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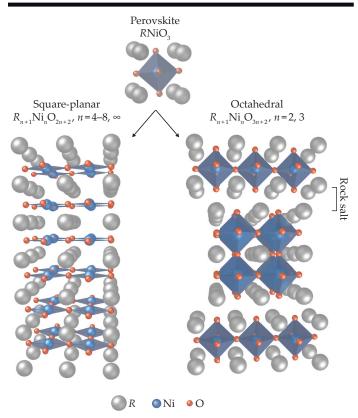


# Nickelates provide answers about high-temperature superconductivity—and raise new questions

Berit H. Goodge and Michael R. Norman

Shortly after researchers synthesized a family of superconducting nickelates in 2019, surprising discoveries were found in related yet distinct nickel compounds.

he search for new superconductors—materials that expel magnetic fields and perfectly transmit electrical current below a critical temperature—has occupied countless physicists, chemists, materials scientists, and engineers for more than a century. So when a group at Stanford University discovered in 2019 that nickel oxides could superconduct,<sup>1</sup> a burst of research ensued to reproduce, improve, and understand their fundamental behavior and their possible technological applications.<sup>2</sup> (For more on the discovery, see Physics Today, November 2019, page 19.)



**FIGURE 1. TWO NICKEL OXIDE MATERIALS** with different atomic crystal structures are capable of superconductivity. The square-planar structure and the octahedral structure are both derived by modifying the cubic perovskite  $R \text{NiO}_3$ , where R is a rare-earth ion. When the number n of nickelate planes approaches infinity, the result is a square-planar  $R \text{NiO}_2$  structure. The octahedral nickelates are formed when blocks of perovskite are stacked and offset. The listed values of n indicate the chemical formulas of both materials that have been shown to exhibit superconductivity.

Although many researchers saw the nickelate superconductivity as an unquestionable breakthrough, in some sense the finding was unsurprising. Nickelate superconductivity had been predicted as early as 1999 because the nickelates are similar to the most widely studied superconducting family in modern condensed-matter physics: the cuprates. In fact, Georg Bednorz and Alex Müller's search for a material that could superconduct at high temperature began with a nickel compound before they found success in 1986 with copper oxide, a discovery for which they received the 1987 Nobel Prize in Physics (see Physics Today, December 1987, page 17).

Cuprate superconductors hold the record for the highest critical temperature  $T_{\rm c}$ —below which superconductivity occurs—under ambient pressure conditions. They are used, for example, to create sensitive magnetometers, powerful electromagnets for particle accelerators, and lossless electrical transmission cables. They are being explored to produce the strong magnetic fields that are needed to contain hot plasma in fusion reactors. From a fundamental perspective, cuprates present a tantalizing puzzle to understand how and why superconductivity emerges in, of all things, ceramics.<sup>3</sup>

The earliest known superconductors were metals. After decades of exploration, researchers built a well-defined theory: Under certain circumstances, electrons in a material experience attractive forces rather than repulsive ones. The attraction causes them to form coherent bound pairs, named Cooper pairs, after Leon Cooper. Cooper's original 1956 paper<sup>4</sup> soon led to a complete theory,<sup>5</sup> known as BCS, developed by John Bardeen, Cooper, and John Schrieffer, who shared the 1972 Nobel Prize for their work (see Physics Today, December 1972, page 73).

In the original BCS theory, an electron attracts positive ions because of its negative charge. But as an electron moves through a material, it takes time for the slower-moving ions to relax, which allows for a second electron to be attracted to the net positive regions left in the wake. The phonons, the collective motions of the positively charged ions in a crystal, provide the "glue" for Cooper pairing.

BCS theory, however, doesn't entirely explain cuprates and other unconventional superconductors, which are derived from magnetic insulators. In such insulators, an electron with an up spin wants to have neighbors with a down spin. The result is an induced attraction between the electrons that can be stronger than the electron–ion interactions described in BCS theory. Physicists are still building a satisfactory fundamental description of unconventional superconductors. A key part of that quest is studying new materials and, hopefully, finding common ground between them.

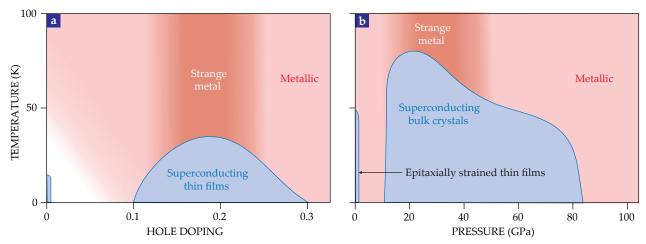
The family of superconducting cuprates is made up of many compounds, each with its own chemical details and crystal structure. Yet a few common traits are found across all of them. In particular, each copper ion has a 3*d*<sup>9</sup> electron configuration: Nine of the 10 electron states of its valence 3*d* shell are occupied. The Cu<sup>2+</sup> ions are coordinated

The nickelates discovered in 2019 share those traits. The nickel ions have a  $3d^9$  electron configuration and are arranged in a square-planar lattice, and each  ${\rm NiO_2}$  plane is separated by rare-earth ions, as shown in figure 1. Because of their similarity to cuprates, nickelates are an enticing experimental platform to test the bounds and validity of our theoretical understanding of unconventional superconductivity.

in a square net of oxygen atoms that bond with the ions.

