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## General trends of electronic structures, superconducting pairing, and magnetic correlations in the Ruddlesden-Popper nickelate $m$ -layered superconductors

### $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$

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We report a comprehensive theoretical analysis of the Ruddlesden-Popper layered nickelates  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to 6) under pressure. These materials have recently received significant attention due to the discovery of superconductivity in some nickelates under pressure. Our results suggest that, while these Ruddlesden-Popper layered nickelates display many similarities, they also show noticeable differences. One of the common features of  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  is that the electronic states near the Fermi level are mainly contributed by Ni  $3d$  orbitals, slightly hybridized with O  $2p$  orbitals. The Ni  $d_{3z^2-r^2}$  orbitals display bonding-antibonding, or bonding-antibonding-nonbonding, characteristic splittings, depending on the even or odd number of stacking layers  $m$ . In addition, the ratio of the in-plane interorbital hopping between  $d_{3z^2-r^2}$  and  $d_{x^2-y^2}$  orbitals and in-plane intraorbital hopping between  $d_{x^2-y^2}$  orbitals was found to be large in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to 6), and this ratio increases from  $m = 1$  to  $m = 6$ , suggesting that the in-plane hybridization will increase as the layer number  $m$  increases. In contrast to the dominant  $s^\pm$ -wave state driven by spin fluctuations in the bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  and trilayer  $\text{La}_4\text{Ni}_3\text{O}_{10}$ , two nearly degenerate  $d_{x^2-y^2}$ -wave and  $s^\pm$ -wave leading states were obtained in the four-layer stacking  $\text{La}_5\text{Ni}_4\text{O}_{13}$  and five-layer stacking  $\text{La}_6\text{Ni}_5\text{O}_{16}$ . The leading  $s^\pm$ -wave state was recovered in the six-layer material  $\text{La}_7\text{Ni}_6\text{O}_{19}$ . In general, at the level of the random phase approximation treatment, the superconducting transition temperature  $T_c$  decreases in stoichiometric bulk systems from the bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  to the six-layer  $\text{La}_7\text{Ni}_6\text{O}_{19}$ , despite the  $m$  dependent dominant pairing. Both in-plane and out-of-plane magnetic correlations are found to be quite complex. Within the in-plane direction, we obtained the peak of the magnetic susceptibility at  $\mathbf{q} = (0.6\pi, 0.6\pi)$  for  $\text{La}_5\text{Ni}_4\text{O}_{13}$  ( $m = 4$ ) and  $\text{La}_7\text{Ni}_6\text{O}_{19}$  ( $m = 6$ ), and at  $\mathbf{q} = (0.7\pi, 0.7\pi)$  for  $\text{La}_6\text{Ni}_5\text{O}_{16}$  ( $m = 5$ ). Along the out-of-plane direction, four layers are coupled as  $\downarrow-\uparrow-\uparrow-\downarrow$  in  $\text{La}_5\text{Ni}_4\text{O}_{13}$ , five layers are coupled as  $\uparrow-\uparrow-\downarrow-\uparrow-\uparrow$  in  $\text{La}_6\text{Ni}_5\text{O}_{16}$ , and six layers are coupled as  $\uparrow-\downarrow-\downarrow-\uparrow-\uparrow-\downarrow$  in  $\text{La}_7\text{Ni}_6\text{O}_{19}$ .

## I. INTRODUCTION

The discovery of superconductivity in infinite-layer Sr-doped  $\text{NdNiO}_2$  ( $d^9$  configuration) films with a  $T_c$  of  $\sim 15$  K [1] started the nickel era of unconventional high-temperature superconductivity [2–9], which are reminiscent of the successful stories of the cuprates [10, 11] and iron-based superconductors [13, 14]. Following the study of infinite-layer nickelates, other  $\text{NiO}_2$  layered materials were found to superconduct, such as quintuple square-planar layered  $\text{Nd}_6\text{Ni}_5\text{O}_{12}$  ( $d^{8.8}$  configuration) with  $T_c \sim 13$  K [15]. Because their electron configuration is close to  $d^9$ , isoelectronic with  $\text{Cu}^{2+}$ , these nickelates are expected to have a  $d_{x^2-y^2}$ -wave superconducting instability [16, 17] as in the cuprates [11,

12]. However, many theoretical and experimental efforts revealed fundamental differences between square-planar layered nickelates and cuprates [6, 18–23]. Very recently, pressure studies found superconductivity in bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  [24] and trilayer  $\text{La}_4\text{Ni}_3\text{O}_{10}$  [25, 26] nickelates with corner-shared  $\text{NiO}_6$  octahedra layers. This rapidly developing new avenue for the study of nickelate superconductors has already attracted much attention in Condensed Matter Physics and Material Sciences [27–44].

$\text{La}_3\text{Ni}_2\text{O}_7$  and  $\text{La}_4\text{Ni}_3\text{O}_{10}$  belong to the Ruddlesden-Popper (RP) perovskite family  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to  $\infty$ ) involving bilayer and trilayer blocks stacking structures, respectively, as displayed in Fig. 1. In contrast to the square-planar layered nickelates  $\text{La}_{m+1}\text{Ni}_m\text{O}_{2m+2}$  ( $m = 1$  to

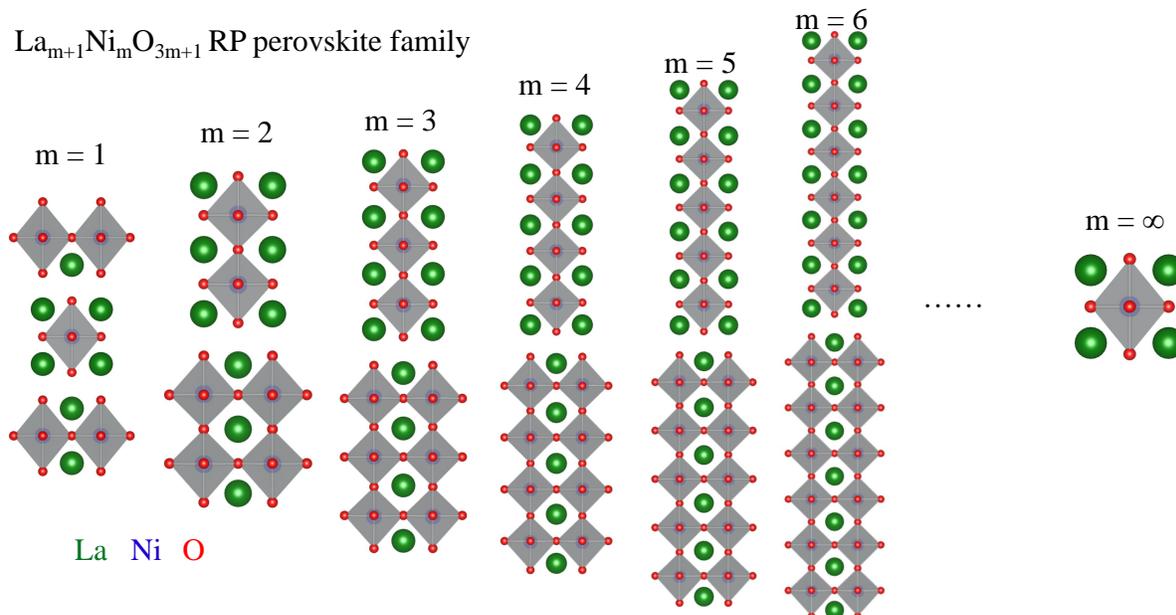


FIG. 1. Schematic crystal structures of the RP perovskite family  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  (green = La; blue = Ni; red = O). The visualization code VESTA is used [110].

