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Prediction of s^{\pm} -wave superconductivity enhanced by electronic doping in trilayer nickelates La₄Ni₃O₁₀ under pressure

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Motivated by the recently reported signatures of superconductivity in trilayer La₄Ni₃O₁₀ under pressure, we comprehensively study this system using *ab initio* and random-phase approximation techniques. Without electronic interactions, the Ni $d_{3z^2-r^2}$ orbitals show a bonding-antibonding and nonbonding splitting behavior via the O p_z orbital inducing a "trimer" lattice in La₄Ni₃O₁₀, analogous to the dimers of La₃Ni₂O₇. The Fermi surface consists of three electron-sheets with mixed e_g orbitals, and a hole and an electron pocket made up of the $d_{3z^2-r^2}$ orbital, suggesting a Ni two-orbital minimum model. In addition, we find that superconducting pairing is induced in the s_{\pm} -wave channel due to partial nesting between the $\mathbf{M}=(\pi,\pi)$ centered pockets and portions of the Fermi surface centered at the $\Gamma=(0,0)$ point. With changing electronic density n, the s^{\pm} instability remains leading and its pairing strength shows a dome-like behavior with a maximum around n = 4.2(~ 6.7% electron doping). The superconducting instability disappears at the same electronic density of La₃Ni₂O₇ does not originate from trilayer- and single-layer structure. Furthermore, we predict an interesting spin-density-wave state in La₄Ni₃O₁₀ with a in-plane (π , π) order and antiferromagnetic coupling bewteen the top and bottom Ni layers, while the middle layer has spin zero.

Introduction.– The discovery of superconductivity in the bilayer Ruddlesden-Popper (RP) perovskite La₃Ni₂O₇ (327-LNO) with a $d^{7.5}$ configuration under high pressure [1] opened a remarkable platform for the study of nickelate-based superconductors [2–8, 10–26]. By increasing pressure, 327-LNO transforms from the Amam to the Fmmm structure, the latter without tilting of oxygen octahedra [1]. Superconductivity was reported in a broad pressure range from 14 to 43.5 Gpa in the Fmmm phase, with the transition temperature $T_c \sim 80$ K [1].

To explore superconductivity in other RP layered nickelates, both theoretical and experimental studies have expanded to single-layer La_2NiO_4 and trilayer $La_4Ni_3O_{10}$ (4310-LNO) systems [6, 27–33], but no superconductivity was found at ambient conditions. In addition, superconductivity was absent also in La_2NiO_4 under pressure [6].

However, very recently, signatures of superconductivity were also reported in another RP perovskite nickelate $La_4Ni_3O_{10}$ (4310-LNO), with T_c about 20 - 30 K above 15 GPa [34-39]. Without pressure, 4310-LNO has a monoclinic P2₁-c structure 14) [28, 32], where the strongly distorted (No. corner-sharing NiO₆ octahedra form a trilayer sublattice stacking along the c-axis (see Fig. 1(a)). Under the influence of hydrostatic pressure, 4310-LNO also shows a structural phase transition from $P2_1$ -c symmetry to a high-symmetry I4/mmm phase without the tilting of oxygen octahedra, similarly to 327-LNO [37].

Thus, considering these developments on 327-LNO and 4310-LNO, several interesting questions naturally arise:

What are the similarities and differences between the bilayer 327-LNO and trilayer 4310-LNO nickelates under pressure? What is the superconducting pairing channel in 4310-LNO? How does superconductivity in 4310-LNO evolve under electron doping?

Trimer vs dimer – Similar to the "dimer" physics in the bilayer lattice [14, 40], the "trimer" physics can also be obtained in the trilayer lattice because the intraorbital coupling is strong and the coupling in between trilayers is weak. Specifically, the $d_{3z^2-r^2}$ orbital would split into antibonding, nonbonding, and bonding states in the trilayer 4310-LNO, as shown in Fig. 1(b). Because the $d_{x^2-y^2}$ orbital is lying in the NiO₆ plane, it remains decoupled among planes, not participating in the formation of the antibonding-nonbonding splitting along the z-axis, resulting in an orbital-selective behavior [41, 42].

