Three-Dimensional Bond Order in Infinite-Layer Nickelates due to Non-Local Quantum Interference Mechanism

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Recently discovered superconducting infinite-layer nickelates $\text{RNiO}_2$ ($R=$Nd, La, Pr) attracts increasing attention as a similar system to cuprates. Both RNiO$_2$ and YBCO cuprates display the three-dimensional (3D) CDW with wave vector $q \sim (2\pi/3, 0, q_z)$, while $q_z$ is non-zero and incommensurate in the former system. Here, we reveal that the characteristic CDW in RNiO$_2$ is naturally explained as the quantum interference between paramagnons, by focusing on the following characteristics of RNiO$_2$: (i) prominent three-dimensionality in the Fermi surface and (ii) large self-hole-doping ($\sim 14\%$). This mechanism predicts the emergence of the $d_{x^2-y^2}$-wave bond order at a secondary 3D nesting vector $q^c \sim (2\pi/3, 0, q_z^c)$ ($q_z^c = 0.2\pi \sim 0.6\pi$). Based on the paramagnon-interference mechanism, we discuss the CDW orders in nickelates and cuprates on the same footing.

The recent discovery of superconducting infinite-layer nickelates with similar electronic structures and phase diagrams to cuprates has stimulated much attention [1–3]. In fact, infinite-layer nickelates RNiO$_2$ ($R=$Nd, La, Pr) have Ni-3$d^9$ configuration, which is the same as Cu-3$d^9$ in cuprates. A high superconducting transition temperature $T_{SC} \lesssim 30\text{K}$ has been reported in RNiO$_2$ [4], which may be due to the electron structure similar to cuprates.

However, there are many differences between RNiO$_2$ and cuprates. The charge-transfer energy estimated by experiments and theories in RNiO$_2$ is larger than cuprates [5–9]. Thus, RNiO$_2$ is close to the Mott-Hubbard regime, away from the charge-transfer regime in Zaanan-Sawatzky-Allen classification [10]. In addition to the charge-transfer energy, significant differences from cuprates are (i) prominent three-dimensionality in the Fermi surface (FS) and (ii) large self-hole-doping. As for (i), the FS composed of Ni $d_{x^2-y^2}$ orbital in NdNiO$_2$ has three-dimensionality, as shown in Figs. 1(a) and (b), while cuprates have two-dimensional FSs. As for (ii), self-hole-doping ($p_{\text{self}} \sim 0.14$) for Ni $d_{x^2-y^2}$ orbital in NdNiO$_2$ is induced since the FSs of the Nd orbitals emerge [6, 7]. We define an effective hole-doping $p_{\text{eff}} = x + p_{\text{self}}$ in Nd$_{1-x}$Sr$_x$NiO$_2$ to make direct comparison with $p_{\text{eff}} = x$ in cuprates without the self-hole-doping.

Very recently, a three-dimensional (3D) CDW at wave vector $q = (q_x, 0, q_z)$ ($q_z \sim 2\pi/3, q_x \neq 0$) has been observed by RIXS measurements in $R_{1-x}$Sr$_x$NiO$_2$ [11–13]. The value of $q_z \sim 2\pi/3$ corresponds to the period-three in $xy$ plane. The value of $q_x$ and the CDW transition temperature ($T_{\text{c,RIXS}}$) observed by RIXS measurements decrease with $p_{\text{eff}}$. Figure 1(c) shows schematic $p_{\text{eff}}$ dependences of $T_{\text{RIXS}}$ observed in nickelates and overdoped cuprates. $T_{\text{RIXS}} \lesssim 400\text{K}$ in nickelates is higher than $T_{\text{RIXS}} \lesssim 200\text{K}$ in cuprates. In both systems, the CDW quantum critical point (QCP) attracts great attention since $T_{SC}$ becomes the maximum around the CDW QCP. In nickelates, critical hole-doping is $p_{\text{eff}}^c \sim 0.25$ [11], while $p_{\text{eff}}^c \sim 0.18$ in YBCO cuprates [14, 15]. Near the CDW QCP, the non-Fermi-liquid transport phenomena such as $T$-linear resistivity and $T$-dependent Hall coefficient have been observed [16–19].

