

Magnetic correlations and pairing tendencies of the hybrid stacking nickelate superlattice $\text{La}_7\text{Ni}_5\text{O}_{17}$ ($\text{La}_3\text{Ni}_2\text{O}_7/\text{La}_4\text{Ni}_3\text{O}_{10}$) under pressure

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Motivated by the recent rapid progress in high- T_c nickelate superconductors, we comprehensively study the physical properties of the alternating bilayer trilayer stacking nickelate $\text{La}_7\text{Ni}_5\text{O}_{17}$. The high-symmetry phase of this material, without the tilting of oxygen octahedra, is not stable at ambient conditions but becomes stable under high pressure, where a small hole pocket γ_0 , composed of the $d_{3z^2-r^2}$ states in the trilayer sublattice, appears. This pocket was identified in our previous work for trilayer $\text{La}_4\text{Ni}_3\text{O}_{10}$ as important to develop superconductivity. Moreover, using random-phase approximation calculations, we find a leading s^\pm pairing state for the high-symmetry phase under pressure with similar pairing strength as that obtained previously for the bilayer $\text{La}_3\text{Ni}_2\text{O}_7$ compound, suggesting a similar or higher superconducting transition temperature T_c , at the random-phase approximation level. In addition, we find that the dominant magnetic fluctuations in the system driving this pairing state have antiferromagnetic structure both in-plane and between the planes of the top and bottom trilayer and bilayer sublattices, while the middle trilayer is magnetically decoupled.

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I. INTRODUCTION

The recent discovery of superconductivity under pressure in the Ruddlesden-Popper (RP) nickelate systems $\text{La}_3\text{Ni}_2\text{O}_7$ [1] and $\text{La}_4\text{Ni}_3\text{O}_{10}$ [2,3] opened a new avenue for the study of unconventional high- T_c superconductivity in the field of condensed matter physics and material sciences [4–11]. $\text{La}_3\text{Ni}_2\text{O}_7$ has the ABAB bilayer (BL) stacking structure [12,13], where superconductivity with a high transition temperature up to 80 K [1] was obtained after a first-order structural pressure-induced transition around 14 GPa [5] that suppresses the tilting of the NiO_6 octahedra [see Fig. 1(a)]. Very recently, Li *et al.* [14] found that superconductivity persists up to 90 GPa. Moreover, they measured the Meissner effect of the superconducting state using the *ac* magnetic susceptibility reporting that the superconducting volume fraction is around 48% [14].

$\text{La}_4\text{Ni}_3\text{O}_{10}$ also has a similar ABAB trilayer (TL) stacking structure [12,15,16]. In this case pressure smoothly suppresses the distortion of the NiO_6 octahedra, yielding a high-symmetry I4/mmm phase [2,3], without the tilting of oxygen octahedra, around 15 GPa [see Fig. 1(b)]. After reaching the high symmetry phase, superconductivity was also found in a very broad pressure range although with a T_c of about 20–30 K [2,3,10,11], lower than in the BL case. In addition, using the *ac* magnetic susceptibility, a $\sim 80\%$ superconducting volume fraction was reported [3].

Density functional theory (DFT) calculations indicate that both systems can be described using a Ni two-orbital BL or TL model [10,17–19], where the Fermi surface (FS) is composed of the Ni's $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbitals [17–21]. In addition, the Ni $d_{3z^2-r^2}$ orbital can form a bonding-antibonding molecular-orbital state [18,19,21] in both systems, while the $d_{x^2-y^2}$ orbital is partially occupied, as shown in Fig. 1. Due to the in-plane hybridization between the e_g orbitals, the induced large interorbital hopping leads to a noninteger population in both orbitals [22,23]. Furthermore, many interesting properties have been unveiled in both theoretical and experimental works, such as spin or charge correlations [24–28], exotic superconducting pairing symmetries [19,29–34], rare-earth effects [35,36], and a prominent role of the Hund coupling [37,38].

Based on these established results, here we propose a very interesting challenge: what will happen if the BL and TL sublattices occur together in a single material? Previous studies in RP iridates reported a single-layer (SL) BL hybrid stacking superlattice structure in $\text{Sr}_2\text{IrO}_4/\text{Sr}_3\text{Ir}_2\text{O}_7$ [39,40], as well as other RP perovskite superlattices [41–43]. Furthermore, similar alternating SL-TL or SL-BL stacking structures were also discussed experimentally in nickelates, by several independent groups [44–48]. Thus, in principle, it appears possible to synthesize a BL-TL stacking structure in a real nickelate material. In this case, several interesting questions naturally arise: What are the electronic and magnetic properties of this BL-TL material? Could it become superconducting? And in this case, what is the pairing channel?

To answer those questions, we systematically investigate the alternating BL and TL hybrid stacking nickelate superlattice $\text{La}_7\text{Ni}_5\text{O}_{17}$ ($\text{La}_3\text{Ni}_2\text{O}_7/\text{La}_4\text{Ni}_3\text{O}_{10}$) by using DFT and

