

Superconductivity in Monolayer-Trilayer Phase of $\text{La}_3\text{Ni}_2\text{O}_7$ under High Pressure

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The discovery of 80 K superconductivity in pressurized bilayer Ruddlesden-Popper (RP) nickelate $\text{La}_3\text{Ni}_2\text{O}_7$ has established a new high-temperature superconductor family [1]. The quest to understand the governing principles of RP nickelate superconductivity has become a central focus in condensed matter physics. Here, we report a critical advance by synthesizing and investigating a distinct structural polymorph of the same compound: the monolayer-trilayer (1313) phase of $\text{La}_3\text{Ni}_2\text{O}_7$. Under high pressure, synchrotron x-ray diffraction and Raman spectroscopy reveal a structural transition from the orthorhombic $Cmmm$ to the tetragonal $P4/mmm$ space group at 13 GPa. Above 19 GPa, the phase shows a clear superconducting transition, confirmed by a zero-resistance state, albeit at a much lower temperature of 3.6 K. The stark contrast with the 80 K transition in the bilayer phase provides a uniquely clean experimental comparison. Our results not only expand the family of RP nickelate superconductors but also deepen the understanding of the superconducting mechanism that governs the transition temperature in these materials.

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I. INTRODUCTION

Since the discovery of superconductivity with a transition temperature T_c approaching 80 K under pressure in the bilayer Ruddlesden-Popper (RP) nickelate $\text{La}_3\text{Ni}_2\text{O}_7$, research on related systems has rapidly attracted widespread attention [1–16]. The chemical formula of RP nickel oxides can be expressed as $A_{n+1}\text{Ni}_n\text{O}_{3n+1}$, where A represents a rare-earth cation and n denotes the number of NiO_6 octahedral layers in the structure. Variations in the number of layers produce different valence states. When $n = 2$, the bilayer $\text{La}_3\text{Ni}_2\text{O}_7$ and its doped compounds with $\text{Ni}^{2.5+}$ exhibit the highest known T_c [17–20]. In these compounds, the strong hybridization between $\text{Ni-}3d_{z^2}$ and $\text{O-}2p_z$ orbitals gives rise to strong interlayer

superexchange interactions, which play a crucial role in the superconducting pairing [21–30]. As n increases, such hybridization weakens due to frustration and T_c decreases [31]. For example, the trilayer $\text{La}_4\text{Ni}_3\text{O}_{10}$ and $\text{Pr}_4\text{Ni}_3\text{O}_{10}$ exhibit maximum T_c values of approximately 40 K under pressure [11–14,32–35]. In these compounds, the Ni ions exhibit a valence state of +2.67 and a $3d^{7.33}$ electronic configuration. On the other hand, both $n = 2$ and 3 compounds undergo structural transitions and suppression of density wave states prior to the emergence of pressure-induced superconductivity [2,32–37]. The structural transition that dominates by straightening of the interlayer Ni-O-Ni bond is expected to enhance the interlayer superexchange coupling, highlighting the important role of lattice degrees of freedom in the superconductivity of RP nickelates.

In RP nickelates, multilayer NiO_6 octahedra and single-layer NiO_6 octahedra can stack in an alternating sequence, forming monolayer-bilayer, monolayer-trilayer, or bilayer-trilayer hybrid phases, namely, the 1212, 1313, and 2323 phases, respectively. These correspond to the chemical formulas $\text{La}_5\text{Ni}_3\text{O}_{11}$, $\text{La}_3\text{Ni}_2\text{O}_7$, and $\text{La}_7\text{Ni}_5\text{O}_{17}$. To date, the 1212 and 1313 phases have been synthesized as single crystals, whereas the 2323 phase has been grown only in thin-film form [16,38–42]. The 1212 phase exhibits

