, phase diagrams and superconducting gaps, and finally make some open discussions for possible pairing symmetries in Ni-based 112 systems.

INTRODUCTION

In 1986, Bednorz and Müller in IBM Zürich Research Laboratory discovered the high-temperature superconductivity (HTS) in Ba-doped insulating cuprate system La$_2$-$_x$Sr$_x$CuO$_4$ and they were awarded the Nobel Prize in Physics for this important discovery. Soon after, the critical temperature ($T_c$) in YBa$_2$Cu$_3$O$_y$ system was found to be as high as 93 K, which broke through the Macmillan limit and the boiling temperature of liquid nitrogen, starting a new era of intensive research on HTS. Scientists discovered more and more cuprate systems and continuously renewed the records of the highest superconducting transition temperatures at ambient pressure. However, the pairing mechanism in HTS remains controversial and elusive. It seems that the superconducting states can coexist or compete with different kinds of emergent intertwined orders, which leads to a complicated phase diagram. Since 2008, the discovery of iron-based superconductors (FeSCs) gives us some new hints for investigating the pairing mechanism of HTS and exploring superconductors with higher transition temperature. For now, cuprate and FeSCs are recognized as the only two systems of HTS under ambient pressure, and it seems to be a long-standing pursuit to realize room temperature superconductors. The most essential consensus that has been achieved is that HTS should have layered structures and 3d orbital electrons of transition metals with appropriate electron correlation. It has been widely perceived that the pairing may be established through exchanging antiferromagnetic (AF) spin fluctuations or superexchange change.

Many experimental and theoretical efforts have been devoted to searching HTS in the analogous compounds without copper. For example, bulk superconductivity was found in Sr$_2$RuO$_4$ with a very low $T_c$ and electron-doped Sr$_2$IrO$_4$ exhibits plausible spectroscopic signatures of superconducting gaps. As shown in Figure 1A, the Ni element locates just between Fe and Cu, and scientists have made great achievements in this fast-developing field. In the following sections of this review, we mainly focus on discussing the representative $NdNiO_2$ system, including the calculated electronic structures with multiband features, spin configurations, magnetic excitations, phase diagrams, and the measurement of single-particle tunneling spectrum; finally, we make some open discussions for the possible pairing symmetries and pairing mechanism in the newly found Ni-based superconductors.

**ELECTRONIC STRUCTURES**

As we know, the parent compounds of cuprates may be described as a Mott insulator with an AF long-range order and superconductivity occurs upon chemical doping. Due to the strong $p-d$ orbitals hybridization, the doped holes enter into the oxygen sites in the CuO$_2$ planes. It was proposed that the doped holes on the p-orbital of oxygens combine with the 3d$_{x^2-y^2}$-$p$ spins of Cu ions to form the Zhang-Rice singlet, moving through the square lattice, and exchange with their neighboring Cu spins. This theoretical proposal sets cuprates into a single band limit and naturally renders an effective two-dimensional (2D) t-J model to describe the low-energy physics of cuprates. In FeSC, the situation becomes more complex because the As/Se atoms locate alternatively above and below the center of the Fe squares. The crystalline environment experienced by Fe atoms is somewhat in between a tetrahedral one, in which the energy of the
t_{2g} orbitals is higher than that of the e_g orbitals, while for an octahedral one as in cuprates, the energy of the t_{2g} orbitals is lower. As a result, the crystal splitting between the orbitals is weakened, and all five d-orbitals give a considerable contribution to the low-lying energy of electronic states. As for the newly found Ni-based superconductors, many papers have made detailed theoretical calculations of the electronic states, demonstrating the multiorbital features in the Nd/La 112 system within the low-energy region and pointing out both similarities and differences between nickelates and cuprates.

Mott insulator, self-doping effect, and possible Kondo coupling

When we compare the NiO_2 layer in nickelate and CuO_2 layer in cuprates, a major difference becomes immediately obvious. As shown by the sketch in Figure 2A, a charge-transfer energy is estimated to be $\Delta = 9$ eV in NiO_2 and $\Delta = 3$ eV in CuO_2. According to the Zaanen-Sawatzky-Allen (ZSA) scheme, cuprates locate in the regime of charge transfer insulator, while nickelates belong to the system of Mott insulator. The holes doped in a Mott insulating NiO_2 layer would reside on the Ni-derived bands, not in the O 2p one. Because Ni^{2+} (3d^9) with $S = 1$ is very common in all the known Ni^{2+} oxides, this makes the appearance of rather high-$T_c$ superconductivity very puzzling and definitely unlike that in cuprates. In experiments, Hepting et al. conduct X-ray absorption spectroscopy (XAS) and emission spectroscopy (XES) measurements near the O K-edge in parent phase RNiO_2 ($R = \text{La, Nd}$), and the results roughly reflect the unoccupied and occupied oxygen partial density of state (PDOS), respectively. As shown in Figure 2B, the oxygen PDOS exhibits a diminished weight near the Fermi energy, especially in the unoccupied states; this indicates that the O 2p orbitals carry less weight in the expected upper Hubbard band by comparison. All of these are consistent with the calculated oxygen PDOS from the calculation of local density approximation (LDA) + U method. In addition, Figure 2C shows the results of resonantinelastic X-ray scattering (RIXS) at the Ni L_2-edge (a core-level 2p to valence 3d transition). The markers A indicate the main absorption peak of LaNiO_3 and NiO_2, which resembles the single peak associated with the 2p^{9}3d^{0}–2p^{8}3d^{1} transition in cuprates. The $\Lambda$' labels highlight the hybridization between the Ni 3d_{x^2-y^2} and R 5d orbitals. In this configuration, the Ni state can have a charge transfer to the rare-earth cation, thus leaving holes in Ni orbitals and electrons in R 5d orbitals, this is the so-called self-doping effect. In NdNiO_2, the similar feature caused by the Nd–Ni hybridization is also revealed by RIXS measurements, but its resonance energy $\Lambda$' almost coincides with the main peak A. Based on above experimental results, one can naturally understand that in parent phase, the singly occupied Ni 3d_{x^2-y^2} orbital with strong correlation may give rise to a local spin 1/2 and a Mott insulator state with an AF long-range order. However, the parent compound NdNiO_2 displays metallic behavior at high temperatures (see Figure 1B) and shows no sign of any magnetic long-range order in the whole measured temperature range. Similar results have also been found previously in LaNiO_3. These experimental observations show an obvious contradiction to the naive expectations. It is therefore important to address what is the nature of the parent compounds and how the AF long-range order is suppressed. Zhang et al. made a detailed analysis of resistivity data as functions of temperature for both parent compounds NdNiO_2 and LaNiO_2.
The Innovation