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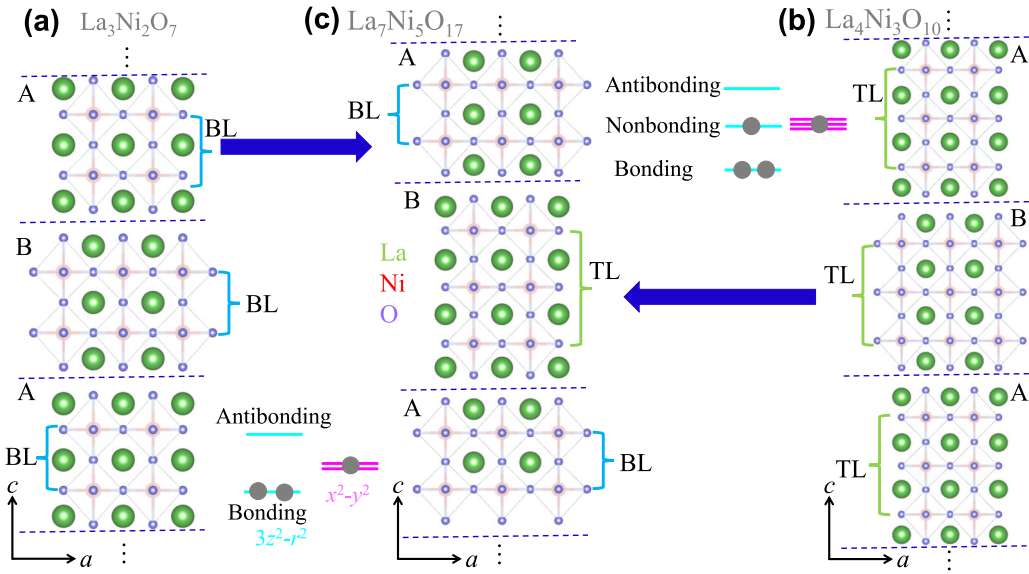


FIG. 1. Schematic crystal structures of (a) BL $\text{La}_3\text{Ni}_2\text{O}_7$, (b) TL $\text{La}_4\text{Ni}_3\text{O}_{10}$, and (c) the alternating BL-TL $\text{La}_7\text{Ni}_5\text{O}_{17}$ proposed here, respectively (green = La; red = Ni; purple = O). Inset: Sketches of electronic states of e_g orbitals in the BL $\text{La}_3\text{Ni}_2\text{O}_7$ and TL $\text{La}_4\text{Ni}_3\text{O}_{10}$. The light blue (pink) horizontal lines represent $d_{3z^2-r^2}$ ($d_{x^2-y^2}$) states. The solid circles represent electrons. The total population n of the e_g electrons considered is $n = 3.0$ and $n = 4.0$, for the BL with two sites and the TL with three sites, respectively.

random phase approximation (RPA) calculations. At ambient conditions, the high-symmetry phase of $\text{La}_7\text{Ni}_5\text{O}_{17}$ without the tilting of oxygen octahedra is not stable it becomes stable under pressure. Similarly to other RP nickelates, $\text{La}_7\text{Ni}_5\text{O}_{17}$ also has a large charge transfer gap and bond-antibonding splittings of $d_{3z^2-r^2}$ orbitals, where its electronic structure resemble the combinations of BL and TL nickelates. Furthermore, our RPA calculations suggest a leading s^\pm pairing state with sizable pairing strength λ similar to that found for the pure BL nickelates, suggesting a similar or higher superconducting transition temperature. Considering previous theoretical and experimental studies on RP nickelate superconductors, we, therefore, believe that superconductivity could also be expected to exist in $\text{La}_7\text{Ni}_5\text{O}_{17}$ for a broad pressure region as well. Moreover, we also find that the dominant magnetic fluctuations driving this pairing state have antiferromagnetic structure both in-plane and between the planes of the top and bottom TL and BL sublattices, while the middle TL layer is magnetically decoupled. However, the antiferromagnetic correlations are much weaker in the BL sublattice, compared to that in the TL sublattice.

II. METHODS

A. DFT Method

In the present work, first-principles DFT calculations were performed using the Vienna *ab initio* simulation package (VASP) code with the projector augmented wave (PAW) method [49–51], where the electronic correlations were considered by the generalized gradient approximation (GGA) and the Perdew-Burke-Ernzerhof (PBE) exchange potential [52]. For the discussion of alternating stacking BL-TL $\text{La}_7\text{Ni}_5\text{O}_{17}$, the atomic positions and crystal lattice were fully relaxed until the Hellmann-Feynman force on each atom was smaller than $0.001 \text{ eV}/\text{\AA}$ for different pressures. Here, the plane-wave

cutoff energy was set as 550 eV and the k -point mesh was $20 \times 20 \times 3$ for the conventional structure. For the phonon spectrum of the P4mm (No. 99) and P4/mmm (No. 123) phases of the alternating BL-TL stacking $\text{La}_4\text{Ni}_3\text{O}_{10}$, a $\sqrt{2} \times \sqrt{2} \times 1$ supercell structure was used in the phonon calculation, by using the density functional perturbation theory approach [53–55], analyzed by the PHONOPY software in the primitive unit cell [56,57]. The evolution of possible phase transitions under pressure and structure at ambient conditions are not the main scope of our present work, thus we leave the issue of adding more pressures to future work. In addition to the standard DFT calculation, the maximally localized Wannier functions (MLWFs) method was also employed to fit the Ni $3d$ bands, by using the WANNIER90 packages [58], to obtain the hopping matrices and crystal-field splittings. All the crystal structures were visualized with the VESTA code [59].

B. Tight-binding method

Based on the hopping matrices and crystal-field splittings obtained from MLWFs, we constructed a ten-band low-energy e_g -orbital BL plus TL tight-binding model, where the overall filling was considered as $n = 7.0$ for five Ni sites, corresponding to 1.4 electrons per Ni site on average. For pedagogical reasons, let us explain from where these numbers arise. In our material of focus $\text{La}_7\text{Ni}_5\text{O}_{17}$, and assuming the valence of oxygen is always 2– and that of Ba is 3+, then charge neutrality requires for Ni a valence 2.6+. Because the $3d$ shell of Ni can accommodate up to 10 electrons, then there are 7.4 electrons per Ni in the $3d$ shell, in average. However, the t_{2g} is assumed fully occupied with 6 electrons/Ni and is not included in our tight-binding model. Thus, in the e_g sector we should have 1.4 electrons, which explains the number used above. Note also that for the 5 Ni's of the formula $\text{La}_7\text{Ni}_5\text{O}_{17}$, then with each Ni contributing 1.4 electrons, for 5 Ni's we

have 7 electrons as expressed above as well. But our calculations indicate that these 7 electrons are split with 3 electrons in the BL sector and 4 electrons in the TL sector.

