Key Issues Review

Superconductivity in infinite-layer nickelates

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Abstract
The recent discovery of the superconductivity in the doped infinite layer nickelates \( R \text{NiO}_2 \) (\( R = \text{La, Pr, Nd} \)) is of great interest since the nickelates are isostructural to doped (Ca, Sr)CuO\(_2\) having superconducting transition temperature (\( T_c \)) of about 110 K. Verifying the commonalities and differences between these oxides will certainly give a new insight into the mechanism of high \( T_c \) superconductivity in correlated electron systems. In this paper, we review experimental and theoretical works on this new superconductor and discuss the future perspectives for the ‘nickel age’ of superconductivity.

Keywords: superconductivity, infinite-layer nickelates, strongly-correlated materials

(Some figures may appear in colour only in the online journal)

1. Introduction

Unconventional superconductivity in strongly correlated electron systems is one of the most central issues in condensed matter physics. In particular, the mechanism of high-temperature superconductivity in cuprates [1] has been the ‘holy grail’ for more than 35 years, and many experimental and theoretical studies have been carried out. One promising strategy to unveil this long-standing puzzle is to compare cuprate superconductors with their variants and clarify which of the features of the cuprates plays a decisive role in superconductivity. However, the search for layered oxides with an electronic state similar to that of copper oxides is a highly non-trivial problem with a long history. In fact, a number of materials that are expected to be compatible with the keywords such as ‘two-dimensional,’ ‘square lattice,’ ‘single orbital,’ and ‘near half-filling’ have been investigated.

The cuprate parent compounds are a Mott insulator with \( d^9 \) filling in the copper 3\( d \) orbital (one hole per site in the hole picture). If there is particle–hole symmetry, a two-dimensional system with one \( d \) electron per site will be promising. Although titanium and vanadium oxides are candidates for a \( d^1 \) analog of cuprates, if we consider a similar crystal structure as cuprates, the electronic structure is not necessarily similar to that of cuprates: in the \( d^1 \) system in a typical octahedral crystal field, the electrons enter nearly degenerate t\(_{2g}\) orbitals, and the ‘single orbital’ condition, one of the important keywords for cuprate superconductivity, is not easily satisfied [2–5]. Furthermore, the d\(_{xy}\) level needs to be lower than that of the d\(_{yz}\) and d\(_{zx}\) orbitals in contrast to the typical octahedral crystal field, and it requires extremely high pressure to realize such a situation [6]. It should also be noted that the t\(_{2g}\) orbitals do not point towards their ligands, so that the electronic structure associated with them, as well as their exchange interactions, is very different from those of the e\(_g\) orbitals.

In the cuprates, the onsite level of the d\(_{x^2−y^2}\) orbital is higher than that of the d\(_{3z^2−r^2}\) orbital. However, by lowering the position of the apical oxygen, the order can be reversed. Then a cuprate-like electronic state can be realized in the d\(_7\) system with one electron in the e\(_g\) shell. Nickel oxides provide
a playground for designing such $d^9$ analogs. However, the problem is how to control the position of the apical oxygen. Although such possibility has been explored considering artificial heterostructures, it seems difficult to achieve the ideal situation [7–9].

We can also consider a $d^9$ electron system where the orbital with the highest level in the $t_{2g}$ shell becomes half-filling. Iridium oxides are such an example, and there are experimental reports suggesting the possibility of superconductivity [10, 11]. When the spin–orbit coupling is strong as in the $5d$ electron systems, the $t_{2g}$ bands split into $j_{\text{eff}} = 1/2$ and $j_{\text{eff}} = 3/2$ bands ($j_{\text{eff}}$: effective total angular momentum). The $j_{\text{eff}} = 1/2$ band becomes half-filling when the filling is $d^9$, and an electronic structure similar to that of the cuprates may be realized. However, the energy splitting between the $j_{\text{eff}} = 1/2$ and $j_{\text{eff}} = 3/2$ bands is smaller than that between the $d_{z^2}$ and $d_{z^2-r^2}$ orbitals in the cuprates, and it is not clear whether iridium oxides can be regarded as a $j_{\text{eff}} = 1/2$ single-orbital system [12–14].

In this way, the hunt for analogs of cuprate superconductors has not been very successful so far. However, recently, there has been significant progress from infinite-layer nickelates. The infinite-layer phase of the nickelates is realized by topotactic reduction, whose first experimental report by Crespin et al. [15, 16] (synthesis of LaNiO$_2$ powder) dates back to 1983, even before the discovery of the superconductivity in cuprates [7–9]. As mentioned in section 1, ‘two-dimensional’, ‘square lattice’, ‘single orbital’, and ‘near half-filling’ were the keywords to search for analogs of cuprates. The infinite-layer nickelate $RNiO_2$ is a candidate that satisfies all of these. The possible similarity between the nickelates and cuprates gives excitement in the field. The ‘nickel age’ of superconductivity has thus started [24, 25].

2. Crystal structure and crystalline electric field

In this section, let us first look into the crystal structure and local electronic configuration of the infinite-layer nickelate $RNiO_2$ ($R = $ La, Pr, Nd) and compare with those of cuprates. The crystal structure of $RNiO_2$ is shown in figure 2(a). It has a layered structure, called an infinite-layer structure, with alternating NiO$_2$ and $R$ layers. The structure is similar to that of the cuprates with a layered structure including CuO$_2$ planes. In fact, the corresponding infinite-layer cuprate ($Sr_1-xCa_x)_{1-y}CuO_2$ also shows superconductivity with $T_c$ of about 110 K [28].

In the infinite-layer structure, the apical oxygens are absent, and the composition changes from the well-known nickel oxide $RNiO_2$. A naïve valence estimate shows that neodymium is a 3+ cation and oxygen is a 2− anion, giving nickel a 1+ valence. The corresponding occupation in the nickel 3d orbitals is $d^9$, which is the same as that of the Cu$^{2+}$ cations in the cuprate parent compounds. The square-planar crystal field [figure 2(b)] without the apical oxygens stabilizes the $d_{z^2}$ orbital; its onsite level becomes comparable to those of the $d_{x^2}$, $d_{y^2}$, and $d_{xy}$ orbitals (called $t_{2g}$ orbitals in the octahedral environment). Therefore, the $d_{z^2}$ orbital is isolated from the other 3d orbitals in the energy diagram. Considering this crystal structure together with the $d^9$ occupation, one expects that the half-filled $d_{z^2}$ orbital system is realized in $RNiO_2$.

As mentioned in section 1, ‘two-dimensional’, ‘square lattice’, ‘single orbital’, and ‘near half-filling’ were the keywords to search for analogs of cuprates. The infinite-layer nickelate $RNiO_2$ is a candidate that satisfies all of these. The possible similarity between the nickelates and cuprates gives excitement in the field. The ‘nickel age’ of superconductivity has thus started [24, 25].

