Optical properties of the infinite-layer La$_{1-x}$Sr$_x$NiO$_2$ and hidden Hund’s physics

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We investigate the optical properties of the normal state of the infinite-layer La$_{1-x}$Sr$_x$NiO$_2$ using DFT+DMFT. We find a correlated metal which exhibits substantial transfer of spectral weight to high energies relative to the density functional theory. The correlations are not due to Mott physics, which would suppress the charge fluctuations and integrated optical spectral weight as we approach a putative insulating state. Instead we find the unusual situation, that the integrated optical spectral weight decreases with doping and increases with increasing temperature. We contrast this with the coherent component of the optical conductivity, which decreases with increasing temperature as a result of a coherence–incoherence crossover. Our optical studies support a picture of a Hund’s metallic state, where dynamical orbital fluctuations are visible at intermediate energies, even if at low energies the Fermi surface has primarily $d_{x^2-y^2}$ character and we propose a low-energy two-band model with atom centered $e_g$ states.

**Introduction**— The recent discovery of superconductivity in the infinite-layer nickelates, Nd$_{1-x}$Sr$_x$NiO$_2$ [1], has attracted intensive interests due to material similarities with high-$T_c$ cuprate superconductors. Several follow-up experiments confirmed the superconductivity [2–6], with some possibly contradictory observations [7, 8]. Nomura et al. estimated the electron-phonon coupling mediated $T_c$ to be $\sim 0.1$ K [17], much less than the observed $T_c \approx 15$ K, showing the mechanism for superconductivity is unconventional thus electron correlations play an important role.

There are many experimental investigations of these systems [1–16]. Besides, multiple theoretical techniques have been applied to study electronic structure of the infinite-layer nickelate [17–28, 30–49]. On the theory side, three different views of this material are emerging. In the first one, the infinite-layer nickelate has a cuprate-like correlated $d_{x^2-y^2}$ band near a Mott transition and an additional uncorrelated “spectator” band near the Fermi level which provides self-doping supported by density functional theory (DFT) [17–21], DFT plus dynamical mean-field theory (DFT+DMFT) [22–24], and model calculations [21, 25, 26]. In the second one, it has been suggested that multiorbital effects are important as for example Hund’s physics, using DFT+DMFT [27], GW+DMFT [28, 29], and model studies [30–33]. A third approach invokes Kondo physics between correlated and uncorrelated bands. This has been supported by DFT [34], DFT+Gutzwiller [35], DFT+DMFT [35, 36], and model calculations [37, 38].

Optical conductivity experiments have been very useful in identifying the origin and nature of electronic correlations in different archetypical systems [50]. Here, we employ DFT+DMFT [51–53] in a broad energy window to investigate the infinite-layer LaNiO$_2$ system and find an optical response is very different from canonical Mott-Hubbard systems. Instead our results support a picture of a Hund’s metallic state. The correlations arise from Hund’s coupling $J_H$, the low-energy bands can be reproduced by a two-band model with atom centered $e_g$ bands, and the valence histogram favors the highest possible spin configuration in the $e_g$ manifold. This Hund’s metal is very anisotropic in orbital space, with a highly correlated $d_{x^2-y^2}$ orbital and an $d_{z^2}$ orbital with weaker correlations.

**Methods**— We perform fully charge self-consistent DFT+DMFT calculations [54] implemented in the all-electron full-potential Wien2k package [55] with exact double counting scheme [56]. We choose a large energy window from -10 eV to 10 eV with respect to the Fermi level $E_F$ in order to describe both low and high energies precisely. The fully rotational invariant form is applied for a local Coulomb interaction Hamiltonian with on-site Coulomb repulsion $U = 5$ eV and Hund’s coupling $J_H = 1$ eV [57]. This approach was recently shown to give results consistent with the occupancies measured in high-energy spectroscopies [14, 27, 28]. The computational details is provided in Supplemental Material [58].

**Results: Optical Conductivity**— The DMFT optical conductivity is computed with the formalism presented in Refs. [54, 58] and is shown in Fig. 1. The DFT optical conductivity is provided for comparison. The optical conductivity consists of a Drude weight and interband transitions at $\sim 3.5$, $\sim 6$, and $\sim 8.5$ eV. The former corresponds to a transition from Ni 3d to La 4f orbitals and the last two correspond to transitions from O 2p to La 4f orbitals [58].