$\infty$ ) [45–47], there are apical oxygens connecting the Ni layers in the RP nickelate family  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to  $\infty$ ). At ambient pressure, bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  has an  $\text{Amam}$  (No. 63) structure [48], and thus, the  $\text{NiO}_6$  octahedra are distorted. By applying hydrostatic pressure, the distortion of  $\text{NiO}_6$  octahedra is suppressed around 14 GPa [28], accompanied by a first-order structural phase transition, resulting in an  $\text{Fmmm}$  (No. 69) phase without a tilting distortion of the  $\text{NiO}_6$  octahedra [24]. By further increasing pressure, a superconducting state appears in a very broad pressure regime [24, 49]. The initial claim of superconductivity in the  $\text{La}_3\text{Ni}_2\text{O}_7$  system was based on measurements of the resistance by a four-terminal device, where the presence of zero resistance and the Meissner effect [24] was deduced by observing a sharp transition and a flat stage in resistance when using potassium bromide as the pressure-transmitting medium and a diamagnetic response in the susceptibility. Later, actual zero resistance [28, 29, 50] and Meissner effect [49] were observed by several groups, establishing the superconducting state in bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$ . The superconducting volume fraction was reported to be around 48% in their high-quality superconducting samples by using the  $ac$  magnetic susceptibility [49]. Very recently, a tetragonal  $\text{I4/mmm}$  (No. 139) phase was proposed to be the correct high-pressure structure of  $\text{La}_3\text{Ni}_2\text{O}_7$  from both theory [51] and experiment [52], instead of the  $\text{Fmmm}$  symmetry. The tilting distortion of the  $\text{NiO}_6$  octahedra is suppressed in both phases. Due to the very small distortion from  $\text{I4/mmm}$  (No. 139) to  $\text{Fmmm}$  (No. 69) in  $\text{La}_3\text{Ni}_2\text{O}_7$  [53], those two phases

are expected to display no fundamental differences, providing the same physics [54].

As in the bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$ , the  $\text{NiO}_6$  octahedra are also distorted in the trilayer nickelate  $\text{La}_4\text{Ni}_3\text{O}_{10}$  with a  $\text{P21/c}$  (No. 14) phase [55–59] at ambient pressure. Under hydrostatic pressure, the distortion of the  $\text{NiO}_6$  octahedra is suppressed, and a high-symmetry  $\text{I4/mmm}$  phase appears at around 15 GPa [25, 26, 60], once again without the tilting of oxygen octahedra. Following this structural transition, the superconducting state was observed in a broad pressure range with a  $T_c$  of about 20–30 K [25, 26, 33, 61], which is lower than in the bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  [24]. The Meissner effect was also observed in  $ac$  magnetic susceptibility experiments with superconducting volume fraction around  $\sim 80\%$  [26]. However, the single-layer RP nickelate  $\text{La}_2\text{NiO}_4$  (see Fig. 1) has never been reported to superconduct at both ambient conditions and high pressure [50].

Theoretical studies have played a crucial role in providing insights into the nature of superconducting states in nickelates [62–94]. Density functional theory (DFT) calculations suggest that both  $\text{La}_3\text{Ni}_2\text{O}_7$  and  $\text{La}_4\text{Ni}_3\text{O}_{10}$  nickelates systems can be described using a two- $e_g$ -orbital model in a bilayer or in a trilayer Ni lattice [33, 95–97]. It is also indicated that the Fermi surfaces are contributed by Ni  $d_{x^2-y^2}$  and  $d_{3z^2-r^2}$  orbitals [56, 95–98], with two-electron sheets with mixed  $e_g$  orbitals and a small hole pocket from the Ni  $d_{3z^2-r^2}$  orbital at high pressure which is absent at ambient pressure. Angle-resolved photoemission spectroscopy experiments confirmed the Fermi surface obtained by DFT calculations for both  $\text{La}_3\text{Ni}_2\text{O}_7$  and  $\text{La}_4\text{Ni}_3\text{O}_{10}$  at

ambient pressure [30, 56].

Based on the two- $e_g$ -orbitals model, theoretical studies suggested that  $s_{\pm}$ -wave pairing is dominant in bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$ , induced by spin fluctuations due to the partial nesting of the Fermi surfaces with wave vectors approximately  $(\pi, 0)$  or  $(0, \pi)$  [62, 64, 66, 69, 99–101]. The  $s_{\pm}$ -wave pairing channel is believed to be driven mainly by strong interlayer coupling via the  $d_{3z^2-r^2}$  orbital, but the  $d_{x^2-y^2}$  orbital also contributes robustly to the superconducting gap functions, comparable in some cases to the contributions of the  $d_{3z^2-r^2}$  orbital [75, 101]. Alternatively, there also appeared other theoretical studies, suggesting a dominant  $d_{x^2-y^2}$ -wave or  $d_{xy}$ -wave superconducting pairing channel driven by the intralayer coupling [71, 73, 102, 103]. In fact, the interplay between the intralayer and the interlayer pairing tendency was discussed many years ago by using  $t - J$  models, where a prevailing interlayer coupling leads to  $s$ -wave pairing while a prevailing stronger intralayer coupling results in  $d$ -wave pairing [104].

Considering these established results for bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  and trilayer  $\text{La}_4\text{Ni}_3\text{O}_{10}$ , it is natural to wonder about the properties of other members of the RP nickelates family  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ : Could the RP higher-order  $m = 4 - 6$  stacking nickelate be superconducting under pressure? In this case, what trends of superconductivity dominate in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ ? What is the dependence of the pairing channel under pressure in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ ? What are the magnetic correlations in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ ? Could we establish a general intuitive picture of the RP nickelates systems?