### Making nickelates into superconductors

The 20-year gap between the prediction and realization of superconducting nickelates was not because of lack of interest but because of the limits of thermodynamics. In the desired  $R\mathrm{NiO}_2$  structure—with R denoting a trivalent (3+) rarearth ion—nickel, with its nine 3d electrons, is monovalent (1+) and thus unstable, so it is impossible to grow crystalline compounds directly. (The rare earths used in experiments so far include lanthanum, praseodymium, neodymium, and samarium.) Nickelates, instead, must be grown first as the cubic perovskite  $R\mathrm{NiO}_3$ . The extra oxygen, which later needs



**FIGURE 2. PHASE TRANSITIONS.** To superconduct, **(a)** square-planar nickelates must have a certain fraction of their trivalent rare-earth ions doped by divalent ions, and **(b)** octahedral nickelates must be at high pressure. In the strange-metal region of phase space, electrical resistivity has a linear temperature dependence—a characteristic seen in cuprate superconductors and also observed in both nickelate families. In many cases, the strange-metal property precedes a superconducting transition. Some groups have reported superconductivity in undoped films (shown on the left in panel a), but the details are still under investigation.

to be chemically extracted to leave behind the desired square-planar form, enables high-quality crystals with nickel to be grown in a more stable  $3d^7$  (3+) configuration.

For superconductivity to emerge in square-planar nickelates, the oxygen-reduced material needs to be doped with extra charge carriers in the nickel band structure. The most common approach is chemical substitution, in which roughly 10–30% of the rare-earth ions are replaced with similarly sized divalent (2+) ions, such as strontium, calcium, and europium, each of which can occupy the same lattice position. To maintain the global charge neutrality of the compound, the nickelion valence adjusts accordingly. The superconducting nickelate material with the highest  $T_{\rm c}$  contains nickel with a configuration near  $3d^{8.8}$  (1.2+), with about 20% doping.

A similar effect can be achieved through structural doping, in which atomically precise engineering is used to insert extra rare-earth planes ( $R_{n+1}\text{Ni}_n\text{O}_{2n+2}$ ) into the crystal structure. The process starts with the Ruddlesden–Popper crystal family ( $R_{n+1}\text{Ni}_n\text{O}_{3n+1}$ ), with n stacked perovskite unit cells separated from the next set of layers by rock-salt blocks ( $R_2\text{O}_2$ ). Then, as before, the extra oxygen atoms are chemically extracted to leave n layers of n layers of n layers of n layers of n layers on tune the nickel valence by controlling the density of the extra planes.

In 2023, a surprising discovery was announced: The lanthanum bilayer member ( $\text{La}_3\text{Ni}_2\text{O}_7$ ) of the Ruddlesden–Popper series also superconducts and at a significantly higher temperature than the square-planar nickelate, although only under very high pressure. Soon after, superconductivity was also found with a lower  $T_c$  in the trilayer version ( $\text{La}_4\text{Ni}_3\text{O}_{10}$ ) under high pressure. Rather than a nickel–oxygen square net, Ruddlesden–Popper nickelates are built of nickel–oxygen octahedra in a framework of rare-earth ions, as shown in figure 1.

How the new octahedral nickelate superconductors relate

to their square-planar nickelate cousins remains unclear, as does how they may be related to cuprates or other high- $T_{\rm c}$  superconductors. The reduced square-planar nickelates have a  $3d^9$  configuration similar to cuprates. The octahedral nickelates, however, have a different electron filling, with nickel configurations of  $3d^{7.5}$  (2.5+) for the bilayer materials and  $3d^{7.33}$  (2.67+) for the trilayer materials. Another distinction is that the octahedral nickelates are tuned into a superconducting state not by chemical doping but by mechanical pressure.

The distinctions between square-planar nickelates and octahedral nickelates manifest far beyond their different crystal structures and 3d electron counts. Most obvious are their critical temperatures, plotted in figure 2. Most square-planar compounds have a  $T_c$  around 15–20 K, although tuning the rare-earth chemistry appears to be a promising route to increasing  $T_c$ , with a recent report nearing 40 K. The octahedral bilayer phase, on the other hand, has already reached 90 K. That temperature is an important benchmark for potential technological applications because the material can be cooled with liquid nitrogen instead of liquid helium, which is expensive and nonrenewable. (For more on helium supply issues, see Physics Today, September 2023, page 18.)