The temperature($T$)-dependent optical conductivity is displayed in Fig. 2(a). The Drude peak develops gradually upon cooling, resulting in a decrease of the resistivity $\rho$ as shown in the inset of Fig. 2(a). The computed $\rho$ follows a $T^2$ behavior, found experimentally at intermediate temperatures [12]. At lower temperatures a resistivity upturn below $T \sim 100$ K is observed in ex-
periments [1, 12, 13] which we ascribe to disorder effects which are not included in the calculations.

Results: Integrated Optical Spectral Weight—To understand the physics of this material we analyze the integrated spectral weight $K(\Omega) = \int_0^\Omega \sigma_1(\omega) d\omega$ as a function of a cutoff frequency $\Omega$ [50]. Figure 2(b) displays $K(\Omega)$ for DMFT normalized with that for DFT to present the kinetic energy ratio between DFT and DMFT. The ratio of $K_{\text{DMFT}}/K_{\text{DFT}}$ decreases upon heating for low cutoff $\Omega$ (less than $\sim 50$ meV) as a result of the broadening of the Drude peak. However the total integral over the Drude peak (up to $\Omega = 0.369$ eV) increases with increasing temperature, which can be understood as a result of the quasiparticles becoming lighter as shown in Fig. 2(c). The total spectral weight of the Drude peak in DMFT is less than in DFT by 0.58 for $T = 116$ K. Part of the lost weight in the Drude peak is transferred to a low-energy interband transition around $\sim 0.5$ eV as shown in Fig. 1.

We see that the ratio of $K_{\text{DMFT}}/K_{\text{DFT}}$ depicted in Fig. 2(b) is less than one. This demonstrates the significance of electronic correlations which reduces the electronic kinetic energy. For LaNiO$_2$, $K_{\text{DMFT}}/K_{\text{DFT}} = 0.5 - 0.6$, thereby suggesting that it is a (moderately) correlated metal. The kinetic energy ratio is comparable to Hund's metal compounds of LaFePO and SrRuO$_3$ [50]. It is noteworthy that $K_{\text{DMFT}}/K_{\text{DFT}} \approx 0$ for cuprates of La$_2$CuO$_4$ and Nd$_2$CuO$_4$, those are charge-transfer insulators, and $\sim 0.2$ for La$_{2-x}$Sr$_x$CuO$_2$ ($x = 0.1, 0.15, 0.2$) [66]. In addition, in the paramagnetic metallic phase of V$_2$O$_3$, which is a prototypical Mott system, $K_{\text{DMFT}}/K_{\text{DFT}} \approx 0.2$ [50]. Based on the values of $K_{\text{DMFT}}/K_{\text{DFT}}$, LaNiO$_2$ is far from a Mott system, but close to a Hund's metal.

Notice that the behavior of $K_{\text{DMFT}}/K_{\text{DFT}}$ of LaNiO$_2$ as a function of temperature (when $\Omega$ is large) is the opposite of what is observed in canonical Mott insulating systems such as V$_2$O$_3$ where $K_{\text{DMFT}}(\Omega)$ (or $K_{\text{DMFT}}/K_{\text{DFT}}$) decreases upon heating (within the paramagnetic metallic phase) [65] (see details in Supplemental Material [58]).

This reflects the fact that the kinetic energy is reduced as an insulating state is approached at higher temperatures. Therefore, LaNiO$_2$ is far from a Mott system and closer to a Hund’s system such as BaFe$_2$As$_2$ [67].

Results: Orbital Character—We now turn to the orbital character of the different optical features. First we analyze the quasiparticle weight $Z$ as a function of $T$ as depicted in Fig. 2(c) [68]. Ni $d_{x^2-y^2}$ has the smallest and drastically different $Z$ from other 3$d$ orbitals that have $Z \approx 0.8$. Besides, it exhibits strong temperature dependence: $Z$ increases linearly upon heating. However, other orbitals have very weak or no temperature depen-
dence as shown in the inset of Fig. 2(c). Hence, the correlated \( d_{x^2-y^2} \) shows strong temperature dependence but the others are almost temperature independent, thereby presenting orbital differentiation clearly.

The Drude peak could be decomposed into two characters: the correlated Ni \( d_{x^2-y^2} \) and an uncorrelated hybridized band which includes Ni \( d_{z^2} \) and \( d_{xz}/dy_z \) orbitals [58]. It illustrates the multiorbital feature of LaNiO\(_2\). The dominant component of the Drude peak is the correlated \( d_{x^2-y^2} \) which exhibits strong temperature dependence as shown in \( Z \). The remaining contribution originates from the uncorrelated hybridized band that are almost temperature independent. Therefore, \( T \)-dependent width of the Drude peak is almost solely determined by the electronic correlation exhibited in the \( d_{x^2-y^2} \) orbital.