To address these questions, here we theoretically study the high-symmetry  $I4/mmm$  phase of RP nickelates  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to 6) without the tilting of the  $\text{NiO}_6$  octahedra under pressure by using first-principles DFT and random phase approximation (RPA) calculations. Our studies find many similarities but also some differences in the RP nickelates with different  $m$ -layer blocks. The  $d_{3z^2-r^2}$  orbital displays the bonding-antibonding, or the bonding-antibonding-nonbonding splitting character, depending on the even or odd number of stacking layers  $m$ . In addition, the in-plane ratio of the interorbital hopping between the  $e_g$  orbitals and intraorbital hopping between  $d_{x^2-y^2}$  orbitals was found to be large in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ , and this ratio increases from  $m = 1$  to  $m = 6$ . In contrast to the  $s^{\pm}$ -wave pairing driven by spin fluctuations in the bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  and trilayer  $\text{La}_4\text{Ni}_3\text{O}_{10}$ , two nearly degenerate  $d_{x^2-y^2}$ -wave and  $s^{\pm}$ -wave instabilities are obtained in  $\text{La}_5\text{Ni}_4\text{O}_{13}$  and  $\text{La}_6\text{Ni}_5\text{O}_{16}$ , while a leading  $s^{\pm}$ -wave state was found in  $\text{La}_7\text{Ni}_6\text{O}_{19}$ . In general, at the level of the RPA treatment, the superconducting transition temperature  $T_c$  was expected to decrease in stoichiometric bulk systems from the bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  to the six-layer  $\text{La}_7\text{Ni}_6\text{O}_{19}$ .

Moreover, in the Ni planes, we obtained the peak of the magnetic susceptibility at  $\mathbf{q} = (0.6\pi, 0.6\pi)$  for  $\text{La}_5\text{Ni}_4\text{O}_{13}$  and  $\text{La}_7\text{Ni}_6\text{O}_{19}$ , and at  $\mathbf{q} = (0.7\pi, 0.7\pi)$  for  $\text{La}_6\text{Ni}_5\text{O}_{16}$ . Furthermore, the out-of-plane magnetic correlations are complex in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ , with details depending on the stacking layer number  $m$ .

The scope of this work is to explore and understand the trends in the RP nickelates family  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  with different  $m$ -layer stacking. Considering previous studies of superconductivity in the bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  and trilayer  $\text{La}_4\text{Ni}_3\text{O}_{10}$ , here we use the high-symmetry  $I4/mmm$  structure (see Fig. 1) without the tilting of  $\text{NiO}_6$  octahedra to perform our study. The paper is organized as follows: in Section II the methods used are described, the results are presented in Section III, and Section IV is devoted to the conclusions and discussions.

## II. METHOD

In this work, first-principles DFT calculations were performed employing the Vienna *ab initio* simulation package (VASP) code by using the projector augmented wave method [105–107] with the generalized gradient approximation and the Perdew-Burke-Ernzerhof (PBE) exchange potential [108]. The plane-wave cutoff energy was set as 550 eV. Both lattice constants and atomic positions were fully relaxed until the Hellman-Feynman force on each atom was smaller than 0.01 eV/Å. In addition, the  $\mathbf{k}$ -point mesh used was  $20 \times 20 \times 2$  for the  $I4/mmm$  phase of  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  at 30 GPa.

To carry out further theoretical analyses, we derive maximally localized Wannier functions (MLWFs) projected on Ni  $3d$  states using the WANNIER90 package [109] and construct low-energy  $e_g$ -orbital tight-binding models, consisting of orbital-dependent hopping matrices and crystal-field splitting. The resulting tight-binding Hamiltonian is expressed as

$$H_k = \sum_{i\gamma\gamma'} t_{\gamma\gamma'}^{\vec{\alpha}} (c_{i\gamma\sigma}^\dagger c_{i+\vec{\alpha}\gamma'\sigma} + H.c.) + \sum_{i\gamma\sigma} \Delta_\gamma n_{i\gamma\sigma}. \quad (1)$$

The first term represents the hopping of an electron from orbital  $\gamma'$  at site  $i + \vec{\alpha}$  to orbital  $\gamma$  at site  $i$ .  $c_{i\gamma\sigma}^\dagger$  ( $c_{i\gamma\sigma}$ ) is the creation (annihilation) operator of an electron at site  $i$ , orbital  $\gamma$  with spin  $\sigma$ .  $\Delta_\gamma$  represents the crystal field of orbital  $\gamma$  with  $n_{i\gamma\sigma} = c_{i\gamma\sigma}^\dagger c_{i\gamma\sigma}$ . The vectors  $\vec{\alpha}$  are along the three directions, defining different hopping neighbors. In addition, the detailed input files of hopping matrices and crystal-field splittings can be found in a separate attachment in the Supplemental Materials [111]. Based on the obtained Fermi energy for the stoichiometric ratio filling of  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to 6) in our tight-binding model calculations, a  $4001 \times 4001$   $\mathbf{k}$ -mesh was used to calculate the Fermi surface.

To discuss the superconducting pairing and magnetic correlations in different  $m$ -layer stacking nickelates under pressure, here we used the many-body RPA method based on a perturbative weak-coupling expansion in the Hubbard interaction, similar to our previous analysis [62, 97]. We considered a multi-orbital Hubbard model, including the kinetic energy  $H_k$  and local interaction  $H_{\text{int}}$  terms. The model is written as  $H = H_k + H_{\text{int}}$ , where  $H_{\text{int}}$  includes the intraorbital Hubbard repulsion  $U$ , the interorbital Hubbard repulsion  $U'$ , the Hund's coupling  $J$ , and the interorbital electron-pair hopping  $J'$ , given by

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

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

In the multi-orbital RPA approach [112–117], the spin susceptibility is obtained from the bare susceptibility (Lindhart function)  $\chi_0(\mathbf{q})$  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  $\lambda_\alpha$  for channel  $\alpha$  and the corresponding gap structure  $g_\alpha(\mathbf{k})$  are obtained from solving an eigenvalue problem of the form

$$\int_{FS} d\mathbf{k}' \Gamma(\mathbf{k} - \mathbf{k}') g_\alpha(\mathbf{k}') = \lambda_\alpha g_\alpha(\mathbf{k}), \quad (3)$$

where the momenta  $\mathbf{k}$  and  $\mathbf{k}'$  are on the Fermi Surface FS and  $\Gamma(\mathbf{k} - \mathbf{k}')$  contains the irreducible particle-particle vertex. While both spin and charge susceptibilities contribute to the pairing interaction, the dominant contribution comes from the RPA spin susceptibility tensor  $\chi_{\ell_1 \ell_2 \ell_3 \ell_4}^s(\mathbf{k} - \mathbf{k}')$ , where  $\{\ell_i\}$  denote the different  $e_g$  orbitals of the RP nickelates, as shown in Fig. 1. For the electronic densities, we use the stoichiometric cases (i.e. 5 electrons for  $m = 4$ , corresponding to 1.25 electrons per site for the two  $e_g$  orbitals).

### III. RESULTS

#### A. Model systems

RP nickelates  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m$  from 1 to  $\infty$ ) have  $m$ -layer blocks of corner-shared  $\text{NiO}_6$  octahedra. Considering the formal valence of  $\text{La}^{3+}$  and  $\text{O}^{2-}$ , the average valence of Ni in RP nickelates is given by  $3 - 1/m$  as a function of  $m$ , leading to the  $3d$  electron density  $n$  varying from 8 at  $m = 1$  to 7 at  $m = \infty$  as shown in

Fig. 2(a). Since Ni  $t_{2g}$  orbitals are fully occupied, these nickelates could be regarded as “effective” two- $e_g$ -orbital systems.

