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Electronic and magnetic structure of infinite-layer NdNiO₂: trace of antiferromagnetic metal

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The recent discovery of Sr-doped infinite-layer nickelate NdNiO₂ offers a new platform for investigating unconventional superconductivity in nickelate-based compounds. Most intriguingly, the resistivity minimum and Hall coefficient drop were identified simultaneously in the experiment, reflecting a novel electronic structure and transport property of NdNiO₂. Driven by this pioneering work, we present a first-principles calculation for the electronic and magnetic structure of undoped parent NdNiO₂. By taking into account experimentally relevant interaction strength, we found that (π, π, π) antiferromagnetic NdNiO₂ is a compensated bad metal with small Fermi pockets. However, due to the small exchange coupling between 3d-electrons of Ni and strong hybridization with 5d-electrons of Nd, the discovered antiferromagnetic ordering is very weak. Crucially, with the decreasing of temperature, there exists a phase transition between good paramagnetic metal and bad AFM metal. The estimated transition temperature is ~70–90 K, which is consistent with that for observing the resistivity minimum and Hall coefficient drop. In this regard, our results provide a plausible physical interpretation for these significant experimental observations.

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INTRODUCTION

Since the discovery of high-temperature (high- T_c) superconductivity in cuprates¹, extensive effort has been devoted to investigate unconventional superconductors, ranging from non-oxide compounds^{2,3} to iron-based materials^{4,5}. Exploring high- T_c materials could provide a new platform to understand the fundamental physics behind high- T_c phenomenon, thus is quite valuable. Very recently, the discovery of superconductivity in Sr-doped NdNiO₂⁶ potentially raises the possibility to realize high- T_c in nickelate family^{7,8}.

One key experimental observation for the infinite-layer NdNiO₂ is that its resistivity exhibits a minimum ~70 K and an upturn at a lower temperature⁶. At the same time that the resistivity reaches minimum, the Hall coefficient drops towards a large value, signaling the loss of charge carriers⁶. Interestingly, no long-range magnetic order has been observed in powder neutron diffraction on NdNiO₂ when temperature is down to 1.7 K⁶. This greatly challenges the existing theories, since it is generally believed that magnetism holds the key to understand unconventional superconductivity^{9–12}. Therefore, it is highly desirable to study the magnetic properties of undoped parent NdNiO₂ and elucidate its experimental indications.

In this work, the electronic and magnetic properties of NdNiO₂ are systemically studied by first-principles calculations combined with classical Monte Carlo (MC) calculations. For the paramagnetic (PM) phase, its Fermi surface includes one large sheet and two electron pockets at Γ and A point, respectively. This can be approximately described by a three-band low-energy effective model that captures the main physics of exchange coupling mechanism, which is in line with previous works^{13–18}. Moreover, by including the Hubbard- U , (π, π, π) anti-ferromagnetic (AFM) ordering is confirmed to be the magnetic ground state. Most significantly, the Fermi surface of AFM phase is simpler than that

of PM phase, demonstrating an interaction induced elimination of Fermi pockets. Before NdNiO₂ enters correlated insulator, it is a compensated metal with one small electron pocket formed by d_{xy} orbital of Nd and four small hole pockets formed by d_{z^2} orbital of Ni. The estimated phase transition temperature (T_N) from PM phase to (π, π, π) AFM phase is ~70–90 K for moderate interaction strength of $U = 5–6$ eV. Through these studies, our results provide a physical understanding of two experimental observations. First, 3d-electrons of Ni tend to form AFM ordering ~70–90 K, coinciding with the minimum in resistivity and drop in Hall coefficient. Second, the (π, π, π) AFM ordering could be weak (compared with cuprates), because of small effective exchange coupling and strong hybridization with itinerant 5d-electrons of Nd. This could be the reason why AFM ordering is missing in previous studies, which calls for more careful neutron scattering measurements on NdNiO₂.