To gain more insight into the optics of the infinite-layer LaNiO\(_2\), the effective plasma frequency \( \omega_p^2 \) and the effective scattering rate \( 1/\tau_{ir}^p \) are extracted from the computed optical conductivity [65]. Recall that the Drude peak could be decomposed into two characters that are the correlated \( d_{x^2-y^2} \) and the uncorrelated hybridized band. The dc conductivity, therefore, can be written as a sum of each band contribution: \( \sigma = \sum_i (\omega_{pi}^2)^2 \tau_{ir}^i / 4\pi \), where \( i \) is a band index. Hence it is the low-frequency analysis (related to the extended Drude analysis) and is free from the cutoff \( \Omega \) in the partial sum rule. Figures 2(d) and (e) show \( (\omega_p^2)^2 \) and \( 1/\tau_{ir}^i \) for each band component as a function of temperature. The temperature dependence in \( (\omega_p^2)^2 \) and \( 1/\tau_{ir}^i \) is directly related to that in \( Z \) and the quasiparticle scattering rate \( 1/\tau_{qp} = -Z\text{Im} \Sigma(i\omega^p) \) as indicated in Ref. [65, 69]. Interestingly, \( (\omega_p^2)^2 \) and \( 1/\tau_{ir}^i \) for the uncorrelated hybridized band are almost temperature independent and those for \( d_{x^2-y^2} \) exhibit strong temperature dependence: particularly \( (\omega_p^2)^2 \) has a linear temperature dependence up to \( T \approx 300 \) K and then a saturation above the temperature. The saturation behavior in \( (\omega_p^2)^2 \) is also found in ruthenates [69] and is explained by coherence-incoherence crossover (see details in Supplemental Material [58]). \( (\omega_p^2)^2 \) increases upon heating until \( T \) is below the coherent temperature of \( T \approx 400 \) K and then shows the saturation behavior above the coherent temperature. In addition, \( 1/\tau_{ir}^i \) for \( d_{x^2-y^2} \) is approximately parabolic in temperature below the coherent temperature, thereby presenting Fermi liquid behavior at the temperature range. However, it shows a deviation from the quadratic behavior above the coherent temperature.

**Results: Doping Dependence**— Now, we turn our attention to Sr-doped LaNiO\(_2\), that is La\(_{1-x}\)Sr\(_x\)NiO\(_2\), where Sr doping provides holes to LaNiO\(_2\). Figure 3(a) shows the doping-concentration\((x)\)-dependent optical conductivity of La\(_{1-x}\)Sr\(_x\)NiO\(_2\). The Drude peak is gradually diminished upon doping and the integrated optical spectral weight \( K(\Omega) \) decreases accordingly as depicted in Fig. 3(b). Since \( K(\Omega) \) is proportional to the electronic kinetic energy, it indicates the decrease of the kinetic energy upon doping. In the case of a Mott system where doping triggers the Mott transition, doping releases localization of electrons, thereby increasing the electronic kinetic energy. Hence, it increases \( K(\Omega) \) upon doping [50, 70]. However, in La\(_{1-x}\)Sr\(_x\)NiO\(_2\), the opposite feature is realized and it is another definite evidence of that La\(_{1-x}\)Sr\(_x\)NiO\(_2\) is far from a Mott system. Note that Mott-like behavior was reported in recent GW+DMFT calculations [28].

The decrease in the kinetic energy originates from the fact that the number of mobile charge carriers \( n_e \) decrease upon doping as presented in Table I. \( n_e \) is estimated from the volume of the Fermi surface by using Luttinger’s theorem. Note that two distinct charge carriers are realized in the Fermi surface, those are the correlated Ni \( d_{x^2-y^2} \) and the uncorrelated hybridized band with Ni \( d_{z^2} \) (see Fig. 4). \( n_e \) for Ni \( d_{x^2-y^2} \) decreases gradually and significantly upon doping. However, \( n_e \) for the uncorrelated hybridized band decreases slowly and its amount is fairly small. From the viewpoint of \( n_e \), the doped holes mostly go to the Ni \( d_{x^2-y^2} \) orbital [71].