The detailed hopping matrices can be found in an attached separate file. The Fermi energy is obtained by integrating the density of states for all ω until the number of electrons $n = 7$ is reached. Based on the obtained Fermi energy, a 4001×4001 k -mesh was used to calculate the Fermi surface.

C. RPA method

To study superconducting pairing properties, the RPA method was used here, based on a perturbative weak-coupling expansion in the Hubbard interaction. It has been shown in many studies that this procedure captures the essence of the physics for Fe-based and Cu-based superconductors (see Ref. [60] as an example). The full Hubbard model Hamiltonian for the BL plus TL discussed here, includes the kinetic energy and interaction terms, and it is written as $H = H_k + H_{\text{int}}$. The electronic interaction portion of the Hamiltonian includes the standard same-orbital Hubbard repulsion U , the electronic repulsion U' between electrons at different orbitals, the Hund's coupling J , and the on-site interorbital electron-pair hopping terms (J'). Formally, it is given by

$$H_{\text{int}} = U \sum_{i\gamma} n_{i\uparrow\gamma} n_{i\downarrow\gamma} + \left(U' - \frac{J}{2} \right) \sum_{i} \sum_{\gamma < \gamma'} n_{i\gamma} n_{i\gamma'} - 2J \sum_{i} \sum_{\gamma < \gamma'} \mathbf{S}_{i,\gamma} \cdot \mathbf{S}_{i,\gamma'} + J \sum_{i} \sum_{\gamma < \gamma'} (P_{i\gamma}^\dagger P_{i\gamma'} + \text{H.c.}), \quad (1)$$

where the standard relations $U' = U - 2J$ and $J' = J$ are assumed, and $P_{i\gamma} = c_{i\downarrow\gamma} c_{i\uparrow\gamma}$.

In the multiorbital RPA approach [60–63], the enhanced spin susceptibility is obtained from the bare susceptibility (Lindhart function) via $\chi_0(\mathbf{q})$ as $\chi(\mathbf{q}) = \chi_0(\mathbf{q})[1 - \mathcal{U}\chi_0(\mathbf{q})]^{-1}$. Here, $\chi_0(\mathbf{q})$ is an orbital-dependent susceptibility tensor and \mathcal{U} is a tensor that contains the intra-orbital U and interorbital U' density-density interactions, the Hund's rule coupling J , and the pair-hopping J' term. The pairing strength λ_α for channel α and the corresponding gap structure $g_\alpha(\mathbf{k})$ are obtained from solving an eigenvalue problem of the form

$$\int_{FS} d\mathbf{k}' \Gamma(\mathbf{k} - \mathbf{k}') g_\alpha(\mathbf{k}') = \lambda_\alpha g_\alpha(\mathbf{k}), \quad (2)$$

where the momenta \mathbf{k} and \mathbf{k}' are on the FS and $\Gamma(\mathbf{k} - \mathbf{k}')$ contains the irreducible particle-particle vertex. In the RPA approximation, the dominant term entering $\Gamma(\mathbf{k} - \mathbf{k}')$ is the RPA spin susceptibility $\chi(\mathbf{k} - \mathbf{k}')$.

In this work, note that we are using RPA to calculate the spin susceptibility and the pairing interaction strength for the model derived from the DFT electronic structure, with no additional self-energy corrections. The values of U , U' , J , and J' are restricted to values below the critical values for which the RPA finds a spin density wave instability and, therefore, can be different from those used in other techniques such as DFT+ U or DFT+DMFT.

III. RESULTS

A. Structural instability

Considering previous work on nickelate superconductors, it has been established that superconductivity exists in the high symmetry phase without the tilting of oxygen octahedra. For this reason, we used the P4/mmm symmetry (No.123) of the high-pressure phase of the SL-TL stacking nickelate [46], without any tilting, to construct the BL-TL stacking structure, as shown in Fig. 1(c). This corresponds to the chemical formula $\text{La}_7\text{Ni}_5\text{O}_{17}$. To better understand the physical properties of this BL-TL $\text{La}_7\text{Ni}_5\text{O}_{17}$ compound, we optimized the crystal structure for different pressures by using first-principles DFT calculations. Using the fully optimized crystal lattice, and a high accuracy tolerance, the space group becomes P4mm (No. 99). Because the distortion from P4/mmm (No. 123) to P4mm (No. 99) is very small, leading to a tiny difference in enthalpy (~ 0.05 meV/Ni), the electronic structures of both symmetries are essentially identical (see Supplemental Material I [64]). This is quite similar to a previous study in the BL $\text{La}_3\text{Ni}_2\text{O}_7$ context, where both lower Fmmm and higher I4/mmm symmetries give the same results [65] for the same reason: only a tiny distortion exists between the I4/mmm and Fmmm phases of $\text{La}_3\text{Ni}_2\text{O}_7$ [66].

Next, we study the structural stability of this BL-TL stacking structure. For this purpose, we calculated the phonon spectrum of the P4mm and P4/mmm phases of $\text{La}_7\text{Ni}_5\text{O}_{17}$ both without and with pressure, by using the density functional perturbation theory approach [53–55] analyzed by the PHONONPY software [56,57]. The phonon dispersion spectrum displays imaginary frequencies appearing at the high symmetry points of $\text{La}_7\text{Ni}_5\text{O}_{17}$, as displayed in Fig. 2(a), suggesting that the P4mm or P4/mmm phase is not stable at 0 GPa. However, both of them become stable under high pressure, namely without showing any imaginary frequency [see the results at 30 GPa in Fig. 2(b)]. This is similar to previous well-discussed nickelate superconductors [19,30], where the untilted structure is not stable at ambient pressure but becomes stable at high pressure.

