Designing nickelate superconductors with d^8 configuration exploiting mixed-anion strategyNaoya Kitamine , Masayuki Ochi , and Kazuhiko Kuroki

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Inspired by a recently proposed superconducting mechanism for a new cuprate superconductor $\text{Ba}_2\text{CuO}_{3+\delta}$, we theoretically design an unconventional nickelate superconductor with a d^8 electron configuration. Our strategy is to enlarge the on-site energy difference between $3d_{x^2-y^2}$ and other $3d$ orbitals by adopting halogens or hydrogen as out-of-plane anions, so that the $3d$ bands other than $d_{x^2-y^2}$ lie just below the Fermi level for the d^8 configuration, acting as incipient bands that enhance superconductivity. We also discuss a possible relevance of the present proposal to the recently discovered superconductor $(\text{Nd}, \text{Sr})\text{NiO}_2$.

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The two families of high- T_c superconductors, cuprates [1] and iron-based [2], are often contrasted as single-orbital versus multi-orbital systems. Namely, in the cuprates, only the $d_{x^2-y^2}$ orbital plays the main role while in the iron-based superconductors, d_{xy} , d_{xz} , and d_{yz} orbitals largely contribute to the electronic structure around the Fermi level (E_F). Recently, however, there has occurred a possible paradigm shift to this picture, owing to some new experimental findings. One such experiment is the discovery of high- T_c superconductivity in $\text{Ba}_2\text{CuO}_{3+\delta}$ [3], where the apical oxygen height is so low that the energy levels of $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ can be reversed compared to conventional cuprates. Another example is a highly overdoped CuO_2 monolayer grown on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ [4], where E_F is significantly lowered so that it reaches the $d_{3z^2-r^2}$ band [5].

For $\text{Ba}_2\text{CuO}_{3+\delta}$, various theoretical studies have been performed [6–12], many of which treat multi-orbital models. In Ref. [12], two of the present authors have introduced a two-orbital model consisting of Wannier orbitals with $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ symmetries, where s -wave superconductivity with a reversed gap sign between the two bands ($s\pm$ -wave pairing) is found to be strongly enhanced near half filling when the energy level offset ΔE between $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals is enlarged so that the bottom of the $d_{3z^2-r^2}$ band is positioned just above E_F . The strong enhancement of superconductivity was explained by transforming the two-orbital model to the bilayer Hubbard model, where the two orbitals of the former model correspond to the bonding and antibonding orbitals of the latter, and hence the orbital level offset ΔE in the former transforms to $2t_\perp$ in the latter, with $t_\perp$ being the vertical hopping between the layers in the bilayer model, as schematically depicted in Fig. 1. A more detailed explanation of this transformation is given in the Supplemental Material [13].

The bilayer Hubbard model has been intensively studied in the past [15–31], and $s\pm$ -wave superconductivity is found to be strongly enhanced near half filling when $t_\perp$ is several times larger than the in-plane hopping and E_F lies in the vicinity of one of the bands [15,20,25–30]. Nowadays, a band sitting just below (or above) E_F is often referred to as an incipient band, and has attracted interest in the study of iron-based superconductors [32–39], bilayer and ladder-type lattices [28–31,40–44], and flat-band superconductivity [45–48]. The two-orbital to bilayer transformation is mathematically exact when there is no hybridization between the two orbitals and also $U = U' = J = J'$ is satisfied, where U , U' , J , and J' are the intra-orbital repulsion, interorbital repulsion, Hund's coupling, and the pair hopping interaction, respectively [12,49]. In reality, $U > U' > J, J'$ and the interorbital hybridization is present, but the analogy between the two models turns out to be approximately valid even in the realistic situation [12].

Actually, a situation where E_F lies close to the $d_{3z^2-r^2}$ band edge is realized in a cuprate $(\text{La}, \text{Sr})_2\text{CuO}_4$. One of the present authors and his colleagues have pointed out that the presence of the $d_{3z^2-r^2}$ band around E_F is the origin of the suppression of T_c of the d -wave superconductivity in this material [50–53] due to the orbital component mixture around the antinodal regime. The difference between $(\text{La}, \text{Sr})_2\text{CuO}_4$ and $\text{Ba}_2\text{CuO}_{3+\delta}$ in Ref. [12] is that in the former, the e_g bands are close to 3/4 filling on average (nearly two electrons in $d_{3z^2-r^2}$ and one electron in $d_{x^2-y^2}$), while the latter is closer to half filling.