**Results: Electronic Structure**— Electronic structure of LaNiO\(_2\) is provided in Fig. 4. The correlated Ni \( d_{x^2-y^2} \) is a dominant character at \( E_F \) and gives a large Fermi surface (FS) shown in Fig. 4(b). The uncorrelated hybridized band gives small FSs at \( \Gamma \) and \( A \). Hence, the

| Table I. The number of mobile charge carriers, \( n_e \), as a function of doping ratio. \( n_e \) is estimated from the volume enclosed by each Fermi surface. 3-dimensional Fermi surface computed with DFT+DMFT is presented in Supplemental Material [58]. |
|---------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| \( x = 0.0 \)                  | 0.98            | 0.93            | 0.85            | 0.76            | 0.66            | 0.56            |
| \( x = 0.1 \)                  | 0.93            | 0.87            | 0.79            | 0.71            | 0.63            | 0.55            |
| \( x = 0.2 \)                  | 0.85            | 0.79            | 0.72            | 0.64            | 0.56            | 0.48            |
| \( x = 0.3 \)                  | 0.76            | 0.71            | 0.64            | 0.56            | 0.48            | 0.40            |
| \( x = 0.4 \)                  | 0.66            | 0.56            | 0.48            | 0.40            | 0.32            | 0.24            |
| \( x = 0.5 \)                  | 0.56            | 0.48            | 0.40            | 0.32            | 0.24            | 0.16            |
multiorbital character is clearly identified in the electronic structure calculations [27, 28, 31]. At $x = 0.2$, the hybridized band with Ni $d_{x^2}$ detaches from $E_F$. As a result, the FS at $\Gamma$ disappears. Upon further doping, at $x = 0.5$, another FS from the hybridized band detaches from $E_F$ as well and the FS from Ni $d_{x^2-y^2}$ is solely realized. Therefore, two distinct Lifshitz transitions are realized upon doping [27, 42].

**Results:** Two-Band Model — The multiorbital character is definitely seen in the DMFT valence histogram as depicted in Fig. 4(c), where the second largest probability of 0.16 comes from the atomic configuration of ($N = 8, e_g$ $S = 1$) with a spin-triplet state within Ni $e_g$ states. Note that ($N = 9, S = 1/2$) with one hole in Ni $d_{x^2-y^2}$ has the largest probability of 0.35. These two largest probabilities are nearly constant over the doping concentration. Since the FS has primarily Ni $d_{x^2-y^2}$, one could interpret in an one-band scenario as a low-energy model [21, 24]. However we find that Hund’s coupling $J_H$ decreases $Z$ and increases $-\text{Im}\Sigma(i0^+)$ for $d_{x^2-y^2}$ [58], which is surprising as the atomic ground state configuration has one hole. This $J_H$ dependence of the correlation strength is the hallmark of a Hund’s metal [72–74]. This is because a metallic state requires fluctuations between $d^9$ and $d^8$, and $J_H$ is important in the latter configuration as seen clearly in the valence histogram in Fig. 4(c). We can think of the crystal field as being frequency dependent, at low energies it leaves $d_{x^2-y^2}$ as the most active orbital, but at intermediate frequencies both $d_{x^2-y^2}$ and $d_{z^2}$ are important [29]. Therefore, the infinite-layer nickelate is a Hund’s metal where Hund’s correlation is hidden at low energies but noticeable at intermediate energies. It is different from the Hund’s metal realized in iron pnictides, chalcogenides, and ruthenates, where a configuration with more than one electron or hole, makes Hund’s correlation prominent in the atomic ground state configuration. This is made explicit by a two-band Wannier construction which is atom centered with the symmetry of the two Ni $e_g$ orbitals, but which exhibits the clear difference of $d_{x^2-y^2}$ and $d_{z^2}$ provided in Supplementary Material [58]. Note that alternative low-energy Wannier constructions have been reported [14, 17, 34–36, 43, 44]. The origin of superconductivity and absence of long-range magnetic order within this kind two-band model is an open problem.