RESULTS

Electronic structure in PM phase

Firstly, we present the band structure of PM phase without Hubbard U . The orbital resolved band structure of PM phase is shown in Fig. 1b. Comparing with typical cuprates CaCuO₂¹³, two significant differences are noted: (1) there is a gap ~2.5 eV between 2p orbitals of O and 3d orbitals of Ni. According to Zaanen–Sawatzky–Allen classification scheme¹⁹, this indicates that the physics of NdNiO₂ is close to Mott-Hubbard rather than charge-transfer^{13,20,21}; (2) there are two bands crossing the Fermi level, in which one is mainly contributed by $d_{x^2-y^2}$ orbital of Ni (called pure-band) and the other one has a complicated orbital compositions (called mixed-band). In $k_z = 0$ plane, the mixed-band is mainly contributed by d_{z^2} orbital of Nd and Ni. The dispersion around Γ point is relatively small, and the pocket at Γ point is

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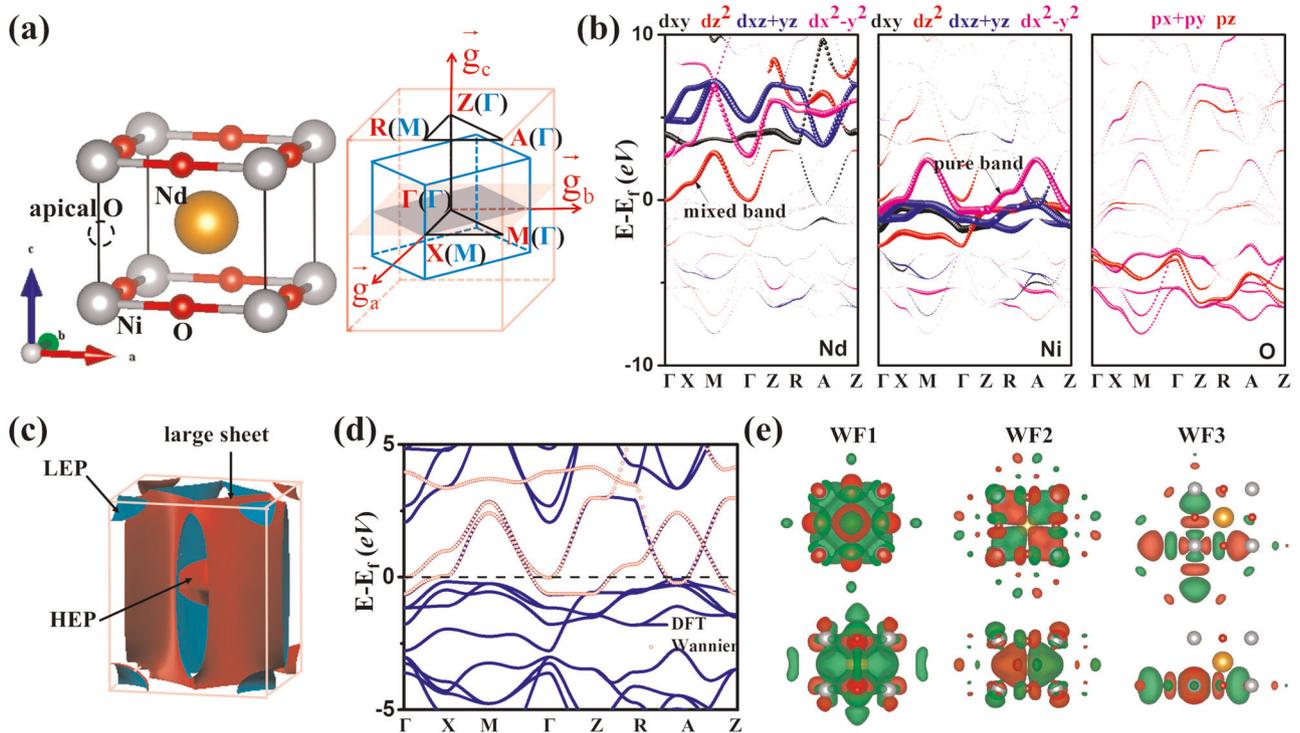


Fig. 1 Atomic structure and electronic structures of PM phase NdNiO₂. **a** Atomic structure of tetragonal NdNiO₂ and first Brillouin zone. The red and blue lines (labels) denotes the first Brillouin zone of PM phase and (π, π, π) AFM phase, respectively. The high symmetric line for band calculation are $\Gamma(0, 0, 0)$ -X(0.5, 0, 0)-M(0.5, 0.5, 0)- Γ -Z(0, 0, 0.5)-R(0.5, 0, 0.5)-A(0.5, 0.5, 0.5)-Z. **b** Orbital resolved band structure of PM phase. The d_{xy} , d_{z^2} , d_{xz+yz} and $d_{x^2-y^2}$ of Nd (spin-up Ni, spin-down Ni) are marked by black, red, blue, and pick-filled circles. The p_z and p_{x+y} of O are marked by red, blue, and pick filled circles. The size of circles represents the orbital weights. **c** Perspective view of Fermi surfaces. LEP and HEP denotes light and heavy electron pocket, respectively. **d** The comparison between first-principles and Wannier-fitting bands around the Fermi-level. **e** Top and side view of three maximally localized Wannier functions.