3. Experimental studies

3.1. Synthesis of superconducting materials

In the first report [18], Li et al. reported the superconductivity in the composition of Nd$_{0.8}$Sr$_{0.2}$NiO$_2$ ($T_c = 9–15$ K). The superconductivity has also been observed in doped PrNiO$_2$ thin film, Pr$_{1-x}$Sr$_x$NiO$_2$, with a maximum $T_c$ of 14 K [19, 20]. All the superconducting samples are thin film samples on SrTiO$_3$ substrates, and the details of the stable synthesis of film samples are discussed in reference [30].

It is worth noting that several other groups have now reproduced superconductivity in doped infinite-layer nickelate film samples [31, 32–35], although, in the early stage, there was a report of failure in reproducing the superconductivity in thin film samples [36]. The phase diagram of Nd$_{1-x}$Sr$_x$NiO$_2$ thin film with different strontium concentration $x$ (figure 3) has been revealed independently by Li et al. [37] and Zeng et al. [31] (the groups of H Y Hwang and A Ariando, respectively). As the latest news, the same two groups have independently observed the superconductivity in doped LaNiO$_2$ thin films [21, 22]. This is remarkable because the initial work
had not succeeded in realizing superconductivity in doped LaNiO$_2$ [18] (see, e.g., reference [38] for the literature debating a possible reason for the absence of superconductivity). Furthermore, reference [21] also reported that there is a sign of superconductivity in undoped LaNiO$_2$ thin films [figure 4(a)]. This is interesting because NdNiO$_2$ and PrNiO$_2$ thin films do not show superconductivity in their parent compounds. In contrast, undoped LaNiO$_2$ film samples in reference [22] are insulating from 300 K to 2 K. It is desirable that the origin of the discrepancy between the two reports be clarified in the future.

As for bulk samples, to the best of our knowledge, there is no success in reproducing superconductivity. We refer to references [39–42] for experimental effort on bulks (see also reference [43] for the discussion on the thermodynamical stability of the bulk infinite-layer nickelates).

### 3.2. Phase diagram

Figure 3 shows the phase diagram of Nd$_{1-x}$Sr$_x$NiO$_2$ thin films on the SrTiO$_3$ substrate. First, the resistivity of the parent material does not show a clear insulating behavior. The term ‘weakly insulating’ refers to the slight upturn of the resistivity at low temperatures. Superconductivity is observed in the region where the strontium doping concentration is $0.125 \lesssim x \lesssim 0.25$, and a dome-shaped $T_c$ is observed. In the overdoped region, where the superconductivity disappears, the resistivity of Nd$_{1-x}$Sr$_x$NiO$_2$ increases at low temperatures, in contrast to the cuprates, which show metallic behavior. The increase of the resistivity may be due to, e.g., disorder, electron correlation, and secondary order parameters. The origin of this behavior is discussed in, e.g., reference [44] (see section 3.3).

What is striking is the difference in the behavior of the parent materials between the nickelate and the cuprates. The cuprate parent compounds are an antiferromagnetic Mott insulator, whereas NdNiO$_2$ is not an insulator. Whereas the presence of the magnetic order in the thin film samples is not clear, the bulk powder NdNiO$_2$ samples show no long-range antiferromagnetic ordering down to low temperatures (1.7 K) [45]. We will discuss magnetism further in section 4.2.2.

Figure 4 shows the comparison of $T_c$ among the superconducting infinite-layer nickelates [21, 22]. One may be able to draw a unified phase diagram like in figure 4(b). It is an important future task to clarify the similarities and differences in more detail among superconducting family members.
Figure 3. (a) Doping ($x$) dependence of the resistivity and (b) and (c) temperature-doping phase diagram of Nd$_{1-x}$Sr$_x$NiO$_2$ thin film samples on SrTiO$_3$ substrate. Reprinted figure with permission from [31, 37], Copyright (2020) by the American Physical Society.

Figure 4. Comparison of the transition temperature among the superconducting infinite-layer nickelates [doped RNiO$_2$ ($R =$ La, Pr, Nd) thin film samples]. (a) and (b) [21] John Wiley & Sons. © 2021 Wiley-VCH GmbH. (c) Reproduced from [22]. CC BY 4.0.

Figure 5. Contour maps of the Hall coefficients of doped NdNiO$_2$, PrNiO$_2$, and LaNiO$_2$ thin films. [21] John Wiley & Sons. © 2021 Wiley-VCH GmbH.

3.3. Experiments to understand electronic structure, superconductivity, and magnetism

Other experimental facts are summarized as follows.

- Hall coefficient measurements of thin film samples suggest a coexistence of electron and hole carriers in Nd$_{1-x}$Sr$_x$NiO$_2$, and the sign of the Hall coefficient changes depending on the strontium concentration $x$ and the temperature [31, 37]. A qualitatively similar sign change is also seen in doped PrNiO$_2$ and LaNiO$_2$ thin films [20–22] (figure 5).

- Experiments using the x-ray techniques and electron energy loss spectroscopy (EELS) reveal that the electronic states of the rare-earth layer show up around the Fermi level [figure 6(a)] and that NdNiO$_2$ is more close to the Mott–Hubbard regime in Zaanen–Sawatzky–Allen classification [46] compared to cuprates [47–50]. It suggests that the holes are mainly doped into nickel 3$d$ orbitals (a signal for the doped holes in the oxygen 2$p$ orbitals is also observed in the O $K$-edge EELS measurement, but the intensity is much smaller than that of the cuprates [49] [figure 6(b)].

- Motivated by the discovery of the superconductivity, the magnetism of the bulk infinite-layer nickelates is reinvestigated using NdNiO$_2$ [48, 51], Nd$_{0.85}$Sr$_{0.15}$NiO$_2$ [52], LaNiO$_2$ [51, 53], and PrNiO$_2$ [51] samples. Recently, a RIXS measurement has been performed on NdNiO$_2$ thin film samples [54]. A quasi-two-dimensional magnetic excitation with a bandwidth of about 200 meV with
large damping is observed (figure 7). It suggests a nonnegligible magnetic exchange interaction of about 64 meV on the NiO$_2$ layer [54], while a Raman experiment using bulk samples gave a smaller value of 25 meV [48]. The origin of the lack of a clear long-range magnetic order is also an open question. See section 4.2.2 for more detailed discussion.

• Scanning tunneling microscope/spectroscopy (STM/STS) experiments on Nd$_{1-x}$Sr$_x$NiO$_2$ film samples observe, depending on the position of the inhomogeneous surface, $V$-shape or full-gap-type single-particle tunneling spectra (figure 8). In some cases, mixed spectra of the two components are observed [32]. The $V$-shape spectrum is compatible with the $d$-wave gap, whereas the full-gap-type spectrum suggests the $s$-wave symmetry. Reference [32] interprets the coexistence of the different tunneling spectra by the different gap symmetries on different Fermi surfaces, while other explanations may also be plausible. The effects of the multi-orbital gap structure, the inhomogeneity, and surface reconstruction remain to be elucidated [55–58].