The electronic states in RNiO$_2$ have been actively studied by using the dynamical mean-field theory (DMFT) [20, 21]. In general, the Mott criticality and mass enhancement are explained by the DMFT, while the CDW formation is nontrivial since the irreducible vertex corrections (VCs) are locally approximated. The pair-density-wave [22–24], the intertwined-order [25, 26], spin-nematic/vestigial-order [27–33], and orbital/bond-order [34–51] scenarios have been proposed to explain the CDW phase. In Refs. [49–52], it was revealed that the bond order and spin current order in cuprates are naturally explained by “the paramagnon-interference mechanism” described by the non-local VCs. The similar theoretical framework has been applied to Fe-based superconductors [38–43], twisted bilayer graphene [53] and kagome metal [54, 55].

In this paper, we study the origin of the 3D CDW in RNiO$_2$ based on the paramagnon-interference mechanism. By focusing on differences (i) prominent three-dimensionality in the FS and (ii) self-hole-doping from cuprates, we analyze the CDW order by using the 3D density-wave (DW) equation. We find that the 3D bond order with wave vector $q^c = (2\pi/3, 0, q_z^c)$ ($0.2\pi \lesssim q_z^c \lesssim 0.6\pi$) emerges, which is consistent with the 3D CDW with $q_z \sim 0.6\pi$ and $q_z \lesssim 0.54\pi$ observed in experiments [13]. The obtained $p_{\text{eff}}$ dependences of the bond order are consistent with experiments [11]. Based on the DW equation, the CDW orders in nickelates and cuprates are understood on the same footing.

We analyze the following 3D three-orbital Hubbard model for NdNiO$_2$, where $d_{x^2-y^2}$ orbital of Ni, $d_{z^2}$ and $d_{xy}$ orbitals of Nd are taken into account:

$$H = H^0 + H^U,$$

(1)

where $H^0$ is the tight-binding model based on the first-principles calculation for NdNiO$_2$ in Ref. [8]. We modify the hopping of Ni $d_{x^2-y^2}$ orbital in order to reproduce the parallel FSs around the M point, which have been...
obtained in other first-principles calculations [9, 56]. Details of the model are explained in Supplementary Material (SM) A [57]. The orbitals 1, 2, and 3 denote Ni $d_{x^2-y^2}$ orbital, Nd $d_{z^2}$ orbital, and Nd $d_{xy}$ orbital, respectively.

Figures 1(a) and (b) show 3D FSs and band dispersion in this model ($x = 0$), where the number of electrons is 1. Since the FSs composed of orbitals 2 and 3 appear, the number of electrons for the Ni $d_{x^2-y^2}$ orbital is about 0.86 (self-hole-doping $p_{\text{eff}} = 0.14$), which is consistent with the self-hole-doping reported in Ref. [6, 7]. In Nd$_{1-x}$Sr$_x$NiO$_2$, the value of $p_{\text{eff}}$ decreases with doping $x$, and $p_{\text{eff}} \sim 0.09$ for $x = 0.15$. Schematic $p_{\text{eff}}$ dependences of $T^\text{RIXS}_c$ observed in nickelates and overdoped cuprates are shown in Fig. 1(c).

Here, we discuss the FSs of the Ni $d_{x^2-y^2}$ orbital, which are important to realize the CDW. Figure 1(d) shows the FSs in $k_z = 0$ plane, where the FS composed of Ni $d_{x^2-y^2}$ orbital is similar to the FS of YBCO cuprates. Figure 1(e) shows the FSs and 3D nesting $q^c \sim (2\pi/3, 0, q^c_z)$ around X point. This secondary nesting between the parallel FSs gives a broad peak structure of the irreducible susceptibility $\chi^s(q)$ along the $q_z$ direction as shown in Fig. S1 in SM A [57], while its height is too small to induce the spin-density-wave order. In addition, the nesting $q^c$ at the FS around R point shown in Fig. 1(f), which is absent in cuprates, assists the secondary nesting. The small instability by the secondary nesting causes the bond-order at $q = q^c \sim (0.2\pi \lesssim q^c_z \lesssim 0.6\pi)$ with the aid of the AL vertex correction in the present theory.