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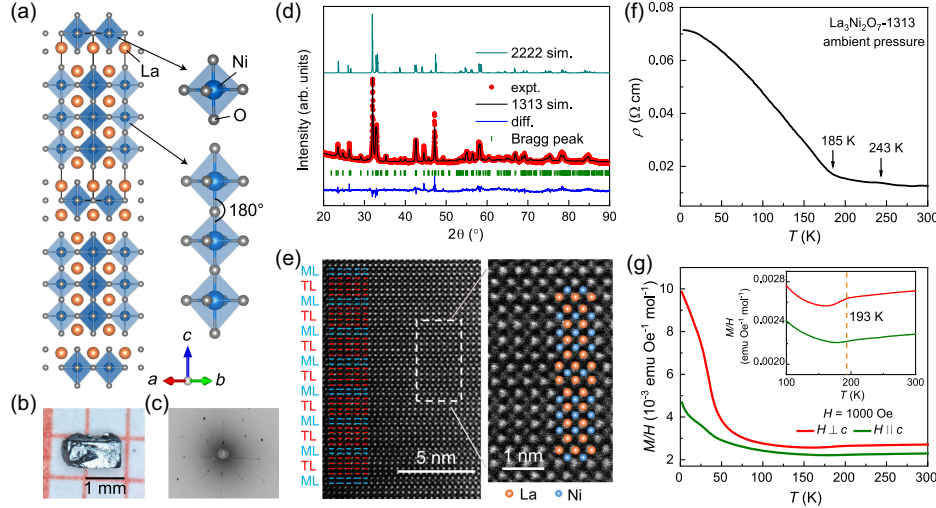


FIG. 1. Structure, resistance, and magnetization of the 1313 $\text{La}_3\text{Ni}_2\text{O}_7$ at ambient pressure. (a) Crystal structure of the 1313 phase with an enlarged view illustrating the alternating stacking of monolayer and trilayer NiO_6 octahedra. The Ni-O-Ni bond angle along the c axis is 180° . (b),(c) Photograph of a single crystal and its corresponding Laue diffraction pattern. (d) Rietveld refinement of the powder XRD pattern using the 1313 structural model. A simulated pattern for the bilayer (2222) $\text{La}_3\text{Ni}_2\text{O}_7$ is shown for comparison. (e) HAADF STEM image of $\text{La}_3\text{Ni}_2\text{O}_7$, with an enlarged view of the dotted area clearly showing the alternating monolayer (ML) and trilayer (TL) sequences. (f) Temperature-dependent resistivity of a 1313 phase single crystal at ambient pressure, showing two distinct anomalies at 243 and 185 K. (g) Temperature-dependent magnetization (M/H) measured at 1000 Oe for magnetic fields applied in plane and out of plane. The inset shows data from 100 to 300 K, revealing an anomaly at 193 K.

pressure-induced superconductivity with a maximum T_c of 64 K, accompanied by the simultaneous collapse of the spin density wave [15,43,44]. A structural transition from orthorhombic to tetragonal occurs prior to the emergence of superconductivity. The 1313 phase shares the same chemical formula as the bilayer $\text{La}_3\text{Ni}_2\text{O}_7$ [38], as shown in Fig. 1(a). Because previously measured 1313 phase samples have limited purity, conclusive evidence for superconductivity in the 1313 phase is lacking. Preliminary investigations have suggested that the 1313 phase may be responsible for the 80 K superconductivity in $\text{La}_3\text{Ni}_2\text{O}_7$, a claim that challenges the conclusion that the bilayer phase exhibits an 80 K T_c and much of the existing theoretical work on superconductivity in RP nickelates [40,45–47]. Recently, researchers have observed superconductivity at ambient pressure in thin films of the bilayer 2222, 1212, and 2323 phases. However, 1313 thin films grown by the same method do not show superconductivity [16]. Given the unique lattice structure of hybrid RP nickelates, investigating the superconducting properties of the pure 1313 phase of $\text{La}_3\text{Ni}_2\text{O}_7$ is of great importance for verifying the superconducting mechanisms of both trilayer and bilayer RP nickelates.

In this work, we successfully synthesized high-purity single crystals of the 1313 phase of $\text{La}_3\text{Ni}_2\text{O}_7$ using the high-pressure floating-zone method. Our measurements reveal an orthorhombic-to-tetragonal structural transition ($Cmmm$ to $P4/mmm$) at 13 GPa and the emergence of superconductivity above 19 GPa, with $T_c^{\text{onset}} = 3.6$ K and $T_c^{\text{zero}} = 2.3$ K at 24.3 GPa. The discovery of

superconductivity in the 1313 phase confirms the universality of pressure-induced superconductivity across the RP nickelate family. More importantly, the significantly lower T_c compared to the bilayer and trilayer phases, despite sharing the same chemical formula as the former and a similar layered motif as the latter, provides compelling evidence that the bilayer NiO_6 structural unit itself is a fundamental prerequisite for high- T_c superconductivity in these materials. Our results establish the number of adjacent, coupled NiO_6 planes as a key structural parameter affecting superconducting pairing strength, offering a crucial design principle for future materials exploration.