• The upper critical field of the doped NdNiO$_2$ thin film samples is investigated by two independent groups [59, 60]. Compared to the cuprates, the upper critical field shows a smaller spatial anisotropy. The analyses of the critical field suggest that the paramagnetic effect is dominant over the orbital effect (Pauli limit).

• When the system is in the Mott–Hubbard regime, the doped configuration becomes mainly $d^8$, rather than $d^9L$ ($L$ denotes a hole in ligand oxygen) in the case of the charge-transfer regime. Reference [61] has investigated the multiplet structure of the $d^8$ configuration of doped NdNiO$_2$ film samples using the XAS and RIXS, and concluded that the orbital-polarized spin-singlet state, where the doped holes reside in the nickel 3$dx^2$−$y^2$ orbital, gives a dominant contribution. See section 5.4 for the detailed discussion on the $d^8$ multiplet structure.

• Reference [62] has reported a diamagnetic response in superconducting Nd$_{1-x}$Sr$_x$NiO$_2$ film samples. The film thickness dependence of the physical observables is also explored. The superconductivity is observed for different thickness (from 4.6 nm to 15.2 nm) samples, and thicker films tend to show a higher $T_c$. The change in the Hall coefficient and the XAS spectra depending on the thickness implies the modulation of the electronic structure due to the strain and interface effects.

• Normal state resistivity of Nd$_{1-x}$Sr$_x$NiO$_2$ film samples is studied in reference [44] by suppressing the superconductivity with out-of-plane magnetic fields. The upturn of the resistivity at low temperatures observed around $x = 0$ outside the superconducting dome persists in the field-induced normal state up to $x \approx 0.225$. At $x \approx 0.225$, a metallic transport is observed, but, above $x \approx 0.225$, the resistivity upturn shows up again. The systematic doping evolution of the transport property implies the intrinsic correlation effect and possible secondary order parameter, for which further investigations are required.

The consistency between these experimental facts and theory will be discussed in the following sections.
4. Electronic structure of infinite-layer nickelates

4.1. Insight from DFT calculations

In section 2, we have discussed that the parent material, RNiO$_2$ ($R = \text{La, Pr, Nd}$), might be a single-orbital strongly-correlated system on a two-dimensional square lattice with half-filled $3d_{x^2-y^2}$ orbital, based on simple valence and crystal-field analyses. However, as we discuss in the following, this picture is not entirely true [63, 64,66–68]. Experimental facts reviewed in section 3 also suggest that there should be a deviation from the simple picture. In this section, we discuss the electronic structure of the infinite-layer nickelates through first-principles calculations.

Here, we restrict ourselves to bulk properties. The film thickness is on the order of 10 nm [30], so that there are several tens of NiO$_2$ layers in the sample. Furthermore, the superconductivity is robustly observed for samples with different thicknesses (from 4.6 nm to 15.2 nm) [62]. However, since the infinite-layer structure consists of charged layers, the reconstruction of the lattice and electronic structures is naturally expected at the interfaces and surfaces. Such effect is studied theoretically in references [58, 69–73] (see also ref-
Fermi surface of NdNiO$_2$. For the labels of high-symmetry points in the Brillouin zone, see the panel (a). Orange, gray, and red spheres indicate the neodymium, nickel, and oxygen atoms, respectively. Yellow surfaces show an isosurface of the electron density. It has large weights around the missing apical oxygen site and neodymium site. Reprinted figure with permission from [63]. Copyright (2019) by the American Physical Society. (c) Fermi surface of NdNiO$_2$. For the labels of high-symmetry points in the Brillouin zone, see the panel (a). The nickel 3$d_{x^2−y^2}$ orbital forms a large quasi-two-dimensional Fermi surface (red). The Fermi pocket near the Γ point (blue) has mainly neodymium 5$d_{z^2−r^2}$ character. The Fermi pocket near the A point (green) is derived from the bonding orbital between the interstitial s orbital and the neodymium 5$d_{xy}$ orbital [see (b)]. The image was drawn using FermiSurfer [65].

Figure 9. Electronic structure of bulk NdNiO$_2$ calculated using DFT with the GGA exchange–correlation functional. The lattice constants are taken from reference [45]. (a) Band structure of NdNiO$_2$ colored by the weight of each orbital (fat band). The neodymium 4$f$ orbitals, which would form localized spins, are treated as ‘frozen core’. (b) Contour plot of the real-space electron density of the Bloch state marked with the $\times$ symbol between the $R$ and $A$ points near the Fermi level in the panel (a). Orange, gray, and red spheres indicate the neodymium, nickel, and oxygen atoms, respectively. Yellow surfaces show an isosurface of the electron density. It has large weights around the missing apical oxygen site and neodymium site. Reprinted figure with permission from [63]. Copyright (2019) by the American Physical Society. (c) Fermi surface of NdNiO$_2$. For the labels of high-symmetry points in the Brillouin zone, see the panel (a). The nickel 3$d_{x^2−y^2}$ orbital forms a large quasi-two-dimensional Fermi surface (red). The Fermi pocket near the Γ point (blue) has mainly neodymium 5$d_{z^2−r^2}$ character. The Fermi pocket near the A point (green) is derived from the bonding orbital between the interstitial s orbital and the neodymium 5$d_{xy}$ orbital [see (b)]. The image was drawn using FermiSurfer [65].

Experimental consideration has also started [62] (see section 3.3).

Figure 9(a) shows the band structure of the parent material NdNiO$_2$ (the first infinite-layer nickelate superconductor) calculated based on the density functional theory (DFT) with the generalized gradient approximation (GGA). In general, the band structure of strongly correlated materials deviates (at least quantitatively) from that of the DFT calculation due to electron correlation effects (mass renormalization, Mott-gap opening, and so on), especially near the Fermi level [75]. However, to see the global energy structure, the DFT-GGA calculations give a good starting point. See section 4.2.1 for the discussion of correlation effects beyond the DFT.

The analysis of the band character near the Fermi level (figure 9) shows that the nickel 3$d_{x^2−y^2}$ orbital creates a large Fermi surface. This is in agreement with the prediction of section 2, and is a common feature with the cuprates. Also, consistently with the crystal-field analysis in section 2, the 3$d_{x^2−y^2}$ orbital has the highest onsite amplitude among the nickel 3$d$ orbitals, and the 3$d_{x^2−y^2}$ band is isolated from the other 3$d$ bands in the energy space.