B. Electronic structures

Let us now discuss the electronic properties of the BL-TL stacking $\text{La}_7\text{Ni}_5\text{O}_{17}$. Similar to other nickelates [67,68], the states of $\text{La}_7\text{Ni}_5\text{O}_{17}$ near the Fermi level mainly arise from the Ni e_g orbitals while the O $2p$ orbitals are located deeper in energy, leading to a large charge-transfer gap between $3d$ and $2p$ states. We then constructed a ten-band e_g -orbital tight-binding model for the BL plus TL stacking structure of $\text{La}_7\text{Ni}_5\text{O}_{17}$ at both 0 GPa and 30 GPa. Here, the overall electron filling is $n = 7$, the ten bands arise from five Ni atoms in a unit cell, each contributing two orbitals, and the model includes the longer-range hoppings linking the BL and TL sublattices obtained from the maximally localized Wannier functions (MLWFs) method based on the WANNIER90 package [58]. Formally, the model's Hamiltonian is written as

$$H_k = \sum_{\substack{i\sigma \\ \bar{\alpha}\gamma\gamma'}} t_{\gamma\gamma'}^{\bar{\alpha}} (c_{i\sigma\gamma}^\dagger c_{i+\bar{\alpha}\sigma\gamma'} + \text{H.c.}) + \sum_{i\gamma\sigma} \Delta_\gamma n_{i\gamma\sigma}. \quad (3)$$

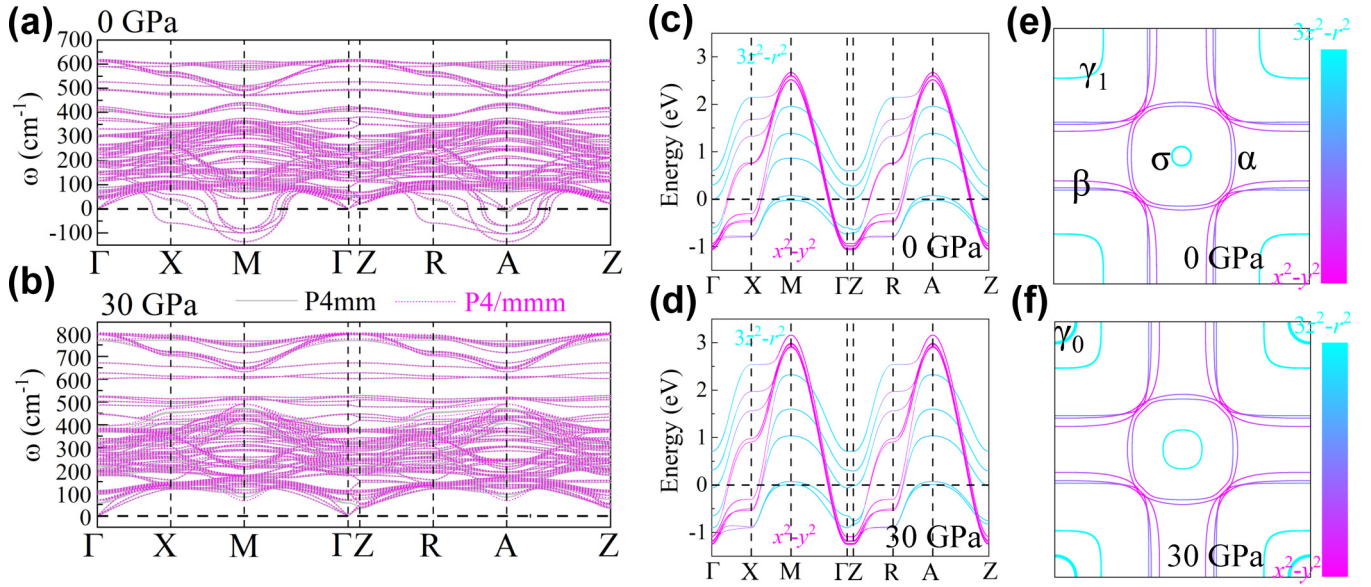


FIG. 2. (a), (b) Phonon spectrum of the BL-TL stacking $\text{La}_7\text{Ni}_5\text{O}_{17}$ compound in the P4mm (No. 99) and P4/mmm (No. 123) phases at 0 GPa, and at 30 GPa, respectively. The results of other pressures can be found in Supplemental Material II [64]. The coordinates of the high-symmetry points in the Brillouin zone (BZ) are $\Gamma = (0, 0, 0)$, $X = (0, 0.5, 0)$, $M = (0.5, 0.5, 0)$, $Z = (0, 0, 0.5)$, $R = (0, 0.5, 0.5)$, and $A = (0.5, 0.5, 0.5)$. Band structures and FSs for (c), (d) 0 and (e), (f) 30 GPa, respectively. Here, a ten-band e_g orbital tight-binding model was considered including long-range hoppings linking the BL and TL substructures, with an overall filling of $n = 7$ (i.e., 1.4 electrons per site).

Here, the first term represents the hopping of an electron from orbital γ' at site $i + \vec{a}$ to orbital γ at site i . $c_{i\sigma\gamma}^\dagger$ ($c_{i\sigma\gamma}$) is the standard creation (annihilation) operator, γ and γ' represent different orbitals, and σ is the z axis spin projection. Δ_γ represents the crystal-field splitting of each orbital γ . The vectors \vec{a} are along the three directions, defining different hopping neighbors (the entire hopping files are in the Supplemental Material [64]).