Before discussing the CDW order, we calculate the spin susceptibility $\chi^s(q)$ because we discuss the spin-fluctuation-driven CDW mechanism. We obtain $\chi^s(q)$ for $q = (q_x, \omega_m = 2m\pi T)$ based on the random-phase-approximation (RPA), which is introduced in SM A [57]. $\chi^s(q) \propto (1 - \alpha_s)^{-1}$, where $\alpha_s$ is the spin Stoner factor. $\alpha_s = 1$ corresponds to spin-ordered state. Hereafter, we fix $T = 60\, \text{meV}$ unless otherwise noted.

Figures 2(a) and (b) show the obtained spin susceptibility for orbital 1 $\chi^s_{1,1,1,1}(q, 0)$, which have rather broad peak around $Q_s = (2\pi/3, 2\pi/3, \pi)$. The value of $\chi^s_{1,1,1,1}(q, 0)$ becomes small away from the $q_z = \pi$ plane, and other orbital components of spin susceptibility is very small. In the present study, we set the Coulomb interaction $U_1 = 1.43\, \text{eV}$ for the orbital 1. At $x = 0$ ($p_{\text{eff}} = 0.14$), rather strong spin fluctuations, $\alpha_s = 0.95$ at $T = 60\, \text{meV}$, are obtained. The value of $\alpha_s$ decreases with $p_{\text{eff}}$. At $x = 0.15$ ($p_{\text{eff}} = 0.24$), moderate spin fluctuations, $\alpha_s = 0.91$ at $T = 60\, \text{meV}$, are obtained. These moderate spin fluctuations correspond to the experimental situation for $x = 0.15$, where $1/T_1 T$ moderately increases for $T \lesssim 100\, \text{K}$ [58]. Thus, $U_1 = 1.43\, \text{eV}$ used in the present study is reasonable.

Next, we analyze the CDW state in NdNiO$_2$ based on the charge-channel DW equation [39, 41, 52]. A rigorous formalism of the DW equation has been constructed based on the Luttinger–Ward (LW) theory in Ref. [59]. The solution of the DW equation gives the minimum of the grand potential in the LW theory, and therefore it is thermodynamically stable. The optimized non-local form factor $f^q(k)$, which describes the DW order parameter, is derived from the following linearized DW equation:

$$
\lambda_\alpha \hat{f}^q(k) = \frac{T}{N} \sum_{k'} \hat{f}^q(k, k') \hat{f}^q(k, k'),
$$

(2)

$\lambda_\alpha$ is the eigenvalue of the form factor $f^q(k)$, $\hat{f}^q(k, k') \equiv G_{lm}(k + \frac{q}{2}) G_{m'\, l'}(k - \frac{q}{2})$, $i\hat{f}^q(k, k')$ is the four-point vertex, and $k = [k, \epsilon_n = (2n + 1)\pi T]$. The charge-channel DW with wave vector $q$ is established when the largest $\lambda_\alpha = 1$. The DW susceptibility
FIG. 2. (a) $q$ dependences of $\chi^s_{1,1,1}(q, 0)$ given by the RPA on $q_z = \pi$ plane, and (b) that on $q_y = \pi$ plane in NdNiO$_2$. (c) Feynman diagram of AL1 term in the DW equation, where the wavy lines represent the spin fluctuations. Broad and incommensurate peak structure of $\chi^s(q, 0)$ at $q \sim Q_x$ gives rise to the charge-channel DW instability at a short wavelength $q^*$. AL1

\[
\frac{f^q_{\text{AL1}}(k)}{p, m} = \frac{1}{k + q/2, m} \quad k = q/2 - \frac{Q}{2}
\]