However, there is a discrepancy from the naive expectation shown in section 2. Besides the large Fermi surface of the nickel 3$d_{x^2−y^2}$ orbital, additional Fermi pockets exist near the $\Gamma$ and $A$ points [figures 9(a) and (c)]. The Fermi pocket around the Γ point is formed by the rare-earth 5$d_{z^2−r^2}$ orbital with nickel 3$d_{x^2−y^2}$ orbital being hybridized [64, 66]. This fact may make nickel 3$d_{x^2−y^2}$ orbital active at the parent compound [64].

The origin of the Fermi pocket around the A point is referred to as the rare-earth 5$d_{xy}$ orbital [67, 68] or the interstitial s orbital at the apical site [76]. In reality, the bonding orbital is formed between these two orbitals [63] [figure 9(b)]: at the A point, there is a large bonding–antibonding energy splitting of more than 10 eV [77], and the bonding part forms the additional Fermi pocket. This bonding orbital can be described either by 5$d_{xy}$-like Wannier orbital centered at the rare-earth site or the s-like orbital centered at the interstitial apical site; therefore, both pictures can be applied [63].

In most materials, interstitial-orbital bands appear far away from the Fermi level on the unoccupied side. However, in NdNiO$_2$, the interstitial orbital at the apical site is stabilized because there is a space to gain the kinetic energy thanks to the absence of apical oxygen, and negatively-charged electrons feel an attraction from the surrounding nickel and neodymium cations. The neodymium 5$d$ and interstitial-s orbitals are both located in the neodymium layer, and will henceforth be referred to collectively as Nd-layer orbitals (or more generally, rare-earth-layer orbitals).

Because the rare-earth-layer orbitals form the additional Fermi pockets, the nickel 3$d_{x^2−y^2}$ orbital deviates from the half-filling even in the parent compound (occupation of the nickel 3$d$ orbitals is not $\frac{1}{2}$) [64]. This ‘self-doping’ effect is also seen in other infinite-layer compounds RNiO$_2$ (see, e.g., references [78, 79] for systematic investigation on the rare-earth element dependence). The self-doping marks one of the major differences from the cuprates, in which only the 3$d_{x^2−y^2}$ band crosses the Fermi level (except for some materials) [64]. The DFT-level estimates suggest that the nickel 3$d_{x^2−y^2}$ orbital is about 10% hole doped at the parent compound [63, 66, 67].

In order to further investigate the similarities and differences between the nickelates and cuprates, we show, in figure 10, the comparison of the band structures of NdNiO$_2$ and CaCuO$_2$, a copper oxide with the same infinite-layer

\begin{footnote}
\textsuperscript{3}The band structure is similar to that of electrides, in which interstitial bands are occupied and the electrons themselves become negative ions.
\end{footnote}
structure as NdNiO₂ (see references [64, 66] for detail). The weights of the nickel/copper 3\(d_{x^2-y^2}\) orbital, the oxygen 2\(p\) orbitals hybridized with the 3\(d_{x^2-y^2}\) orbital, and the neodymium/calcium-layer orbitals are also shown as ‘fat bands’.

In the case of CaCuO₂, the copper 3\(d\)-orbital band is close to the oxygen 2\(p\)-orbital band, and the charge-transfer energy (the potential energy difference between the copper 3\(d\) orbital and the oxygen 2\(p\) orbital, which is the energy required for a hole to move from the copper site to the oxygen site) is small. On the other hand, in the case of NdNiO₂, the valence of the nickel cation is about 1\(^+\) and the attraction from the nucleus is small, which lifts the energy level of the 3\(d\) orbitals and makes the charge transfer energy larger than that of the cuprates. Due to the higher energy level of the 3\(d\) orbitals, the 3\(d_{x^2-y^2}\) band overlaps with the conduction band on the unoccupied side (originating from the Nd-layer orbitals). The self-doping has thus occurred. A larger charge transfer energy makes the hybridization between the nickel 3\(d_{x^2-y^2}\) and oxygen 2\(p\) orbitals smaller. As a result, the bandwidth of 3\(d_{x^2-y^2}\) band (more precisely, the antibonding band between the nickel 3\(d_{x^2-y^2}\) and oxygen 2\(p\) orbitals) in NdNiO₂ is several tens percent smaller than that of CaCuO₂ (figure 11).

In this section, we mainly discuss the electronic structure of NdNiO₂. Importantly, a larger charge transfer energy compared to the cuprates and the existence of the self-doping are common features among LaNiO₂, PrNiO₂, and NdNiO₂. Indeed, if we assume that the rare-earth 4\(f\) orbitals are localized and treat them as ‘frozen core’, the three compounds show qualitatively similar band structures [78, 79]. However, we note that the role of the rare-earth 4\(f\) orbitals has not been settled; a coupling between the rare-earth 4\(f\) orbitals and the orbitals around the Fermi level may affect the electronic structure around the Fermi level [80–83]. We will come back to this point in section 5.2.

4.2. Correlation effect on the electronic structure

Here, we discuss the reconstruction of the electronic structure due to the correlation effects.

4.2.1. Calculations beyond DFT. Many-body effects beyond the DFT level have been investigated using GW-type approach...
[77, 84, 85], DMFT (dynamical mean-field theory [86, 87])-type approach [38, 76, 88–103], and a combination of GW- and DMFT-type approaches [104, 105]. The main effects of the many-body correlations are the mass renormalization of the correlated orbitals as well as the relative shift of the orbital on-site levels. Frequency-dependent self-energy gives rise to incoherent parts in the spectral function (figure 12). Whereas the GW-type approach is good at describing spatially nonlocal correlations, the DMFT-type approach captures the spatially local correlations well. Within the DMFT, the Mott–Hubbard and charge-transfer physics, as well as the local multiplet structure arising from the crystal field and Hund’s coupling, can be studied.

It seems that most of the studies show a qualitative agreement in that the self-doping band robustly remains even with the many-body correlation effects. However, unfortunately, so far, there is no consensus on the role of rare-earth-layer orbitals and the hole-doped electron configuration. Some works support the presence of Kondo physics arising from the coupling between the itinerant rare-earth-layer orbitals and correlated nickel 3d orbitals [76, 92]. Some papers propose the importance of Hund’s coupling and insist that the multi-orbital nature in the nickel 3d manifold is essential [89, 91–93, 100, 102, 104, 105]. On the other hand, references [90, 95, 103] argue that the important correlation effect lies in the nickel 3d\textsubscript{2}\textsubscript{−}\textsubscript{2} single band. The situation might be intermediate between these two [101]. Furthermore, reference [94] proposes that the charge-transfer physics is important as in the cuprates, while references [90, 95, 103] are based on the Mott–Hubbard-type picture. These discrepancies may arise from different conditions (interaction strength, orbital basis, double-counting correction, and so on) employed in the DMFT calculations [96]. For the controversy on active degrees of freedom, we give an extended discussion in section 5.

4.2.2. Magnetism. Another effect of the electron correlation would be inducing some symmetry breaking: magnetism, stripe order, and so on (as for the superconductivity, we discuss in section 5). Among them, magnetic instability is of great importance when we compare the infinite-layer nickelates with the cuprates.