**Conclusion** — To summarize, we have computed the temperature and doping dependence of the optical properties of the normal state of La$_{1-x}$Sr$_x$NiO$_2$ within DFT+DMFT. The ratio of $K_{\text{DMFT}}/K_{\text{DFT}}$ is less than one, indicating electronic correlations but the trends in the evolution of the optics with temperature and doping are opposite to those of established Mott systems. The results suggest that an interpretation in terms of the two-band model at low energies, contain-
ing the strongly differentiated $d_{2−z}^2$ which exhibits the coherence-incoherence crossover and the second band which is less correlated and can be seen as a hybrid of several orbitals with $d_{2}$ character. At very low energies and temperatures, the former is dominant but contributions from the second is seen at intermediate energies and temperatures. While there are some studies of spinful two-orbital models with occupancy near one (or three) [80, 81] the large differences between the two band structures of spinful two-orbital models with occupancy near one $d_{2}$ character. At very low energies and temperatures, the former is dominant but contributions from the second is seen at intermediate energies and temperatures. While there are some studies of spinful two-orbital models with occupancy near one (or three) [80, 81] the large differences between the two bands makes the infinite-layer nickelates a new prototype of strongly correlated material.

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[58] See Supplemental Material, which includes Refs. [11, 54-56, 59-65, 72, 75-79], for the computational details, DFT electronic structure, optics for a prototypical Mott system of V$_2$O$_3$, the decomposition of the Drude peak, the quasiparticle weight $Z$ and the quasiparticle scattering rate for all Ni 3d orbitals, 3-dimensional Fermi surfaces of La$_{1-x}$Sr$_x$NiO$_2$ computed with DFT+DMFT, the DMFT valence histogram as a function of Hund’s coupling $J_H$, coherence-incoherence crossover, and the two-band model based on the Wannier interpretation.

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**Supplemental Material:**

Optical properties of the infinite-layer La$_{1-x}$Sr$_x$NiO$_2$ and hidden Hund’s physics

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**COMPUTATIONAL DETAILS**

Fully charge self-consistent DFT+DMFT calculations implemented in Wien2k package [S1] are performed with formalisms described in Ref. [S2]. The experimental lattice constants of $a = b = 3.871\text{Å}$ and $c = 3.375\text{Å}$ are adopted for the calculations [S3]. The muffin-tin radii are 2.50, 1.95, and 1.68 Bohr radius for La, Ni, and O, respectively.

We choose a wide hybridization energy window from -10 eV to 10 eV with respect to the Fermi level $E_F$. All the five Ni-3$d$ orbitals are considered as correlated ones and the fully rotational invariant form is applied for a local Coulomb interaction Hamiltonian with on-site Coulomb repulsion $U = 5$ eV and Hund’s coupling $J_H = 1$ eV. The Coulomb parameters $U$ and $J_H$ are confirmed by a constraint LDA (cLDA) calculation, where we constructed a $2 \times 2 \times 2$ supercell and put a constrained number of Ni 3$d$-electrons, $n_d = 9$, in the core state. It gives the effective Coulomb parameter of $U_{\text{eff}} = F_{\text{th}}^0 = U - J = 4.11$ eV, which verifies our $U$ and $J_H$ values in the DFT+DMFT calculations.

The continuous time quantum Monte Carlo (CTQMC) [S4, S5] is adopted for an impurity solver. We use a generalized gradient approximation (GGA) [S6] for the exchange-correlation functional and subtract double counting (DC) term with an exact DC scheme invented by Haule, which eliminates the DC issues in correlated materials [S7]. The modified Gaussian method [S2] was used for analytical continuation to obtain the self-energy on the real frequency.

The Drude component in the optical conductivity. In the DFT+DMFT method, the real part of the optical conductivity is computed as follows [S54]:

$$
\sigma^{\mu\nu}(\omega) = \frac{\pi e^2}{V_0} \sum_k \int d\omega (-\frac{\partial f}{\partial \omega}) \text{Tr}[v_\mu(k)A(k,\omega)v_\nu(k)A(k,\omega)],
$$

where $e$, $V_0$, $f$, $v_\mu(k)$, and $A(k,\omega)$ are the elementary charge, the volume of the unit cell, the Fermi-Dirac distribution function, the Fermi velocity along $\mu$-direction, and the spectral function, respectively.

Since Eq. (S1) contains the $k$-sum in the Brillouin zone, the Drude peak could be decomposed into each band character if the all band characters are well separated spatially in the Brillouin zone. Figure S2 demonstrates that all distinct band characters of the Fermi surface are well separated in the Brillouin zone. Note that the hybridized band of Ni $d_{xz}$ and La $d_{xy}$ gives the Fermi surface at $\Gamma$ and that of Ni $d_{yz}$ gives the Fermi surface at La $d_{xy}$.

**DECOMPOSITION OF THE DRUDE PEAK**

Since three band characters cross $E_F$, those are Ni $d_{xz}$, the hybridized band of Ni $d_{xz}/d_{yz}$ and La $d_{xy}$, and that of Ni $d_{yz}$ and La $d_{xz}$, they contribute the Drude component in the optical conductivity.