A crucial trade-off, however, is that the octahedral nickelates must be squeezed, using diamond-anvil cells, to about 15 GPa—higher than the pressure necessary to form diamond and more than 100 times as much pressure found at the bottom of the Mariana Trench. The square-planar compounds superconduct without any applied pressure. But they must be exceptionally thin films—so far, no more than 10 nm thick—because the only way to make square-planar nickelates is with the two-step process of growth and reduction. The superconducting octahedral compounds can be formed directly as bulk crystals. Each of those requirements—thin films for square-planar nickelates and high pressure for the

octahedral compounds—carries its own limitations for experimental measurements.

Another milestone was achieved in late 2024: Thin-film versions of the octahedral bilayer La<sub>3</sub>Ni<sub>2</sub>O<sub>7</sub> grown on a carefully chosen substrate were shown to exhibit superconductivity under ambient pressure conditions.<sup>10</sup> Rather than a high-pressure diamond-anvil cell, the thin film bonds to the substrate on which it forms—a concept known as epitaxial strain engineering.<sup>11</sup> The squeezing of the atomic lattice is sufficiently similar to putting it under high pressure. The demonstration of superconducting thin films has opened the door to various experiments that couldn't be done with high-pressure diamond-anvil cells and will hopefully lead soon to rapid advances in experimental investigations of octahedral nickelates.

## Finding family ties

At a more fundamental level, the electronic, magnetic, and other characteristics of superconductors should help guide and validate theoretical models. Some parameters, such as the atomic arrangement and the average valence state of a given ion, can be probed directly through experiments. Early spectroscopic studies of square-planar nickelates, for example, showed that although the nickel ions have the same formal  $3d^9$  configuration as superconducting cuprates, the relative positions of the transition metals' 3d energy levels differ between the two because of their different nuclear charges.

The differences lead to distinctions in the electronic structure of square-planar nickelates and cuprates, shown in figure 3. In cuprates, the Coulomb repulsion U—the energy separation between occupied and unoccupied copper 3d states—is larger than the charge-transfer gap  $\Delta$ , which is the energy separation between the 3d states and the oxygen 2p states. Most of the doped holes, therefore, are on the oxygen sites. For the square-planar nickelates,  $\Delta$  is larger because the 3d levels float to higher energy, and as a consequence, most of the spectral weight of the doped holes is on the nickel sites.

In both cases, the oxygen ions exchange electrons with the

transition-metal ions—the latter ions thus experience a strong induced interaction between each other. The interaction is known as superexchange, which was developed from a theory by the Nobel laureate Philip Anderson. Because of the large  $\Delta$  in square-planar nickelates, their superexchange interaction is about half that of the cuprates. Whether that difference is connected to the smaller  $T_{\rm c}$  in nickelates is a matter of debate. In addition, the floating of the 3d levels to a higher energy pushes them closer to the nominally unoccupied rare-earth 5d energy levels. As a result, the 5d states self-dope the square-planar nickelates, which means that, unlike their cuprate counterparts, undoped square-planar nickelates are not magnetic insulators.

# Identifying the pairing symmetry

Neither the nickelates' distinct electronic landscape nor the additional rare-earth 5*d* bands' contribution to superconductivity are fully understood. Oxygen-mediated superexchange in cuprates, for example, has been proposed as a fundamental origin of the cuprates' unconventional *d*-wave pairing symmetry.<sup>3</sup> The superconducting pairing symmetry is reflected in the energy gap that opens when the electrons condense to form Cooper pairs. In a conventional BCS superconductor, the gap is isotropic in momentum space. The pairing symmetry, therefore, is labeled as *s*-wave because it's similar to the spherical symmetry of a hydrogen atom's *s* orbital.

Unconventional superconductors, on the other hand, can have order parameters that vary strongly not only in magnitude as a function of momentum but also in sign—where the sign changes, the energy gap is zero. Pairing symmetries are again classified similarly to hydrogen-like orbitals: *p*-wave and *d*-wave, or combinations thereof, depending on the lattice symmetry.

The pairing symmetry in square-planar nickelates has not yet been definitively identified because most experimental measurements are particularly challenging to implement in thin films. But various indirect measurements can reduce the number of plausible options. To date, several groups have

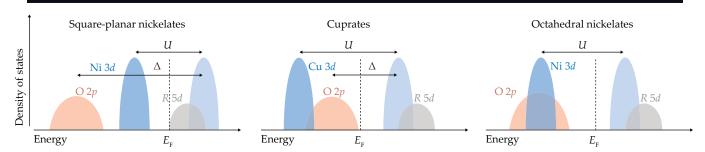


FIGURE 3. THE ELECTRONIC STRUCTURES OF VARIOUS SUPERCONDUCTING MATERIALS are apparent in Zaanen-Sawatzky-Allen diagrams. <sup>18</sup> Each diagram shows the number of possible electron states and their relative energy range for atomic orbitals of interest near the Fermi energy  $E_F$ . The value  $\Delta$  is the charge-transfer energy gap between the oxygen 2p and metal 3d states, and U is the Coulomb repulsion between the 3d electrons, which results in an energy gap between the filled and unfilled d states below and above  $E_F$ . The actual position of  $E_F$  is sensitive to chemical doping, oxygen stoichiometry, and pressure. The significant differences in the electronic structures of the three families of materials may help explain why they each superconduct under different temperature and pressure conditions.