So far, there exists no clear evidence for the magnetic long-range order in the parent compounds NdNiO\textsubscript{2} [45] and LaNiO\textsubscript{2} [106]. On the other hand, various theoretical studies have found magnetic solutions [64, 66, 76, 88, 93, 94, 97–100, 107–110] (figure 13). Although we do not go into detail further because of the lack of experimental evidence for the magnetic order in the parent compounds (for doped bulk Nd\textsubscript{0.85}Sr\textsubscript{0.15}NiO\textsubscript{2}, an NMR study suggests the presence of short-range antiferromagnetic ordering below 40 K [52]), we emphasize that understanding the magnetism in the nickelates is one of the most urgent and important future tasks.

Another interesting related issue is the strength of the magnetic exchange coupling \( J \), which may be one of the key factors for the high-\( T_c \) superconductivity [111]. The cuprates are a charge-transfer material, and \( J \) becomes large (~130 meV) due to the superexchange mechanism [111].

It is an interesting question whether the nickelates, which have larger charge-transfer energy and are more close to the Mott–Hubbard regime, exhibit large \( J \) or not. The \( J \) value for the infinite-layer nickelates is not settled: a Raman experiment using NdNiO\textsubscript{2} bulk samples estimated \( J \) to be much smaller (25 meV) than that of the cuprates [48]; on the other hand, a recent RIXS experiment on NdNiO\textsubscript{2} thin film samples gave a larger value of \( J = 64(3) \) meV [54]. The \( J \) value is also scattered in theoretical estimates [78, 82, 88, 97–99, 107–109, 112–115]. One of the reasons for the discrepancy in theoretical estimates is ascribed to the ambiguity in calculating \( J \): because the infinite-layer nickelate is not a Mott insulator due to the self-doping, there is ambiguity in mapping the system into the spin models. However, there seems to be an overall consensus that the \( J \) value is smaller than that of the cuprates. The problem is how small it is. This is an important question to be clarified in the future because it might be related to the difference in \( T_c \) between the nickelates and cuprates.

5. What are essential degrees of freedom for superconductivity?

In section 4, we see that, although the self-doping and the large charge-transfer energy make a distinction between the nickelates and cuprates, they are similar in that the strongly correlated 3d\textsubscript{2}\textsubscript{−}\textsubscript{2} orbitals have large Fermi surfaces at the DFT level. Then, the most interesting and fundamental question would be the pairing mechanism of superconductivity.

Reference [63] calculated, from first principles, the electron–phonon coupling constant of NdNiO\textsubscript{2} and estimated the transition temperature \( T_c \) assuming the phonon Bardeen–Cooper–Schrieffer [116] mechanism. The estimated \( T_c \) is less than 1 K. Thus, the phonon mechanism cannot explain the experimental \( T_c \) on the order of 10 K. This suggests that the superconductivity in doped NdNiO\textsubscript{2} originates from an unconventional mechanism (we note that the electron–phonon interactions may contribute to the superconductivity in cooperation with other mechanisms). Indeed, unconventional
mechanisms have been proposed theoretically from the early stage [67, 68, 117]. Experimentally, the STM/STS experiment has observed a $d$-wave-like gap in some regions of the inhomogeneous surfaces of doped NdNiO$_2$ films [32] (see also section 3.3).

When discussing unconventional mechanisms, it is helpful to construct an effective lattice Hamiltonian, such as the Hubbard model, $t$-$J$ model, $d$-$p$ model, or periodic Anderson model, for the electronic degrees of freedom near the Fermi level and analyze the superconductivity based on it. The effective Hamiltonian should reflect the crystal and electronic structures of the system. Then, what is the minimum model to describe the superconductivity in the infinite-layer nickelates (different models incorporate different physics such as Mott, Hund, and Kondo physics, as we discuss below)? In other words, what are the essential degrees of freedom for describing superconductivity? In this section, we will discuss this question.

So far, many different proposals have been made. However, there seems to be a consensus that the nickel 3$d_{x^2-y^2}$ orbital is one of the essential degrees of freedom. Candidates for other key degrees of freedom include

(a) The itinerant rare-earth-layer orbitals that form the additional Fermi pockets.
(b) The rare-earth 4$f$ orbitals.
(c) The oxygen 2$p$ orbitals that hybridize with 3$d_{x^2-y^2}$ orbitals on the NiO$_2$ plane ($p_x$ orbital of the oxygen in the $x$-direction and $p_y$ orbital of oxygen in the $y$-direction).
(d) Nickel 3$d$ orbitals other than the 3$d_{x^2-y^2}$ orbital.

Each of these is discussed in the following.

5.1. Itinerant rare-earth-layer orbitals forming additional Fermi pockets

As we see in section 4, the rare-earth-layer orbitals form Fermi pockets around the $\Gamma$ and $A$ points. The x-ray experiments on film samples consistently suggest that the rare-earth-layer orbitals are partially occupied [47]. Therefore, when discussing the symmetry of the superconductivity gap, we need to consider the gap functions on these Fermi surfaces. A key question, however, is whether the superconducting gaps on these bands are a byproduct of the superconductivity in the 3$d_{x^2-y^2}$ band or whether they play an intrinsic role in the emergence of superconductivity. To put it a little differently, in discussing the properties such as superconductivity and magnetism, are the rare-earth-layer orbitals more than 'charge reservoir' controlling the filling of the nickel 3$d_{x^2-y^2}$ orbitals?

If the hybridization between the nickel 3$d$ and rare-earth-layer orbitals is substantial, the rare-earth-layer orbitals are not only a charge reservoir, but they might give Kondo-like physics [113, 118]: the rare-earth-layer electrons have large bandwidth and couple to localized spins at nickel sites as itinerant conduction electrons. In this case, the increase in the electrical resistivity seen in NdNiO$_2$ from around 70 K [18] is interpreted as the Kondo effect [113, 118]. Following this picture, effective Hamiltonians that include both nickel 3$d$ and rare-earth-layer orbitals have been proposed [47, 76, 113]. Reference [56] has analyzed superconductivity based on the model proposed in reference [113] and shown pairing instabilities towards $d$, $d+i$s, and $s$ waves depending on the parameter region (figure 14).

The presence of Kondo physics is under debate: the recent experiments on the resistivity of magnetic-field-induced normal state using Nd$_1-x$Sr$_x$NiO$_2$ film samples have reported the resilience of the resistivity upturn against magnetic field and the positive magnetoresistance [44]. Reference [44] poses a question on the Kondo scenario, whose effect is expected to be suppressed by the magnetic field. On the other hand, reference [119] observes negative magnetoresistance for LaNiO$_2$ film samples. The Kondo temperature is affected by the density of states and Fermi energy of the conduction bands and the strength of the Kondo coupling between the local spins and itinerant electrons [120]. The density of states of the rare-earth-layer orbital is not large [63]. As for the hybridization between the nickel 3$d_{x^2-y^2}$ and rare-earth-layer orbitals giving the Kondo coupling, some works argue that the hybridization is weak [90, 94], whereas others emphasize its importance [76, 92].