II. STRUCTURE AND PROPERTIES UNDER AMBIENT PRESSURE

Figures 1(b) and 1(c) show the picture of an as-grown 1313 phase single crystal and its Laue diffraction pattern, which proves the high-quality crystallinity of the single crystal. The Laue diffraction pattern coincides with the (001) plane. X-ray diffraction (XRD) results for the powder ground from single crystals are shown in Fig. 1(d). The data can be well refined using the 1313 structure model with the orthogonal $Cmmm$ space group, consistent with previously reported structures [38–40]. Under this symmetry, the angle of the out-of-plane Ni-O-Ni bond within a trilayer NiO_6 octahedron remains 180° , as shown in the details of Fig. 1(a). None of the featured peaks of the bilayer phase were detected [Fig. 1(d)]. The 1313 alternating stacking structure is also confirmed by the high-angle annular dark-field scanning transmission electron microscopy (HAADF STEM) image

shown in Fig. 1(e) herein and Supplemental Material Fig. S1 [48]. In the enlarged image of the dotted box area, La and Ni cations can be distinguished individually, as marked in the right-hand part of Fig. 1(e). The stacking arrangement is exactly consistent with the 1313 structure shown in Fig. 1(a).

The temperature dependence of resistivity and magnetization was measured at 1000 Oe under ambient pressure, as shown in Figs. 1(f) and 1(g). The resistivity exhibits a semiconductinglike behavior, consistent with earlier reports [39]. Two resistivity anomalies are observed at 243 and 185 K. The magnetization shows significant anisotropy between the in-plane and out-of-plane directions, indicating anisotropic magnetic correlations between Ni cations. The upturn in magnetization below 50 K may be attributed to minor oxygen vacancies, as previously observed in $\text{La}_3\text{Ni}_2\text{O}_7$ [49,50]. A kink can also be identified at 193 K in the magnetization [inset of Fig. 1(g)], close to the 185 K anomaly in resistivity. The anomalies observed in both resistivity and magnetization are similar to those in other RP nickelates, such as $(\text{La}/\text{Pr})_4\text{Ni}_3\text{O}_{10}$ [13,34], bilayer $\text{La}_3\text{Ni}_2\text{O}_7$ [2,49], and the 1212 phase $\text{La}_5\text{Ni}_3\text{O}_{11}$ [15], suggesting a possible spin density wave transition in the 1313 phase. The weak anomaly at 243 K cannot be observed in all measurements. It may originate from the monolayer NiO_6 units, inherited from La_2NiO_4 , or from a charge density wave transition of the 1313 phase [51,52]. Determining the

precise origins will require further diffraction measurements. We also observed an aging effect in the 1313 phase. For finely ground powder samples, XRD measurements revealed a transformation to the bilayer phase after 12 days. For cracked single crystals of several hundred micrometers in size, both single-crystal XRD and high-pressure transport measurements confirmed the presence of the bilayer phase after 8 months of aging, as shown in Supplemental Material Fig. S2 [48].

III. PRESSURE-INDUCED SUPERCONDUCTIVITY

Figure 2 presents high-pressure electrical transport measurements on a single crystal (S1) of the 1313 phase of $\text{La}_3\text{Ni}_2\text{O}_7$. At 1.0 GPa, two subtle kinks appear around 240 and 181 K, consistent with the anomalies measured at ambient pressure [Fig. 1(f)]; these features are suppressed at 11.3 GPa. A sharp drop in resistance emerges below $T_c^{\text{onset}} = 3.6$ K at 19.0 GPa [Fig. 2(b)]. This drop evolves into a clear zero-resistance state below $T_c^{\text{zero}} = 2.3$ K at 24.3 GPa, confirming the emergence of superconductivity [Fig. 2(c)]. While T_c^{onset} remains relatively constant, T_c^{zero} increases to 3.2 K at 35.0 GPa. The pressure-independent T_c^{onset} in 1313 $\text{La}_3\text{Ni}_2\text{O}_7$ below 35.0 GPa may be ascribed to the fact that the trilayer blocks within the 1313 structure are responsible for superconductivity, and the interspace