As shown in Figs. 2(c) and 2(d), the electronic structure of $\text{La}_7\text{Ni}_5\text{O}_{17}$ appears to arise from a combined behavior between $\text{La}_3\text{Ni}_2\text{O}_7$ (BL) and $\text{La}_4\text{Ni}_3\text{O}_{10}$ (TL): the $d_{3z^2-r^2}$ orbitals display the bonding-antibonding, or the bonding-antibonding-nonbonding splitting behavior characteristic of the BL and TL sublattices, respectively, while the $d_{x^2-y^2}$ orbital remains decoupled among the planes. The bandwidth of the e_g orbitals increases by $\sim 20\%$ from 0 GPa to 30 GPa, suggesting an enhancement of itinerant properties under pressure. However, the TL bonding state involving the $d_{3z^2-r^2}$ orbital does not touch the Fermi level at 0 GPa, but crosses the Fermi level at 30 GPa. This results in a small hole pocket γ_0 in $\text{La}_7\text{Ni}_5\text{O}_{17}$ at 30 GPa, which is *absent* at 0 GPa, as displayed in Figs. 2(e) and 2(f), which is fitting well with DFT FSs (see Supplemental Material I [64]). This small hole pocket from the TL sublattice is important for the superconductivity as argued in the following discussion, and as found in our previous literature on the TL $\text{La}_4\text{Ni}_3\text{O}_{10}$ [19]. There is a larger hole γ_1 pocket composed by the $d_{3z^2-r^2}$ orbitals from the BL sublattice as well, but we find that it does not play a role in pairing. Finally, the α and β sheets of the Fermi surface have a multilayer splitting behavior, as displayed in Figs. 2(e) and 2(f).

Different from the SL-TL stacking nickelate (SL: $n = 1.77$ and TL: $n = 4.23$) [66], we do not observe a robust “charge

transfer” effect between the BL (with two Ni sites) and TL (with three Ni sites) layers at both 0 GPa (BL: $n = 2.96$ and TL: $n = 4.04$) and at 30 GPa (BL: $n = 3.00$ and TL: $n = 4.00$), respectively. In addition, both Ni sites of the top and bottom layers of the BL and TL sublattices [see Fig. 3(a)] have a very small on-site energy difference (< 1 meV), as well as a very small hopping difference between Ni3-Ni4 and Ni4-Ni5 (< 1 meV). Furthermore, the in-plane hybridization ($t_{x/y}^{12}/t_{x/y}^{22}$) between the $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals is slightly enhanced in the BL sublattice (~ 0.512) in $\text{La}_7\text{Ni}_5\text{O}_{17}$, compared with that in the BL $\text{La}_3\text{Ni}_2\text{O}_7$ (~ 0.475).

C. Pressure effect

As displayed in Fig. 3(b), the $d_{3z^2-r^2}$ orbitals from the BL sublattice have slightly lower on-site energy than that of the bottom and top layers from the TL sublattice, while the $d_{3z^2-r^2}$ orbitals of the Ni4 site from the middle layer of the sublattice has much higher on-site energy. In addition, we also show several key crystal-field splitting energies for different pressures in Fig. 3(c). As pressure increases, the values of the crystal splittings between the e_g orbitals (Δ_1 , Δ_3 , and Δ_5) also increase, as do the values of both the interorbital and intraorbital hopping matrix elements. However, the hopping ratios $t_z^{11}/t_{x/y}^{22}$ for the nearest-neighbor Ni-Ni along the interlayer direction do not change much for different Ni sites under pressure, as shown in Fig. 3(d). Moreover, the different ratios of in-plane hybridizations ($t_{x/y}^{12}/t_{x/y}^{22}$) of e_g orbitals do not change much either, but this hybridization is stronger in the TL sublattice than that in the BL sublattice, at least in the pressure region we studied [see Fig. 3(e)]. The slight differences between the Ni1 and Ni2 sites, as well as between the Ni3 and Ni5 sites, arise from the lower-symmetry P4mm (No. 99) phase, in which