An important consequence of  $m$ -layer stacking in RP nickelates is the bonding-antibonding splitting of Ni  $d_{3z^2-r^2}$  orbitals. Due to the single-layer blocks stacking,  $d_{3z^2-r^2}$  orbitals do not have bonding-antibonding splitting in  $\text{La}_2\text{NiO}_4$  ( $m = 1$  case), as shown in Fig. 2(b). For the multiple layer stacking geometries in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ , the  $d_{3z^2-r^2}$  orbital shows the bonding-antibonding, or the bonding-antibonding-nonbonding splitting, depending on the even or odd number of stacking layers. Similar to the “dimer” physics in the  $m = 2$  RP nickelate, the  $m = 4$  and  $m = 6$  compounds show an orbital-selective behavior [118, 119], where the  $d_{3z^2-r^2}$  orbital splits into bonding and antibonding states, while the  $d_{x^2-y^2}$  orbital remains decoupled among planes, as shown in Fig. 2(c). Similar to the “trimer” physics in the  $m = 3$  RP nickelate, the  $d_{3z^2-r^2}$  orbitals have bonding-antibonding and nonbonding splitting for  $\text{La}_6\text{Ni}_5\text{O}_{16}$  ( $m = 5$  case), as displayed in Fig. 2(d). At the other limit ( $m = \infty$ ), i.e. the cubic perovskite  $\text{LaNiO}_3$ , the  $d_{3z^2-r^2}$  orbital does not have bonding-antibonding splitting as well (see Fig. 2(e)). However, due to the strong in-plane hybridization between the  $e_g$  orbitals, both  $d_{3z^2-r^2}$  and  $d_{x^2-y^2}$  orbitals are not integer-filled, leading to a “self-doping” effect in those two orbitals. Thus, this effect is also expected in all other  $m$ -layer stackings  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ .

Note that the scope of this work is to explore and understand the trends in the RP nickelates family  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  with different  $m$ -layer stacking. Considering previous studies on the bilayer and trilayer nickelate bulk superconductors where superconductivity was found in the high-symmetry phase under pressure where the tilting of  $\text{NiO}_6$  octahedra is absent, here we use the high-symmetry I4/mmm structure [47] (see Fig. 1) to mimic the uniform lattice structure expected for RP nickelates at 30 GPa. Since the electronic states near the Fermi level of RP nickelates  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to 6) are mainly contributed by Ni  $3d$  orbitals while the O  $2p$  orbitals are located deeper in energy, a large charge-transfer gap between Ni  $3d$  and O  $2p$  states is expected as a common feature in these systems.

#### B. Tight-binding band structures and Fermi surfaces

To explore and understand the trends in the RP nickelates family with different  $m$ -layer stacking, we considered two  $e_g$ -orbitals for each  $m$ -layer system, i.e., the model has  $2m$  bands. Based on the optimized crystal structures at 30 GPa, we obtained hoppings and crystal-field splittings by using the MLWFs method in the WANNIER90 packages [109], and constructed an

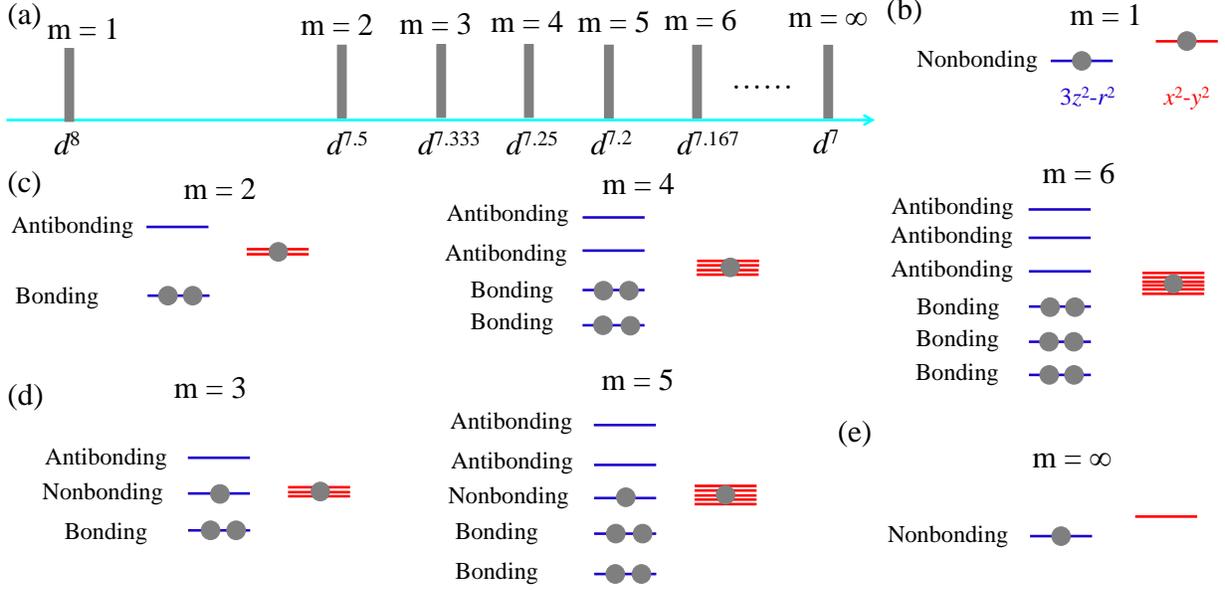


FIG. 2. (a) Schematic electronic densities of 3d-electrons per Ni for different  $m$ -layer stacking RP nickelates  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ . (b-e) Sketches of electronic states for two “active”  $e_g$  orbitals in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  with layers (b)  $m = 1$ , (c)  $m = 2, 4, 6$ , and (d)  $m = 3, 5$  and (e)  $\infty$ , respectively. The light blue and pink horizontal lines represent  $d_{3z^2-r^2}$  and  $d_{x^2-y^2}$  states, respectively. The solid circles indicate the occupied electrons of  $e_g$  orbitals in the stoichiometric ratio  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ .

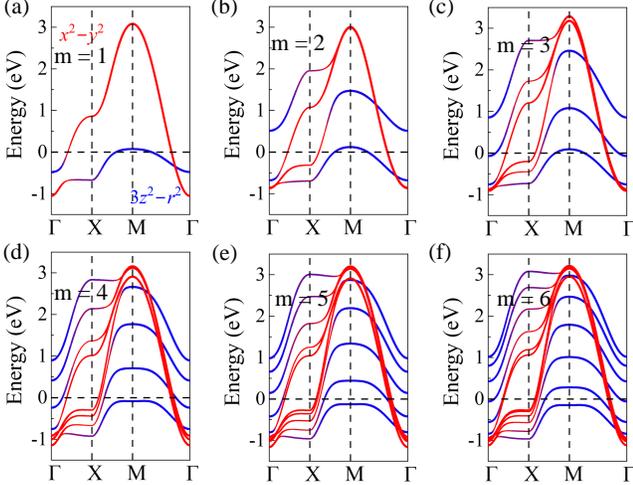


FIG. 3. Band structures of the tight-binding models for  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to 6) at 30 GPa. The coordinates of the high symmetry points in the Brillouin zone are  $\Gamma = (0, 0, 0)$ ,  $X = (0, 0.5, 0)$ , and  $M = (0.5, 0.5, 0)$ . Here, two  $e_g$  orbitals were considered in the tight-binding models with an overall filling of  $n = 2$  to 7 for  $m = 1$  to 6 (e.g., 1.2 electrons per site for  $m = 6$ ).