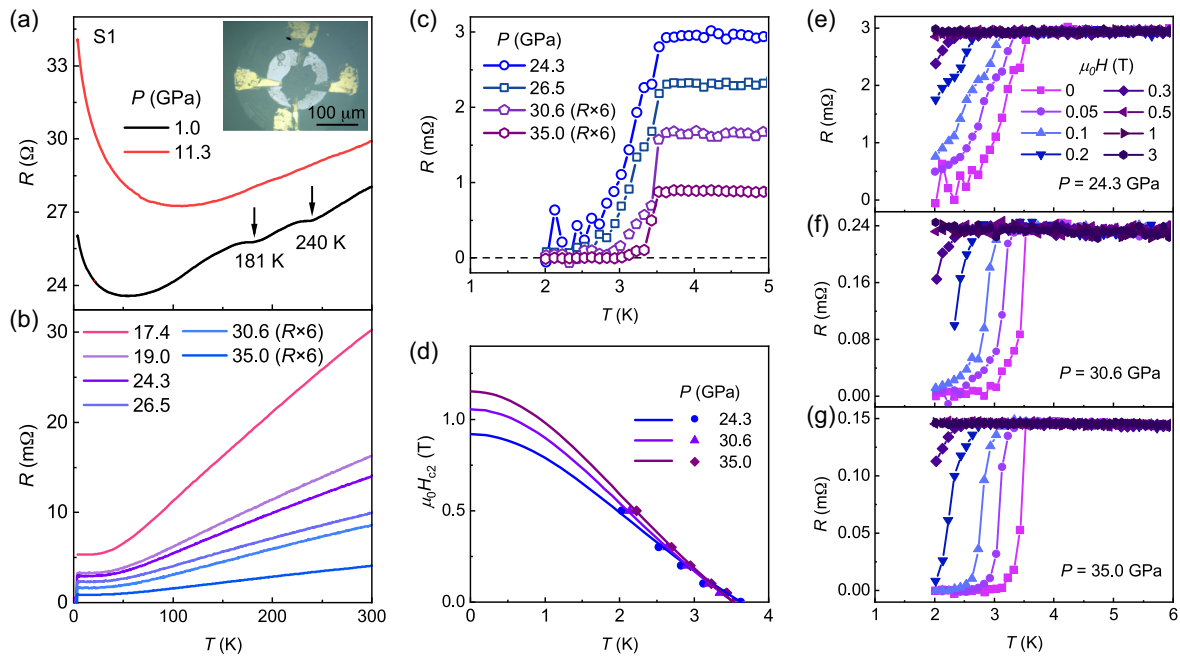


FIG. 2. Electrical transport properties of the 1313 $\text{La}_3\text{Ni}_2\text{O}_7$ under high pressure. (a),(b) Temperature-dependent in-plane resistance for sample S1 at various pressures. Two anomalies are visible at 240 and 181 K at 1.0 GPa. A sharp superconducting transition emerges at $T_c^{\text{onset}} = 3.6$ K and 19.0 GPa. The inset of (a) shows the sample configured for high-pressure transport measurements. (c) Detailed view of the low-temperature resistance of S1 at 24.3, 26.5, 30.6, and 35.0 GPa, demonstrating the development of a zero-resistance superconducting state with increasing pressure. (d) Temperature dependence of the upper critical field $\mu_0 H_{c2}(T)$ at 24.3, 30.6, and 35.0 GPa, with solid lines representing fits using the Ginzburg-Landau model. (e)–(g) Evolution of the superconducting transition with applied magnetic field at selected pressures.

between the single-layer and trilayer units may effectively relieve a significant portion of the applied pressure. Another possibility is that the trilayer structure is more metallic than the bilayer, which may render the superconductivity less sensitive to pressure. To further verify the superconducting state, we measured the field-dependent resistance at 24.3, 30.6, and 35.0 GPa [Figs. 2(e)–2(g)]. The superconductivity is completely suppressed by a magnetic field of 0.5 T at 2 K. The upper critical field $\mu_0 H_{c2}(0)$, estimated from Ginzburg-Landau fitting using the T_c^{onset} , where the resistance deviates from the nearly constant normal-state resistance, is approximately 1 T across these pressures [Fig. 2(d)].