called heavy electron pocket (HEP). In $k_z = 0.5$ plane, the mixed-band is mainly contributed by d_{xy} (d_{xz} , d_{yz} and d_{z^2}) orbital of Nd (Ni). The dispersion around A point is relatively large, and the pocket at A point is called light electron pocket (LEP). As a comparison, one notices that there is only one pure-band crossing the Fermi level in CaCuO₂¹³. The Fermi surface of PM phase is shown in Fig. 1c. There is a large sheet contributed by the pure-band, as the case in CaCuO₂¹³. This Fermi surface is obviously two-dimensional (2D), because of the weak dispersion along Γ -Z. In addition, there are two electron pockets residing at Γ and A point, respectively, showing a feature of three-dimensional (3D) rather than 2D (see labels HEP and LEP in Fig. 1(c)). Therefore, the 3D metallic state will be hybridized with the 2D correlated state in NiO₂ plane, suggesting NdNiO₂ to be an “oxide-intermetallic” compound^{20,22}.

The existence of mixed-band also reflects the inherent interactions between Nd 5d and Ni 3d electrons. To explore the low-energy physics of NdNiO₂, a three-band model consisting of Ni $d_{x^2-y^2}$, Nd d_{z^2} and Nd d_{xy} orbitals is constructed by Wannier90 package. As shown Fig. 1d, one can see the good agreement between first-principles and Wannier-fitting bands near the Fermi level. The little deviation between DFT and Wannier-fitting bands above the Fermi level between Z and R is due to the hybridization between Nd 5d and Ni 3d orbitals. Such a hybridization causes the gap open which is missing in Wannier-fitting bands. The corresponding three maximally localized WFs are shown in Fig. 1e, demonstrating the main feature of d_{z^2} (WF1) and d_{xy} (WF2) orbital of Nd, and $d_{x^2-y^2}$ (WF3) orbital of Ni. However, these WFs still have some deviations from standard atomic orbitals, that is, WF1 and WF2 are mixed with d_{z^2} orbital of Ni, and WF3 is mixed with $p_{x/y}$ orbital of O in the NiO₂ plane. According to the classical

Goodenough–Kanamori–Anderson rules^{23–25}, these deviations (or hybridizations) will give clues for the magnetic properties.

Magnetic properties

To determine the magnetic ground state of NdNiO₂, six collinear spin configurations are taken into account in a $2 \times 2 \times 2$ supercell, that is, AFM1 with $\mathbf{q} = (\pi, \pi, \pi)$, AFM2 with $\mathbf{q} = (\pi, \pi, 0)$, AFM3 with $\mathbf{q} = (0, 0, \pi)$, AFM4 with $\mathbf{q} = (\pi, 0, 0)$, AFM5 with $\mathbf{q} = (\pi, 0, \pi)$, and FM with $\mathbf{q} = (0, 0, 0)$, as shown in Fig. 2a. Within all Hubbard U ranges, we found that AFM1 configuration always has the lowest energy, as shown in Fig. 2b, indicating a stable (π, π, π) AFM phase with respect to electron–electron interactions and is in accordance with random phase approximation treatment¹⁶. This can be attributed to the special orbital distributions around the Fermi level. The intralayer NN exchange coupling is the typical 180° typed Ni–O–Ni superexchange coupling, that is, the coupling between $d_{x^2-y^2}$ orbital of Ni is mediated by $p_{x/y}$ orbital of O (see WF3), preferring a (π, π) AFM phase in NiO₂ plane. The interlayer NN exchange coupling is due to the superexchange between the Ni d_{z^2} orbitals mediated by Nd d_{z^2} orbital as shown in WF1, preferring a (π, π) AFM phase between NiO₂ planes. Therefore, the superexchange coupling results in a stable (π, π, π) AFM phase in NdNiO₂. Moreover, the noncollinear magnetic states are further checked by including the spin-orbit coupling (SOC). We found that the spin moment prefers along c direction with the magnetic anisotropic energy of ~ 0.5 meV/Ni. Thus, the tiny SOC effect can be safely neglected and the collinear magnetic states are used in the following phase transition temperature calculations.

