Proceedings of CONF-MPCS 2026 Symposium: Theoretic Physics and Plasma Physics
DOI: 10.54254/2753-8818/2026.DL33826

Nickelate Superconductors: A Gateway to High-Temperature Superconductivity

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Abstract. The recent discovery of superconductivity in nickelate materials has generated significant excitement in the condensed matter physics community, representing the first new family of unconventional superconductors that share structural and electronic similarities with cuprate high-temperature superconductors since their discovery over three decades ago. This comprehensive review systematically examines the crystal and electronic structure, synthesis methods, physical properties, and current research status of nickelate superconductors. This paper analyzes the superconducting parameters, including critical temperature and upper critical field values reported across various nickelate systems, with particular emphasis on the infinite-layer nickelates ($RNiO_2$, R=rare earth) and the newly discovered bilayer $La_3Ni_2O_7$ under high pressure. Ultimately, this review concludes that while infinite-layer and pressure-induced bilayer nickelates offer a vital comparative platform for decoding unconventional superconductivity and strong electron correlations, the field is fundamentally bottlenecked by phase metastability and thin-film synthesis constraints. Overcoming these specific material challenges to enable ambient-pressure bulk superconductivity, coupled with systematic comparative studies against cuprates, constitutes the essential pathway for unveiling the universal mechanisms of high-temperature superconductivity.

Keywords: High-temperature superconductors, Nickelate superconductors, Unconventional superconductivity mechanisms

1. Introduction

The pursuit of high-temperature superconductivity has been a central focus of condensed matter physics for decades, with cuprate superconductors remaining the benchmark for elevated transition temperatures since their discovery in 1986 [1]. The recent emergence of nickelate superconductors represents a significant breakthrough, offering a new platform to explore unconventional superconductivity mechanisms and potentially achieve higher transition temperatures [2, 3]. Nickelates share structural similarities with cuprates but possess distinct electronic characteristics that may provide crucial insights into the fundamental principles governing high-temperature superconductivity [4, 5].

The discovery of superconductivity in infinite-layer $Nd_{0.8}Sr_{0.2}NiO_2$ films with Tc ≈ 9-15 K in 2019 marked a watershed moment [6], followed by the more recent discovery of pressure-induced

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DOI: 10.54254/2753-8818/2026.DL33826

superconductivity in $La_3Ni_2O_7$ with Tc up to 80 K [7, 8]. These developments have revitalized the field and prompted extensive theoretical and experimental investigations. This review aims to synthesize the current understanding of nickelate superconductors, highlighting both the progress made and the challenges that remain in realizing their full potential.

2. Crystal and electronic structure

2.1. Crystal structure

Nickelate superconductors primarily crystallize in layered perovskite-derived structures. The infinite-layer nickelates ($RNiO_2$, where R is a rare earth element such as Nd, La, or Pr) feature a simple tetragonal structure (space group P4/mmm) with alternating NiO_2 planes separated by rare-earth ions [2, 9]. This structure is obtained through topotactic reduction of the perovskite precursor $RNiO_3$, which removes oxygen atoms from the apical positions and results in a square-planar coordination of Ni ions [10, 11].

The crystal structure of infinite-layer nickelates is characterized by two key parameters: the in-plane lattice constant a and the out-of-plane constant c. For $NdNiO_2$, the lattice parameters are a = 3.92 Å and c = 3.37 Å [12], while $LaNiO_2$ exhibits slightly larger parameters (a = 3.96 Å, c = 3.37 Å) due to the larger ionic radius of La^{3+} compared to Nd^{3+} [13, 14]. The reduced coordination of Ni ions in the infinite-layer structure creates a two-dimensional electronic environment like the CuO_2 planes in cuprates, but with distinct differences in orbital hybridization and electronic correlation effects.

More recently, bilayer nickelates such as $La_3Ni_2O_7$ have attracted attention due to their significantly higher superconducting transition temperatures under high pressure [7, 8]. These materials feature a more complex crystal structure with bilayers of NiO_2 planes separated by rock-salt LaO layers, creating a different interlayer coupling environment compared to the infinite-layer compounds [15].

2.2. Electronic structure

The electronic structure of nickelates distinguishes them from cuprates and provides crucial insights into their superconducting behavior. In infinite-layer nickelates, the formal Ni^+ oxidation state (d^9 configuration) appears analogous to Cu^{2+} (d^9) in cuprates, but important differences emerge due to the extended nature of Ni 3d orbitals and their hybridization with rare-earth 5d states [16].

First-principles calculations reveal that the electronic structure of $RNiO_2$ compounds consists of a two-dimensional Mott-Hubbard system derived from Ni 3d_{x^2-y^2} orbitals coupled to a three-dimensional metallic band originating from rare-earth 5d states [4, 17]. This multi-band character contrasts with the single-band description often applied to cuprates and introduces additional complexity to the superconducting mechanism.