For type-II superconductors, the upper critical field is given by $\mu_0 H_{c2}(T) = \Phi_0 / 2\pi \xi^2(T)$, where Φ_0 is the magnetic flux quantum and $\xi(T)$ is the superconducting coherence length. At 35.0 GPa, we obtain a coherence length $\xi(0) \approx 169$ Å. Given that $\xi(T) \propto v_F / \Delta$, where v_F is the Fermi velocity and Δ is the superconducting gap, the observed increase in $\mu_0 H_{c2}$ from 24.3 to 35.0 GPa suggests a moderate enhancement of the pairing gap. This occurs in bilayer $\text{La}_3\text{Ni}_2\text{O}_{7-\delta}$ with different oxygen stoichiometries, where T_c^{onset} remains nearly constant but the $\mu_0 H_{c2}$ is quite different [53,54].

We performed similar high-pressure electronic transport measurements on a second sample, S2 (Supplemental Material Fig. S3 [48]). The data from S2 exhibit improved metallic behavior at low pressures and indicate the emergence of a superconducting state below approximately 3.0 K at pressures above 24.5 GPa. However, the resistance does not drop to zero, which may be due to poorer pressure homogeneity, a phenomenon also observed in other bilayer and trilayer RP nickelate phases [33,55,56]. Notably, no signature of superconductivity near 80 K was detected in either sample up to the maximum pressure of 35.0 GPa in our experiments. Factors such as oxygen deficiency may influence the T_c of the 1313 phase, although significant oxygen deficiency is unlikely, as it would suppress superconductivity. Indeed, superconductivity at ~ 5 K has been reported for trilayer intergrowths [54].

Among the superconducting RP nickelates, the 1313 phase exhibits the most fragile superconductivity. The maximum ratio $\mu_0 H_{c2}(0)/T_c \sim 0.33$ is significantly lower than that of bilayer (~ 1.5 – 2.5) and trilayer (~ 1.5 – 1.8) single crystals [1,11,13,17,18,34]. Interestingly, this value is comparable to the ratio of ~ 0.44 observed in the 1212 phase [15]. We note that pressurized $\text{La}_2\text{NiO}_{4.06}$ does not exhibit superconductivity up to 29.4 GPa [57]. The substantially reduced $H_{c2}(0)$ relative to T_c in both hybrid phases indicates that the coupled NiO_6 layers are responsible for pressure-induced superconductivity, whereas the monolayer NiO_6 octahedra do not contribute. Instead, they serve to separate the coupled superconducting NiO_6 layers and suppress $H_{c2}(0)$, in contrast to the self-doping effect

theory, which emphasizes the role of superconducting monolayers [47]. Resolving this issue conclusively will require measurements of the anisotropic $H_{c2}(0)$.

IV. STRUCTURAL TRANSITION UNDER HIGH PRESSURE

To investigate the structural evolution under pressure, we performed synchrotron XRD and Raman spectroscopy measurements. Figure 3 summarizes the high-pressure powder XRD patterns and Raman spectra acquired at room temperature. The XRD pattern at 4.6 GPa is consistent with the ambient-pressure $Cmmm$ structure [Fig. 3(a)], in agreement with the as-grown single-crystal structure [38–40]. At 13.4 GPa, the adjacent (0 2 0) and (2 0 0) diffraction peaks merge [Fig. 3(b)], indicating a transition to a higher-symmetry tetragonal phase. This transition is further supported by the pressure dependence of the peak positions (Supplemental Material Fig. S4 [48]). Rietveld refinement confirms that the high-pressure phase is well described by the tetragonal $P4/mmm$ space group [Fig. 3(a)], consistent with a previous high-pressure study [40]. The pressure evolution of the lattice constants and the in-plane Ni–O–Ni angle are summarized in Figs. 3(d) and 3(f), whose raw data are shown in Supplemental Material Fig. S4 [48]. Precise determination of the structural transition pressure from XRD is challenging because oxygen atoms have a weak x-ray scattering cross section.