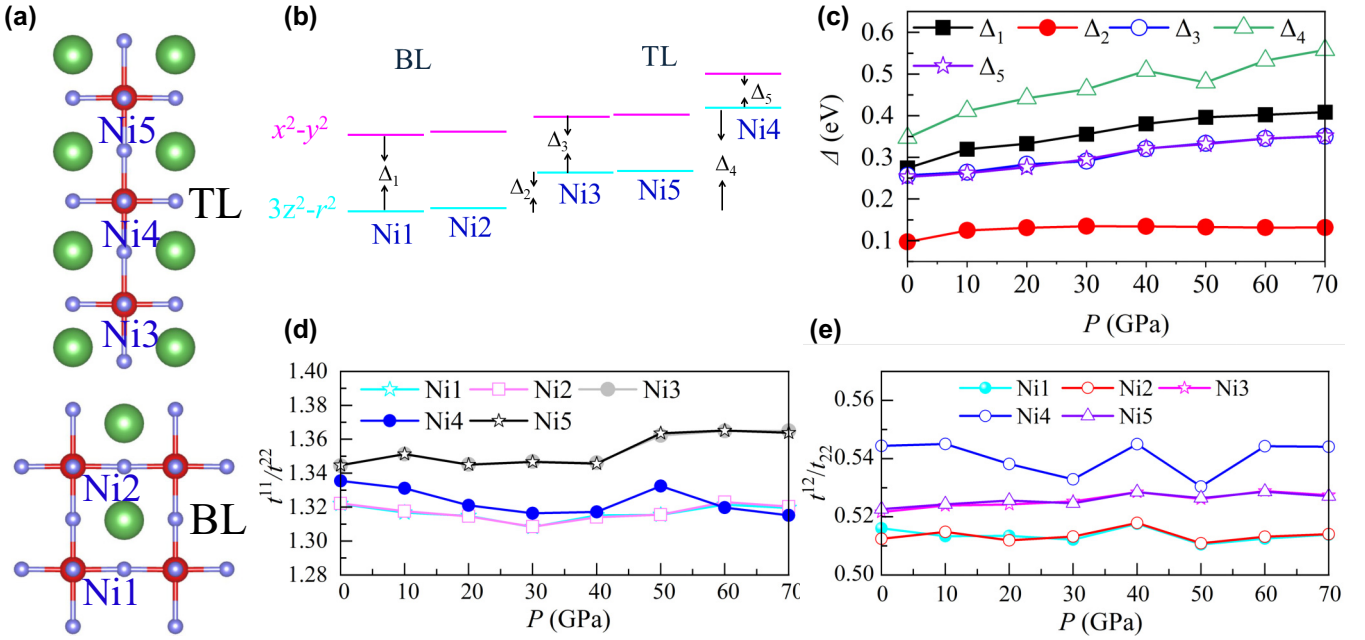


FIG. 3. (a) Schematic crystal structure of the BL-TL stacking of La₇Ni₅O₁₇, where the different Ni sites are marked. (b) Crude sketches of the crystal-field splitting of the *e_g* orbitals for different Ni sites. (c) Crystal-field splitting energies as a function of pressure. (d) Ratio of hoppings t_z^{11}/t_x^{22} , and (e) ratio of hoppings t_x^{12}/t_y^{22} , vs pressure. The $\gamma = 1$ and $\gamma = 2$ orbitals correspond to the $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals, respectively. Here, all the results are obtained in the P4mm phase.

the Ni-O bonds connecting the upper and lower Ni layers are slightly different in the BL or TL blocks (see Supplemental Material III for more details [64]).

D. Pairing symmetry under pressure

Next, we discuss the possibility of superconductivity under pressure in the La₇Ni₅O₁₇ system of our focus. To study the superconducting pairing tendencies in this BL-TL stacking La₇Ni₅O₁₇, we have used multiorbital RPA calculations for the tight-binding model in Eq. (3) including the BL and TL sublattices by using hopping and crystal-field splittings obtained for 30 GPa. In the previous study of BL nickelate systems [69], pairing was found to be potentially sensitive to the real-space hopping cutoff. In this work, we include long-range hoppings for all Ni-Ni distances less than 10 Å. Here, we have used $U = 0.8$ eV, $U' = U/2$, $J = J' = U/4$, as in previous literature to facilitate comparison and study trends. $U = 0.8$ eV was selected to obtain a sizable λ , while still avoiding a spin-density-wave instability.

In the RPA, a spin-density-wave instability will always occur with increasing U . For the La₃Ni₃O₇ and La₄Ni₃O₁₀ cases [19,30], $U = 0.95$ eV could be used because for these systems the critical U at which the spin-density-wave occurs is higher than for La₇Ni₅O₁₇. However, in La₇Ni₅O₁₇, $U = 0.95$ eV is already beyond the spin-density-wave instability. As shown in the DFT calculations (see Fig. S3), the small pocket closes along the z axis, whereas in our two-orbital model, this pocket remains open. Thus, investigating this interesting paradox within a full three-dimensional calculation would be very interesting. This discrepancy may suggest that the two-orbital model is insufficient to resolve this particular issue. Addressing this topic would require adopting a

more realistic five-orbital model. However, implementing a five-orbital model across five layers effectively results in a 25-orbital system, which is computationally demanding for pairing in RPA calculations and beyond our current resources. Thus, this is a valuable direction and we leave this issue to further study.

As shown in Fig. 4(a), we find that the leading eigenvector $g_0(\mathbf{k})$ of the gap Eq. (2) has a sign-switched s^\pm structure and eigenvalue $\lambda_s = 0.2$ [70]. This calculation was performed for the model in Eq. (3) for the BL-TL stacking La₇Ni₅O₁₇ with overall filling $n = 7.0$ at 30 GPa and a temperature $T = 0.01$ eV. The gap is largest on the inner Γ pocket σ and the order parameter has opposite sign on the small M-centered pocket γ_0 . We find that this leading instability is well separated from the next leading gap structures that have an order of magnitude smaller eigenvalues. For $T = 0.01$ eV, the pairing strength $\lambda_s = 0.20$ is already larger than the pairing strength $\lambda_s = 0.14$ we obtained for the bilayer 327-LNO at 25 GPa and the same interaction strength, but at lower $T = 0.002$ eV [30], suggesting that the pairing correlations in the BL-TL stacking system are stronger and could potentially lead to higher T_c than in the bilayer. In addition, we also would like to note that RPA calculations are based on a perturbative weak-coupling expansion which has its own shortcomings. Our predictions are obtained at the weak-coupling perturbative RPA level and in this framework the system enters a spin-density-wave state beyond a critical U .

The gap distribution features of the BL-TL stacking La₇Ni₅O₁₇ [see Fig. 4(a)] are close to that in the TL stacking La₄Ni₃O₁₀ [19], but the pairing strength λ is strongly enhanced in La₇Ni₅O₁₇. The enhanced pairing strength is due to the better nesting between the inner σ hole pocket and the γ_0 pocket in the BL-TL stacking La₇Ni₅O₁₇. These pockets