5.2. Rare-earth 4$f$ orbitals

At the moment, the role of the rare-earth 4$f$ orbitals is an open question. Since the 4$f$ orbitals are spatially localized, the bandwidth of 4$f$ orbitals becomes small. Because of the correlation effect, the 4$f$ electrons tend to be localized and form local magnetic moments. The disordered localized spins on the rare-earth layer may affect the transport property on
5.3. Oxygen 2p orbitals

It is of great importance to consider the role of oxygen 2p orbitals when comparing the cuprates and nickelates. A problem is to which orbitals the doped holes go (see figures 15 and 16). In the cuprates classified as a charge-transfer insulator in the Zaanen–Sawatzky–Allen phase diagram [46], the doped holes mainly enter the oxygen 2p orbitals because the charge-transfer energy $\Delta_{dp}$ is smaller than the 3d-orbital Hubbard interaction $U_{dd}$. Therefore, the electron configuration of the hole-doped cuprates is mainly $d^xL^y$. And the physics of Zhang–Rice singlet emerges [121].

On the other hand, in the case of the infinite-layer nickelates $R$NiO$_2$, $\Delta_{dp}$ is larger than that of the cuprates (see section 4). This suggests that $R$NiO$_2$ is a Mott–Hubbard type material. The doped holes mainly go to the nickel 3d orbitals, and the doped electron configuration is mainly represented by $d^9$ (doping is expected to make the rare-earth-layer Fermi pockets smaller and weaken the self-doping effect). Note, however, that the hybridization between the nickel 3d$_{x^2-y^2}$ orbital and the oxygen 2p orbitals is nonzero. Therefore, some holes should go to oxygen 2p orbitals [94, 109]. In fact, the O K-edge EELS measurement has detected a signal of holes in the oxygen 2p orbitals, but the intensity is much smaller than that of the cuprates [49] (see also section 3.3).

5.4. Other 3d orbitals

Based on the above discussion, the undoped $d^9$ configuration and the $d^9$ configuration with holes in the nickel 3d orbitals would be important in describing the superconductivity in the infinite-layer nickelates. When we follow this picture, an effective Hamiltonian would consist of nickel 3d orbitals (note that, in this case, the hybridization of the oxygen 2p orbitals with the nickel 3d$_{x^2-y^2}$ orbital is taken into account by considering 3d$_{x^2-y^2}$-like Wannier orbitals centered at nickel sites with oxygen 2p tails). This raises a question: which model is more appropriate, the single-orbital or multi-orbital 3d model?

This can be understood as a competition between Hund’s coupling and the crystal-field splitting, where the former (latter) favors the high-spin (low-spin) configuration [figure 15(b)]. If the crystal-field splitting between the 3d$_{x^2-y^2}$ orbital and the other 3d orbitals is sufficiently large, the holes stay in the 3d$_{x^2-y^2}$ orbitals. Then, the doped $d^9$ configuration takes the low-spin state ($S = 0$), and the single-orbital picture would be justified. If Hund’s coupling induces the high-spin ($S = 1$) $d^9$ configuration, the multi-orbital model becomes indispensable. Several theoretical studies have argued that the multi-orbital nature cannot be ignored [89, 91–93, 100, 102, 104, 105, 110, 112, 122–124] (figure 17). On the other hand, references [90, 95, 103] argue a single-orbital picture (for example, a recent DFT + DMFT study [103] has well reproduced the XAS, XPS, and RIXS spectra of NdNiO$_2$ in references [47, 48, 61] with the low-spin ground state). An intermediate picture also exists: while the low-spin state is realized at low energy, the trace of Hund’s coupling can be observed as dynamical orbital fluctuations at high frequencies [101]. Experimentally, reference [61] performed RIXS
Figure 15. (a) Schematic figure showing the difference between a Mott–Hubbard insulator and a charge-transfer insulator. UHB and LHB are the upper Hubbard and lower Hubbard bands of the $3d$ orbitals, respectively. (b) Schematic energy diagram for the hole doping. There is a large energy scale competition between $d^9$ and $d^8$ ($U_{dd}$ vs $\Delta_{dp}$). Within the $d^8$ configuration, a smaller energy scale competition exists between the low-spin ($S = 0$) and high-spin ($S = 1$) states ($\Delta$ vs $J_H$). $\Delta$ is the crystal-field splitting between the $3d_{x^2-y^2}$ orbital and the other $3d$ orbitals [see figure 2(b)]. $J_H$ is Hund’s coupling of the nickel $3d$ orbitals.

Figure 16. Schematic figure comparing the cuprates and nickelates assuming the charge-transfer energy $\Delta_{dp}$ (denoted as $\Delta$ in the figure) in the nickelates is much larger than that of the cuprates. The blue bands show Hubbard bands, while the red bands are oxygen $2p$ bands with the $pd$ hybridization being switched off. Reprinted figure with permission from [112], Copyright (2020) by the American Physical Society.

and XAS measurements using the doped NdNiO$_2$ film samples and suggested a low-spin character of the doped configuration (see also reference [125] and references therein for the situations for other $d^8$ nickelates with the square-planar environment).

5.5. Brief summary

To summarize the above discussion, most of the works view that the correlation effects in the nickel $3d_{x^2-y^2}$ orbital are important. Indeed, ab initio estimates of the effective Coulomb interactions within the nickel $3d$ manifold have suggested that the infinite-layer nickelates are strongly correlated systems [63, 67]. When one considers the nickel $3d_{x^2-y^2}$ orbital to be the most essential degrees of freedom, the $d$-wave pairing might be plausible: theoretically, spin-fluctuation-induced $d$-wave pairing for the nickel $3d_{x^2-y^2}$ orbital has been proposed [67, 68] (figure 18). From the standpoint of the single-orbital picture, the experimentally observed dome-like $T_c$ [20–22, 31, 37] would be related to the change in superconducting instability as a function of the occupation of the nickel $3d_{x^2-y^2}$ orbital [90] (figure 19).

On the other hand, if we take a position that the multi-orbital nature is important, a comparison with iron-based superconductors may be interesting [100]. Enhanced dynamical spin fluctuations around spin-freezing (a phenomenon where the local moment is very slowly fluctuating, i.e., almost frozen) crossover induced by physical and effective Hund’s coupling may be relevant to the superconductivity [89]. Strong-coupling-expansion type approach using the $3d$ multi-orbital model may help understanding the unconventional pairing [122–124]. Even when the Fermi surfaces of the other $3d$ orbitals do not exist, if other $3d$ bands exist just below the Fermi level (incipient band), such incipient bands can play a role in enhanced the superconductivity [57].