noninteracting Fermi surfaces and random phase approximation (RPA) spin susceptibilities are calculated within a weak coupling scenario. It can help us to obtain the electronic structure of the parent phase (solid lines) and 20% hole doping (dashed lines). The main Fermi surface in the system named by $\Gamma$ and $A$ points, which are contributed mainly from the La-derived orbitals. These self-doping bands are crucially originated from the hybridization between La $5d$ and Ni $3d$ orbitals. More specifically, $\beta$ pocket around the $A$ point has the mixture of La $5d_S$ and Ni $3d_{yz}$, while $\gamma$ pocket around the $A$ point has the mixture of La $5d_y$ and Ni $3d_{x^2-y^2}$ orbitals. The similar band structure of Ni$_2$O$_3$ can be obtained in density functional theory (DFT), as shown in Figure 3B. The DFT calculation suggests that with hole doping, $R$ 5d band rises up much faster than Ni $3d_{x^2-y^2}$ band. Therefore, at the doping level around 20%, the $\beta$ pocket has disappeared almost completely, and it leaves a diminished $\gamma$ pocket around the $A$ point in both compounds.

We want to emphasize that the band structures and Fermi surfaces discussed above are calculated within a weak coupling scenario. It can help us to obtain the noninteracting Fermi surfaces and random phase approximation (RPA) spin susceptibility. We need also notice that the parent phase RNiO$_2$ starts with the description of self-doping Hubbard-Mott insulator, which should be naturally categorized into a strong coupling regime. Therefore, it becomes necessary to calculate the more correlated electronic structures within the scheme of LDA + $U$, in which the strength of interaction is mainly determined by the value of Hubbard $U$. Many theoretical papers have put forward that the multiorbital picture in RNiO$_2$ is not only depicted based on the localized Ni $3d_{x^2-y^2}$ and $d$-dominated self-doping bands, but also the Ni $3d_{yz}$ band in a Hund-assisted manner, so $U$ is defined as the on-site Hubbard interaction of Ni $3d_{x^2-y^2}$/3$d_{yz}$ orbitals. Lechermann$^{95}$ pioneers the doping-dependent competition between Ni $3d_{x^2-y^2}$ versus Ni $3d_{yz}$ and first describes the flat band feature of Ni $3d_{yz}$ states at the Fermi level with hole doping. In the large-$U$ limit (larger than 10 eV), such a model at a quarter filling by holes (three-electron filling) is expected to exhibit a Mott insulator for the Ni $3d_{yz}$ band. Because the self-doped character remains robust up to large interaction strengths, the nickelate compound avoids an insulating state for even larger $U = 15$ eV. Compared to the LDA result in the undoped case, the size of the electron pocket around $A$ is reduced, but the many-body calculation intensifies the stronger Ni $3d_{yz}$/Ni $3d_{xy}$ hybridized electron pocket around $A$. At about 0.2 hole doping, the role of the self-doping band is depleted already. Therefore, Ni $3d_{yz}$ and $3d_x$ compete at energies close to the Fermi level, and the spectral weight close to $\epsilon_F$ is strongly Ni $e_g$ orbitals dominated.

To conclude, for the mother compound, a small number of holes are self-doped into the Ni $3d_{yz}$ orbital because of the presence of electron pockets. These electron pockets may have relevance to the metallic behavior observed experimentally, while the electronic state of the $d_{yz}$-$d_{xy}$ band with a hole self-doping may be close to that of the heavily underdoped cuprates with no magnetism or superconductivity. Furthermore, the superconductivity in thin films of Nd$_{1-x}$Sr$_x$NiO$_2$ may share some similarities like that of the single-orbital cuprates, while more distinctions are expected, providing a new fascinating platform on the research of unconventional correlated superconducting materials.