is proportional to $(1 - \lambda_q)^{-1}$ [59]. Therefore, $\lambda_q$ represents the strength of the DW instability. In the DW Eq. (2), the Maki–Thompson (MT) terms and Aslamazov–Larkin (AL) terms are included in the four-point vertex, as we explain in SM A [57]. When $\chi^s(q)$ is large, the AL term in Fig. 2(c) is strongly enhanced in proportion to $\sum_p \chi^s(p + q^*)\chi^s(p)$. As a result, the quantum-interference mechanism causes the charge-channel DW order at $q^* \approx Q - Q'$, where $Q$ and $Q'$ are close to the paramagnon wave vector $Q_s$ [39, 41, 52].

Figures 3(a) and (b) show the $q$ dependence of the obtained $\lambda_q$, which shows the highest value at the 3D nesting vector $q^* \approx (2\pi/3, 0, q^*_z)$ $(0.2\pi \lesssim q^*_z \lesssim 0.6\pi)$. From the Fourier transformation of $f^q(k)$ shown in SM B [57], the obtained order is identified as the $d_{xy}$-wave bond order. The bond order modulated period-three in $xy$ plane as shown in Fig. 3(c) due to the $q_x$ component of $q^*$ ($q^*_x \approx 2\pi/3$). In the bond-ordered state, the hopping integrals are modulated (the hopping integrals increase at the red bonds, and those decrease at the blue bonds). The obtained period-three bond order is consistent with the experiments [11–13]. Since $q^*$ has $q_z$ component $q^*_z$, the bond order also has a modulation along the $z$ direction, which is consistent with the 3D CDW with $q_z \approx 0.6\pi$ and $q_x \lesssim 0.54\pi$ observed in Ref. [13].

In the following, we explain the decisive role of the non-locality of the irreducible four-point vertex on the 3D bond order [38–44, 52]. As discussed in Ref. [52], in the presence of moderate spin fluctuations, the AL terms give strong attraction between the Fermi momenta $k$ and $k' = \pm k$ in the DW Eq. (2), which leads to the relation $f^{\text{AL1}}_q(k)f^{\text{AL1}}_q(k') > 0$ for $k' = \pm k$. Thus, the AL terms strongly enhance the instability of various even-parity ($f(k) = f(-k)$) DW states. In addition, the MT term gives moderate repulsion between the Fermi momenta $k$ and $k' = k + Q_s$, which leads to the relation $f^{\text{MT}}_q(k)f^{\text{MT}}_q(k + Q_s) < 0$ at $Q_s \sim (2\pi/3, 2\pi/3, \pi)$ [52]. The MT term favors the $d$-wave form factor with sign reversal, as shown in Figs. S2(a) and (b) in SM B [57]. Due to the cooperation between the attraction by the AL terms and the repulsion by the MT term, the $d_{xy}$-$d_{xz}$-wave bond order is naturally realized at high transition temperature. We note that the non-local and non-$s$-wave DW states cannot be obtained when locally approximated $f^{\text{local}}_q(\epsilon_n, \epsilon_m) = \sum_{k, k'} f^q(k, k')$ is applied, even if the AL and MT terms are taken into account [60].

Here, we also explain the importance of the secondary 3D nesting with short wavelength shown in Fig. 1(e) and (f). This secondary nesting assists the $q^*$ bond order. $\tilde{g}^{q}_{1,1,1}(k)$ in the DW Eq. (2) becomes large when both $k + \frac{Q}{2}$ and $k - \frac{Q}{2}$ locate on the FSs. The existence of this secondary nesting is well recognized in a broad peak structure of $\chi^s_{1,1,1}(q)(x \sum_k \tilde{g}^{q}_{1,1,1}(k))$, as we show in Fig. S1 in SM A [57]. The existence of the secondary nesting stabilizes the 3D CDW in NdNiO$_2$.