In 4310-LNO, the total electronic density of Ni is n = 7.33, corresponding to Ni^{2.67} on average, leading to partially filled e_g orbitals and three fully occupied t_{2g} states. In this case, the $d_{3z^2-r^2}$ orbital is nearly half-filled, and the $d_{x^2-y^2}$ orbital is close to one-third occupied. In 327-LNO, the system is "self-doped" caused by the in-plane interorbital hopping between the e_g states, leading to a non-integer electronic population of both orbitals [15]. Thus, this behavior of the e_g orbitals is also expected in the trilayer 4310-LNO.

To better understand these broad issues, using first-principles density functional theory (DFT) [1–4], we have studied the trilayer 4310-LNO in detail. Without pressure, our DFT results find that the P2₁-c phase has an energy lower by about -48.26 meV/Ni than



FIG. 1. (a) Schematic crystal structure of the conventional cells of trilayer 4310-LNO for the P2₁-c and I4/mmmm phases without pressure and at high pressure, respectively (green = La; gray = Ni; red = O). Note that the local z-axis is perpendicular to the NiO₆ plane towards the top O atom, while the local x- or y-axis are along the in-plane Ni-O bond directions. All crystal structures were visualized using the VESTA code [5]. (b) Sketches of the $d_{3z^2-r^2}$ orbital in the trilayer and bilayer nickelates with two electrons. (c) Phonon spectrum of the I4/mmm phase of trilayer 4310-LNO at 30 GPa.

the I4/mmmm phase. By introducing pressure, the monoclinic distortion is gradually suppressed, leading to a high-symmetry I4/mmmm phase at high pressure. Furthermore, the phononic calculations indicate that the I4/mmm phase of 4310-LNO is stable without any imaginary frequency at 30 GPa [48], as displayed in Fig. 1(c), by using the density functional perturbation theory approach [7, 8], analyzed by the PHONONPY software [9, 10]. Thus, the pressure effect is quite similar in 4310-LNO and 327-LNO [15], where the spontaneous suppression of octahedral distortion under pressure leads to a phase transition from low to high symmetry.

Electronic structures of LNO – Near the Fermi level, the main contributions to the electronic density of states are from the Ni 3d orbitals hybridized with the O porbitals with a large charge-transfer energy $\Delta = \varepsilon_d - \varepsilon_p$, sharing the common character of other nickelates [15]. Using the maximally localized Wannier functions [6] by fitting DFT and Wannier bands of the non-magnetic state of the I4/mmm phase of 4310-LNO at 30 GPa, we find that both e_q orbitals of the outer layer Ni have lower onsite energies than that in inner layer Ni. The nearest-neighbor (NN) hopping of the $d_{3z^2-r^2}$ orbital along the z-axis for 4310-LNO ($\sim 0.694 \text{ eV}$) is slightly larger than that in 327-LNO ($\sim 0.640 \text{ eV}$) [8, 14]. In the Ni plane, the largest hopping is the intraorbital hopping of the $d_{3x^2-y^2}$ orbital (~ 0.519/0.511 eV for inner and outer layer Ni). Furthermore, we also obtain a large *interorbital* hopping between $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals in 4310-LNO, caused by the ligand "bridge" of the in-plane O p_x or p_y orbitals connecting those two

orbitals.

Next, we constructed a six-band e_g -orbital tight binding (TB) model on the trilayer lattice for the I4/mmmm phase of 4310-LNO at 30 GPa with overall filling n = 4 by using the NN and next nearest-neighbor (NNN) hoppings. The kinetic hopping component is:

$$H_k = \sum_{\substack{i\sigma\\\vec{\alpha}\gamma\gamma'}} t^{\vec{\alpha}}_{\gamma\gamma'}(c^{\dagger}_{i\sigma\gamma}c_{i+\vec{\alpha}\sigma\gamma'} + H.c.) + \sum_{i\gamma\sigma} \Delta_{\gamma}n_{i\gamma\sigma}.$$
 (1)

The first term represents the hopping of an electron from orbital γ' at site $i + \vec{\alpha}$ to orbital γ at site $i. c^{\dagger}_{i\sigma\gamma}(c_{i\sigma\gamma})$ 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 $\vec{\alpha}$ are along the three trilayer-lattice directions, defining different neighbors of hoppings (see more details in Supplementary Material [55]).