Transport measurements and phase diagram

Considering the difficulty in synthesizing the high-quality superconducting nickelate thin films, we just make a brief review on the transport data and the related phase diagram. Li et al.$^{104}$ have made a detailed study on resistivity and Hall effect measurements on the typical system of infinite-layer Nd$_{1-x}$Sr$_x$NiO$_2$. Figure 4A displays the temperature-dependent in-plane resistivity $\rho_{xx}$ across the doping series. The films with $x = 0.15, 0.175, 0.2$, and $0.225$ show varying $T_C$ while for $x = 0.1, 0.125$, and $0.25$, a weakly insulating feature emerges at low temperatures, as shown in the upper panel of Figure 4B. These data indicate a superconducting dome that is similar to hole-doped cuprates$^{105,106}$ in the lower panel, but approximately half as wide in the doping regime. Furthermore, an approximately linear $T$ dependence of $\rho_{xx}$ is observed across a wide temperature range above $T_C$ similar to that found in cuprates (known as the "strange metal" phase$^{107-109}$) and other strongly correlated systems, suggesting a similar possible origin for $\rho_{xx}(T)$ despite different underlying electronic structures.$^{110}$ As for the noticeable upturn in $\rho_{xx}(T)$ for nonsuperconducting compositions ($x = 0.1, 0.125, 0.25$), it is not simply identified as weak localization. Also, the lack of strongly insulating behavior has already been widely reported for the undoped compounds.$^{46}$ It can be possibly attributed to the self-doping bands and Kondo effect. They further find that the "overdoped" regime does not appear to approach the Fermi liquid ending point commonly understood in the hole-doped cuprates.$^{111-114}$ Overall, the relevance of "hole doping" remains an open question for Nd$_{1-x}$Sr$_x$NiO$_2$. They also measure the evolution of the normal state Hall effect in this series of samples. Figure 4C
that although the 112 phase has a better crystallinity, the insulating behavior seems to be even stronger, and superconductivity remains absent. Despite a lot of challenges, it is still worth making more efforts to figure out the unsettled issue.

**SPIN CONFIGURATIONS AND MAGNETIC EXCITATIONS**

As we know, the parent phase of cuprates is depicted as a charge transfer insulator with an AF order, where the Cu$^{2+}$ ions have one active d$_{xy}$ orbital hybridized with the p orbitals of the neighboring in-plane oxygens. Upon hole doping, because of the negative charge-transfer gap, the holes predominantly go to the O-p orbitals. Thus, it will give rise to the spin configuration of 3d$^9$ Cu$^{2+}$, surrounded by a ligand hole on oxygen. This is the formation of Zhang-Rice singlets. In inelastic neutron-scattering measurements, a square-shaped continuum of excitations peaked at incommensurate positions, and an "hourglass" shape of the magnetic dispersions in the superconducting state of cuprates has been extensively observed. These excitations are regarded as a general property of cuprates and a promising candidate for magnetically mediated electron pairing. Moreover, when revising the phase diagrams of temperature versus doping level both in cuprates and FeSC, we can find that the occurrence of superconductivity is intimately correlated with the disappearance of AF long-range order and the occurrence of AF spin fluctuations. As a result, it may lead to the unconventional pairing state of $d$ wave in cuprates and $s^\pm$ in FeSC. Accordingly, it is highly desirable to address the spin configurations and magnetic excitations in nickel-based superconductors.

**Competition between HS and LS states**

Because the charge-transfer energy is much larger in the nickelates, one may expect the doped holes to reside dominantly on the Ni site, rather than in the O-p band. Moreover, many theoretical papers have proposed that the two orbitals of Ni 3d$^{9}_{xy}$/3d$_{yz}$/3d$_{zx}$ are essential for the multiorbital process in the nickelate system because of Hund's coupling. For example, Werner and Hoshino use dynamical mean field theory (DMFT) calculations for the two-orbital Ni 3d$^{9}_{xy}$/3d$_{yz}$/3d$_{zx}$ system and argue that a multiorbital description of nickelate superconductors is warranted. Lechermann pioneers the doping-dependent competition between Ni 3d$^{9}_{xy}$/3d$_{yz}$/3d$_{zx}$ versus Ni 3d$_{xy}$ orbitals. Accordingly, in a square lattice environment as depicted in Figure 5A, the $e_g$ states of Ni$^{2+}$ may be energetically arranged in two different ways, which depends on the limit whether the Hund's rule coupling is larger than the crystal-field splitting between the two $e_g$ orbitals. For the high-spin (HS) state, the electron from the d$_{xy}$ orbital is removed, in which case the resultant states are $t_{2g}^3d_{xy}^1$ with $S = 1$; for the low-spin (LS) state, the electron from the d$_{xy}$ orbital is removed, in which case the configuration is $t_{2g}^2d_{yz}^2$ with $S = 0$.

Concerning that the Ni$^{2+}$ ions are commonly found to be spin-triplet state in most of the nickel compounds, it has recently been argued in several theoretical works that hole-doping Nd$_{1−x}$Sr$_x$NiO$_2$ should produce Ni$^{2+}$ with spin $S = 1$. Based on the HS state, Zhang et al. propose a variant of t-J model and find two distinct mechanisms for $d$-wave superconductivity. However, as early as in 1959, Ballhausen and Liehr have discussed the $S = 0$ spin singlet state in the planar Ni(ii) complexes based on the theory of ligand field. Recently, Jiang et al. first point out that the $S = 1$ state may be incompatible with robust superconductivity, and a number of many-body calculations using LDA and DMFT pointed to the formation of intraorbital singlets. However, if the first-principles calculations of explicit Sr doping in RNO$_2$ (R = La, Nd) superconductors, an LS state is favored. From experimental measurements, Rossi et al. use a combination of high-resolution XAS and RIXS, and find that doped holes are primarily introduced in the Ni 3d$_{xy}$/3d$_{yz}$/3d$_{zx}$ Hubbard band in a LS configuration. Wan et al. discuss the solutions of an effective two-band model, including Ni 3d$_{xy}$/3d$_{yz}$/3d$_{zx}$ orbitals on the basis of DMFT. As shown in Figure 5B, whether an $S = 0$ or 1 state emerges depends on a precise value of the intraatomic Hund's coupling $J_H$ in the vicinity of its commonly accepted range of values 0.5–1 eV. Until now, we cannot make a definite conclusion about whether the $S = 0$ or 1 scenario is realized for doped nickelates. The key issue is the competition between Hund’s coupling and the crystal field splitting. Because of the considerable hybridizations between the itinerant Ni 5d and Ni 3d orbitals, the values of $J_H$ and crystal field splitting based on DFT simulations may be questionable. Therefore, the spin state of Ni$^{2+}$ in hole-doping Nd$_{1−x}$Sr$_x$NiO$_2$ remains highly debatable and worthy of more attention.