We stress that the $q = \mathbf{0}$ DW orders have been realized in various systems such as cuprates, Fe-based superconductors, and twisted-bilayer graphene, and they are naturally explained by the paramagnon-interference mechanism. The $q$ dependence of DW order is sensitive to the structure of FSs. The three-dimensionality in FSs
might be important to understand the absence of \( q = 0 \) order. Clarifying the presence or absence of \( q = 0 \) order in \( RNiO_2 \) is an important future problem.

Here, we discuss the doping \( x \) dependence of the CDW in \( Nd_{1-x}Sr_xNiO_2 \). In this study, the hole-doping \( x \) is introduced by the rigid-band shift. Figure 4(a) shows \( x \) dependence of \( q_x^{\text{max}} \). \( q_x^{\text{max}} \) is defined as \( q_x \) component, where \( \lambda_q \) becomes maximum. The value of \( q_x^{\text{max}} \) decreases with hole-doping \( x \) since the distance of the two FSs around the M point, which is related to the 3D nesting \( q^z \), decreases with \( x \). This \( x \) dependence is consistent with the experimental results [11].

In order to derive the theoretical transition temperature \( T_{c}^{\text{DW}} \), we calculate the \( T \) dependences of the maximum value of \( \lambda_q \) (= \( \lambda_{\text{max}} \)) and \( \alpha_s \). As shown in Fig. 4(b), \( \lambda_{\text{max}} \) reaches 1 at \( T_{c}^{\text{DW}} = 67\text{meV} \) for \( x = 0 \) \( (p_{\text{eff}} = 0.14) \), while the spin-ordered state is absent because \( \alpha_s < 1 \).

In the present 3D calculation, we employ \( N = 48^3 \) \( k \) meshes and 512 Matsubara frequencies. In this case, the calculation accuracy is ensured for \( T \geq 60\text{meV} \), which is higher than the experimental \( T_{c}^{\text{RIXS}} \). However, \( T_{c}^{\text{DW}} \lesssim 60\text{meV} \) can be obtained correctly by extrapolating the accurate results for \( T \geq 60\text{meV} \).

Figure 4(c) shows the obtained \( T \) dependence of \( \lambda_{\text{max}} \) for each \( x \) for \( T \geq 60\text{meV} \). \( T \)-linear behavior of \( \lambda_{\text{max}} \) is well reproduced, which is characteristic behavior ensured in the DW Eq. [59]. Particularly, very accurate \( T \)-linear behaviors of \( \lambda_{\text{max}} \) are obtained for \( x \leq 0.05 \). By extrapolating the \( T \)-linear \( \lambda_{\text{max}} \) shown by dotted line, we obtain reliable \( T_{c}^{\text{DW}} \) for each \( x > 0 \).

Figure 4(d) shows the obtained \( x \) dependence of \( T_{c}^{\text{DW}} \). This \( x \) dependence of long-range order \( T_{c}^{\text{DW}} \) well reproduces \( T_{c}^{\text{RIXS}} \) observed in the RIXS measurements [11–15]. At the bond-order QCP \( (T_{c}^{\text{DW}} = 0) \), the non-Fermi-liquid transport phenomena and the strong pairing interaction are induced by the bond-order fluctuations [54]. In fact, non-Fermi-liquid transport phenomena [16, 17] and the enhancement of \( T_{\text{SC}} \) and \( H_{c2} \) have been observed near the bond-order QCP.

In the present study, the obtained \( q_z^c \) of the 3D CDW is incommensurate. In contrast, \( q_z = 0 \) of the 3D CDW has been observed in YBCO cuprates under a large magnetic field and a uniaxial strain [61–65]. This significant difference is understood by the presence or absence of the 3D secondary nesting. We note that the RIXS peak \( q_z \sim \pi \) in the 2D CDW phase in YBCO is much broader along the \( q_z \) direction than that of \( NdNiO_2 \) [13, 63–65]. This fact means the realization of the 3D CDW in \( NdNiO_2 \).