When the rare-earth-layer orbitals are active, they lead to another type of unconventional pairing through the Kondo coupling [56]. Even when they are not the main player in the development of superconductivity, they do form Fermi pockets. Thus, the superconducting gap should also be finite on these additional bands. The interactions between the nickel $3d$ orbitals and the rare-earth-layer orbitals would be an important factor in determining a gap structure for the additional Fermi pockets [55]. It is an important future task for experiments to reveal the gap structure on multiple Fermi pockets (a starting point is given by reference [32], see section 3.3).

6. Searching for a new addition to the family

As we describe in section 3, rare-earth infinite-layer nickelates show superconductivity for rare-earth elements of La, Pr, and Nd. Superconductivity may also appear for other rare-earth
element cases, which is an interesting future task to be elucidated. Another intriguing issue is whether the superconductivity appears only in the infinite-layer structure or not. Here, we discuss several possible candidates for a new member of the family of nickel-based superconductors.

6.1. Multi-layer nickelates

The most natural extension of the infinite-layer nickelates would be the multi-layer square-planar nickelates [27, 126]. This is because the rare-earth infinite-layer nickelates are a special case (\(n = \infty\)) of the multi-layer square-planar nickelates \(R_{n+1}\text{Ni}_n\text{O}_{3n+1}\). In the case of the cuprates, superconductivity appears both in infinite-layer and multi-layer structures [127].

Multi-layer square-planar nickelates with \(n = 2\) and \(3\) had already been synthesized using the reduction process from the Ruddlesden–Popper parent compound \(R_{n+1}\text{Ni}_n\text{O}_{3n+1}\) (see, e.g., reference [26] for a review). This situation is similar to the infinite-layer case, where \(RNiO_2\) is obtained by the reduction from \(RNiO_3\). An average nominal valence of the nickel ions in the \(n = 3\) compound is 1.33+ (i.e., \(d^{10}\)), which corresponds to the overdoped regime in the cuprates (see figure 20 for the crystal structure and the DFT band dispersion). Indeed, multi-layer nickelates \((n = 3)\) do not show superconductivity [128]. However, there is a signature for a large magnetic exchange coupling about 70 meV in the trilayer nickelate \(La_2Ni_3O_8\), which makes multi-layer nickelates interesting reference systems between the cuprates and infinite-layer nickelates [129]. Increasing \(n\) would put the filling of nickel 3\(d\) orbital of multi-layer nickelates in the right place for possibly realizing superconductivity [24, 27]. In this respect, a notable experimental advance is a recent report of the epitaxial growth of the parent compound before the reduction, the Ruddlesden–Popper \(R_{n+1}\text{Ni}_n\text{O}_{3n+1}\), up to \(n = 5\) (\(R = La [130]\) and \(R = Nd [131]\)).

6.2. \(d^9\) nickelates without self-doping

One of the major differences between the nickelates and cuprates is the presence of self-doping in the former. If the self-doping is eliminated, the Fermi surface topology of the nickelates will become more similar to the cuprates.

Reference [77] systematically proposed layered nickelates by changing the composition of the layers between the NiO\(_2\) layers (‘block layer’ [132], in the case of RNiO\(_2\), the rare-earth layer corresponds to the block layer). Reference [133] also introduced a similar idea: indeed, one of the materials studied in reference [133] is also investigated in reference [77]. In order to avoid charge transfer between the block and NiO\(_2\) layers, the material design was carried out using elements in the 1–3 groups that prefer closed shells. Furthermore, to keep a large crystal field splitting in the nickel 3\(d\) manifold, block layers without apical oxygens were considered. The structural stability of the designed nickelates was investigated from first-principles [77].

The designed nickelates tend to suppress the self-doping. Some of them are completely free from the self-doping, and only the nickel 3\(d_{z^2−r^2}\) band crosses the Fermi level in the DFT band structure [77]. The crystal and band structures of two such materials are shown in figure 21. In contrast with RNiO\(_2\), these nickelates may satisfy all the keywords for cuprate analogs listed in section 1, ‘two-dimensional,’ ‘square lattice,’ ‘single orbital,’ and ‘near half-filling’.

Since the strongly-correlated nickel 3\(d_{z^2−r^2}\) orbital becomes half-filling, the proposed self-doping-free \(d^9\) nickelates are expected to become a Mott insulator, unlike the infinite-layer nickelates, which could not become a Mott insulator due to the self-doping [114]. The effective model for a Mott insulator is the spin-1/2 Heisenberg model with magnetic exchange interaction \(J\). A first-principles estimate of the \(J\) value of the \(d^9\) nickelates gives \(J = 80–100\) meV [114], which is not far smaller than that of the cuprates (~130 meV [111]). The new nickelates showing a large
Figure 18. (a) Temperature evolution of the $d_{3z^2-r^2}$-wave pairing instability at 20% hole doping for effective models of LaNiO$_2$. $\lambda$ is the eigenvalue of the Eliashberg equation. Three different models for LaNiO$_2$ are compared with the five orbital model of HgBa$_2$CuO$_4$ (see reference [67] for details). Reference [67] argues that the superconducting instability in the nickelates is weaker than that of the cuprates because of a larger interaction and the resulting self-energy renormalization effect. The insets show $\lambda$–$T$ log–log plot (bottom left) and the eigenfunction of the Eliashberg equation at $k_z = 0$ (top right). Reproduced from [67]. CC BY 4.0. (b) The superconducting gap of $d_{3z^2-r^2}$-wave pairing at 20% hole doping in the $t$-$J$ model. Reprinted figure with permission from [68], Copyright (2020) by the American Physical Society.

Figure 19. The superconducting transition temperature $T_c$ of the $d$-wave pairing as a function of Sr-doping. The calculation is performed for a single-orbital Hubbard model using the dynamical vertex approximation (DΓA). In the blue-shaded region, a single-orbital Hubbard model description is argued to be possible. Reproduced from [90]. CC BY 4.0.

Figure 20. The crystal structure (left) and the DFT band dispersion (right) of a trilayer nickelate $R_4Ni_3O_8$. In the DFT calculation, $R =$ La is employed. The bands with dominant nickel $3d_{x^2-y^2}$-orbital character are highlighted in red. Reprinted figure with permission from [126], Copyright (2020) by the American Physical Society.

magnetic exchange coupling $J$ would provide an interesting playground to search for new superconductors close to the Mott–Hubbard regime in Zaanen–Sawatzky–Allen diagram [114].

In addition, the $d^8$ nickelates may provide a rare example of realizing a two-dimensional square-lattice Hubbard model, if the holes are doped into the $3d_{x^2-y^2}$ orbital [114]. Since the phase diagram of the doped Hubbard model on the square lattice is being reinvestigated thanks to a recent rapid advance in numerical techniques [134–136], the $d^8$ nickelates are of great interest as 'testbed' materials [114].