In cuprates, the Fermi surface is unstable with electron–electron interactions, making its parent phase to be an AFM insulator. However, this is apparently not the case in NdNiO₂, because of the

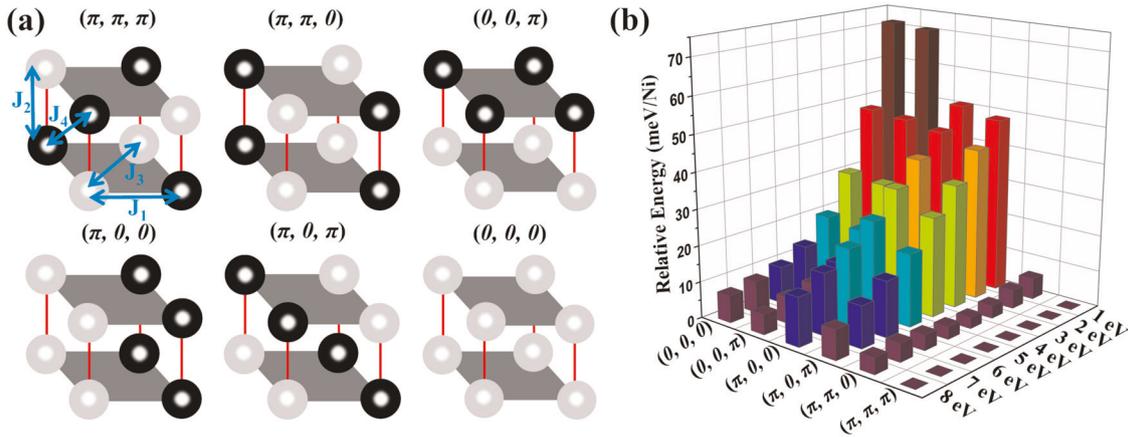


Fig. 2 Antiferromagnetic configurations and relative energies. **a** Illustration of six collinear spin configurations. The white and black ball represents local up and down spin moment, respectively. The four exchange coupling parameters are indicated by the blue arrows. **b** Energy comparison for the six collinear spin configurations with different values of Hubbard U . Energy of (π, π, π) AFM is set to zero.

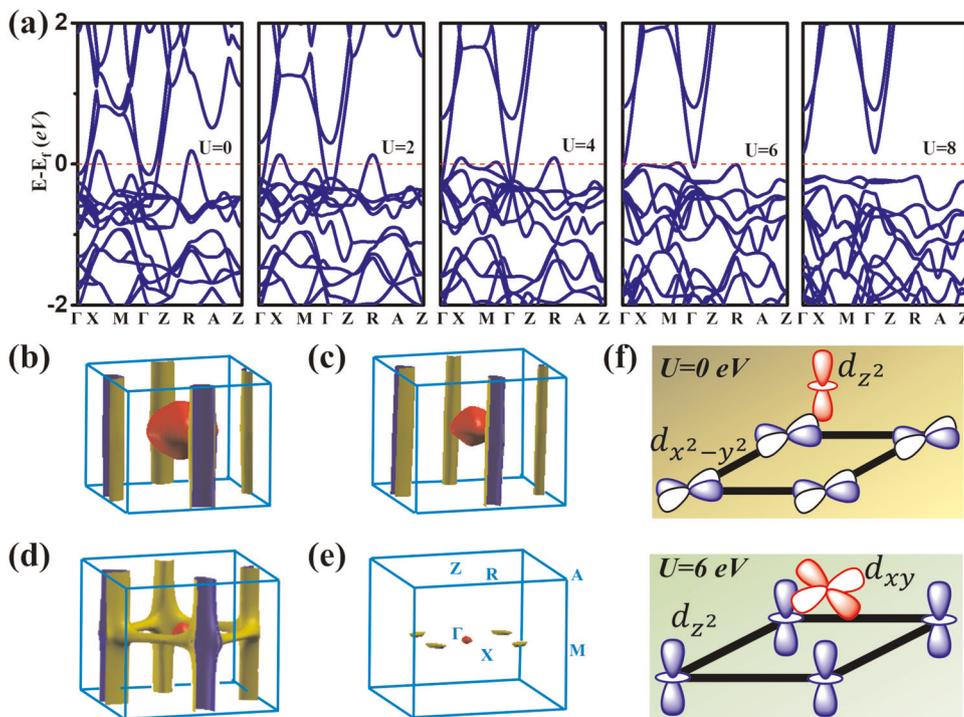


Fig. 3 Band structure and Fermi surface of (π, π, π) AFM phase. **a** Band structures of (π, π, π) AFM phase with different values of Hubbard U . **b–e** Perspective view of the Fermi surfaces of (π, π, π) AFM with $U = 0, 2, 4,$ and 6 eV. The two hole pockets in **b–d** are degenerated and can not be distinguished from this picture. The high symmetry k -points in **e** are labeled to guide the eye. **f** Schematic diagram of major self-doping channel at $U = 0$ and $U = 6$ eV. The red/blue color represents d orbital of Nd/Ni respectively.