Hereafter, we discuss differences in the CDW quantum critical behaviors between \( RNiO_2 \) and cuprates. The CDW in cuprates has been identified as the bond order by the paramagnon-interference mechanism [49–51]. Experimentally observed \( T_{c}^{\text{RIXS}} \lesssim 400K \) and \( p_{\text{eff}}^{\text{eff}} \lesssim 0.25 \) in \( RNiO_2 \) are larger than \( T_{c}^{\text{RIXS}} \lesssim 200K \) and \( p_{\text{eff}} \lesssim 0.18 \) in cuprates as shown in Fig. 1(c). Thus, the CDW instability at a fixed \( p_{\text{eff}} \) in \( RNiO_2 \) is stronger than that in cuprates. These differences are understood by the strength of the Coulomb interaction: \( U_1 = 3.8eV \) in \( RNiO_2 \) given by the first-principles calculation is larger than \( U_1 = 2.6eV \) in Hg cuprates [9]. In fact, cuprates belong to the charge-transfer regime, while \( RNiO_2 \) belongs to the Mott-Hubbard regime [10]. In addition, the 3D bond order in \( RNiO_2 \) is stabilized by the 3D nesting \( q^z \) around R point shown in Fig. 1(f), which is absent in cuprates. For these reasons, the CDW (bond-order) instability in \( RNiO_2 \) is stronger than that in cuprates.

Note that the obtained long-range-order \( T_{c}^{\text{DW}} \) may be overestimated when the self-energy is not taken into account. In FeSe, the nematic transition temperature \( \sim 100K \) is well reproduced by introducing the self-energy [59]. It is an important future problem to discuss the self-energy effect for \( RNiO_2 \). By considering the self-energy, the required model parameter \( U_1 \) will approach the first principles value 3.8eV [41–43].

We comment on the recent developments of theories. The cluster DMFT and the density matrix renormalization group theories have recently developed and solved exactly finite-size systems [66, 67]. These theories can take various quantum interference effects into account, and therefore they are complementary to the present field-theoretical approach.

In summary, we studied the origin of the 3D CDW in \( RNiO_2 \) based on a realistic 3D Hubbard model. We found that the 3D CDW is identified as the \( d \)-wave bond

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**FIG. 4.** (a) \( x \) dependences of \( q_x^{\text{max}} \) in \( Nd_{1-x}Sr_xNiO_2 \). (b) \( T \) dependences of \( \lambda_{\text{max}} \) and \( \alpha_s \) for \( x = 0 \) \( (p_{\text{eff}} = 0.14) \). (c) Obtained \( T \) dependences of \( \lambda_{\text{max}} \) for \( x = 0, 0.025, 0.05, 0.075, 0.1 \), where the dotted lines are fitted by the least squares. (d) \( x \) dependences of \( T_{c}^{\text{DW}} \) and \( T_{c}^{\text{RIXS}} \). Lines are calculation results, while green dots represent experimental results in Ref. [11].
order with the wave vector \( \mathbf{q}^c \sim (2\pi/3, 0, q^c_z) \) \((0.2 \pi \lesssim q^c_z \lesssim 0.6 \pi)\). This 3D bond order with the period-three in \( xy \) plane is driven by the paramagnon-interference mechanism, and it is further stabilized by the secondary 3D short-wavelength nesting shown in Figs. 1(e) and (f). The doping dependences of the CDW order have been well reproduced by the present mechanism. Based on the DW equation, the CDW orders in nickelates and cuprate are understood on the same footing.

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A: Model Hamiltonian of NdNiO$_2$, formalism of the RPA and the DW equation

First, we introduce a tight-binding model for NdNiO$_2$ by referring the first-principles tight-binding model in Ref. [1]. We modify the hoppings of Ni $d_{x^2-y^2}$ orbital in order to reproduce the parallel FSs along the $z$-direction around the M point, which have been obtained in other first-principles calculations [2, 3]. We modify the intralayer next-nearest-neighbor hopping $t_2$, the intralayer third-nearest one $t_3$, and interlayer nearest one $t_z$ to $t_2/t_1 = -1/3$, $t_3/t_1 = 0.2$, and $t_z/t_1 = 2/3$ ($t_1$ is the nearest-neighbor hopping), respectively. These intralayer hoppings are similar to those in YBCO cuprate.