Figure 3(c) displays the high-pressure Raman spectra. At 1.5 GPa, five distinct peaks are observed in the 300–600 cm^{-1} range. With increasing pressure, the two prominent peaks P2 and P3 gradually converge, eventually merging into a single peak, P2'. The intensity and position of these peaks reveal a structural transition at approximately 13.0 GPa [Fig. 3(e) herein and Supplemental Material Figs. S4(e)–S4(g) [48]]. Combined, the XRD and Raman measurements demonstrate that the orthorhombic-to-tetragonal structural transition occurs at 13.0 GPa, preceding the emergence of superconductivity at 19 GPa. The similar structural-transition pressures for the bilayer, trilayer, and hybrid phases may be attributed to the common multilayer NiO_6 octahedral framework. The Ni–O–Ni bond angle along the c axis has been proposed as a critical parameter for superconductivity in RP nickelates. In the orthorhombic $Cmmm$ structure of the 1313 phase $\text{La}_3\text{Ni}_2\text{O}_7$ at ambient pressure, this angle is 180° , satisfying the proposed criterion. The separation between the structural and superconducting transition pressures in the 1313 and 1212 phases, in contrast to their coincidence in the bilayer and trilayer phases, highlights the crucial role of the 180° Ni–O–Ni bond angle along the c axis, rather than a tetragonal structure, in the emergence of superconductivity in RP nickelates [13,15,17].