As shown in Fig. 2(a), $d_{3z^2-r^2}$ displays the bonding-antibonding, and nonbonding splitting behavior, while the $d_{x^2-y^2}$ orbital remains decoupled among planes, in agreement with our discussion in the previous section. Compared with the bilayer 327-LNO (see Fig. 2(b)), the bandwidth of the e_q orbitals increases by about $\sim 8\%$. The calculated electronic densities are 2.085 and 1.915 for the $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals (0.695 and 0.638 per site), respectively, in the trilayer TB model of 4310-LNO, while they are 1.682 and 1.318 for 327-LNO (0.814 and 0.659 per site). Considering the average valences of the Ni ions in 4310-LNO and 327-LNO, holes are favored to enter the $d_{3z^2-r^2}$ orbitals.



FIG. 2. TB band structures and FS's for (a,c) trilayer 4310-LNO, and (b,d) bilayer 327-LNO, respectively. (a,c) The six-band e_g orbital TB model was considered with three NN and NNN hoppings in a trilayer lattice for the overall filling n = 4 (4/3 electrons per site). (b,d) The four-band e_g orbital TB model was considered in a bilayer lattice for the overall filling n = 3 (1.5 electrons per site), where the hoppings used from a previous study [8].

Five bands are crossing the Fermi level in 4310-LNO at high pressure, contributing to the Fermi surface (FS) as displayed in Fig. 2(c), namely, bands α , β_1 β_2 , γ and δ , respectively. Similarly to the FS of 327-LNO (see Fig. 2(d), the hole pocket γ is made up by the $d_{3z^2-r^2}$ orbital, while the three electron sheets α , β_1 and β_2 originate from mixed $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals. In addition, an electron pocket δ made up by the nonbonding $d_{3z^2-r^2}$ orbital is obtained for 4310-LNO.

RPA pairing tendencies – Next, we have used multi-orbital random phase approximation (RPA) calculations to assess the bilayer TB models for their superconducting behavior. The RPA is based on a perturbative weak-coupling expansion in the Coulomb interaction [11–13, 59]. The pairing strength λ_{α} for channel α and the corresponding gap structure $g_{\alpha}(\mathbf{k})$ are obtained by solving an eigenvalue problem of the form

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

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

By solving the eigenvalue problem in Eq. (S6) for the RPA pairing interaction of 4310-LNO (n = 4.0), we find that the s^{\pm} gap structure is the leading pairing symmetry caused by spin-fluctuation. The gap is large and switches sign between the small electron pocket at Γ and the small hole pocket at M, which are separated by (π , π) (see Fig. 3(a)). The calculated pairing strength



FIG. 3. (a) The calculated RPA superconducting gap structure $g_{\alpha}(\mathbf{k})$ for momenta \mathbf{k} on the FS of 4310-LNO with s_{\pm} -wave symmetry at n = 4.0. The sign of the gap is indicated by the colors (red = positive, blue = negative), and the gap amplitude by its intensity. (b) The RPA calculated pairing strength λ for the s^{\pm} , $d_{x^2-y^2}$ and d_{xy} instabilities versus electronic densities for the trilayer model. (c-d) The calculated RPA (c) superconducting gap structure $g_{\alpha}(\mathbf{k})$ with s_{\pm} -wave symmetry and (d) the pairing interaction $\Gamma(k, k_0)$ with k_0 indicated by the black diamond for n = 4.2. The RPA calculations used U = 0.95, U' = U/2 and J = J' = U/4 in units of eV (J is the Hund coupling, J' the pair hopping), with NN and NNN hoppings from the I4/mmm phase of 4310-LNO.

 λ (~ 0.202) of the s^{\pm} -wave gap structure is smaller than that in 327-LNO at the same U = 0.95 (~ 0.39). Since in our RPA treatment, the pairing strength λ enters exponentially in the equation for T_c , i.e. $T_c = \omega_0 e^{-1/\lambda}$ with a spin-fluctuation cut-off frequency ω_0 , this comparison suggests a lower T_c for 4310-LNO to the extent that ω_0 is similar in both systems.