The above consideration brings us to an idea of realizing incipient-band-enhanced superconductivity in *nickelates*, where the e_g bands become half filled on average for the natural Ni^{2+} valence, namely, the d^8 electron configuration. Actually, if all the bands other than $d_{x^2-y^2}$ sink below E_F for the d^8 configuration, a pair of $d_{x^2-y^2}$ and any other d orbital is half filled (two electrons per two orbital on average), so if a small amount of electrons is doped into such systems, we may expect enhanced $s\pm$ -wave superconductivity. To enlarge the energy level offset between $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbitals, we adopt a mixed-anion strategy [54], namely, we chose halogens or hydrogens instead of oxygens as out-of-plane anions

FIG. 1. A schematic image of the equivalence between the two-orbital model and the bilayer model.

(which corresponds to enhancing $t_{\perp}$ in the bilayer system), for which we end up with the composition of $A_2\text{NiO}_2X_2$ ($A = \text{Ca}, \text{Sr}; X = \text{H}, \text{F}, \text{Cl}, \text{Br}, \text{I}$) [55].

We note that similar materials have been proposed in Ref. [56] in the context of a recently discovered superconductor $(\text{Nd}, \text{Sr})\text{NiO}_2$ [57], but there, the aim was to construct ideal single-band systems with a d^9 electron configuration, where bands other than $\text{Ni } d_{x^2-y^2}$ do not intersect E_F for the mother compound. In fact, here we later conversely discuss a possible relevance of the present d^8 proposal to the superconductivity of $(\text{Nd}, \text{Sr})\text{NiO}_2$.

We consider $T(\text{K}_2\text{NiO}_4)$ and T' (Fig. 2) structures as candidates for possible crystal structures [58]. We also take La_2NiO_4 (T structure) as a reference with a d^8 configuration. We perform a structural optimization adopting the Perdew-Burke-Ernzerhof generalized gradient approximation (PBE-GGA) exchange-correlation functional [59] and the projector augmented-wave method [60]. We use the Vienna *ab initio* simulation package (VASP) [61–64]. A $12 \times 12 \times 12$ k mesh and a plane-wave cutoff energy of 550 eV were used. After the structural optimization, we perform a first-principles band-structure calculation using the WIEN2K code [65]. We adopt $RK_{\text{max}} = 7$ (6 for oxyhydrides), and take a $12 \times 12 \times 12$ k mesh in the self-consistent-field calculations. From the calculated band structures, we extract the Wannier functions [66,67] of five $\text{Ni } 3d$ orbitals using the WIEN2WANNIER [68] and WANNIER90 [69] codes. Throughout the study, the spin-orbit coupling is neglected.

In Fig. 2(b), we plot the total energy difference between the two structures for all choices of A and X elements. It is found that for $X = \text{Cl}, \text{Br}$, and I , the T structure has

FIG. 2. (a) T and T' crystal structures. (b) The total energy difference per formula unit between T and T' structures for various choices of elements.

FIG. 3. The band structures of (a) La_2NiO_4 , (b) $\text{Ca}_2\text{NiO}_2\text{Cl}_2(T)$, and (c) $\text{Ca}_2\text{NiO}_2\text{H}_2(T')$. The band dispersion of the five-orbital model is superposed to the first-principles bands.

lower energy ($E_T - E_{T'} < 0$), while the difference between the two is small for $X = \text{H}, \text{F}$. In fact, the phonon calculation of $\text{Ba}_{0.5}\text{La}_{0.5}\text{NiO}_2\text{F}_2$ in Ref. [56] shows the appearance of imaginary modes for the T structure, but not for T' , which suggests stability of the latter. The optimized lattice constants presented in the Supplemental Material exhibit a trend where the in-plane lattice constant a becomes larger as the ion radius of element X or A is increased [13].

The first-principles band structures obtained for the optimized lattice structures of La_2NiO_4 (T), $\text{Ca}_2\text{NiO}_2\text{Cl}_2$ (T), and $\text{Ca}_2\text{NiO}_2\text{H}_2$ (T') are shown in Fig. 3. The energy dispersion of the five-orbital model is superposed to the first-principles bands for each material. The band structures of materials with other elements are presented in the Supplemental Material [13]. It can be noticed that in the mixed-anion materials, the $d_{3z^2-r^2}$ band totally sinks below E_F despite the d^8 electron configuration with the $d_{x^2-y^2}$ band partially filled, in sharp contrast to the case of La_2NiO_4 . The entire d_{xy} band also lies below E_F , while the top of the $d_{xz/yz}$ bands intersects E_F .