Angle-resolved photoemission spectroscopy (ARPES) studies on $La_{0.8}Sr_{0.2}NiO_2$ have identified a Fermi surface comprising electron pockets with mixed Ni 3d and Nd 5d character [18], as well as a prominent hole-like band near the Brillouin zone center. The measured band dispersion shows significant mass enhancement compared to density functional theory calculations, indicating substantial electron correlation effects [18].

The role of rare-earth 4f orbitals in the electronic structure remains an active area of investigation. While these orbitals are typically considered localized and inert, evidence suggests they may participate in hybridization with Ni 3d states, potentially influencing superconducting

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pairing [5]. This distinguishes nickelates from cuprates, where the analogous rare-earth ions have fully occupied 4f shells that do not hybridize significantly with Cu 3d orbitals.

3. Materials synthesis

The synthesis of nickelate superconductors presents significant challenges that have limited progress in the field. The primary synthesis route involves a two-step process: first, epitaxial growth of perovskite precursor films ($RNiO_3$) using techniques such as pulsed laser deposition (PLD) or molecular beam epitaxy (MBE) [11], followed by topotactic reduction to achieve the infinite-layer structure.

Pulsed laser deposition has emerged as the most successful technique for growing high-quality nickelate thin films. $SrTiO_3$ (001) substrates are most used due to their suitable lattice matching and stability under reducing conditions.

The reduction process represents the most critical step in achieving superconductivity. The reduction process must be carefully controlled to avoid incomplete reduction or over-reduction, both of which detrimentally affect superconducting properties. Recent advances have explored alternative reduction methods, including atomic hydrogen treatment in ultra-high vacuum, which offers more precise control over the reduction process.

Superconductivity in these materials is then induced through the application of external pressure using diamond anvil cells, typically in the range of 14-40 GPa [8].

A significant challenge in nickelate synthesis is the formation of extended defects, particularly Ruddlesden-Popper-type stacking faults [11], which can disrupt the crystal structure and degrade superconducting properties. Additionally, the infinite-layer structure is metastable and susceptible to degradation upon exposure to air or moisture [10, 11], necessitating careful handling and in situ characterization.

4. Physical properties and current research status

4.1. Superconducting parameters

Nickelate superconductors exhibit a range of superconducting parameters that provide insights into their pairing mechanisms and potential applications. The critical temperature (Tc) varies significantly across different nickelate families. Infinite-layer nickelates typically show Tc values in the range of 9-18 K [6], with $Nd_{0.8}Sr_{0.2}NiO_2$ exhibiting Tc ≈ 15 K [6] and $La_{0.8}Sr_{0.2}NiO_2$ reaching Tc≈ 18.8 K [19]. Remarkably, the bilayer nickelate $La_3Ni_2O_7$ demonstrates substantially higher Tc values under high pressure, with reports of onset temperatures up to 80 K at pressures above 14 GPa [8].

The upper critical field (Hc2) is another important parameter that reflects the strength of superconducting pairing and robustness against magnetic fields. For infinite-layer nickelates, Hc2 values show significant anisotropy between directions parallel and perpendicular to the NiO_2 planes. $La_{0.8}Sr_{0.2}NiO_2$ exhibits Hc2 (0) ≈ 52-55 T for H // ab and Hc2 (0) ≈ 40 T for H // c [14, 19], exceeding the Pauli limit and suggesting non-spin-singlet pairing mechanisms. $La_3Ni_2O_7$ under high pressure shows even more impressive Hc2 values, estimated to exceed 70 T based on extrapolations of resistivity measurements in high magnetic fields.

Other key superconducting parameters include the coherence length, which is typically around 2-3 nm for infinite-layer nickelates, and the London penetration depth, which measurements suggest is on the order of 200-300 nm [14], indicating moderate carrier density. The superconducting energy

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gap, as measured by scanning tunneling spectroscopy, shows values consistent with weak to moderately strong coupling, with 2Δ/kBTc ratios ranging from 3.5 to 4.5 [20].

4.2. Key experimental findings

Recent experimental studies have revealed several remarkable properties of nickelate superconductors. Transport measurements show signature characteristics of unconventional superconductivity, including linear-in-temperature resistivity in the normal state and the anomalous Hall effect [14, 21]. The resistance-temperature curves typically exhibit sharp superconducting transitions with a width $\Delta Tc < 2$ K in optimal samples, indicating high sample quality.

Magnetic characterization confirms the presence of bulk superconductivity through clear diamagnetic shielding signals in magnetization measurements [6, 22]. The Meissner effect is observed, though the shielding fraction varies significantly between samples, likely due to the presence of defects or secondary phases. The magnetic penetration depth shows power-law temperature dependence at low temperatures, consistent with nodal superconductivity.