To understand doping effects, we also studied the dependence of the RPA pairing strength λ on the electron density n in the trilayer model, as shown in Fig. 3(b). One sees that the s^{\pm} state remains leading over the entire density range we have studied, while $d_{x^2-y^2}$ and d_{xy} states are subleading. The pairing strengths λ for all three states show a dome-like doping dependence with a peak at n = 4.2 (~ 6.7% electron doping). At hole doping near n = 3.6 or electron doping near n = 4.5, the calculated RPA pairing strength λ becomes negligible, indicating that a superconducting instability may be absent beyond the central dome. The leading s^{\pm} gap for the optimal density n = 4.2 is shown in Fig. 3(c), and the corresponding pairing interaction $\Gamma(\mathbf{k} - \mathbf{k}_0)$ for this case in Fig. 3(d). Here, \mathbf{k}_0 is fixed at the Fermi momentum on the inner Γ -centerd pocket indicated by the black diamond, and **k** runs along all the Fermi surface points. One sees that $\Gamma(\mathbf{k} - \mathbf{k}_0)$ is large and peaked for

a momentum transfer of $\mathbf{q} \sim (\pi, \pi)$ that connects states on the inner Γ pocket and on the *M*-centered pocket. This pair scattering drives the leading s^{\pm} state which has a large gap with opposite signs on these Fermi surface sheets as seen in Fig. 3(c).

In addition, we find that the superconducting pairing strength at n = 4.5, corresponding to 1.5 electron per site, is almost zero. For this case we find that the hole γ pocket is absent, as shown in Fig. 4(a). In addition, very recently, several groups independently reported a new phase of La₃Ni₂O₇ with alternating monolayer and trilayer structures [60-62]. Note that the electronic density of the e_q orbitals of La₃Ni₂O₇ is also 1.5 per Ni. We therefore calculated the band structure of the P4/mmm phase of this new phase of $La_3Ni_2O_7$ by using the experimental structure under high pressure [63]. Figure 4(b) indicates that the γ pocket of the $d_{3z^2-r^2}$ orbital contributed by the trilayer structure is absent in this new phase of $La_3Ni_2O_7$. Our RPA results show that the superconductivity instability disappears at n = 4.5 correlated with the absence of the γ pocket while a superconductivity instability was obtained for the 327-LNO with same electronic density [15]. Thus, our results suggest that the previously discovered superconductivity in La₃Ni₂O₇ does not originate from an alternating monolayer and trilayer stacking structure.

Magnetic tendency – We also studied the static RPA enhanced spin susceptibility $\chi'(\mathbf{q}, \omega = 0)$ that is obtained from the Lindhart function $\chi_0(\mathbf{q})$ as

$$\chi(\mathbf{q}) = \chi_0(\mathbf{q})[1 - \mathcal{U}\chi_0(\mathbf{q})]^{-1}.$$
(3)

 $\chi_0(\mathbf{q})$ is an orbital-dependent susceptibility tensor and \mathcal{U} is a tensor involving the interaction parameters [12].

 $\chi(\mathbf{q})$ for n = 4.2 presents a strong peak at $\mathbf{q} = (\pi, \pi, \pi/2)$, as displayed in Fig. 4(c). This spin density wave fluctuation corresponds to a G-type antiferromagnetic state in which the top and bottom layers are coupled antiferromagnetically both in-plane and between the planes and where the middle layer has zero spin density.

To confirm the RPA results, we also studied the magnetic properties by using the DFT+U+Jformalism within the Liechtenstein formulation with a double-counting term to deal with the onsite Coulomb interactions [16], where U is fixed at 4 eV, following recent DFT studies of niceklelate [10, 15]. Here, we considered several possible magnetic structures of the Ni trilayer spins with spin-zero in the middle layer as input: (1) A-AFM or ferromagnetic (FM) with in-plane wavevector (0, 0) where top and bottom are AFM or FM coupled; (2) G-AFM or C-AFM with in-plane wavevector (π, π) where top and bottom layers are AFM or FM coupled; (3) Stripe-AFM or stripe-FM with in-plane wavevector $(\pi, 0)$, where the top and bottom layers are AFM or FM coupled.

As displayed in Fig. 4(d), the G-AFM state has the lowest energy when J < 1 eV among all the candidates.



FIG. 4. (a) TB FS for n = 4.5 with the absence of the γ pocket. (b) The DFT band structure for La₃Ni₂O₇ with alternating monolayer (ML) and trilayer (TL) structures using the experimental structure at high pressure. (c) The RPA calculated static spin susceptibility $\chi'(\mathbf{q}, \omega = 0)$ versus q_x , q_y for $q_z = \pi/2$ for the two-orbital trilayer TB model for n = 4.2. (d) The DFT+U+J calculated energies for different J's of different magnetic configurations at U = 4 eV.