In the DFT+DMFT method, the real part of the optical conductivity is computed as follows [S54]:

$$
\sigma^{\mu\nu}(\omega) = \frac{\pi e^2}{V_0} \sum_k \int d\omega (-\frac{\partial f}{\partial \omega}) \text{Tr}[v_\mu(k)A(k,\omega)v_\nu(k)A(k,\omega)],
$$

where $e$, $V_0$, $f$, $v_\mu(k)$, and $A(k,\omega)$ are the elementary charge, the volume of the unit cell, the Fermi-Dirac distribution function, the Fermi velocity along $\mu$-direction, and the spectral function, respectively.

Since Eq. (S1) contains the $k$-sum in the Brillouin zone, the Drude peak could be decomposed into each band character if the all band characters are well separated spatially in the Brillouin zone. Figure S2 demonstrates that all distinct band characters of the Fermi surface are well separated in the Brillouin zone. Note that the hybridized band of Ni $d_{xz}$ and La $d_{xy}$ gives the Fermi surface at $\Gamma$ and that of Ni $d_{yz}$ gives the Fermi surface at La $d_{xy}$.
Fermi surface at $A$. Both are uncorrelated and correlations exhibited in them are almost temperature independent. Therefore, the low-energy Drude peak could be decomposed into two band components: the correlated Ni $d_{x^2-y^2}$ and the remaining uncorrelated hybridized band. Then, decomposition of the Drude peak is accomplished by performing the $k$-sum in each discretized Brillouin zone indicated in Fig. S2.

Figure S3 shows the decomposition of the Drude peak computed within DFT+DMFT at a broad temperature range. The effective plasma frequency ($\omega_p^*$) and the effective scattering rate $1/\tau_{qp}$ for each band component are extracted from the data presented in Fig. S3 and they are depicted as a function of temperature in the main text.

**OPTICAL CONDUCTIVITY OF A PROTYPICAL MOTT SYSTEM OF $V_2O_3$**

Figure S4(a) shows the optical conductivity of the paramagnetic metallic phase of $V_2O_3$ computed with DFT+DMFT, which is adopted from Ref. [S9]. From the data, we compute the integrated optical spectral weight $K = \int_0^\Omega \sigma_1(\omega) d\omega$ and provide it as a function of integration cutoff value $\Omega$ in Fig. S4(b). Since the electronic kinetic energy is proportional to the integrated optical spectral weight $K$, Fig. S4(b) demonstrates that the kinetic energy decreases upon heating. This reflects the Mott behavior that the kinetic energy is reduced as an insulating state is approached at higher temperatures. This optical response upon heating is opposite to the case of LaNiO$_2$, where the kinetic energy increases upon heating as shown in the main text. Based on the optical response, the infinite-layer LaNiO$_2$ is, therefore, far from a Mott system.

**LOW-ENERGY PHYSICAL QUANTITIES**

Figure S5 shows the imaginary part of self-energy for Ni $d_{x^2-y^2}$ on the real axis. The modified Gaussian method [S2] is adopted for analytical continuation. Upon cooling, $\text{Im}\Sigma(0)$ approaches to zero and $\text{Im}\Sigma(\omega)$ exhibits a quadratic behavior at low frequencies. Hence, LaNiO$_2$ clearly shows the Fermi liquid behavior.

**TABLE S1.** Quadratic fitting $(aT^2 + b)$ in the quasiparticle scattering rate $1/\tau_{qp}$ provided in Fig. S6(b). The coefficients $a$ and $b$ are provided for each Ni orbital.

| Orbital     | $a$ (eV/K$^2$) | $b$ (eV) |
|------------|----------------|----------|
| $d_{x^2-y^2}$ | $2.029 \times 10^{-7}$ | $6.124 \times 10^{-5}$ |
| $d_{z^2}$    | $4.563 \times 10^{-9}$ | $2.532 \times 10^{-4}$ |
| $d_{xy}$     | $1.753 \times 10^{-9}$ | $2.045 \times 10^{-4}$ |
| $d_{xz}$     | $2.393 \times 10^{-9}$ | $2.117 \times 10^{-4}$ |