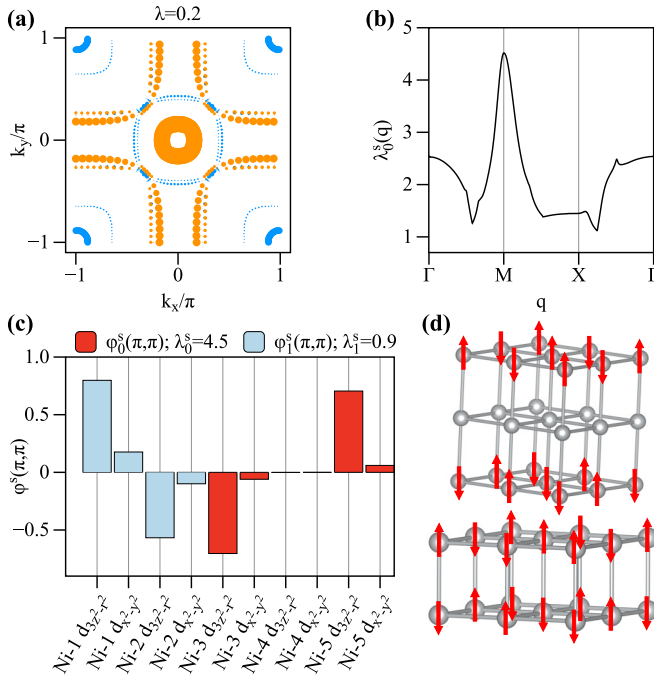


FIG. 4. (a) The RPA calculated leading superconducting singlet gap structure $g(\mathbf{k})$ for momenta \mathbf{k} on the FS of $\text{La}_7\text{Ni}_5\text{O}_{17}$ with pairing strength $\lambda = 0.2$ at 30 GPa for $n = 7.0$. The sign of $g_\alpha(\mathbf{k})$ is indicated by the color (orange = positive, blue = negative), and its amplitude by the point sizes. Here we used Coulomb parameters $U = 0.8$ eV, $U' = U/2$, and $J = J' = U/4$, and the calculation was performed for a temperature of $T = 0.01$ eV. (b) Leading eigenvalue $\lambda_0^s(\mathbf{q})$ of the spin susceptibility tensor $\chi_{\ell_1 \ell_1 \ell_2 \ell_2}^s(\mathbf{q})$, where ℓ_i are the ten Ni- d orbitals included in the model, along a high-symmetry path in the Brillouin zone. (c) Leading and subleading eigenvectors $\varphi_\alpha^s(\pi, \pi)$ of $\chi_{\ell_1 \ell_1 \ell_2 \ell_2}^s(\mathbf{q})$ for its maximum at $\mathbf{q} = (\pi, \pi)$ with corresponding eigenvalues $\lambda_0^s = 4.5$ and $\lambda_1^s = 0.9$, respectively. Here, the red bars are only finite on Ni3 and Ni5 sites, and the blue bars are only finite on Ni1 and Ni2 sites. (d) Schematic of the magnetic structure of the BL-TL $\text{La}_7\text{Ni}_5\text{O}_{17}$, where spin-up and spin-down are marked by red arrows. In the middle layer of the TL sublattice, the spin is zero.

are very well nested in terms of shape. Moreover, the Fermi velocities are also small because *the pockets are shallow*. This means that the density of states is high. Furthermore, both pockets are completely $d_{3z^2-r^2}$, so the scattering between them involves only intra-orbital scattering, which is always much stronger than interorbital scattering. As discussed in the $\text{La}_4\text{Ni}_3\text{O}_{10}$ case [19], the pairing strength can be significantly enhanced by electronic doping due to better nesting behaviors. In the BL-TL stacking case, the $d_{3z^2-r^2}$ orbital has different electronic densities in both the two following cases: 2.181 electrons (~ 0.727 per Ni site) in trilayer sublattice in the 7517 material and 2.085 electrons (~ 0.695 per Ni site) in $\text{La}_4\text{Ni}_3\text{O}_{10}$. Compared to the $\text{La}_4\text{Ni}_3\text{O}_{10}$ cases, the $d_{3z^2-r^2}$ orbital is self-doped in the trilayer sublattice in the BL-TL stacking $\text{La}_7\text{Ni}_5\text{O}_{17}$, thus the pairing strength is stronger in the $\text{La}_7\text{Ni}_5\text{O}_{17}$ system. However, in the BL $\text{La}_3\text{Ni}_2\text{O}_7$ system [30], the nesting is between the γ_0 pocket and the β sheet, which also involves the $d_{x^2-y^2}$ orbital. Thus, there is interorbital scattering involved in the process, which is weaker than

the interlayer scattering of intra $d_{3z^2-r^2}$ orbitals. Therefore, the pairing strength λ is much greater in $\text{La}_3\text{Ni}_2\text{O}_7$ than in $\text{La}_4\text{Ni}_3\text{O}_{10}$.

For comparison, we do not find any singlet pairing solution with sizable λ for the system at 0 GPa, suggesting the importance of pressure effects for superconductivity. Moreover, the FSs of other pressures look quite similar to the 30 GPa in the whole pressure range (see Supplemental Material V [64]) for which the high-symmetry phase is stable (up to 70 GPa). Thus, it is reasonable to expect that superconductivity can exist over a broad pressure range, considering previous studies of nickelate superconductors [1].

E. Magnetic correlations under pressure

Finally, to study the spin fluctuations driving the leading pairing instability in the BL-TL stacking $\text{La}_7\text{Ni}_5\text{O}_{17}$ system, we analyze the RPA enhanced spin susceptibility tensor $\chi(\mathbf{q}, \omega = 0)$ that is obtained from the Lindhart function tensor $\chi_0(\mathbf{q})$ as

$$\chi(\mathbf{q}) = \chi_0(\mathbf{q})[1 - \mathcal{U}\chi_0(\mathbf{q})]^{-1}. \quad (4)$$

Here, all the quantities are rank-four tensors in the orbital indices $\ell_1, \ell_2, \ell_3, \ell_4$ and \mathcal{U} is a tensor involving the interaction parameters [60]. The physical spin susceptibility is obtained by summing the pairwise diagonal $\chi_{\ell_1 \ell_1 \ell_2 \ell_2}(\mathbf{q})$ over ℓ_1, ℓ_2 .