In addition, we also considered the cases with nonzero spin in the middle layer. Those spin states were found to have higher energy than the cases with spin zero in the middle layer. Considering the previously calculated J for other layered nickelates (~ 0.61 - 0.68 eV) [10, 65, 66], our DFT+U+J calculations also found the in-plane (π , π) order with AFM coupling between top and bottom Ni layers, while the middle layer has spin zero, in agreement with the RPA calculations.

Conclusions.– In summary, we have unveiled clear similarities and differences between the trilayer nickelate and the recently much-discussed bilayer 327-LNO nickelates. (1) Similar to 327-LNO, pressure spontaneously suppresses the octahedral distortion in the trilayer, leading to a phase transition from a lowto a high-symmetry phase in 4310-LNO as well as to a large in-plane interorbital hopping between the e_g states. (2) The Ni $d_{3z^2-r^2}$ orbital shows a bonding-antibonding splitting, but also has a nonbonding state in 4310-LNO due to the geometry of the Ni trilayer lattice. (3) The 4310-LNO Fermi surface contains three electron sheets formed by mixed e_q orbitals, and a hole and an electron pocket made of the $d_{3z^2-r^2}$ orbital, establishing that a minimum two e_q orbital model per Ni is needed. (4) We also found a leading spin-fluctuation driven s^{\pm} -wave pairing state in 4310-LNO, where the gap is largest and has opposite signs on the small electron pocket at Γ and the small hole pocket at M, which are separated by (π, π) . (5) Under variation of the electron density n, the pairing strength displays dome-like behavior and is strongly enhanced for n = 4.2 before it becomes negligibly weak at n = 4.5, correlated with the disappearance of the *M*-centered γ pocket. (6)We also found an interesting spin density wave state with in-plane (π, π) spin order, zero spin density in the middle layer, and AFM coupling between the top and bottom layers in 4310-LNO.

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Supplementary Material of "Prediction of s^{\pm} -wave pairing superconductivity enhanced by electronic doping in trilayer nickelates La₄Ni₃O₁₀ under pressure"

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I. METHOD

software in the primitive unit cell [9, 10].

A. DFT Method

In this work, first-principles density functional theory (DFT) calculations were implemented based on the Vienna *ab initio* simulation package (VASP) code by using the projector augmented wave (PAW) method [1-3]. In addition, the electronic correlations were considered by the generalized gradient approximation (GGA) and the Perdew-Burke-Ernzerhof (PBE) exchange potential [4]. Here, the plane-wave cutoff energy was set as 550 eV and the k-point mesh was appropriately modified for different structural phases to render the k-point densities approximately the same in reciprocal space (i.e., $16 \times 16 \times 2$ for the conventional cell of the I4/mmm phase of $La_4Ni_3O_{10}$). Furthermore, the atomic positions and crystal lattices were fully relaxed until the Hellman-Feynman force on each atom was smaller than 0.001 eV/Å. In our present work, we only considered two pressures: 0 GPa and 30 GPa. Note that the purpose of this work is to study the high-pressure I4/mmm phase of La₄Ni₃O₁₀. At 30 GPa, the monoclinic distortion is already fully suppressed, leading to the high-symmetry I4/mmm phase at 30 GPa. The evolution of possible phase transitios under pressure is not the scope of our present work, thus we leave the issue of adding more pressures to future work. All the crystal structures were visualized with the VESTA code [5].

In addition to the standard DFT calculation discussed thus far, the maximally localized Wannier functions (MLWFs) method was employed to fit Ni 3d bands by using the WANNIER90 packages [6]. Here, we constructed five disentangled Wannier functions for the Ni five 3d orbitals of the I4/mmmm phase of the trilayer La₄Ni₃O₁₀ at 30 GPa, with results that are fitted very well with the DFT band in the low-energy region. Then, we obtained the crystal-field splitting Δ and hopping matrix $t_{\gamma\gamma'}$ of the five Ni 3d orbital. For the phonon spectrum of the I4/mmm phase of the trilayer La₄Ni₃O₁₀, a 2 × 2 × 1 conventional cell structure of the I4/mmm phase was used in the phonon calculation, by using the density functional perturbation theory approach [7, 8], analyzed by the PHONONPY

B. TB method

Furthermore, a six-band low-energy trilayer tight-binding (TB) model was introduced to calculate the band structures and Fermi surface. Filling n = 4 was used, corresponding to 4/3 electrons per Ni site. The kinetic hopping component of the Hamiltonian is:

$$H_k = \sum_{\substack{i\sigma\\\vec{\alpha}\gamma\gamma'}} t^{\vec{\alpha}}_{\gamma\gamma'}(c^{\dagger}_{i\sigma\gamma}c_{i+\vec{\alpha}\sigma\gamma'} + H.c.) + \sum_{i\gamma\sigma} \Delta_{\gamma}n_{i\gamma\sigma}.$$
 (S4)