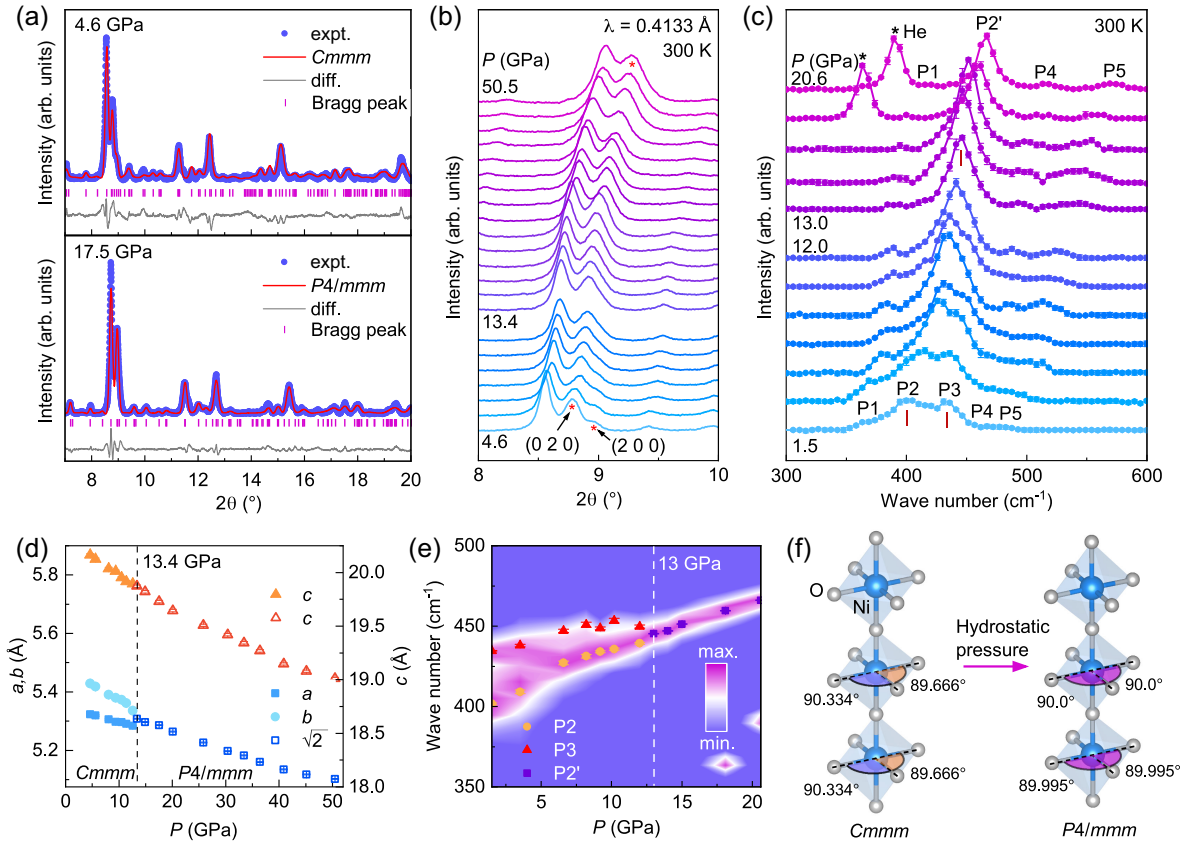


FIG. 3. High-pressure structural evolution of the 1313 $\text{La}_3\text{Ni}_2\text{O}_7$. (a) Rietveld refinements of synchrotron XRD patterns for the 1313 phase at 4.6 and 17.5 GPa, measured at room temperature. The refinements correspond to the orthorhombic $Cmmm$ and tetragonal $P4/mmm$ space groups, respectively. (b) Pressure evolution of selected reflection peaks between 8° and 10° , showing the merging of the $(0\ 2\ 0)$ and $(2\ 0\ 0)$ peaks, which signals the transition to the tetragonal phase. Neon was used as the pressure-transmitting medium. (c) Raman spectra of the 1313 phase at room temperature under various pressures. Helium was used as the pressure-transmitting medium. (d) Pressure dependence of the lattice constants a , b , and c . (e) Color map of the Raman intensity as a function of pressure and wave number. The positions of peaks P2, P3, and P2', determined by Lorentzian fits, are overlaid. The intensity of peak P2 is normalized to unity at each pressure for comparison. The dashed line at 13 GPa marks the merging of two peaks, indicating a structural transition. (f) Schematic of the trilayer structure unit in the 1313 phase before and after the structural transition.

V. PHASE DIAGRAM

We summarize the electronic transport and structural properties of the 1313 phase of $\text{La}_3\text{Ni}_2\text{O}_7$ from 0 to 36 GPa in the phase diagram of Fig. 4, where the color scale represents the normalized resistance $R/R_{50\text{ K}}$. The orthorhombic-to-tetragonal structural transition is accompanied by a substantial change in resistance, demonstrating a strong coupling between the electronic and lattice degrees of freedom. Superconductivity in the 1313 phase emerges within the tetragonal structure, mirroring its trilayer counterpart. A systematic comparison across the RP nickelate family reveals a dramatic hierarchy in superconducting transition temperatures: from the bilayer phase ($T_c \sim 80$ K) to the trilayer phase ($T_c \sim 30$ K), and further to the 1212 ($T_c \sim 64$ K) and 1313 ($T_c \sim 3$ K) phases. Structurally, the trilayer can be viewed as a frustrated bilayer, where the nonbonding $\text{Ni}-3d_{z^2}$ state introduces frustration and weakens the correlations established by the bonding and antibonding

states. The 1212 phase is a separated bilayer, and the 1313 phase is both frustrated and separated. Both the bilayer and trilayer blocks of the hybrid phases are compressed in plane and stretched out of plane more than their counterparts (Supplemental Material Fig. S5 [48]). The pronounced suppression of T_c from the bilayer to the trilayer structure, and to their hybrid phases, underscores the critical role of interlayer electronic and magnetic exchange couplings. Our results, therefore, establish the pristine, coupled bilayer as the essential structural motif for achieving high- T_c superconductivity in RP nickelates, providing a fundamental principle for guiding the search for new superconducting materials.

VI. METHODS

A. Sample synthesis

Single crystals of the 1313 phase of $\text{La}_3\text{Ni}_2\text{O}_7$ were grown using the high-pressure floating-zone method. Feed and seed rods were prepared by mixing predried La_2O_3