extra electron pockets and the inherent interaction between Nd $5d$ and Ni $3d$ electrons. At $U = 0$ eV, there are two electron pockets at Γ point and two hole pockets along X – R direction as shown in Fig. 3a, b. Physically, the origin of these four pockets can be easily understood through the comparison of orbital resolved band structures between PM phase (Fig. 1b) and (π, π, π) AFM phase (Fig. 4). Because of the Zeeman field on Ni, its spin-up and -down bands are split away from each other. The original pure-band ($d_{x^2-y^2}$ orbital of Ni) in PM phase becomes partially occupied in spin-up channel (forming two hole pockets) and totally unoccupied in spin-down channel. Hence, the two hole pockets in AFM phase are inherited from large sheet in PM phase, showing a 2D

character with negligible dispersion along Γ – Z direction. For the electron pockets at Γ point, the heavier one is mainly contributed by d_{z^2} orbital of Nd and Ni, so it comes from the HEP at Γ point of PM phase. While for the lighter one, it comes from the LEP at A point of PM phase which is folded into the Γ point of (π, π, π) AFM phase [see Fig. 1a]. The orbital composition can also be used to check this folded band, which is contributed by d_{xy} orbital of Nd, $d_{xz/yz}$ (d_{z^2}) orbital of spin-down (-up) Ni.

These pockets have a different evolution with the increasing value of Hubbard U . For electron pockets, the heavier one is very sensitive to Hubbard U and disappears at $U = 1$ eV. Meanwhile the lighter one does not disappear until $U = 6$ eV. In addition, the

PM-AFM transition

In order to quantitatively describe such a phenomenon, the phase transition temperature is further calculated. For (π, π, π) AFM phase, the magnetic momentum of Ni increases from $0.58 \mu_B$ ($U = 0$ eV) to $1.04 \mu_B$ ($U = 8$ eV) and becomes gradually saturated, as shown in Fig. 5a. This is also consistent with the fact that $d_{x^2-y^2}$ orbital of Ni is closer to single occupation with the increasing value of Hubbard U . Therefore, Ni is spin one half ($S = 1/2$) in infinite-layer NdNiO₂, just like the case in cuprates. To extract the exchange coupling parameters of J_1, J_2, J_3 , and J_4 (as labeled in Fig. 2a), the total energy of five AFM configurations obtained from DFT + U calculations are mapped onto the Heisenberg spin Hamiltonian. In the $2 \times 2 \times 2$ supercell, there are 8 Ni atoms and the total energy of different AFM configurations are:

$$\begin{aligned} E_{AFM1} &= E_0 - 16J_1S^2 - 8J_2S^2 + 16J_3S^2 + 32J_4S^2 \\ E_{AFM2} &= E_0 - 16J_1S^2 + 8J_2S^2 + 16J_3S^2 - 32J_4S^2 \\ E_{AFM3} &= E_0 + 16J_1S^2 - 8J_2S^2 + 16J_3S^2 - 32J_4S^2 \\ E_{AFM4} &= E_0 + 8J_2S^2 - 16J_3S^2 + 32J_4S^2 \\ E_{AFM5} &= E_0 - 8J_2S^2 - 16J_3S^2 - 32J_4S^2 \end{aligned} \quad (1)$$

where E_0 is the reference energy without magnetic order. Only (π, π, π) and $(\pi, \pi, 0)$ are stable at $U = 0$ eV, and we ignore results at $U = 0$ eV in the following discussions. The calculated exchange coupling parameters as a function of Hubbard U are shown in Fig. 5b. We would like to make several remarks here: (1) the NN intralayer exchange coupling (J_1), mediated by $d_{x^2-y^2}$ orbital of Ni, demonstrates a $1/U$ law; (2) the NN interlayer exchange coupling (J_2) is ~ 10 meV with little variation. The positive value of J_2 indicates an AFM coupling between NiO₂ planes; (3) the next NN intralayer exchange coupling (J_3), mediated by d_{xy} orbital of Nd, is comparable to J_1 at large value of Hubbard U , which is dramatically different to that in infinite-layer SrFeO₂^{32–34}. This large value could be attributed to the relative robustness of lighter electron pocket and orbital purification; (4) the next NN interlayer exchange coupling (J_4) is ~ 0 meV, indicating the validity of our Hamiltonian up to the third NN; (5) J_1, J_2 , and J_3 have the same strength at large U , suggesting NdNiO₂ is a 3D magnet rather than 2D magnet.