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Figure 3. Orbital resolved band structures of LaNiO$_2$ and NdNiO$_2$ in parent phase and 20% hole doping (A) Band structure of LaNiO$_2$ from the first-principles (solid lines). The band structure of the seven-orbital model is superposed, where the Wannier-orbital weight is represented by the thickness of lines with color-coded orbital characters. Adapted from Sakakibara et al. (B) DFT band structure of NdNiO$_2$. Adapted from Lechermann et al. Right panels display cross sections of the Fermi surfaces at $k_z = 0$ (left) and $k_z = \pi$ (right), where the red and blue lines depict Ni and La/Nd-dominant Fermi surfaces, respectively. Undoped and 20% hole doped cases are plotted by solid lines and dashed lines, respectively. The Fermi surfaces of La-112 and Nd-112 are adapted from Sakakibara et al. and Petocchi et al., respectively.
AF magnetic excitation

Most intriguingly, one key experimental observation for the infinite-layer NdNiO$_2$ is that its resistivity exhibits a minimum at about 70 K and an upturn at a lower temperature. Meanwhile, the Hall coefficient drops toward a larger value, signaling the loss of charge carriers. More interestingly, no long-range magnetic order has been observed in powder neutron diffraction on LaNiO$_2$ and NdNiO$_2$ when temperature is lowered down to 5 and 1.7 K, respectively. This greatly challenges the existing theories, because it is generally believed that magnetism is essential for the emergence of unconventional superconductivity. Therefore, it is highly desirable to study the magnetic properties of undoped parent NdNiO$_2$ and elucidate its experimental indications.

In theoretical aspects, there are still some debates on the estimation of exchange coupling strength $J$ in nickelates; some theories suggest $J$ to be one order of magnitude smaller than in cuprates because of the large charge transfer energy, whereas some others argue it could be comparable to that in cuprates. In nickelates, the calculated exchange coupling parameters as a function of Hubbard $U$ are shown in Figure 5D. The main finding is that the estimated magnetic exchange interaction is around $J_{1} \sim 10$ meV, $J_{2} \sim 10$ meV. This indicates that effective exchange interactions in NdNiO$_2$ are about one order of magnitude smaller than those of cuprates $\sim 112$ meV. Also, it results in a relatively weaker magnetic ordering and lower Néel temperature, $T_N$, in NdNiO$_2$ compared with cuprates. Furthermore, the infinite-layer nickelate is believed to be a worse metal compared to elemental nickel with Hubbard $U$ about 3 eV, which can be considered as a lower boundary of $U$. The Coulomb interaction in infinite-layer nickelates should be smaller than that in the charge-transfer insulator NiO with $U$ about 8 eV, which can be considered as an upper boundary of $U$.

Therefore, a reasonable value of $U$ in NdNiO$_2$ can be estimated around 5–6 eV. As shown in Figure 5E, there could exist a transition from normal metal to bad AFM metal around $T_N \sim 70$–90 K, which provides a plausible understanding of minimum of resistivity and drop of Hall coefficient in infinite-layer NdNiO$_2$.

From experimental aspects, Cui et al. report the $^{1}$H nuclear magnetic resonance (NMR) measurements on powdered Nd$_{0.95}$Sr$_{0.05}$NiO$_2$ samples by taking advantage of the enriched proton concentration after hydrogen annealing. The spin-lattice relaxation rate $T_{1}^{-1}$ is a sensitive probe of low-energy spin fluctuations. The temperature-dependent $T_{1}^{-1}$ under various fields is plotted in Figure 6A. From 2 K, the $T_{1}^{-1}$ first increases dramatically, then forms a peaked feature at $T \sim 40$ K, and finally flattens out at temperatures above 100 K. A sharp peak in $T_{1}^{-1}$ is usually a signature of magnetic phase transition, while the broad one at about 40 K suggests the onset of a short-range glassy AF order in bulk Nd$_{0.95}$Sr$_{0.05}$NiO$_2$, which is similar to underdoped cuprate superconductor. Furthermore, the plot of a log-log scale is shown in Figure 6B. Below 40 K, $T_{1}^{-1} \propto T^{\alpha}$, following a power-law behavior with a low-power-law exponent $\alpha = 2$, much smaller than that caused by AF spin-wave excitations ($\alpha = 5$). This indicates the onset of low-energy spin fluctuations and remaining spin excitations extending to much higher temperatures. The finding of strong AF fluctuations reveals the strong electron correlations in bulk Nd$_{0.95}$Sr$_{0.05}$NiO$_2$ and paves the way for understanding the relationship between magnetism and superconductivity in nickelates. Lu et al. measure the dispersion of magnetic excitations in undoped NdNiO$_2$ by using RIXS at the Ni L$_3$-edge. Figure 6C plots a summary of fitted magnetic mode energy $\epsilon$ (filled red circles) and damping factor $\gamma$ (empty red circles) versus in-plane momentum transfers $q_{xy}$ along high-symmetry directions. NdNiO$_2$ possesses a branch of dispersive excitations with a bandwidth of approximately 200 meV, which can be fitted to the linear spin wave theory of a 2D AF Heisenberg model with $J_{1} \sim 63.6$ meV, $J_{2} \sim 103.7$ meV. These results can clarify that the exchange interaction in NdNiO$_2$ is comparable to that in cuprates. Besides, the significant damping and rather constant $\gamma_0$ of these modes indicate that rare-earth itinerant electrons play a role here, which are highly coupled to Ni 3d orbitals.