In Fig. 4, we plot the on-site energy (ΔE) of the d orbitals with respect to that of $d_{x^2-y^2}$ in the five-orbital model. Let us focus on the crystal structure having the lower total energy

FIG. 4. The on-site energy of the d orbitals other than the $d_{x^2-y^2}$ orbital, measured from that of $d_{x^2-y^2}$. (a) $A = \text{Ca}; T$, (b) $\text{Ca}; T'$, (c) $\text{Sr}; T$, and (d) $\text{Sr}; T'$. Here, the solid (dashed) symbols denote the lattice structure with lower (higher) total energy. As a reference, the values of ΔE for La_2NiO_4 are plotted in (a).

FIG. 5. Eigenvalue of the Eliashberg equation λ at $T = 0.01$ eV against the band filling for (a) $A = \text{Ca}$ and (b) Sr . In (a), also the results for La_2NiO_4 are plotted as a reference. (c) Gap function of $\text{Ca}_2\text{NiO}_2\text{Cl}_2$ for $n = 4.125$ in the orbital representation. (d) Similar plot of λ obtained for various one-, three-, and four-orbital models (see text) of $\text{Ca}_2\text{NiO}_2\text{Cl}_2$. The horizontal axis is the band filling (for the single-orbital to the five-orbital model from top to bottom).

(denoted by the solid symbols). $|\Delta E|$ tends to be larger (i.e., the energy level is lowered since $\Delta E < 0$) as the ion radius of X is reduced within $X = \text{Cl}, \text{Br}$, and I . This is due to the smaller in-plane lattice constant (shorter Ni-O distance), which pushes up the $d_{x^2-y^2}$ energy level. For the T' structure, the on-site energy of the $d_{3z^2-r^2}$ orbital is strongly reduced due to the absence of the apical anions, but the t_{2g} orbitals are pushed up compared to those in the T structure. If we compare $A = \text{Sr}$ and Ca , $|\Delta E|$ of the t_{2g} orbitals are reduced in the former compared to the latter, while that of $d_{3z^2-r^2}$ is less affected. This is because the lattice constants a and c are shorter for $A = \text{Ca}$ than for Sr .

We now analyze superconductivity based on the obtained five-orbital models. We assume on-site intra- and interorbital interactions, U , U' , J , and J' , and the many-body study of this model is performed within the fluctuation exchange approximation (FLEX) [70]. We mainly adopt $U = 4$ eV, $J = J' = U/8$, $U' = U - 2J$, but also discuss the interaction dependence in the Supplemental Material [13]. A relatively large U is taken in accord with a study for LaNiO_2 [71]. We obtain the renormalized Green's function by solving the Dyson's equation in a self-consistent calculation. The obtained Green's function and the pairing interaction mediated mainly by spin fluctuations are plugged into the linearized Eliashberg equation. Since the eigenvalue λ of the equation reaches unity at $T = T_c$, here we adopt λ , obtained at a fixed temperature of $T = 0.01$ eV, to measure how close the system is to superconductivity. The eigenfunction of the Eliashberg equation will be called the gap function. In the FLEX calculation, a $16 \times 16 \times 4$ (k_x, k_y, k_z) mesh and 2048 Matsubara frequencies were taken.

In Figs. 5(a) and 5(b), we plot the eigenvalue of the Eliashberg equation against the band filling, varied assuming a rigid band, for various materials with the crystal structure (T or T') having a lower total energy. In all mixed-anion compounds, λ is peaked at around $n = 4.1$ – 4.2 , which corresponds to 10%–20% electron doping starting from the stoichiometric band filling of $n = 4$. The compounds having the largest maximum λ are $\text{Ca}_2\text{NiO}_2\text{Cl}_2$ and $\text{Ca}_2\text{NiO}_2\text{H}_2$. These maximum values of λ are much larger compared to that of La_2NiO_4 [72], and they are in fact even larger than that of $\text{HgBa}_2\text{CuO}_4$, a $T_c = 100$ K superconductor, obtained in the same manner [71]. It is also worth mentioning that the Stoner factor for magnetic ordering remains far from unity for the situation when λ of superconductivity is optimized, which is typical for the incipient-band-enhanced superconductivity (see Supplemental Material [13]).

The gap function presented in Fig. 5(c) for the case of $\text{Ca}_2\text{NiO}_2\text{Cl}_2$ with $n = 4.125$ shows that the four bands other than $d_{x^2-y^2}$ have the same sign of the gap, opposite to that of the $d_{x^2-y^2}$ band (see the Supplemental Material for the contour plot of the gap [13]). In this sense, this is $s\pm$ -wave pairing with the four bands other than $d_{x^2-y^2}$ being incipient, namely, located below E_F , with the $d_{xz/yz}$ band top being the closest to E_F . In order to see the role played by each orbital in more detail, we extract one, three, or four orbitals from the five-orbital model, and apply FLEX to those models to obtain λ in a similar manner, as depicted in Fig. 5(d). The comparison among the models is made at the same electron doping rates, since the Fermi surfaces are the same among the multiorbital models that contain the $d_{xz/yz}$ orbitals. The $d_{x^2-y^2}$ Fermi surfaces of the multiorbital models are also (nearly) the same as that of the single-band model in the relevant band-filling regime, as explained in more detail in the Supplemental Material [13].