Here, we explain the Coulomb interaction introduced in the present study. Ni $d_{x^2-y^2}$ orbital, Nd $d_{xy}$ orbital, and Nd $d_{yz}$ orbital are included in our model. Ni has a single orbital, while Nd has two orbitals. We introduce the on-site Coulomb interaction $U_1$ for Ni $d_{x^2-y^2}$ orbital. The Coulomb interaction for the spin channel in the Nd orbitals 2 and 3 is

$$ (\Gamma^a)_{l_1l_2l_3l_4} = \begin{cases} U, & l_1 = l_2 = l_3 = l_4 \\ U', & l_1 = l_3 \neq l_2 = l_4 \\ J, & l_1 = l_2 \neq l_3 = l_4 \\ J, & l_1 = l_4 \neq l_2 = l_3 \\ 0, & \text{otherwise}, \end{cases} \tag{S1} $$

and that in the orbital 1 is $\Gamma^a_{1111} = U_1$. Furthermore, the Coulomb interaction for the charge channel in the orbitals 2 and 3 is

$$ (\Gamma^c)_{l_1l_2l_3l_4} = \begin{cases} -U, & l_1 = l_2 = l_3 = l_4 \\ U' - 2J, & l_1 = l_3 \neq l_2 = l_4 \\ -2U' + J, & l_1 = l_2 \neq l_3 = l_4 \\ -J, & l_1 = l_4 \neq l_2 = l_3 \\ 0, & \text{otherwise}, \end{cases} \tag{S2} $$

and that in the orbital 1 is $\Gamma^c_{1111} = -U_1$. By using the Coulomb interaction, the spin (charge) susceptibility in the RPA is given by

$$ \hat{\chi}^{s(c)}(q) = \hat{\chi}^{0}(q)[1 - \hat{\Gamma}^{s(c)}(q)]^{-1}, \tag{S3} $$

where the irreducible susceptibility is

$$ \chi^{0}_{l,l';m,m'}(q) = -\frac{T}{N} \sum_{k} G_{l,m}(k + q)G_{m',l'}(k). \tag{S4} $$

$\hat{G}(k)$ is the multiorbital Green function without self-energy $\hat{G}(k) = [(i\epsilon_n - \mu)\mathbb{1} - \hat{h}^{0}(k)]^{-1}$ for $= (2n + 1)\pi T$. Here, $h^{0}(k)$ is the matrix expression of $H^{0}$ and $\mu$ is the chemical potential.

Figure S1 shows $q_z$ dependence of $\chi^{0}_{1,1,1,1}(q_x,0,q_z)$ for each $q_z$. $\chi^{0}_{1,1,1,1}(q_x,0,q_z)$ has a broad peak at $(2\pi/3,0,q_z)$ for $q_z < 2\pi/3$, which is induced by the secondary nesting in Fig. 1(e).

![Figure S1. $q_z$ dependence of $\chi^{0}_{1,1,1,1}(q_x,0,q_z)$ for each $q_z$ for $x = 0$.](image)

The spin (charge) Stoner factor $\alpha_{s(c)}$ is defined as the maximum eigenvalue of $\Gamma^{s(c)}(q)\hat{\chi}^{0}(q,0)$.

In the present study, we set $U_1 = 1.43eV, U/U_1 = 2/3$, $J/U = 0.15$, and $U' = U - 2J$. The values of $U/U_1 = 2/3$ and $J/U = 0.15$ are consistent with the first-principles calculations [2, 4]. The value of $U_1 = 1.43eV$ is smaller than $U_1 = 3-4eV$ obtained by the first-principles calculations [2, 4] since the self-energy is not included in the present study. In order to decide the value of $U_1$, we set the value of $\alpha_{s} \sim 0.95$, which represents the strength of the spin fluctuations. We use $N = 48 \times 48 \times 48$ $k$ meshes and 512 Matsubara frequencies.