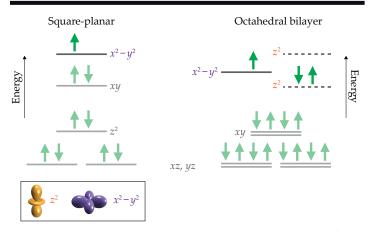


FIGURE 4. THE PARTIALLY FILLED 3D ELECTRONIC STATES contribute to the superconducting behavior in two nickelate materials. The electronic states differ because of each material's particular crystalline structure. Electrons fill the  $z^2$  orbitals in the square-planar material, but they only partially fill the  $z^2$  orbitals in the octahedral bilayer material. The splitting of the  $z^2$  orbitals in the octahedral material arises from the coupling between the two nickelate layers.

reported that square-planar nickelates are, like the cuprates, consistent with d-wave pairing. 14 Other experimental and theoretical groups have put forth alternative hypotheses. Whether the pairing symmetry is similar to or distinct from cuprates and how that emerges from key similarities or differences in superexchange or other characteristics-will help clarify the critical ingredients for high-temperature superconductivity.

The octahedral nickelates are a different beast entirely (see figure 4). Unlike cuprates, in which the active states are the  $3d_{x^2-y^2}$  orbitals, octahedral nickelates have active  $3d_{x^2-y^2}$ and active  $3d_{z^2}$  orbitals, with the latter strongly bonded to apical oxygen atoms-those above and below the nickel ions. The strong bonds lead to paired spins, called singlets, in the  $3d_{x^2}$  states between nickel layers. The  $3d_{x^2}$  singlets don't occur in cuprates and square-planar nickelates because the apical oxygen atoms are missing from the square-planar structure. The first theory proposals for octahedral nickelates focused on the nickel  $3d_{z^2}$  orbitals and suggested that high pressure would enhance their overlap by compressing the octahedral layers.

Early models of the octahedral bilayer material suggested an unconventional  $s_{+-}$  pairing symmetry, in which the order parameter is isotropic but with opposite signs on the two layers. More recent modeling, however, indicates that solutions of  $s_{+-}$  or d-wave pairing are extremely sensitive to a given model's parameters, which themselves depend on subtle differences in the bonding and crystalline environments. 15

Some models have proposed that the  $3d_{z^2}$  orbitals are inert, which would mean that the octahedral nickelates are more like cuprates in that only the  $3d_{x^2-y^2}$  states are relevant for superconductivity.16 Alternatively, multiple superconducting phases—or even the novel possibility of a superconductor with tunable pairing symmetry-could exist in octahedral nickelates. Some of the relevant parameters are measurable for octahedral bilayer thin films that superconduct at ambient pressure, so experiments with those materials should hopefully drive progress.

## Linking the nickelates

The connection between the two nickelate familiesthe square-planar and octahedral materials-remains a fascinating piece of the puzzle of what makes certain materials superconduct. Despite their differences, the two families are linked by atomic structure. Removing oxygen from the trilayer octahedral nickelates that superconduct under pressure, for example, yields a trilayer version of the structurally doped square-planar nickelates, which behave similarly to the nonsuperconducting cuprates.<sup>17</sup> Could the reduced trilayer version also superconduct if it were doped in the other direction or if it were pressurized?

Hopefully, the rich phase space between the two families of nickelates can be studied through several intermediate nickelate compounds. Some promising possibilities include the reduced bilayer structure with a nickel configuration of 3d85; other naturally occurring square-planar

nickelates with 3d8 configurations, similar to some other cuprates; and chemically doped octahedral nickelates.

Both the square-planar and octahedral nickelate families stand as triumphs of collaboration between physicists, chemists, and materials scientists. Continued advances in the materials' synthesis and engineering will improve them further. As high-quality samples become more widely available, the experimental community will hopefully continue to grow and quickly build a foundation of robust knowledge to guide the theory of superconductivity. Similarly, new theoretical insights and frameworks will elucidate key mechanisms and predict promising new routes of experimentation and exploration. Such back-and-forth will accelerate progress across the fields and advance our fundamental understanding of nickelates and, more generally, unconventional superconductivity.

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