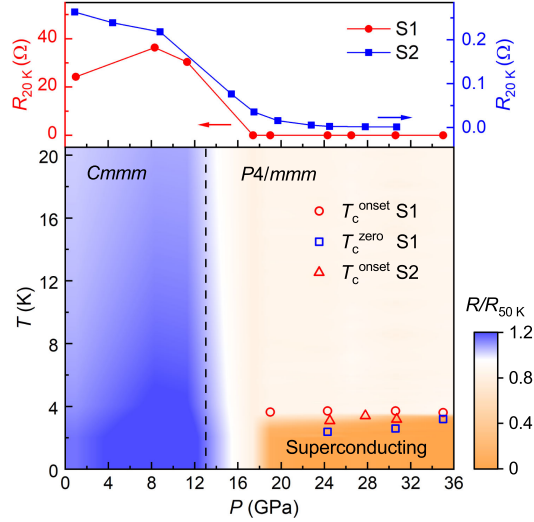


FIG. 4. Phase diagram of the 1313 $\text{La}_3\text{Ni}_2\text{O}_7$. Upper panel: pressure dependence of the resistance at 20 K for samples S1 and S2. Lower panel: temperature-pressure phase diagram for the 1313 phase. The superconducting onset temperature T_c^{onset} and zero-resistance temperature T_c^{zero} for S1, along with T_c^{onset} for S2, are indicated. The pressure-induced structural transition from the orthorhombic $Cmmm$ phase to the tetragonal $P4/mmm$ phase at 13 GPa is marked by a vertical dashed line. The background color represents the normalized resistance $R/R_{50\text{ K}}$ for S1, highlighting the superconducting region and the pronounced resistance change at the structural transition.

(99.99%) and NiO (99.99%) powders in a molar ratio of 3:4. The mixture was ground, pressed into pellets, and sintered at 1100 °C for 48 h. We repeated this grinding and sintering process 2–3 times. The resulting powder was then ground again, pressed into rods, and sintered at 1200 °C for 72 h to form feed and seed rods 10 cm long and 5 mm in diameter.

We performed crystal growth in a vertical optical floating-zone furnace (HKZ, SciDre) under an oxygen pressure of 15 bar. The feed and seed rods were counter-rotated at a relative speed of 35 rpm, with a growth rate of 3 mm/h. Throughout the growth process, the molten zone width was minimized to maximize contact between the reactant and oxygen. The resulting crystal was approximately 3 cm in length. Pure-phase $\text{La}_3\text{Ni}_2\text{O}_7$ -1313 single crystals were obtained at the position close to the growth finish end after mechanically removing a ~ 1 -mm-thick surface layer.

B. Sample characterization at ambient pressure

We characterized the crystal structure of the 1313 phase of $\text{La}_3\text{Ni}_2\text{O}_7$ at ambient pressure by XRD (Rigaku), Laue diffraction (Photonic Science), and HAADF STEM. HAADF STEM experiments were conducted using a double aberration-corrected JEOL ARM200F microscope operated at 200 kV. We performed dc magnetic susceptibility and

electrical resistance measurements using a superconducting quantum interference device (SQUID) and a physical property measurement system (PPMS), respectively.

C. High-pressure electronic transport, x-ray diffraction, and Raman measurements

High-pressure experiments were conducted using DACs. For electronic transport measurements, we used a DAC with 300- μm culet anvils. Single crystals of dimensions $80 \times 80 \times 10 \mu\text{m}$ were loaded into a sample chamber of 160 μm diameter and approximately 30 μm thickness. Daphne oil 7373 was used as the pressure-transmitting medium (PTM) to ensure a hydrostatic pressure environment. Resistance measurements were performed using a PPMS.

High-pressure synchrotron XRD experiments were conducted at the ID31 high-pressure beamline of the Beijing High Energy Photon Source. Powder samples, obtained by grinding single crystals, were loaded into a DAC with 250- μm culet anvils using neon gas as the PTM.

High-pressure Raman spectroscopy measurements employed a DAC with 250- μm culet anvils and helium gas as the PTM. A single crystal, measuring $50 \times 50 \times 10 \mu\text{m}$, was loaded for measurement. Raman spectra were collected in a confocal geometry (MoniVista CRS3) using a Princeton Instruments HRS-500 spectrometer. We used a 532-nm laser with power below 3 mW for excitation. Pressure calibration below 10.0 GPa was performed using the fluorescence spectrum of a 10- μm ruby sphere; for pressures above 10.0 GPa, the high-frequency edge of the diamond Raman spectrum was utilized.

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M. W. and H. S. conceived and designed the project. C. H., D. H., and M. H. grew and characterized the single crystals. J. L., H. Z., and X. C. performed the high-pressure measurements and analyzed the data. X. H. and Z. C. conducted the STEM measurements.

DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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