$e_g$ -orbital tight-binding model for  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to 6).

In Fig. 3, we plot the tight-binding band structures of the I4/mmm phase for different  $m$ -layer stacking at 30 GPa. By increasing  $m$  from 1 to 6, the bandwidth

of  $e_g$  orbitals is slightly increased by about 3%. In bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  and trilayer  $\text{La}_4\text{Ni}_3\text{O}_{10}$ , a small hole pocket was found at the M point due to the bonding  $d_{3z^2-r^2}$  orbitals. Such a small hole pocket is absent in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  with  $m = 4$  to 6, because the lowest bonding  $d_{3z^2-r^2}$  orbital does not touch the Fermi level.

Figure 4 summarizes the Fermi surfaces of RP nickelates. As shown in Fig. 4(a), there are two bands crossing the Fermi level in the single-layer  $\text{La}_2\text{NiO}_4$  at 30 GPa, leading to an electronic sheet made of the mixed  $d_{3z^2-r^2}$  and  $d_{x^2-y^2}$  orbitals and a hole sheet originating from the  $d_{3z^2-r^2}$  orbital. As mentioned above,  $\text{La}_3\text{Ni}_2\text{O}_7$  and  $\text{La}_4\text{Ni}_3\text{O}_{10}$  have small hole pockets around the M point (see Figs.4(b) and (c)), and such a hole pocket is missing in  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 4$  to 6) at 30 GPa as displayed in Figs. 4 (d-f). In addition, a hole sheet from the  $d_{3z^2-r^2}$  orbital was found near the M point in  $\text{La}_6\text{Ni}_5\text{O}_{16}$  (see  $\gamma_5$  in Fig.4(e)) and  $\text{La}_7\text{Ni}_6\text{O}_{19}$  (see  $\gamma_6$  in Fig.4(f)). Similar to the trilayer  $\text{La}_4\text{Ni}_3\text{O}_{10}$  case [97], an electron  $\sigma$  pocket made up of a  $d_{3z^2-r^2}$  orbital was also obtained in  $\text{La}_7\text{Ni}_6\text{O}_{19}$ , as in Fig.4(f). The tight-binding Fermi surfaces of the  $m = 4$  to  $m = 6$  cases discussed here are also in agreement with previous DFT calculations in the tetragonal I4/mmm phase of high-order RP nickelates [47].

We also notice the characteristic trend of hopping intensities. In the single-layer  $\text{La}_2\text{NiO}_4$  ( $\sim 0.333$ ) at 30 GPa, 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 significantly small, compared to the bilayer and trilayer nickelates ( $\sim 0.475$

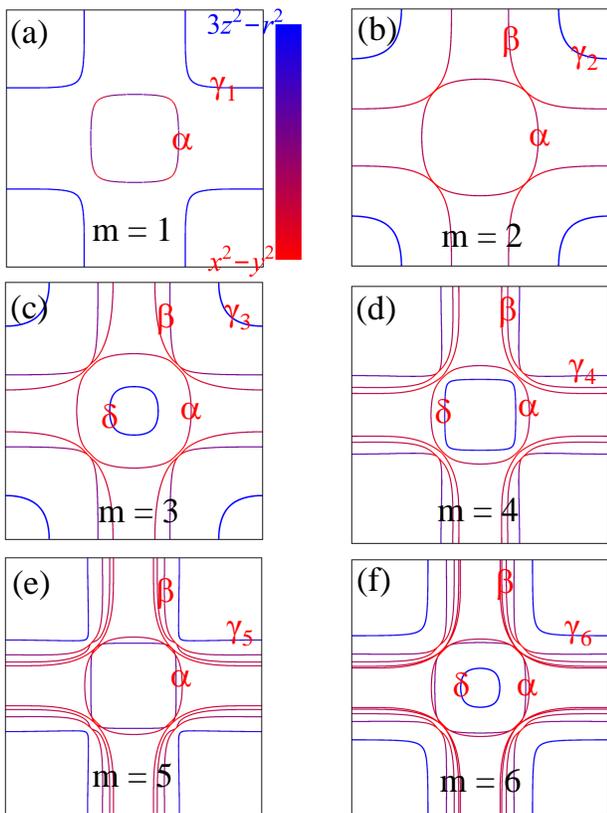


FIG. 4. Fermi surfaces of the tight-binding models for  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 1$  to  $6$ ) at 30 GPa. Here, two  $e_g$  orbitals were considered in the tight-binding models with an overall filling of  $n = 2$  to  $7$  for  $m = 1$  to  $6$  (e.g., 1.2 electrons per site for  $m = 6$ ).

for  $\text{La}_3\text{Ni}_2\text{O}_7$  and  $\sim 0.532/0.543$  for  $\text{La}_4\text{Ni}_3\text{O}_{10}$ ) [62, 97]. This value  $t_{x/y}^{12}/t_{x/y}^{22}$  continues to increase with the increase of high-order layer stacking at 30 GPa:  $\sim 0.543/0.564$  for  $\text{La}_5\text{Ni}_4\text{O}_{13}$ ,  $\sim 0.545/0.573/0.580$  for  $\text{La}_6\text{Ni}_5\text{O}_{16}$ , and  $\sim 0.544/0.568/0.584$  for  $\text{La}_7\text{Ni}_6\text{O}_{19}$ , suggesting that the in-plane hybridization between the  $d_{3z^2-r^2}$  and  $d_{x^2-y^2}$  orbitals and the layer number  $m$  has a positive correlation.

### C. Superconducting pairing tendency

Next, we study the superconducting pairing tendencies in the RP nickelates. Here, we carry out the RPA technique to investigate the pairing instability of the multi-orbital Hubbard model,  $H = H_k + H_{int}$ , where  $H_k$  is the single-particle part of the Hamiltonian with hopping matrix and crystal-field splittings from the I4/mmm phase of  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  at 30 GPa, given by Eq. (1). The RPA calculations of the pairing vertex are based on a perturbative weak-coupling expansion with respect to the electron-electron interaction part of the

Hamiltonian (see Method section),  $H_{int}$ , given by Eq. (2), where the local Coulomb interaction matrix involves the intra-orbital ( $U$ ), inter-orbital ( $U'$ ), Hund's rule coupling ( $J$ ), and pair-hopping ( $J'$ ) terms [112–115].