To reconcile the discrepancy between the observed paramagnetic metallic state (only the remaining AF excitations) of RNiO$_2$ and the measured considerable exchange interaction $J$ along with the theoretically predicted AF transition nearly 90 K, one fact that needs to be noticed is that the estimation of $T_N$ in Figure 5E has not taken the influence of conduction electrons into account. As Gu et al. point out, the hybridization between Ni-$d_{xy}$ orbital and itinerant electrons in RNiO$_2$ is substantially stronger than previously thought. Because of that, Ni local moment is screened by itinerant electrons, and the critical $U_{Ni}$ for long-range magnetic ordering is increased. As a result, the local magnetic AF...
ordering below the $T_N$ should be very weak, which may be hard to be visualized in the magnetic related measurements, such as NMR and neutron scattering. Instead, only the remaining AF fluctuations can be detected.

**SUPERCONDUCTING STATE**

The foregoing sections are mainly focused on the electronic structures and magnetic excitations of normal state in RNiO$_2$, now we will turn to discussing the superconducting properties of this newly discovered superconducting nickelate Nd$_x$Sr$_{2}$NiO$_{4+}$ thin film, including upper critical field, superconducting gap, and possible pairing symmetries.

**Isotropic Pauli-limited pairing state**

As we know, in most superconductors, the superconducting gap ($\Delta$) and the pairing strength may be linked to the upper critical field $H_{c2}$ via the Pippard relation, $\xi = h\omega_c/\pi v_F$ and $\mu_B H_{c2} = \Phi_0/2\pi r^2$ (orbital de-pairing), where $\xi$ is the coherence length, $v_F$ is the Fermi velocity, and $\Phi_0$ is the flux quantum. However, in a few superconductors, Cooper pairs can be broken mainly because of the Zeeman-split effect, and thus the upper critical field is dominated by Pauli paramagnetic limit $\mu_B H_{c2} = \sqrt{2}d/\mu_B g$. Here, $\mu_B$ is the Bohr magneton, and $g$ is the Landé factor. Therefore, it is worthwhile to measure the upper critical field to obtain the information of the pairing strength for this new superconducting system. More generally, the Nd$_x$Sr$_{1-x}$NiO$_2$ thin film system can be regarded as layered superconductor, in which case one may naturally wonder whether there is some anisotropy in $H_{c2}$ relative to the presence of disorders. Meanwhile, a very large value of $\alpha$ indicates the possible existence of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state in the high-magnetic-field region at low temperatures. Because the FFLO state is fragile in the presence of disorder, the existence of this state in the Nd$_x$Sr$_{1-x}$NiO$_2$ thin film requires further investigation via high-magnetic-field experiments. Second, the value of $\alpha$ approaches to 1 when the temperature is well below $T_N$, namely, the very small anisotropy between $\xi_{ab}$ and $\xi_c$, (equivalent to the effective mass ratio of $m_{c}/m_{ab}$). This provides strong evidence that the existence of two 3D electron pockets makes the electronic structures more isotropic, when compared to the only one single band of quasi 2D Ni 3$d$-$3$p$.

Xiang et al. extract $H_{c2}(T)$ and $H_{c2.1}(T)$ from $\rho(T)$ curves under different magnetic fields in Nd$_0.7$Sr$_{0.3}$NiO$_2$ thin films by using two different criteria of 95% $\rho_c(T)$ and 98% $\rho_c(T)$, where $\rho_c(T)$ is the linear extrapolation of the normal-state resistivity. Because the normal-state residual resistivity is large for this film, they attempt to use the Werthamer, Helfand, and Hohenberg (WHH) theory in the dirty limit for a superconductor with a single s-wave gap to fit the data, as shown in Figure 7A. They achieve several interesting experimental observations. First, the Maki parameter $\alpha$ ranges from 13 to 42, when the magnetic field is applied within ab plane or along c axis and under different criteria. The obtained very large $\alpha$ here is very rare, even much larger than that in the most well-known Pauli-limited systems, such as heavy-fermion and organic superconductors. Meanwhile, a very large value of $\alpha$ indicates the possible existence of the FFLO state in the high-magnetic-field region at low temperatures. Because the FFLO state is fragile in the presence of disorder, the existence of this state in the Nd$_x$Sr$_{1-x}$NiO$_2$ thin film requires further investigation via high-magnetic-field experiments.