Figure 4(b) plots the leading eigenvalue $\lambda_0^s(\mathbf{q})$ of $\chi_{\ell_1 \ell_1 \ell_2 \ell_2}(\mathbf{q})$ for momenta \mathbf{q} along high-symmetry directions in the Brillouin zone. The strong peak at $\mathbf{q} = (\pi, \pi)$ (M-point) indicates strong in-plane antiferromagnetic correlations. Focusing on the corresponding eigenvector $\varphi_0^s(\mathbf{q})$ (red bars in panel c) for the maximum at $\mathbf{q} = (\pi, \pi)$, one sees the antiferromagnetic (π, π) peak mainly arises from scattering between the Ni-3 and Ni-5 $d_{3z^2-r^2}$ states in the TL subsystem. The magnetic correlations are antiferromagnetic between the top and bottom layer, due to the opposite sign between the Ni-3 and Ni-5 sites, while the middle layer does not contribute to the (π, π) correlations. To assess the correlations in the bilayer subsystem, we also plot the subleading eigenvector $\varphi_1^s(\pi, \pi)$ [blue bars in Fig. 4(c)]. This eigenvector has opposite contributions from the bottom (Ni-1) and top (Ni-2) $d_{3z^2-r^2}$ states and a much smaller eigenvalue $\lambda_1^s = 0.9$ than the leading eigenvector ($\lambda_0^s = 4.5$), indicating much weaker antiferromagnetic correlations in the BL subsystem. The resulting magnetic correlations are illustrated in Fig. 4(d), showing that the top and bottom layers are antiferromagnetically correlated both in-plane and between the planes in both the BL and TL sublattices, while the middle layer of the TL sublattice has zero spin density.

Previously, for the pure BL $\text{La}_3\text{Ni}_2\text{O}_7$ system, we found that the peak in the magnetic susceptibility was near $q = (\pi, 0)$ or $(0, \pi)$ rather than at (π, π) . This difference can be intuitively understood. Compared to the pure BL stacking case with orbital site occupations of 0.9 for $d_{3z^2-r^2}$ and 0.6 for $d_{x^2-y^2}$, the electronic densities are 0.85 and 0.65 per site for the $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals in the BL sublattice of the BL-TL stacking $\text{La}_7\text{Ni}_5\text{O}_{17}$, respectively. Thus, it is reasonable to expect that the ferromagnetic tendency along one in-plane direction due to the recently proposed half-empty mechanism [71–73] should be reduced, leading to the G-AFM

state despite the filling of 1.5 electrons per site in the BL sublattice.

IV. CONCLUSION

In summary, we have systematically studied the alternating BL-TL stacking nickelate superlattice $\text{La}_7\text{Ni}_5\text{O}_{17}$ ($\text{La}_3\text{Ni}_2\text{O}_7/\text{La}_4\text{Ni}_3\text{O}_{10}$) by using DFT and RPA calculations. Similarly to other RP nickelates, the high-symmetry phase of $\text{La}_7\text{Ni}_5\text{O}_{17}$ without the tilting of oxygen octahedra is not stable at ambient conditions but becomes stable under pressure. The electronic structure of this BL-TL stacking $\text{La}_7\text{Ni}_5\text{O}_{17}$ resemble the combinations of BL and TL nickelates. Based on RPA calculations, we obtain a leading s^\pm pairing state with sizable pairing strength λ similar to that found for the pure BL nickelates. Considering previous theoretical work on nickelate superconductors, we therefore predict that superconductivity should also be expected to exist in $\text{La}_7\text{Ni}_5\text{O}_{17}$ for a broad pressure region. Moreover, we find that the dominant magnetic fluctuations driving this pairing state have antiferromagnetic structure both in-plane and between the planes of the top and bottom TL and BL sublattices, while the middle TL layer is magnetically decoupled. However, the antiferromagnetic correlations are much weaker in the BL sublattice, compared to that in the TL sublattice. To obtain this alternating BL-TL stacking nickelate superlattice $\text{La}_7\text{Ni}_5\text{O}_{17}$, a possible way is to grow a sample layer-by-layer by using molecular beam epitaxy growth, as obtained in SL-BL stacking iridate $\text{Sr}_2\text{IrO}_4/\text{Sr}_3\text{Ir}_2\text{O}_7$ [39,40]. Furthermore, by controlling experimental conditions, the single-crystal sample of the SL-TL $\text{La}_3\text{Ni}_2\text{O}_7$ ($\text{La}_2\text{NiO}_4/\text{La}_4\text{Ni}_3\text{O}_{10}$) [44–47] or SL-BL $\text{La}_5\text{Ni}_3\text{O}_{11}$ ($\text{La}_2\text{NiO}_4/\text{La}_3\text{Ni}_2\text{O}_7$) [48] nickelates have been also achieved already, showing that this procedure is

realistic. Thus, our results provide clear predictions for future experiments on this specific material and other these promising Ni-oxide superconductors.

Note added in proof. During the review process of our present work, we became aware of an independent study [74] that also proposed the “bilayer–trilayer” stacking sequence in Ruddlesden–Popper nickelate as a promising candidate for high- T_c superconductivity, albeit using different approaches.

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DATA AVAILABILITY

The dataset of the main findings of this study is openly available in the Zenodo Repository [76]. In addition, the hopping and crystal field parameters for our Tight-binding and RPA calculations are available in a separate file of the Supplementary Materials and Zenodo Repository [76] for reproducing our results, as well as the crystal structural files for the DFT calculations. The *Ab initio* calculations are done with the code VASP [77]. Simulation RPA codes are available at [78].

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