The quasiparticle weight $Z$ and the quasiparticle scattering rate $1/\tau_{qp} = -Z\text{Im}\Sigma(i0^+)$ are depicted in Fig. S6. Ni $d_{x^2-y^2}$ has the lowest $Z (0.4 \sim 0.5)$ among the others ($Z \approx 0.8$), indicating that Ni $d_{x^2-y^2}$ is the correlated orbital and the others are almost uncorrelated ones. $1/\tau_{qp}$ for all Ni $3d$ orbitals exhibit a quadratic behavior in temperature, thereby presenting the Fermi liquid behavior. In the case of Ni $d_{x^2-y^2}$, deviation from the quadratic behavior in $1/\tau_{qp}$ is recognized around $T \sim 600$ K. The coherent temperature is $T_{coh} \sim 450K$ as demonstrated in Fig. S6(c), hence deviation from the Fermi liquid behavior is apparent above the coherent temperature. It is noteworthy that the only Ni $d_{x^2-y^2}$ orbital shows coherence-incoherence crossover and the others are still coherent even at high temperature. The orbital-differentiated coherence-incoherence crossover is one of the hallmark of a Hund’s metal.

Table S1 provides coefficients of the quadratic fitting $(aT^2 + b)$ on $1/\tau_{qp}$ used in Fig. S6(b). For Ni $d_{x^2-y^2}$, $b$ is relatively small and $\Gamma/k_BT$ depicted in Fig. S6(c) follows $aT + b/T \approx aT$, that is a linear behavior. On the other hand, the others have relatively large $b$, so that they exhibit $b/T$ at low temperature, but $aT$ at high temperature as demonstrated in Fig. S6(c).

**ELECTRONIC STRUCTURE OF THE INFINITE-LAYER NICKELATE UPON STRONTIUM DOPING**

Figure S7 shows the 3-dimensional Fermi surface of the infinite-layer La$_{1-x}$Sr$_x$NiO$_2$ for several doping concentrations $x$ computed with DFT+DMFT. $\text{Im}\Sigma(\omega)$ is set to be zero in Fig. S7, which leads to obtain the quasiparticle Fermi surface. At $x = 0.1$, the small electron pocket at $\Gamma$ disappears. Upon further doping, another electron pocket at $A$ diminishes and disappears eventually at $x = 0.5$. Therefore, two distinct Lifshitz transitions are realized from $x = 0.0$ to $x = 0.5$.

The number of mobile charge carriers $n_c$ for each band component contributing the intraband transitions is estimated from the volume enclosed by the corresponding Fermi surface presented in Fig. S7 and is provided in the main text. The size of the Ni $d_{x^2-y^2}$ Fermi surface shrinks upon doping and its $n_c$ decreases accordingly. It reduces the electronic kinetic energy as shown in Fig. 3 in the main text. It is another definite evidence of that the infinite-layer La$_{1-x}$Sr$_x$NiO$_2$ is far from a Mott system, where doping releases localization of electrons and increases the electronic kinetic energy correspondingly.
HUND’S RULE CORRELATION IN THE INFINITE-LAYER NICKELATE

In order to gain more insight into the role of Hund’s rule correlation in the infinite-layer LaNiO$_2$, we provide the DMFT valence histogram as a function of Hund’s coupling $J_H$ as depicted in Fig. S8(a). Note that DMFT simulations with $J_H = 0.3$ and 0 eV along with the exact double counting scheme give different occupation numbers of Ni 3d orbitals, $n_d = 8.29$, and 8.17, respectively. Recall that $n_d = 8.58$ for $J_H = 1.0$ eV. Therefore, in order to balance the $n_d$ for $J_H = 1.0$ eV and that for other $J_H$ values, we fix the double-counting energy obtained in the $J_H = 1.0$ eV simulation and perform DMFT calculations with other $J_H$ values. It helps to obtain the similar $n_d$. As demonstrated in Fig. S8(a), the most probable atomic configuration in a $N = 8$ sector is the spin-triplet state ($S = 1$) in Ni $e_g$ orbitals and its probability decreases as $J_H$ becomes smaller. It clearly shows the Hund’s rule correlation. The Hund’s rule correlation is clearly exhibited in Figs. S8(b) and (c), where the quasiparticle weight $Z$ and the electronic coherence scale $-\text{Im}\Sigma(i0^+)$ change significantly as a function of Hund’s coupling $J_H$. Figure S8(c) particularly shows that Hund’s coupling $J_H$ increases $-\text{Im}\Sigma(i0^+)$, thereby reducing the coherence scale drastically. It leads to lower the coherent temperature $T_{coh}$. In Hund’s metal systems such as ruthenates and iron pnictides [S12–S14] and chalcogenides [S15], $T_{coh}$ is defined by $\Gamma/k_B T = 1$ and it is estimated as $T_{coh} \sim 450$ K. The deviation from the $T^2$ behavior is visible above the coherence temperature. The same behavior could be found in $1/\tau^*_B$ for Ni $d_{x^2−y^2}$ as shown in Fig. 2(e) of the main text. Note that the coherence scale (or $T_{coh}$) is severely diminished by Hund’s coupling $J_H$ as demonstrated in Fig. S8(c). The coherence-incoherence crossover is already realized in other Hund’s metal systems such as ruthenates [S10, S11] and iron pnictides [S12–S14] and chalcogenides [S15], where Hund’s coupling $J_H$ drastically reduces the coherence scale.