The first term represents the hopping of an electron from orbital γ at site *i* to orbital γ' at a nearest-neighbor site $i + \vec{\alpha}$. $c_{i\sigma\gamma}^{\dagger}(c_{i\sigma\gamma})$ is the standard creation (annihilation) operator, γ and γ' represent the different orbitals, and σ is the *z*-axis spin projection. Δ_{γ} represents the crystal-field splitting of each orbital γ . The unit vectors $\vec{\alpha}$ are along the three trilayer-lattice directions, defining different site neighbors for the hoppings.

C. RPA Method

In addition, the random phase approximation (RPA) method was used to study the full model regarding its superconducting and magnetic behavior 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. The full Hamiltonian for the bilayer Hubbard model discussed here, includes the kinetic energy and interaction terms, and it is written as $H = H_{\rm k} + H_{\rm int}$.

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 inter-orbital electron-pair hopping terms (J'). Formally, it is given by:

$$H_{\text{int}} = U \sum_{i\gamma} n_{i\uparrow\gamma} n_{i\downarrow\gamma} + (U' - \frac{J}{2}) \sum_{\substack{i \\ \gamma < \gamma'}} n_{i\gamma} n_{i\gamma'}$$
$$-2J \sum_{\substack{i \\ \gamma < \gamma'}} \mathbf{S}_{i,\gamma} \cdot \mathbf{S}_{i,\gamma'} + J \sum_{\substack{i \\ \gamma < \gamma'}} (P_{i\gamma}^{\dagger} P_{i\gamma'} + H.c.), \quad (S5)$$

where the standard relation U' = U - 2J and J' = J are assumed, and $P_{i\gamma} = c_{i\downarrow\gamma} c_{i\uparrow\gamma}$. Thus, there are only two free parameters: U and J.

In the multi-orbital RPA approach [11–13], 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 inter-orbital 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

$$\int_{FS} d\mathbf{k}' \, \Gamma(\mathbf{k} - \mathbf{k}') g_{\alpha}(\mathbf{k}') = \lambda_{\alpha} g_{\alpha}(\mathbf{k}) \,, \qquad (S6)$$

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 $\chi(\mathbf{k} - \mathbf{k}')$.

II. MORE RESULTS FOR $LA_4NI_3O_{10}$ UNDER PRESSURE

Near the Fermi level, the electronic density is mainly contributed by Ni 3d states, as displayed in Fig. S1(a). The hybridization of O 2p and Ni 3d states is also quite weak in La₄Ni₃O₁₀, similar to other nickelates [14, 15]. The Ni 3d states of the I4/mmm phase of La₄Ni₃O₁₀ are mainly located in the range of energy from -3 to 3 eV, leading to a large bandwidth (~ 6 eV). In addition, the O 2p bands of La₄Ni₃O₁₀ extend over a broad range of energy from ~ -8.5 eV to ~ 3.5 eV (not shown here). Figure S1(b) indicates that there are two orbitals crossing the Fermi level for this material: Ni $d_{x^2-y^2}$ and Ni $d_{3z^2-r^2}$. In this case, this system can be regarded as a two-orbital model system.

As shown in Fig. S2, there are five bands crossing the Fermi level in La₄Ni₃O₁₀ at 30 GPa, contributing to the Fermi surface: α , β_1 , β_2 , γ and δ , respectively. The hole pocket γ is made of the $d_{3z^2-r^2}$ orbital, while the three electron sheets α , β_1 and β_2 originate from mixed $d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals. In addition, an electron pocket δ contributed by the nonbonding $d_{3z^2-r^2}$ orbital was obtained in La₄Ni₃O₁₀.