From Fig. 5(d), it can be seen that λ in the $d_{x^2-y^2} + d_{xz/yz}$ model is strongly enhanced compared to the single-orbital model consisting only of $d_{x^2-y^2}$, indicating the important role played by the incipient $d_{xz/yz}$ bands. Adding d_{xy} to the $d_{x^2-y^2} + d_{xz/yz}$ model somewhat enhances λ , but not drastically. Our interpretation for this is that the interaction between d_{xy} and $d_{x^2-y^2}$ orbitals enhances superconductivity because the signs of the gap are opposite, but the interaction between d_{xy} and $d_{xz/yz}$ gives a negative contribution to superconductivity because of the same sign of the gap. On the other hand, adding the $d_{3z^2-r^2}$ orbital to the $d_{x^2-y^2} + d_{xz/yz}$ model even slightly suppresses λ . An argument similar to d_{xy} should apply also to $d_{3z^2-r^2}$, but, in addition, we find that the hybridization between $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ (note that the $d_{x^2-y^2}$ orbital is hybridized only with $d_{3z^2-r^2}$) gives a negative contribution to superconductivity, similarly to what has already been known for the cuprates [50–53]. It is also interesting to note that the $d_{3z^2-r^2}$ gap function has the largest value among the incipient bands, despite the negative contribution of $d_{3z^2-r^2}$ to λ , and also despite the $d_{3z^2-r^2}$ band lying somewhat away from E_F compared to the top of the $d_{xz/yz}$ bands. We believe that this is a consequence of small $|\Delta E|$ for the $d_{3z^2-r^2}$ orbital in $\text{Ca}_2\text{NiO}_2\text{Cl}_2$, which implies that this band lies closest to E_F among the incipient bands as far as the center of gravity of the band is concerned. More detailed analyses on these issues are given in the Supplemental Material [13].

Since λ is dominated by the position of the $d_{xz/yz}$ bands, the materials having a larger energy difference between the $d_{x^2-y^2}$ and $d_{xz/yz}$ orbitals, corresponding to larger $t_{\perp}$ in the bilayer Hubbard model [12], tend to exhibit larger λ . On the other hand, it should be stressed that the strong reduction of λ for La_2NiO_4 compared to mixed-anion materials mainly comes from the small $|\Delta E|$ for the $d_{3z^2-r^2}$ orbital; the $d_{3z^2-r^2}$ band intersecting E_F gives rise to the strong development of low-energy spin fluctuations, which works destructively against superconductivity, as in the case of the bilayer Hubbard model with small $t_{\perp}$ [26,27,29,30]. A further analysis on the role played by each orbital is given in the Supplemental Material [13].

We note that all of our calculations assume a low-spin state, while nickelates with a d^8 configuration can take high-spin states when the d -level splittings are small. In fact, the reference system in our study La_2NiO_4 is actually well known to be in the high-spin state, and also $\text{Sr}_2\text{NiO}_2\text{Cl}_2$, synthesized in Ref. [73], was found to be in a high-spin state. On the other hand, a hypothetical infinite layer nickelate SrNiO_2 , without apical anions, was theoretically shown to have a low-spin ground state [74]. Hence, large $|\Delta E|$ is not only preferable for enhancing T_c , but it is also favorable from the viewpoint of realizing a low-spin ground state. In this sense, $A = \text{Ca}$ rather than Sr and also $X = \text{Cl}$ rather than Br or I (T structure) is likely to be the best choice from the viewpoint of both realizing a low-spin state and enhancing T_c because of the smaller in-plane lattice constant and hence larger $|\Delta E|$. Our result indicates that superconductivity in $\text{A}_2\text{NiO}_2\text{X}_2$ is optimized by electron doping, which may be realized in actual materials by, e.g., partially substituting Ca^{2+} (or Sr^{2+}) with La^{3+} . In fact, a pure d^8 configuration (band filling of $n = 4$) may result in an insulating state not predicted within the present calculation. Electron doping should prevent the material from being an insulator, and hence may also stabilize the preferred low-spin state in actual materials. In total, the materials we propose here as candidates for new unconventional superconductors have the composition forms $\text{Ca}_{2-x}\text{La}_x\text{NiO}_2\text{Cl}_2$ and $\text{Ca}_{2-x}\text{La}_x\text{NiO}_2\text{H}_2$.