Based on the above exchange coupling parameters, the phase transition temperature (T_N) is calculated by classical Monte Carlo method in a $12 \times 12 \times 12$ supercell based on the classical spin Hamiltonian:

$$H = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \quad (2)$$

where the spin exchange parameters J_{ij} have been defined above. First, we calculate the specific heat (C) after the system reaches equilibrium at a each given temperature (T), as shown in Fig. 5c. Then, T is extracted from the peak position in the curve of $C(T)$, as shown in Fig. 5c. For $U = 1$ eV, T_N is as high as 220 K, which can be ascribed to the large value of J_1 . With the increasing value of Hubbard U , T_N gradually decreases and becomes ~ 70 K at $U = 6$ eV. To further check the effect of interlayer exchange coupling on 3D magnet, an additional Monte Carlo calculation is performed without J_2 and J_4 . As shown in Fig. 5e, the $C(T)$ vs T plot shows a broaden peak at a lower temperature. Since Mermin-Wagner theorem prohibit magnetic order in 2D isotropic Heisenberg model at any nonzero temperatures³⁵, the broad peak in $C(T)$ vs T plot implies the presence of short-range order. Regarding the small drop of T_N (~ 30 K in Fig. 5f), our MC simulations indicate that the weak interactions between NiO₂ planes.

Experimental relevance

To address the relevance of the current calculations and experimental observations, we first estimate the value of Hubbard interaction strength U . Although the exact value of Hubbard U cannot be directly extracted from the first-principles calculations, its value range can still be estimated based on similar compounds. The infinite-layer nickelate is believed to be a worse metal compared to elemental nickel with $U \sim 3$ eV³⁶, which can be considered as a lower bound of Hubbard U . The Coulomb interaction in infinite-layer nickelates should be smaller than that in the charge-transfer insulator NiO with $U \sim 8$ eV³⁷, which can be considered as an upper bound of Hubbard U . Therefore, a reasonable value of Hubbard U in NdNiO₂ will be between 3 eV and 8 eV. In the following discussion, we would like to use the median value, $U \sim 5$ –6 eV.