Wang et al. have conducted similar measurements to probe the anisotropy in Nd$_0.77$Sr$_{0.23}$NiO$_2$ with a slightly lower $T_N$. Figure 7B plots superconducting $H_{c2}$ phase diagrams for magnetic fields along the $c$ axis and in the ab plane, including many intriguing phenomena. First, the key observation is a $T$-linear dependence of $H_{c2.1}$ and a $(T - T_\text{c})^{1/2}$ dependence of $H_{c2}$. First of all, they rule out the possibility of 2D superconductivity. The main reason is that they deduce a thickness to be 2.3–3 times larger than the observed one, if adopting the linearized Ginzburg–Landau model of 2D superconductor. Another plausible origin is that a Pauli-limited superconductor can also give a $(T - T_\text{c})^{1/2}$ dependence of $H_{c2}$ and high values of Maki parameter $\alpha$ for both

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**Figure 5. Spin configurations and possible AF state** (A) Left panel: spin configurations of Ni 3$d$ in a square-planar environment. High-spin state with $S = 1$ and low-spin state with $S = 0$. Right panel: crystal field splitting on Nd. (B) Calculated probabilities for the three-electron $S = 1/2$, two-electron $S = 1$ and states as a function of Hund’s coupling $J_H$. Adapted from Wan et al. (C) Exchange interaction of RNiO$_2$ calculated by 4$^5$U$^4$ based on different schemes of theoretical methods DFT, Wannier, and linear response theory, compared with the cuprate exchange interactions that are taken from some experimental results. Adapted from Been et al. (D) The Hubbard $U$ dependent of exchange coupling parameters. (E) The estimated $T_N$. Adapted from Liu et al.
orientations are obtained, which indicates the presence of the paramagnetic pairing breaking effect. Third, the phase diagram shows a crossover from superconductivity limited by orbital pair-breaking to Pauli-limited one. The upturn of $H_{c2}(T)$ deviates from the WHH fits, suggesting an increasing role of the paramagnetic effect at the low-temperature region. This anomalous feature may also indicate the occurrence of two-band superconductivity. The high values of $\alpha$ suggest possible existence of FFLO state at low temperatures and high magnetic fields, which seems to be consistent with the upturn of $H_{c2}$ data below 4 K. However, the fact that the film lies in the dirty limit is inconsistent with anticipated expectations.

To draw a conclusion about the magnetotransport measurements of the two works above, they both point out the isotropic Pauli-limited superconductivity with unconventional pairing in the newly found nickelate superconducting film. This provides strong evidence that two small 3D electron-like pockets $\beta$ and $\gamma$ contributed mainly from Nd-derived orbitals to play a non-negligible role here. In addition, a very large value of Maki parameter $\alpha$ indicates possible existence of the very interesting FFLO state in the high-magnetic-field region at low temperatures under the condition that the system can be pushed into a clean limit.

**Single-particle tunneling spectrum measurement**

Concerning the pairing mechanism of nickel-based 112 systems, the core issue is to know the superconducting gap function that measures the pairing interaction of the two electrons of a Cooper pair. However, the research is very rare on physical properties of this material, especially for angle-resolved photo-emission spectroscopy (ARPES) and scanning tunneling microscopy (STM) measurements. Thus, it is urgent to conduct spectroscopic measurements to directly determine the gap structure. In this section, we introduce the first investigation of single-particle tunneling measurements on the superconducting Nd$_{1-x}$Sr$_x$NiO$_2$ thin films. Figure 9A shows the surface of the film just after annealing by the soft-chemistry method. One can see that the surface is not atomically flat, showing a roughness of about 1–2 nm. This large roughness may be induced by a drastic reaction of the 113 film with hydrogen during the post-annealing process. However, if we take a long time vacuum annealing (at about 180 °C in a vacuum of $10^{-9}$ torr for 12 h) on the film with this type of rough surface, some areas of the surface show layer-by-layer structure with terraces, and a typical surface morphology is shown in Figure 8D. Now the roughness becomes much smaller, in which case the tip can have a better stability during the tunneling process. We have conducted measurements of scanning tunneling spectroscopy (STS) on the surfaces with these two different morphologies, one is called rough surface and another one is called smooth surface. We find that the superconducting spectra predominantly show two types of features on the rough surfaces. One type shows a typical V-shape feature, which is shown in Figure 8B. By doing the Dynes model fitting, as displayed by the red curve, we find that the spectrum can be nicely fitted with a $d$-wave gap of $\Delta = 3.9$cos$2\theta$ (meV). The other one shows a full gap feature plotted in Figure 8C, with a Dynes fitting of $\Delta = 2.35(0.85 + 0.15$cos$4\theta$) (meV). A slight anisotropy (about 15% weight of the differential conductivity) is added to the gap function in order to have a good fit. This may suggest that at least one of the bands is fully gapped. On the smooth surface, we find dominant V-shape spectra as shown in Figure 8E. It needs to be emphasized that this type of full gap is hardly observed on the smooth surface. Instead, we can easily observe a mixture of the two-gap features on the spectra, as shown in Figure 8F. The Dynes fitting is $\Delta_1 = 5.3(0.8$cos$2\theta + 0.2$cos$6\theta$) (meV), taking a dominant weight of $\rho_1 = 85\%$ and $\rho_2 = 2$ meV.

Furthermore, one can note that the topographic image in Figure 9A shows strong roughness, which provides the possibility for the STM tip to detect tunneling behavior along different directions at different positions. This may give us the advantage to detect the superconducting gap features derived from different bands, which could be the reason for us to see two distinct gap structures at different positions. The same situation occurs in the STM measurements of MgB$_2$ bulk film. The STM tip can detect the gap with a magnitude of about 7.1 meV on the $\sigma$-band on some grains and can also measure the gap on the $\pi$-band with the value of 2.3 meV on other grains. However, as mentioned above, on the smooth surface, it is hard to observe a "clean" full gap feature. Most times the spectrum shows a mixture of the two, and a robust V-shape feature appears near zero bias. This may be understood in the way that, now the tunneling current mainly goes along $\sigma$ axis direction, with a reduced side tunneling component that would occur in the measurements on the rough surface. Based on the above discussion and the multiband features on the smooth surface, one may naturally conclude that the two kind of features on the measured spectra correspond to the gaps on different Fermi surfaces.