Now, if we increase the amount of electron doping in the present materials, they approach the materials with a d^9 electron configuration studied in Refs. [56,75] in the context of a recently discovered superconductor $(\text{Nd}, \text{Sr})\text{NiO}_2$ [57]. Then, conversely, it is interesting to see how the d^9 and d^8 states are connected in $(\text{Nd}, \text{Sr})\text{NiO}_2$ by hypothetically removing the electrons. Here, we consider LaNiO_2 adopting the experimentally determined lattice parameters of NdNiO_2 to avoid the ambiguity regarding the treatment of the f electrons in Nd [71]. Since a previous study has shown that the $\text{La } 5d$ orbitals have a small effect on spin-fluctuation-mediated superconductivity [71], we construct a five $\text{Ni } 3d$ -orbital model similar to those for $\text{A}_2\text{NiO}_2\text{X}_2$, as shown in Fig. 6(a). The band structure somewhat resembles that of $\text{A}_2\text{NiO}_2\text{Cl}_2$ in that the position of the $d_{3z^2-r^2}$ band is lowered, which is because the apical anions are absent in this infinite layer material, but the difference lies in that the $d_{3z^2-r^2}$ band exhibits strong three dimensionality. We perform a similar FLEX analysis of superconductivity for this model by hypothetically varying the band filling assuming a rigid band. As expected from our results on $\text{A}_2\text{NiO}_2\text{X}_2$, there appears a peak in λ close to $n = 4$, i.e., the d^8 configuration,

FIG. 6. (a) The first-principles band structure of LaNiO_2 with the five-orbital model superposed. (b) λ against the band filling for the model of LaNiO_2 . $\leftrightarrow$ shows the band-filling regime corresponding to the Sr content where superconductivity is observed in Refs. [82,83] assuming no residual hydrogens are present.

in addition to the already known d -wave superconductivity around $n = 4.5$, the d^9 configuration.

Although we believe that the mechanism similar to the cuprates, where only the $d_{x^2-y^2}$ band plays an important role, is a strong candidate for the superconducting mechanism of $(\text{Nd}, \text{Sr})\text{NiO}_2$ [71,76–78], the present analysis suggests that their might be an alternative scenario where $\text{Ni } 3d$ orbitals other than $d_{x^2-y^2}$ play the role of incipient bands. In fact, as studied in Ref. [79], $(\text{Nd}, \text{Sr})\text{NiO}_2\text{H}$ may form during the reduction process by CaH_2 , which would reduce the number of d electrons [80]. Another theoretical study shows that a similar situation can occur also at the SrTiO_3 substrate interface [81]. In Fig. 6(b), we indicate the band-filling regime corresponding to the Sr content where superconductivity is observed in Refs. [82,83] assuming no residual hydrogens are present. This region will be shifted by $p/2$ toward the smaller band-filling regime if residual hydrogens are present in the form $(\text{Nd}, \text{Sr})\text{NiO}_2\text{H}_p$. It will then be an interesting future problem to carefully examine the Ni valence in $(\text{Nd}, \text{Sr})\text{NiO}_2$. Also, the gap function would be of interest since the d wave should be realized for the d^9 pairing mechanism, whereas the s wave is expected for the d^8 scenario. In fact, recently, a tunneling spectroscopy experiment has been reported [84], where s -wave and d -wave-like gaps have been observed, depending on the position on the sample. The experiment has been interpreted in terms of the gap with a different symmetry opening on different Fermi surfaces, which was suggested theoretically [85]. From our viewpoint, the position dependence of the pairing symmetry might originate from the inhomogeneity of the residual hydrogen remained during the reduction process [86].

To conclude, we have designed unconventional nickelate superconductors with a nearly d^8 electron configuration, where superconductivity is enhanced by the bands other than $d_{x^2-y^2}$ playing a role of the incipient bands. The key idea is to exploit the mixed-anion strategy to enlarge the on-site energy difference between $d_{x^2-y^2}$ and other d orbitals. This corresponds to increasing the interlayer hopping $t_{\perp}$ of the bilayer Hubbard model, which is known to exhibit $s\pm$ -wave superconductivity strongly enhanced by the incipient-band effect for large $t_{\perp}$. The possible relevance of the present proposal to the observation of superconductivity in $(\text{Nd}, \text{Sr})\text{NiO}_2$ is an interesting future problem, where a non-rigid-band variation of the incipient-band dispersion due to residual hydrogens [79] should be taken into account.

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