In addition, we also considered the cases with spin-nonzero in the middle layer, by using the



FIG. S1. (a) DOS near the Fermi level using the non-magnetic states for the I4/mmm phase of La₄Ni₃O₁₀ at 30 GPa. (b) Projected band structures of the I4/mmm phase of La₄Ni₃O₁₀ for the non-magnetic state at 30GPa. The Fermi level is shown with horizontal dashed lines. The weight of each orbital is represented by the intensity of the lines. The coordinates of the high-symmetry points of the Brillouin zone are $\Gamma = (0, 0, 0), X = (0, 0, 0.5), P = (0.25, 0.25, 0.25), N = (0, 0.5, 0), and M = (0.5, 0.5, -0.5).$



FIG. S2. DFT Fermi surface for the I4/mmm phase of trilayer $\rm La_4Ni_3O_{10}$ at 30 GPa.



FIG. S3. The DFT+U+J calculated energies for different J's and different magnetic configurations, all at U = 4 eV. Here, we used the A-AFM state discussed in the main text as the reference energy. This A-AFM state has the in-plane wavevector (0, 0) while top and bottom are FM coupled and the middle layer has spin zero.

DFT+U+Jformalism within the Liechtenstein formulation with the double-counting item to deal with the onsite Coulomb interactions [16], where U was fixed to be 4 eV and J was changed from 0.4 to 1 eV. Here, several possible magnetic structures of the Ni trilayer spins were considered: (1) A-AFM-TL: ferromagnetic (FM) coupling in the NiO_2 layer plane and antiferromagnetic (AFM) coupling between the Ni layers; (2) FM-TL: FM coupling along both the NiO_2 laver plane and between the Ni lavers; (3) G-AFM-TL: AFM coupling along both the NiO_2 layer plane and between the Ni layers; (4) C-AFM-TL: AFM coupling along the NiO_6 layer plane and FM coupling between the layers; (5) Stripe-AFM-TL: AFM in one of the in-plane directions and FM in the other, while the coupling along the Ni layers direction is AFM. (6) Stripe-AFM-TL: AFM in one of the in-plane directions and FM in the other, while the coupling along the Ni layers direction is FM.

As displayed in Fig. S3, the energies of those magnetic configurations are higher than the dominant spin-density-wave G-AFM state discussed in the main text that has an in-plane AFM order and AFM coupling between the top and bottom Ni layers, while the middle layer has spin zero. Here, we used the same reference energy (A-AFM) as discussed in the main text, where the A-AFM has in-plane FM coupling while top and bottom are FM coupled and the middle layer has spin zero.



FIG. S4. The hoppings used for the 326-LNO system.

III. TRILAYER LATTICE AND HOPPINGS

The crystal-field splitting between the $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbitals are 0.058 and 0.291 eV for the inner and outer layer, respectively. The crystal-field splitting between $d_{3z^2-r^2}$ of inner layer and $d_{x^2-y^2}$ of outer layer is 0.027 eV. As shown in Figure S4, we considered several nearest-neighbor and next nearest-neighbor hopping matrices in our minimum TB model with basis $\{d_{3z^2-r^2}, d_{x^2-y^2}\}$. The hopping amplitudes are (in eV):

$$t_{\vec{x}}^{inner} = \begin{bmatrix} -0.162 & 0.282\\ 0.282 & -0.519 \end{bmatrix},$$
 (S7)

$$t_{\vec{y}}^{inner} = \begin{bmatrix} -0.162 & -0.282\\ -0.282 & -0.519 \end{bmatrix},$$
 (S8)

$$t_{\vec{x}}^{outer} = \begin{bmatrix} -0.144 & 0.272\\ 0.272 & -0.511 \end{bmatrix},$$
 (S9)

$$t_{\vec{y}}^{outer} = \begin{bmatrix} -0.144 & -0.272\\ -0.272 & -0.511 \end{bmatrix},$$
 (S10)

$$t_{\vec{z}} = \begin{bmatrix} -0.694 & 0.000\\ 0.000 & -0.007 \end{bmatrix},$$
(S11)

$$t_{\vec{x}+\vec{y}}^{inner} = \begin{bmatrix} -0.020 & 0.000\\ 0.000 & 0.061 \end{bmatrix},$$
 (S12)

$$t_{\vec{x}+\vec{y}}^{outer} = \begin{bmatrix} -0.020 & 0.000\\ 0.000 & 0.065 \end{bmatrix},$$
 (S13)

$$t_{\vec{x}+\vec{z}} \begin{bmatrix} 0.033 & -0.041 \\ -0.041 & 0.002 \end{bmatrix},$$
(S14)

$$t_{\vec{y}+\vec{z}} = \begin{bmatrix} 0.033 & 0.041\\ 0.041 & 0.002 \end{bmatrix},$$
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