**Robust $d$-wave pairing and possible explanations for full gap**

Considering the common feature of V-shape spectra measured on superconducting thin films, we first discuss the possibility of a $d$-wave gap. It has been extensively demonstrated in many theoretical papers that the Nd$_{1-x}$Sr$_x$NiO$_2$ system has a robust $d$-wave pairing, in both limits using a weak-coupling approach of RPA and strong coupling of the $t-J$ model. Wu et al. found that, in all instances, the dominant pairing tendency is in the $d_{x^2-y^2}$ channel. In analogy with cuprates, heavy-fermion superconductors, superconductivity in the present compound is assumed to be mediated by spin fluctuations. Based on the multiorbital fluctuation exchange approximation, the effective pair interaction vertex $t(k,\mathbf{q})$ is determined by RPA susceptibility. By solving the dimensionless pairing strength functional, the largest pairing eigenvalue $\lambda$ will lead to the highest transition temperature, and its eigenfunction determines the symmetry of the gap. The dominant peaks locate around the $M$ and $A$ points, indicating intrinsic AF fluctuations. The prominent features in the orbital-resolved susceptibility are that the peaks around $M$ and $A$ are mainly attributed to the Ni $d_{x^2-y^2}$ orbital, which will further promote the dominant $d_{x^2-y^2}$-wave pairing state. As the effective low-energy interaction parameters remain undetermined for Nd$_{1-x}$Sr$_x$NiO$_2$, a large region of parameter space has been calculated within RPA calculations. The obtained pairing eigenvalues as a function of interaction strength $U_{xy}$ for $n = 0.8$ are displayed in Figure 9B. One can find that the $d_{x^2-y^2}$ pairing state is dominant.
over other competing ones, for example, for the $d_{xy}$ state. This is consistent with the fact that both the dominant density of states and pairing interactions reside on the Ni $d_{x^2-y^2}$ orbital.

The nickelate system seems to belong to the intermediate coupling one; it thus becomes important to resolve the pairing mechanism. From a strong-coupling perspective, concerning the nickelate system, the $t$-$J$ model can be reduced to one single Ni $3d_{x^2-y^2}$ orbital for simplicity. Wu et al.\textsuperscript{95} investigate the pairing state with an extended range of doping levels. Figure 9C plots the representative superconducting gap of the $d_{x^2-y^2}$ pairing versus different dopings. It shows that there is a superconducting dome, and the gap reaches the maximum at a doping of 0.1 hole/Ni. Moreover, the 3D gap function of the obtained $d_{x^2-y^2}$-wave pairing is displayed in Figure 9D at 0.2 hole doping, without considering the contribution of Nd orbitals. The main findings from a $t$-$J$ model analysis are consistent with the RPA analysis.

Now it can be understood that the measured V-shape spectra that featured a $d$-wave gap mainly originated from the Ni $3d_{x^2-y^2}$ orbital; meanwhile, several theoretical assumptions are proposed to explain the observed full gap. The first picture is that this full gap may just simply reflect the gap function on the hybridized orbitals of the Ni $3d_{x^2-y^2}$, $3d_{z^2}$ mixed with the Nd $5d_{x^2}$, $5d_{y^2}$, namely, on the $\beta$ and $\gamma$ Fermi pockets. However, if we just simply follow the $d_{x^2-y^2}$ notation for the gaps in the whole momentum space, the nodal line will cut the two electron pockets, which is not consistent with our observation of a full gap. Theoretically, Adhikary et al.\textsuperscript{66} propose a two-orbital model consisting of Ni $3d_{x^2-y^2}$ and an axial orbital that is constructed out of Nd $5d_{x^2}$, Ni $3d_{z^2}$, and Ni $s$ characters. Considering the crucial role of the interorbital Hubbard interaction in superconductivity, it turns out to be orbital selective electron pairing in Nd$_{1-x}$Sr$_x$NiO$_2$. Furthermore, Bandypadhyay et al.\textsuperscript{96} point out that the $f$-orbital of R element has an important role in the electronic structure of RNiO$_2$. The forbital can pull down the $d$-orbital to the Fermi level in Nd- and Pr-based nickelates compared with in the La-one. This difference shows up in computed doping-dependent superconducting properties of the three compounds within a weak coupling theory, which finds two-gap superconductivity for NdNiO$_2$ and PrNiO$_2$ and the possibility of a single-gap superconductivity for LaNiO$_2$.