The four-point vertex $i^{q}(k,k')$ in the DW Eq. (2) is given as

$$ I_{l,l';m,m'}^{q}(k,k') = \sum_{b=s,c} \frac{a_{b}^{2}}{2} V_{l,m,l',m'}^{b}(k-k') + \frac{T}{N} \sum_{p,l_1,l_2,m_1,m_2} \frac{a_{b}^{2}}{2} V_{l_1,m_1,l_2,m_2}^{b} \left( p \frac{q}{2} \right) V_{m_1,l_2,m_1}^{b} \left( p \frac{q}{2} \right). \tag{S5} $$

[Supplementary Material]

Three-Dimensional Bond Order in Infinite-Layer Nickelates due to Non-Local Quantum Interference Mechanism

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\[ \times G_{l_1m_1}(k - p)G_{l_2m_2}(k' - p) + \frac{T}{N} \sum_{p,j_1,j_2,m_1,m_2} a^b \frac{V^b_{j_1j_2m_1'}(p + \frac{q}{2})}{V^b_{m_2m_1}} \times G_{l_1m_1}(k - p)G_{l_2m_2}(k' + p)], \] where \( a^s = 3, a^c = 1, p = (p, \omega), \) and \( \hat{V}^{s(c)}(q) = \hat{\Gamma}^{s(c)} + \hat{\chi}^{s(c)}(q)\hat{\Gamma}^{s(c)}. \)

In Eq. (S5), the first line corresponds to the MT term, and the second and third lines give the AL1 and AL2 terms, respectively. In the MT term, the first-order term with respect to \( \hat{\Gamma}^{s,c} \) gives the Hartree–Fock (HF) term in the mean-field theory.

**B: Detailed results of the form factor**

Here, we explain details of the obtained \( q^c = (2\pi/3, 0, 0.4\pi) \) form factor. Figure S2(a) and (b) show the form factors of orbital 1 \( f^{q_0^c}_{11}(k) \), which is derived from the analytic continuation of \( f^{q_0^c}_{11}(k) \). FSs shifted by \( \pm \frac{q_0^c}{2} \) are shown by green and orange lines. The form factor has a large value at \( k \), where the two shifted FSs overlap since the \( g^{q_0^c}_{11,11}(k') \) in the kernel function is enlarged there. From the \( k \)-dependence of the form factor, we find that the form factor corresponds to the \( d_{x^2-y^2} \)-wave bond order. In order to study the real space structure of the bond order, we calculate a Fourier transformed form factor \( \tilde{f}_{11}(r) \) given as

\[ \tilde{f}_{11}(r) = \sum_k f^{q^c}_{11}(k)e^{-ik\cdot r}. \] (S6)

The obtained \( \tilde{f}_{11}(r) \) is shown in Figs. S2(c) and (d). The values at the nearest neighbors from \( r = 0 \) are dominant. Sign of \( \tilde{f}_{11}(\pm 1, 0, 0) \) is opposite to that of \( \tilde{f}_{11}(0, \pm 1, 0) \), which means the \( d_{x^2-y^2} \)-wave bond order in \( xy \) plane. Since the magnitude of \( \tilde{f}_{11}(0, 0, \pm 1) \) along \( z \) direction is similar to that of \( \tilde{f}_{11}(\pm 1, 0, 0) \), the bond order has 3D structure. In addition, the small value of \( \tilde{f}_{11}(0) \neq 0 \) corresponds to the slight charge order.

The modulation of hopping \( \delta t_{i,j} \) is given by

\[ \delta t_{i,j} = 2\tilde{f}_{11}(r_i - r_j) \cos \left[ \frac{q^c}{2} \cdot (r_i + r_j) \right]. \] (S7)

The obtained period-three \( \delta t_{i,j} \) bond order around \( z = 0 \) plane is shown in Fig. 3(c). Since \( q^c \) has \( q_z \) component \( 0.2\pi \lesssim q_z^c \lesssim 0.6\pi \), the bond order also has a modulation along the \( z \) direction.

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