As discussed in previous work, the bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  [62] and trilayer  $\text{La}_4\text{Ni}_3\text{O}_{10}$  [97] have leading  $s^\pm$ -wave pairing instabilities driven by spin-fluctuations with different nesting vectors:  $\mathbf{q} = (\pi, 0)$  or  $(0, \pi)$  for  $\text{La}_3\text{Ni}_2\text{O}_7$  and  $\mathbf{q} = (\pi, \pi)$  for  $\text{La}_4\text{Ni}_3\text{O}_{10}$ . For the high-order nickelates ( $m = 4 - 6$ ), starting from the high-symmetry I4/mmm phase obtained from the previous study [47], we fully relaxed the atomic positions and lattice constants at 30 GPa. In the following, we focus on  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  with  $m = 4$  to  $6$  and explore their superconducting pairing tendencies.

Figure 5 shows our numerical results for the leading pairing instability in the high-order stacking  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 4$  to  $6$ ), obtained by solving the eigenvalue problem in Eq. (3). Here, we used Coulomb parameters  $U = 1.4$  eV,  $U' = U/2$ , and  $J = J' = U/4$ , and the calculation was performed at a temperature  $T = 0.01$  eV.

For the four-layer  $m = 4$  case, we find nearly degenerate  $d_{x^2-y^2}$ -wave and  $s^\pm$ -wave leading states with almost identical eigenvalues  $\lambda_d = 0.1114$  and  $\lambda_s = 0.1113$ , respectively. For both the  $d_{x^2-y^2}$ -wave and the  $s^\pm$  states, the gap is largest on the inner  $\delta$  pocket with mainly  $d_{3z^2-r^2}$  character and the  $\beta$  sheets with mixed  $e_g$  character (see Fig. 5(a)).

For the  $s^\pm$ -wave state, the phase of the gap changes signs between these two sheets. The five-layer  $m = 5$  case also has nearly degenerate  $d_{x^2-y^2}$ -wave and  $s^\pm$ -wave leading solutions with similar pairing strengths,  $\lambda_d = 0.1089$  and  $\lambda_s = 0.101$ , respectively. In contrast to the 4-layer case, here the gap is largest on the inner  $\alpha$  sheet with mixed  $e_g$  character and the  $\gamma_5$  sheet with  $d_{3z^2-r^2}$  character. The sign changes in or between these two sheets, as displayed in Fig. 5(b). The six-layer  $m = 6$  case shown in Fig. 5(c) has a slightly reduced  $\lambda$  for the leading  $s^\pm$ -wave state and an even lower  $\lambda$  for the  $d_{x^2-y^2}$ -wave channel. For both these states, the gap is largest on the inner  $\delta$  pocket and the inner  $\gamma_6$  sheet.

We note that the leading pairing strength  $\lambda_d = 0.1114$  for four-layer stacking  $\text{La}_5\text{Ni}_4\text{O}_{13}$  at  $U = 1.4$  eV is already much smaller than the pairing strength  $\lambda_s = 0.202$  we obtained for the trilayer system  $\text{La}_4\text{Ni}_3\text{O}_{10}$  at 30 GPa but with a smaller interaction strength  $U = 0.95$  eV [62], suggesting that the pairing correlations in the trilayer stacking system are much stronger. Moreover, we found previously that the trilayer has smaller pairing strength than the bilayer 327-LNO at the same  $U = 0.95$  ( $\lambda_s \sim 0.39$ ). Since in our RPA treatment, the pairing strength  $\lambda$  enters exponentially in the equation for  $T_c$ , i.e.  $T_c = \omega_0 e^{-1/\lambda}$  with a spin-fluctuation cut-off frequency  $\omega_0$ , this, and the new results in Fig. 5 suggests that  $T_c$  is expected to decrease in stoichiometric bulk systems as

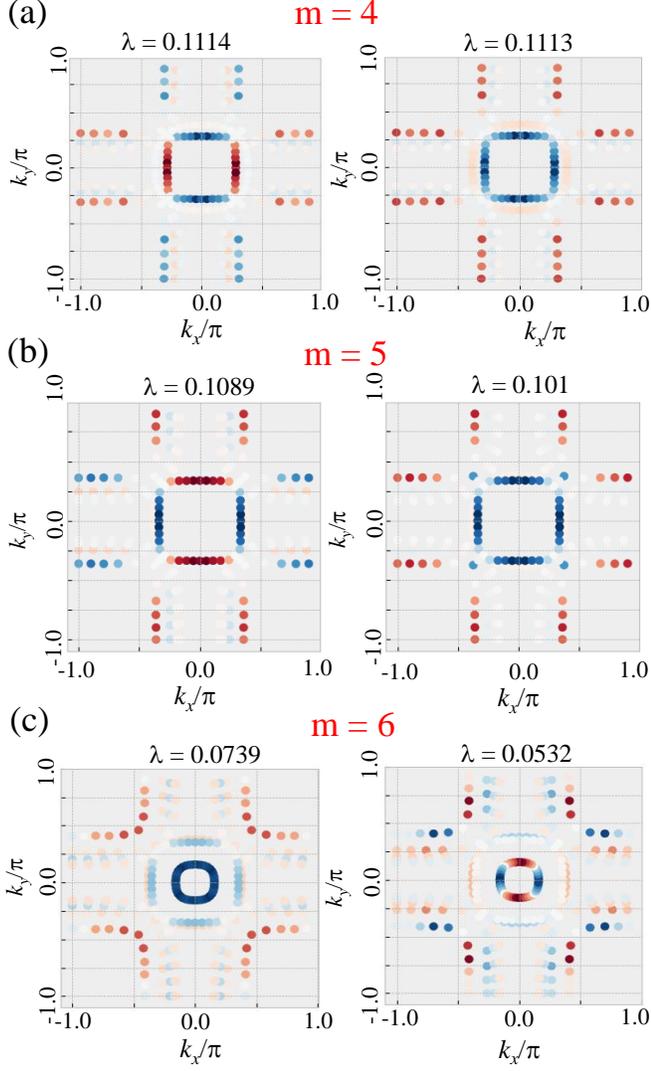


FIG. 5. The RPA calculated leading superconducting singlet gap structures  $g_\alpha(\mathbf{k})$  for momenta  $\mathbf{k}$  on the Fermi surfaces for (a)  $\text{La}_5\text{Ni}_4\text{O}_{13}$ , (b)  $\text{La}_6\text{Ni}_5\text{O}_{16}$ , and (c)  $\text{La}_7\text{Ni}_6\text{O}_{19}$  with corresponding pairing strengths  $\lambda$  at 30 GPa. The sign of  $g_\alpha(\mathbf{k})$  is indicated by the color (red = positive, blue = negative), and its amplitude by the color darkness. We used Coulomb parameters  $U = 1.4$  eV,  $U' = U/2$ , and  $J = J' = U/4$ . The calculation was performed at  $T = 0.01$  eV.

the layer stacking  $m$  increases.