Based on the proposal of the orbital selective pairing, we can plot a cartoon picture for the Fermi surfaces and the gap structure on different cuts of $k_z$ in the weak coupling scenario, as depicted in Figure 10. Concerning the interorbital Hubbard interaction between different orbitals, we expect not only a $d$-wave gap on the $\alpha$ pocket but also full gaps on the $\beta$ and $\gamma$ ones with opposite gap signs. The latter is a bit like the $s^\pm$ pairing in many pnictides. This scenario is quite interesting and tells that not only the intra-pocket interaction but also the inter-pocket interaction plays an important role here, leading to the orbital selective pairing. The second one to interpret this full gap relies on a recent theoretical calculation about the doping-dependent phase evolutions of the pairing symmetries.\textsuperscript{187} This phase diagram is established based on the proposed picture of self-doped Mott insulator,\textsuperscript{89} which shows an evolution from a $d$-wave dominant region to an s-wave region with the intermediate phase of $d$-is wave. To satisfy this model, we need to postulate that the doping level is not homogeneous in the film; thus, somewhere the system shows a $d$-wave gap, somewhere $s$-wave, and somewhere a mixture of the two. This seems compatible with our results; however, according to our experience of MBE growth, the doping level of Sr may not vary too much in the deposition region ($\delta \times 5 \text{ mm}^2$) of the thin film, but rather the local clustering or reconstruction may give more influential effects. The third one suggests that the full gap of the tunneling spectrum may arise from the NiO$_2$ terminated surface, which has natural buckling planes of NiO$_2$.\textsuperscript{188} This picture is also interesting, which can be checked out on a surface with atomically resolved morphology. The fourth one proposes that the experimental observation can be simply explained within a pairing scenario characterized by a $d_{x^2-y^2}$-wave gap structure with lowest harmonic on the Ni-band and higher harmonics on the Nd-band.\textsuperscript{189} This scenario can be tested in future STM experiments with improved sample quality where the position of STM tip with respect to the Nd$_{1-x}$Sr$_x$NiO$_2$ unit cell can be precisely determined. Last but not the least, the full gap feature may just be induced by the tunneling matrix problem. The symmetry of superconductor gap function refers to its transformation under crystal symmetry operations. In this general point of view, even a $p$-wave or a time-reverse-symmetry breaking $d$-wave would also produce the spectrum with a full gap feature. Because the V-shape spectra have been widely observed in our experiment, we thus believe that the $d$-wave gap should be a dominant one. And this is consistent with many theoretical calculations.

Intuitively, the pairing form in Nd$_{1-x}$Sr$_x$NiO$_2$ may serve as a bridge between the cuprates and the FeSC. Because the former has a single band feature and only the intraorbital interaction as the driving force for pairing, leading to the $d$-wave gap, while the latter is a multiband system, one needs not only intraband but also the interorbital interaction for pairing, resulting in the orbital selective pairing and $s$-wave gaps with opposite signs on different Fermi pockets.\textsuperscript{190–193} At the moment, we do not know whether the gaps on the hybridized-orbital-derived $\beta$ and $\gamma$ pockets are really fully gapped and whether these gaps have opposite signs. The direct experimental evidence...
of the \(d\)-wave gap on the \(\alpha\) pocket is also lacking. To resolve this issue, we need to do further phase-referenced quasiparticle interference experiments\(^{194–197}\) on single-crystal samples when they are available, which has been conducted successfully in FeSC\(^{190,198–200}\) and cuprates.\(^{201}\) Clearly, more efforts are desired in order to pin down the assignment of the superconducting gaps on different Fermi pockets.

Figure 8. STM measurement of topography and tunneling spectra on \(\text{Nd}_x\)-\(\text{Sr}_x\)-\(\text{NiO}_2\) film (A) 3D illustration of the topographic image after topotactic reaction method; one can see the surface roughness about 1–2 nm. (B) A tunneling spectrum with a V-shape, which is measured on the rough surface. The Dynes model fitting yields a gap function \(\Delta = 3.9\cos 2\phi\) (meV). (C) A tunneling spectrum with full gap feature, which is measured on the rough surface. The Dynes model fitting is \(\Delta = 2.35(0.85 + 0.15\cos 4\phi)\) (meV). (D) 3D illustration of the topographic image after a long-time vacuum annealing. The roughness becomes much smaller. We can see a clear step with the height about 0.17 nm, being consistent with half of the unit cell height. (E) A typical V-shape spectrum measured on the smooth surface. A gap function \(\Delta = 3.95(0.95\cos 2\phi + 0.05\cos 6\phi)\) (meV) is used in the fitting. (F) A mixture of the two-gap features on the spectrum is measured on the smooth surface. Dynes fitting results, \(\Delta_1 = 5.3(0.8\cos 2\phi + 0.2\cos 6\phi)\) (meV), \(\Delta_2 = 2\) meV, \(p_1 = 85\%\), and \(p_2 = 15\%\). Adapted from Gu et al.\(^{174}\)

Figure 9. Robust \(d\)-wave pairing (A) Bare susceptibility for \(n = 0.8\). (B) The pairing eigenvalues as a function of the interaction \(U_{\text{Ni}}\) for \(n = 0.8\). (C) The \(d_{x^2-y^2}\)-wave gap as a function doping with \(J_1 = J_2 = 0.1\) eV. Positive (negative) values relate to electron (hole) doping. (D) 3D illustration of \(d_{x^2-y^2}\)-wave gap. Adapted from Wu et al.\(^{90}\)
Meanwhile, we also point out both similarities and distinctions between nickelate and cuprates. Therefore, we need to focus on whether there are emergent intertwined electronic structures, magnetic excitations, and superconducting pairing. Meanwhile, we also point out both similarities and distinctions between nickelates and cuprates. Furthermore, bulk samples with the same structure and proper doping have no trace of superconductivity. It remains highly debatable and worthy of more attention.

As for a future perspective, despite the discovery of superconductivity in all the three kinds of Nd, Pr, La based 113 systems, $T_c$ is still too low compared to that of cuprates. Strictly speaking, nickelates may not be categorized into the family of high-temperature superconductors for now. We need to find out the reason and make an effort to increase $T_c$.

In conclusion, at the moment, it is just the beginning of research on superconductivity in the nickelate system. The phenomena observed so far show some similarities with cuprates, but more distinctions. Thus, the nickelate system provides a new platform for exploring unconventional superconductivity. The establishment and understanding of superconductivity in nickelates will shed new light on resolving the elusive pairing mechanism in high-temperature superconductors.

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AUTHOR CONTRIBUTIONS

Q.G. and H.W. wrote the manuscript. All authors read and approved the final manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.