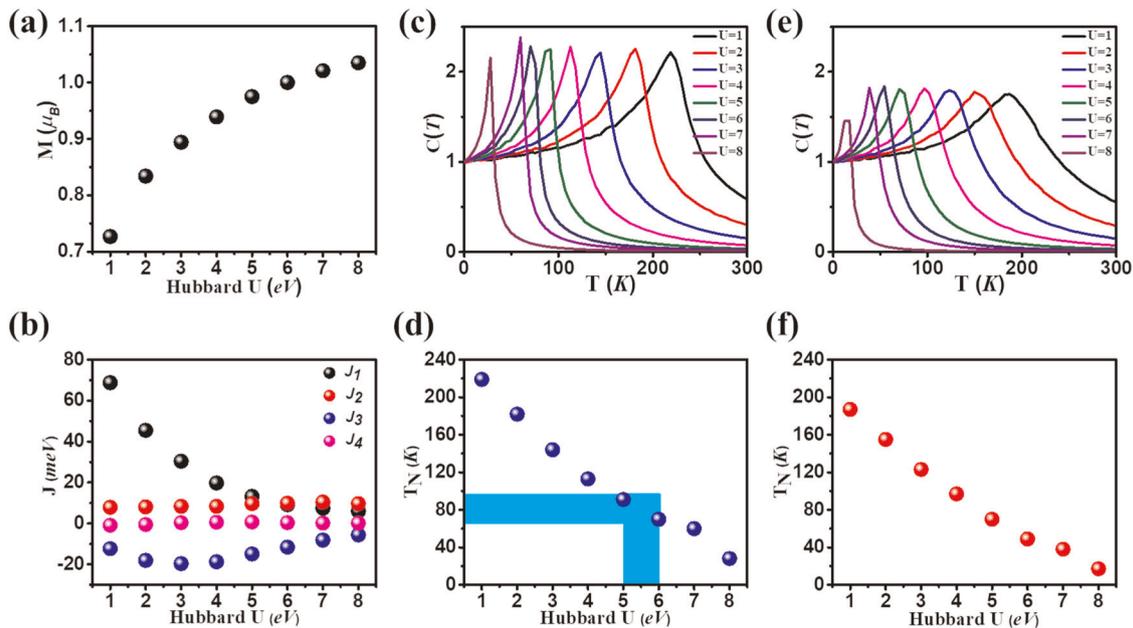


Fig. 5 Dependence of magnetic properties on Hubbard U . The Hubbard U dependence of **a** the magnetic momentum of Ni in (π, π, π) AFM phase, **b** the exchange coupling parameters, **c** specific heat (C) vs T with four J 's, **d** the estimated T_N with four J 's, **e** specific heat C vs T with J_1, J_3 only and **f** the estimated T_N with J_1, J_3 only. The shaded region in **d** highlights the possible T_N of 70–90 K with a reasonable $U = 5$ –6 eV.

Second, if the interaction strength is around $U \sim 5\text{--}6\text{ eV}$, the estimated magnetic exchange interaction is around $J_1 \sim 10\text{ meV}$, $J_2 \sim 10\text{ meV}$, $J_3 \sim -13\text{ meV}$ (see Fig. 5b). This indicates that, effective exchange interactions in NdNiO_2 are about one-order smaller than those of cuprates $\sim 112\text{ meV}$ ^{21,38–44}. Also it results in a relative weaker magnetic ordering and lower Neel temperature T_N in NdNiO_2 , compared with cuprates (see below).

Third, to further discuss the magnetic transition temperature, we show the Neel temperature dependence on interaction strength in Fig. 5d. Within the estimated interaction range $U \sim 5\text{--}6\text{ eV}$, we find the Neel temperature, which separates the PM phase from the (π, π, π) AFM phase, is near $70\text{--}90\text{ K}$. Interestingly, this coincides with the minimum in resistivity and the drop in Hall coefficient⁶. It provides a novel understanding of minimum in resistivity, i.e. partial electrons tend to localize and form magnetic order, therefore the conducting carriers decreases. Thus, our calculation indicates that, the resistivity minimum and the drop in Hall coefficient relates to the magnetic phase transition at the Neel temperature.

Fourth, previous power neutron scattering measurement on NdNiO_2 did not find the signal of AFM ordering^{45,46}. This could be attributed to the following reason. The estimation of Neel temperature (in Fig. 5d) does not include the influence of conduction electrons. We notice that, there are small Nd $5d$ -electron pockets near the Fermi surface at the critical interaction strength (Fig. 3e). By concluding the hybridization effect between Nd $5d$ -electrons and Ni $3d$ -electrons, there are at least two influences. On one hand, it is expected that the local magnetic AFM ordering below the Neel temperature is further weakened. On the other hand, the magnetic phase transition is replaced by a crossover from normal metal to bad AFM metal around $T_N \sim 70\text{--}90\text{ K}$, which is more likely to occur in the experiment⁶. Both of the above effects lead to further weaken the AFM ordering²⁶, which could be hard to detect in neutron scattering measurement.

Lastly, some recent experiments fail to find bulk superconductivity in NdNiO_2 systems, and the parent samples show strong insulating behaviors⁴⁷. The insulating behavior could be attributed to strong inhomogeneous disorder or improper introduction of H during the reaction with CaH_2 ⁴⁸. Especially, it is worth noting, these experiments cannot rule out the possibility of weak AFM ordering, due to the presence of Ni impurities in their samples. (Actually this problem has been pointed out before^{45,46}.) The strong ferromagnetic order from elemental Ni would dominate over and wash out the weak signal of AFM ordering from NdNiO_2 as we suggested in this work.

Taken as a whole, we conclude that, our calculations provide a self-consistent understanding on the experimental observation⁶: the resistivity minimum and drop in Hall coefficient is related to the crossover from normal metal to bad AFM metal around $T_N \sim 70\text{--}90\text{ K}$. The AFM ordering at lower temperature is weak due to the hybridization or self-doping effect, which is hard to detect in the previous measurements. In this regarding, the upcoming inelastic neutron scattering on high-quality samples is highly desired.