We note a remarkable experimental advance also in synthesizing $d^8$ nickelates as well as multi-layer square-planar nickelates. Independently from the theoretical proposal [77], an experimental work [137] published around the same time has reported a successful synthesis of T'-type structure La$_2$NiO$_3$F. The same material has been investigated in reference [77] and is predicted to be dynamically stable; thus, the experiment and theoretical prediction show a nice agreement. There is also an experimental report of the synthesis of a $d^9$ nickelate Sr$_2$NiO$_2$Cl$_2$ [138]. This is of great interest because Sr$_2$NiO$_2$Cl$_2$ has the same T-type structure as a theoretically proposed $d^9$ nickelate $A_2$NiO$_2$Br$_2$ ($A$ is a 2.5+-valent cation) in figure 21(c), though the nickel valence is different (for the discussion of $d^9$ nickelates, see section 6.3.1).

Another route for realizing $d^9$ nickelates would be nickel fluorides instead of nickel oxides [139]. Reference [139] investigated infinite-layer fluoro-nickelates ANiF$_2$ with
A = Li, Na, K, Rb, and Cs. Because fluorine is the most electronegative element, the energy difference between the nickel 3d and fluorine 2p orbitals becomes large. This places the fluoro-nickelates well inside the Mott–Hubbard regime and makes the nickel 3d_2−2^2 bandwidth smaller (more strongly correlated).

There is also a proposal for nickel chalcogenides [140]. Reference [140] argues that, compared to nickelates, nickel fluorono nickelates well inside the Mott–Hubbard regime and makes the nickel 3d_2−2^2 bandwidth smaller (more strongly correlated).

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6.3. Other variants

6.3.1. d^9 nickelates. The superconductivity in the infinite-layer nickelates emerges close to nickel d^9 filling, and thus the relation with the cuprates has been discussed intensively. Recently, there was a report on the superconductivity in nominally heavily-overdoped compound Ba_3CuO_3+d [141]. Then, it would be interesting to expand the space for materials search. References [57, 142] have investigated the layered nickelates around d^9 filling and discussed possible superconductivity.

6.3.2. Palladium and silver compounds. As is discussed in section 4, the bandwidth of nickel 3d_2−2^2 orbital (or more precisely, anti-bonding orbital between nickel 3d_2−2^2 orbital and oxygen 2p orbitals) is smaller compared to that of the cuprates. This makes the kinetic energy scale for the superconductivity smaller. This gets a larger kinetic energy scale, palladium oxides (palladates), in which palladium 4d orbitals are more extended in space than nickel 3d orbitals, might be promising. Layered palladates around d^9 filling have been theoretically investigated in references [77, 143].

Silver-based compounds may also be interesting. A difficulty in using silver is that silver cation strongly favors 1+ valence. Using elements more electronegative than oxygen, Ag^{2+} can be realized. Reference [144] studied the property of AgF_2 and shown that AgF_2 can be a cuprate-analog d^9 material with a large magnetic exchange coupling, reaching about 70% of that of cuprates.

7. Summary and outlook

The discovery of superconductivity in the doped infinite-layer nickelate Nd_{0.8}Sr_{0.2}NiO_2 was reported in August 2019. As a review at the initial stage of the research, we have reviewed the basics of the bulk electronic state.

In section 4, we have discussed that the infinite-layer nickelate RNiO_2 is similar to the cuprates in that the nickel 3d_2−2^2 orbital forms a strongly correlated system on the two-dimensional square lattice near the half-filling. However, a crucial difference from the cuprates is the presence of the self-doping: rare-earth-layer orbitals form additional Fermi pockets. In this sense, the parent compounds of the infinite-layer nickelates could be viewed as a material that fails to be a Mott insulator because of the self-doping. The superconductivity is most likely unconventional, and the correlation effect will play an important role.

Then, a question is what kind of correlation physics is essential (section 5). If the itinerant rare-earth-layer orbitals are active, they would give rise to Kondo-like physics. If the holes are doped into oxygen 2p orbitals, the physics of Zhang–Rice singlet may emerge (Mott–Hubbard vs charge-transfer). When the holes reside within 3d manifold (Mott–Hubbard type), there should be a competition between the crystal-field splitting and Hund’s coupling (low-spin vs high-spin). If the high-spin state is favored, Hund’s physics comes into play. On the other hand, if the low-spin state is favored, Mott physics possibly masked by the self-doping would be important. We need more comprehensive
experimental and theoretical investigations to clarify these points. As for a future perspective, as one of the most important tasks, bulk superconducting samples are highly desired. If the superconductivity can only be realized in thin films, a difference between thin film and bulk samples needs to be discussed more seriously.

Another crucial question to be elucidated is the possibility of increasing $T_c$ (currently, maximum $T_c$ is about 15 K) in nickel-based superconductors. In this respect, the relation between superconductivity and other possible long-range-order instability needs to be clarified in the nickelates. In particular, the relation between superconductivity and magnetism would be one of the most urgent tasks to be investigated, considering the fact that the cuprates have an extraordinarily large magnetic exchange coupling of more than 100 meV. Although experiments detect a signature of magnetic fluctuations, so far, there is no clear evidence for long-range magnetic order in the infinite-layer parent compound $RNiO_2$ (see section 3). The presence of the self-doping might be the origin of the absence of long-range order (note that the long-range order is quickly suppressed in the hole-doping side of the cuprate phase diagram [145]). In this respect, $d^9$ nickelates without self-doping (see section 6), if realized, would be an ideal material to investigate magnetism in the nickelates and compare it with that of the cuprates. An expected Mott insulating behavior eliminates the ambiguity in the mapping to spin model, and the magnetic exchange coupling is well defined.

To conclude, for sure, one needs more comprehensive experimental and theoretical efforts to get a complete picture of nickelate superconductivity. The nickelates nicely expand the playground for exploring unconventional superconductivity around the cuprate high-$T_c$ superconductivity and provide excellent references to be compared with other unconventional superconductors. The establishment and understanding of superconductivity in nickelates will shed new light on the unresolved challenge of elucidating the superconducting mechanism in the correlated materials.

**Note added**

After submitting the manuscript, we became aware of intriguing experimental progress. Recently, superconductivity has been discovered in film samples of a quintuple-layer nickelate $Nd_5Ni_3O_{12}$ [146]. This is the first report of nickelate superconductivity other than in infinite-layer compounds (see section 6.1). Also, charge-order and charge-density-wave instabilities in infinite-layer nickelates are discussed in recent works [147–149]. Interestingly, charge stripes, which couple to the spin degrees of freedom, had been found in a tri-layer nickelate $La_4Ni_3O_8$ [150, 151]. Elucidating the systematic phase diagram of multi-layer and infinite-layer nickelates is a major challenge for the future.

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**Data availability statement**

All data that support the findings of this study are included within the article (and any supplementary files).

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