#### D. Magnetic correlations

Finally, we discuss the magnetic correlations in the RP nickelates  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  with different layer blocks  $m$ . To this end, 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

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

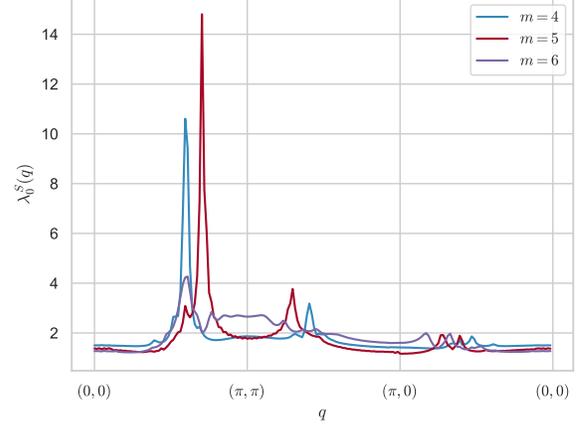


FIG. 6. The RPA calculated leading eigenvector  $\lambda_0^S(\mathbf{q})$  of the spin-susceptibility matrix  $\chi_{\ell_1\ell_2\ell_2\ell_2}$  for  $\text{La}_5\text{Ni}_4\text{O}_{13}$  ( $m = 4$ ),  $\text{La}_6\text{Ni}_5\text{O}_{16}$  ( $m = 5$ ), and  $\text{La}_7\text{Ni}_6\text{O}_{19}$  ( $m = 6$ ) for the  $I4/mmm$  phase at 30 GPa. The Coulomb parameters  $U = 1.4$  eV,  $U' = U/2$ , and  $J = J' = U/4$ , and the calculation was performed for a temperature of  $T = 0.01$  eV.

where 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 [112]. The physical spin susceptibility is obtained by summing the pairwise diagonal tensor  $\chi_{\ell_1\ell_2\ell_2\ell_2}(\mathbf{q})$  over  $\ell_1, \ell_2$ .

As displayed in Fig. 6, the leading eigenvalue  $\lambda_0^S(\mathbf{q})$  of  $\chi_{\ell_1\ell_2\ell_2\ell_2}(\mathbf{q})$  for momenta  $\mathbf{q}$  along a high-symmetry path in the Brillouin zone has the strongest in-plane peak at  $\mathbf{q} = (0.6\pi, 0.6\pi)$ ,  $\mathbf{q} = (0.7\pi, 0.7\pi)$ , and  $\mathbf{q} = (0.6\pi, 0.6\pi)$  for  $m = 4, 5$ , and  $6$ , respectively. For the single-layer  $\text{La}_2\text{NiO}_4$  case, the leading eigenvalue  $\lambda_0^S(\mathbf{q})$  has a maximum at  $(\pi, 0)$  or  $(0, \pi)$ . This is different from the bilayer and trilayer systems [62, 97], for which we previously found that the in-plane peak is near  $(\pi, 0)$  or  $(0, \pi)$  and  $(\pi, \pi)$ , respectively. In the RPA treatment, this difference arises from differences in the Fermi surface and the respective nesting wavevectors. This may be a consequence of the intraorbital and interorbital hopping mechanisms with different electronic densities, as discussed in previous studies [120–122].

Figure 7 shows the corresponding eigenvector  $\varphi_0^S(\mathbf{q})$  at the in-plane  $\mathbf{q}$  for which its eigenvalue  $\lambda_0^S(\mathbf{q})$  has a maximum. One sees that the main contributions to the magnetic correlations come from the  $d_{3z^2-r^2}$  orbitals from outer Ni layers. In addition, the out-of-plane, inter-layer magnetic correlations determined by the relative sign of the eigenvector contributions also strongly depend on the number of layers  $m$ . In the 4- and 5-layer systems, the outer Ni layers are coupled ferromagnetically, but in the 6-layer system, they have antiferromagnetic coupling. For  $\text{La}_5\text{Ni}_4\text{O}_{13}$ , the four layers are coupled as  $\downarrow\text{-}\uparrow\text{-}\uparrow\text{-}\downarrow$  and the five layers

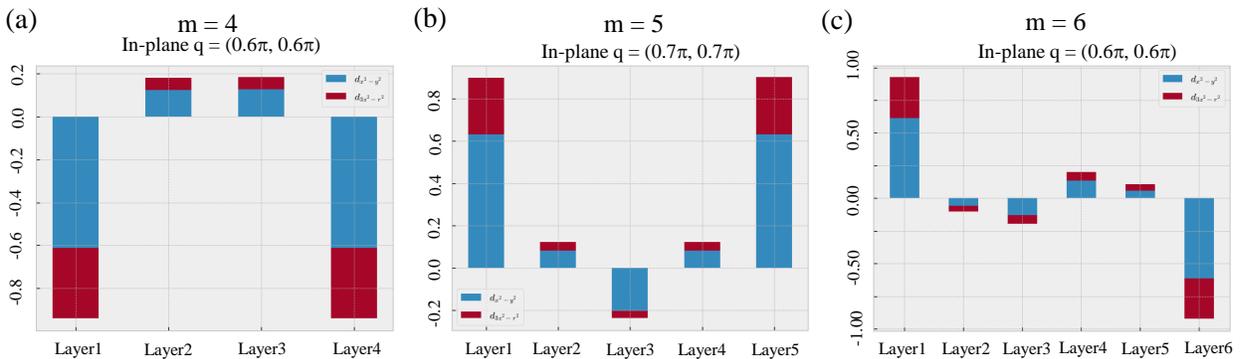


FIG. 7. Leading eigenvector of the susceptibility matrix  $\chi_{\ell_1\ell_1,\ell_2\ell_2}(q,\omega=0)$  at the in-plane  $q$ -vector for which its leading eigenvalue  $\lambda_0^S(q)$  has a maximum, for (a)  $\text{La}_5\text{Ni}_4\text{O}_{13}$ , (b)  $\text{La}_6\text{Ni}_5\text{O}_{16}$ , and (c)  $\text{La}_7\text{Ni}_6\text{O}_{19}$ . The contributions of  $d_{3z^2-r^2}$  and  $d_{x^2-y^2}$  orbitals are shown by blue and red colors. The Coulomb parameters are  $U = 1.4$  eV,  $U' = U/2$ , and  $J = J' = U/4$ , and the calculation was performed for temperature  $T = 0.01$  eV.

are coupled as  $\uparrow-\uparrow-\downarrow-\uparrow-\uparrow$  along the out-of-plane directions, while the  $\text{La}_7\text{Ni}_6\text{O}_{19}$  has an  $\uparrow-\downarrow-\downarrow-\uparrow-\uparrow-\downarrow$  coupling. In contrast to the trilayer case, we do not find a spin-zero Ni layer in high-order  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  with  $m = 4$  to 6.