DISCUSSION

In this work, we present a systematic study of the electronic and magnetic properties of parent compound NdNiO_2 by first-principles calculations combined with classical Monte Carlo calculations. The Fermi surface of PM phase is quite large with two 3D-like electron pockets, which is consistent with the previous work^{13–16,18}. By tuning the interaction strength, before it enters correlated insulator, NdNiO_2 is a compensated metal with one small electron pocket formed by d_{xy} orbital of Nd and four small hole pockets formed by d_{z^2} orbital of Ni. We identify the interaction selects a (π, π, π) AFM order as the correct magnetic ground state. In the reasonable estimation $U = 5\text{--}6\text{ eV}$ ^{20,37}, we find

several conclusions related to the experiment⁶: (1) the exchange-coupling parameters are $\sim 10\text{ meV}$, which is one order smaller than cuprates^{21,38–44} and results in a low T_N compared with cuprates; (2) the self-doping effect from $5d$ orbital of Nd and $3d$ orbital of Ni may screen the local magnetic momentum in $d_{x^2-y^2}$ orbital of Ni, which gives a small magnetic momentum less than $1\mu_B$ and makes the long-range AFM order unstable^{6,45,49,46}; (3) with the decreasing of temperature, there is a phase transition from PM phase to (π, π, π) AFM phase near $70\text{--}90\text{ K}$. Therefore, there could exist a transition from normal metal to bad AFM metal around $T_N \sim 70\text{--}90\text{ K}$, which provides a plausible understanding of minimum of resistivity and Hall coefficient drop in infinite-layer NdNiO_2 ⁶. We envision that our calculations will intrigue intensive interests for studying the magnetic properties of high-quality infinite-layer NdNiO_2 samples.

METHODS

The first-principles calculations are carried out with the plane wave projector augmented wave method as implemented in the Vienna ab initio simulations package (VASP)^{50–52}. The Perdew–Burke–Ernzerhof (PBE) functionals of generalized gradient approximation (GGA) is used for PM phase⁵³. To incorporate the electron–electron interactions, DFT + U is used for AFM phase, which can reproduce correctly the gross features of correlated-electrons in transition metal oxides^{54–56}. There are two kinds of strongly localized orbitals in NdNiO_2 : $4f$ of Nd and $3d$ of Ni. The $4f$ electrons of Nd are expected to display the local magnetic moment as Nd^{3+} in Nd_2CuO_4 ⁵⁷. The hybridization between f electrons and superconducting plane is very complicated. In $\text{EuRbFe}_4\text{As}_4$ ⁵⁸, superconductivity in FeAs layers and ferromagnetism in Eu layers can coexist. In $\text{PrBa}_2\text{Cu}_3\text{O}_7$, where the local environment of the $4f$ ion is the same as in NdNiO_2 , the antibonding coupling of the $\text{Pr } f_{z(x^2-y^2)}$ orbital to the O $2p$ orbitals makes $\text{PrBa}_2\text{Cu}_3\text{O}_7$ not superconducting⁵⁹. Recently, it has been proposed that the intra-atomic $4f\text{--}5d$ exchange coupling of Nd may change the understanding of the superconducting properties in NdNiO_2 ⁶⁰. Since the local magnetic moment of Nd^{3+} is weakly coupled in NdNiO_2 ⁶⁰, here we ignore the possibility of local moment formed by $4f$ electron of Nd^{3+} in our calculation by treating them as the core-level electrons and focus only on the magnetic order formed by Ni. The Hubbard U ($0\text{--}8\text{ eV}$) term is added to $3d$ electrons of Ni. The energy cutoff of 600 eV , and Monkhorst–Pack k point mesh of $11 \times 11 \times 11$ and $18 \times 18 \times 30$ is used for PM and AFM phase, respectively. The maximally localized Wannier functions (WFs) are constructed by using Wannier90 package^{61,62}. The structure of infinite-layer NdNiO_2 is shown in Fig. 1a, including NiO_2 layers sandwiched by Nd, which can be obtained from the perovskite NdNiO_3 with reduction of apical O atoms in c direction^{45,46}. Due to apical O vacancies, the lattice constant in c direction shrinks (smaller than a direction) and the space group becomes $P4/mmm$. The experimental lattice constant $a = b = 3.92\text{ \AA}$ and $c = 3.28\text{ \AA}$ are used in our calculations.

DATA AVAILABILITY

The numerical datasets used in the analysis in this study, and in the figures of this work, are available from the corresponding author on reasonable request.

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AUTHOR CONTRIBUTIONS

W.Z. and J.Y. proposed the idea. Z.L. carried out the calculations and drafted the article. All authors contribute to the analysis of date, writing, and revision of the manuscript.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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