Spectroscopic techniques have provided crucial insights into the electronic structure and pairing symmetry. ARPES measurements, though challenging due to surface degradation issues, have revealed Fermi surfaces with mixed orbital character and evidence of band renormalization [18]. Scanning tunneling microscopy has identified gap features consistent with d-wave symmetry in some nickelates [20], though the situation appears more complex than in cuprates.

Resonant inelastic X-ray scattering (RIXS) studies have uncovered strong magnetic excitations in nickelates [5, 14], with dispersive features reminiscent of those in cuprates but with important differences in energy scale and momentum dependence. These magnetic modes may play a role in mediating superconducting pairing, similar to proposals in cuprate superconductors.

Nuclear magnetic resonance (NMR) measurements have revealed Knight shifts that decrease below Tc, confirming spin-singlet pairing [9], and spin-lattice relaxation rates showing power-law behavior consistent with unconventional superconductivity. However, the detailed interpretation of NMR data remains challenging due to the multi-band nature of these materials.

5. Challenges and future directions

5.1. Challenges in materials synthesis

The synthesis of high-quality nickelate superconductors remains a significant challenge that limits experimental progress. The metastable nature of the infinite-layer structure makes it susceptible to degradation upon exposure to air or moisture [10, 11], necessitating sophisticated in-situ synthesis and characterization capabilities. Reproducibility issues persist across different research groups, with success rates for obtaining superconducting samples often below 50% despite carefully controlled synthesis parameters [12].

The topotactic reduction process is particularly problematic, as slight variations in temperature, duration, or reducing agent distribution can lead to incomplete reduction or the formation of secondary phases. The interface between the nickelate film and substrate introduces additional complications, including strain effects, interdiffusion, and the possible formation of oxygen-deficient layers that affect electronic properties.

Future improvements in synthesis will require a better understanding of the reduction kinetics and the development of more controlled reduction techniques. Atomic layer deposition with in-situ reduction capabilities may offer better uniformity and control [23]. Alternatively, direct growth of

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infinite-layer nickelates without the need for topotactic reduction would represent a major advance, though this has proven extremely challenging due to the metastability of the phase.

5.2. Core challenges in mechanism studies

Understanding the superconducting mechanism in nickelates represents a fundamental challenge with implications beyond this specific materials class. The multi-band nature of nickelates introduces complexity beyond the single-band Hubbard model often applied to cuprates. The role of rare-earth bands in the superconducting mechanism remains controversial, with theories ranging from passive bystander to active participant in the pairing.

The symmetry of the superconducting order parameter is not yet definitively established, with conflicting evidence from different experiments. Whether nickelates exhibit clean d-wave symmetry like cuprates or more complex order parameter symmetry remains an open question. The relationship between magnetism and superconductivity appears different from cuprates, with no clear evidence of antiferromagnetic order in the parent compounds.

Future mechanism studies will require improved sample quality to obtain clearer experimental signatures. Theoretical advances are needed to handle the multi-orbital nature of these materials and their complex correlation effects. Systematic studies across different rare-earth nickelates may help disentangle material-specific effects from universal behavior [9, 24]. Development of better experimental techniques for probing superconducting properties in high-pressure environments will be crucial for studying bilayer nickelates.

6. Conclusion

Nickelate superconductors represent a significant breakthrough in the field of high-temperature superconductivity, offering a new materials platform with both similarities to and differences from the celebrated cuprates. The past five years have witnessed remarkable progress in synthesizing these challenging materials, characterizing their properties, and developing theoretical frameworks to understand their behavior.

The infinite-layer nickelates $RNiO_2$ (R = Nd, La, Pr) provide a relatively simple system for studying unconventional superconductivity, with Tc values up to 18 K and evidence of strong electron correlations and possible d-wave pairing symmetry. The more recent discovery of pressure-induced superconductivity in $La_3Ni_2O_7$ with Tc up to 80 K has dramatically expanded the potential of nickelates, suggesting that higher transition temperatures may be achievable in this material family.

Significant challenges remain in materials synthesis, understanding the superconducting mechanism, and developing practical applications. The metastability of these compounds, difficulties in achieving phase-pure samples, and limitations in sample form (currently restricted to thin films) present obstacles to both basic research and application development. However, the rapid progress since the initial discovery in 2019 suggests that these challenges may be overcome with continued research effort.

The study of nickelate superconductors has already provided valuable insights into the general principles of unconventional superconductivity and promises to continue doing so in the coming years [25]. Particularly valuable is the opportunity to compare and contrast with cuprates, potentially identifying which features are essential for high-temperature superconductivity and which are material-specific. As synthesis techniques improve and theoretical understanding advances,

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nickelates may indeed prove to be a gateway to both higher-temperature superconductivity and a deeper understanding of this remarkable quantum phenomenon.

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