#### IV. CONCLUSIONS AND DISCUSSIONS

In this work, studying the high-symmetry  $I4/mmm$  phase under pressure, we systematically explored physical trends in the RP  $m$ -layer stacking nickelates  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$ . Near the Fermi surface, the states are mainly contributed by Ni  $3d$  orbitals, because O  $2p$  orbitals are located deeper in energy, suggesting a common picture in nickelate superconductors involving a large charge-transfer gap between  $3d$  and  $2p$  states. Furthermore, the  $d_{3z^2-r^2}$  orbital exhibits the bonding-antibonding or the bonding-antibonding-nonbonding splitting characters, depending on whether  $m$  is even or odd. Due to the in-plane interorbital hopping between the  $d_{3z^2-r^2}$  and  $d_{x^2-y^2}$  orbitals, the electronic occupations of both orbitals is not an integer, leading to a “self-doping” situation, as observed in other RP nickelate superconductors. Furthermore, the ratio of the in-plane interorbital hopping between the  $e_g$  orbitals and in-plane intraorbital hopping between  $d_{x^2-y^2}$  orbitals is found to increase with  $m$ .

For the single-layer  $m = 1$  case, we found a strong spin-density-wave instability around  $(\pi, 0)$  or  $(0, \pi)$  due to the perfect nesting between two nearly flat  $\gamma_1$  pockets. For the bilayer  $m = 2$  and trilayer  $m = 3$  cases [62, 97], the spin-fluctuation-driven  $s^\pm$ -wave state is dominant in both cases. For  $m = 4$  and  $m = 5$  cases, we find two nearly degenerate  $d_{x^2-y^2}$ -wave and  $s^\pm$ -wave leading states. However, in  $\text{La}_7\text{Ni}_6\text{O}_{19}$ , this six-layer stacking system has a slightly reduced  $\lambda$  for the leading  $s^\pm$ -wave

state and an even lower  $\lambda$  for the  $d_{x^2-y^2}$ -wave channel. In general, at the level of the RPA treatment, *the superconducting transition temperature  $T_c$  decreases as the layer stacking number  $m$  increases in stoichiometric bulk systems.*

Similar to the pairing instability, magnetic correlations are found to be quite complex in the RP nickelates, depending on the stacking layer number  $m$ . As mentioned above, single-layer  $\text{La}_2\text{NiO}_4$  has a strong stripe instability characterized by  $\mathbf{q} = (\pi, 0)$  or  $(0, \pi)$ , similar to our previous finding on bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  [62]. For the trilayer  $\text{La}_4\text{Ni}_3\text{O}_{10}$  [97], the peak of the magnetic susceptibility was found at  $\mathbf{q} = (\pi, \pi)$ . For  $\text{La}_5\text{Ni}_4\text{O}_{13}$  ( $m = 4$ ) and  $\text{La}_7\text{Ni}_6\text{O}_{19}$  ( $m = 6$ ), the peak position is shifted to  $\mathbf{q} \approx (0.6\pi, 0.6\pi)$ , and for  $\text{La}_6\text{Ni}_5\text{O}_{16}$  ( $m = 5$ ) the peak position is at  $\mathbf{q} \approx (0.7\pi, 0.7\pi)$ . Moreover, the out-of-plane magnetic correlations strongly depend on the number of layers  $m$ .

Figure 8 summarizes our findings. We hope that our comprehensive study of the  $m$  dependence of magnetic and pairing properties will stimulate experimental efforts to verify our theoretical predictions. The thin film of high-order  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 4$  and  $m = 5$ ) [123] and  $\text{Nd}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 4$  and  $m = 5$ ) [124] has been stabilized in experiments using reactive molecular-beam epitaxy. Very recently, Y.-F. Zhao et. al. [125] found that tensile strain can induce a small  $\gamma$  pocket from the lowest bonding state of  $d_{3z^2-r^2}$  orbitals near M points in the  $I4/mmm$  phase of four-layer stacking  $\text{La}_5\text{Ni}_4\text{O}_{13}$  and five-layer stacking  $\text{La}_6\text{Ni}_5\text{O}_{16}$ , while a compressive strain can not induce this pocket for both cases, suggesting that the pressure may not induce the  $\gamma$  pocket as well. In the previous context of bilayer and trilayer nickelates, this pocket is believed to be crucial for superconductivity. Thus, the next step is to investigate in the high-order stacking nickelates  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  ( $m = 4$  to 6) the connection between superconductivity and the hole pocket from the lowest bonding state of  $d_{3z^2-r^2}$  orbitals.

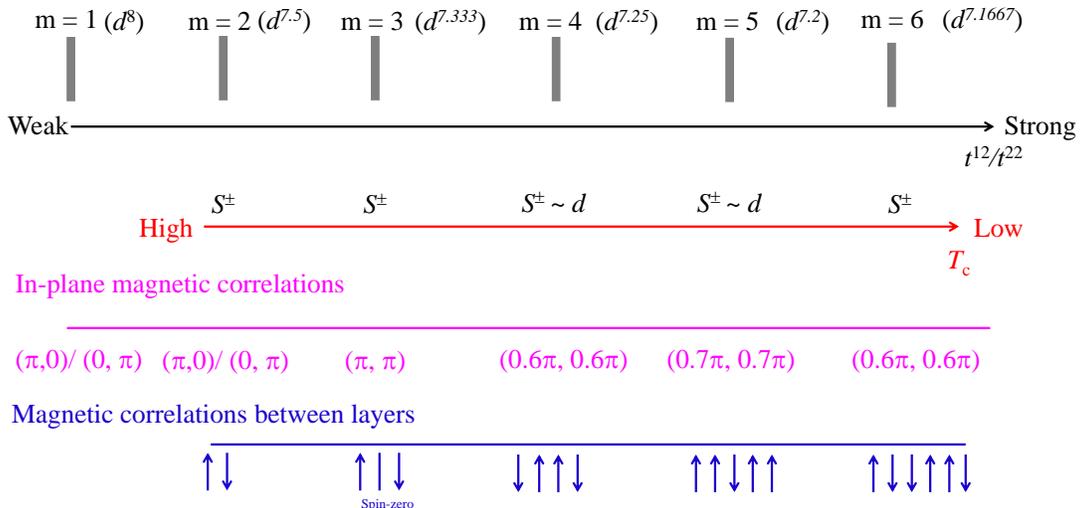


FIG. 8. Summary of our main findings for the  $I4/mmm$  phase of the  $\text{La}_{m+1}\text{Ni}_m\text{O}_{3m+1}$  family with different layer stacking number  $m$  under pressure.

Especially, to explore if the superconductivity can be enhanced in those high-order La-based systems or in other rare-earth RP nickelates ( $m = 4-6$  case) when the small  $\gamma$  pocket from the lowest bonding state of  $d_{3z^2-r^2}$  orbitals exists near the M point.

#### ACKNOWLEDGMENTS

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#### DATA AVAILABILITY

The input parameters for the kinetic energy term in our calculations are available for reproduction of our results in a separate file in the Supplemental Materials. Any additional data are also available from the authors upon reasonable request.

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