The coherence temperature $T_{coh}$ manifests itself also in the $T$-dependent Fermi surface (FS), displayed in Fig. S9(b). Below $T_{coh}$, quasiparticles are well-defined, thereby providing apparent FSs. Above $T_{coh}$, Ni 3d orbitals except for $d_{x^2−y^2}$ are still coherent even at very high temperature, however the Ni $d_{x^2−y^2}$ gets incoherent. As a result, the large FS of Ni $d_{x^2−y^2}$ diminishes. $T_{coh}$ is identified in $(\omega_p^*)^2$ as well presented in Fig. 2(d) of the main text, where $(\omega_p^*)^2$ for Ni $d_{x^2−y^2}$ increases linearly upon heating below $T_{coh}$ and then shows the saturation behavior above $T_{coh}$. $(\omega_p^*)^2$ for the uncorrelated hybridized band does not show temperature dependence. The orbital-differentiated coherence-incoherence crossover is one of the main characteristic of the Hund’s metal physics [S10, S14].

COHERENCE-INCOHERENCE CROSSOVER

Figure S9(a) shows $\Gamma/k_B T$ as a function of $T$, where $\Gamma$ is the quasiparticle scattering rate with $\Gamma = -Z\text{Im}\Sigma(i0^+)$. The Fermi-liquid behavior of $\Gamma \propto T^2$ is identified up to ~400 K. The coherence temperature $T_{coh}$ is defined by $\Gamma/k_B T = 1$ and it is estimated as $T_{coh} \sim 450$ K. The deviation from the $T^2$ behavior is visible above the coherence temperature. The same behavior could be found in $1/\tau^*_B$ for Ni $d_{x^2−y^2}$ as shown in Fig. 2(e) of the main text. Note that the coherence scale (or $T_{coh}$) is severely diminished by Hund’s coupling $J_H$ as demonstrated in Fig. S8(c). The coherence-incoherence crossover is already realized in other Hund’s metal systems such as ruthenates [S10, S11] and iron pnictides [S12–S14] and chalcogenides [S15], where Hund’s coupling $J_H$ drastically reduces the coherence scale.

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TWO-BAND MODEL

We investigate the effective low-energy Hamiltonian of the infinite-layer LaNiO$_2$ based on the Wannier interpretation [S16, S17]. Since band characters that cross the Fermi level are the correlated Ni $d_{x^2−y^2}$ and the uncorrelated hybridized band including Ni $d_z$, we choose two Wannier bases of Ni $d_{x^2−y^2}$ and $d_z$ orbitals. During the process of Wannier minimization, we found that a center of the Wannier wave-function of Ni $d_z$ is shifted toward a La site and the spread of the Wannier wave-function develops gradually due to a significant hybridization with La $d_{x^2}$ orbital. Hence, in order to keep an atom centered Ni $d_z$ orbital, we performed an one-shot Wannier minimization computation and the results are presented in Fig. S10. As shown in Fig. S10, the two-band Wannier band dispersion explains the DFT one near $E_F$, thereby suggesting that the effective low-energy Hamiltonian is described with atom centered Ni $e_g$ orbitals.

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FIG. S1. DFT electronic structure of the infinite-layer LaNiO$_2$. (a) DFT band dispersion in a broad energy window. The orange arrows correspond to interband transitions identified in the optical conductivity presented in the main paper. (b) DFT Fermi surface. Three Fermi surfaces are realized and their characters are indicated. (c) DFT band dispersions with orbital characters. (left) La $5d$, Ni $3d$, and O $2p$ (middle) La $d_{z^2}$, $d_{xy}$, $d_{x^2-y^2}$, and $d_{xz}/d_{yz}$ (right) Ni $d_{z^2}$, $d_{xy}$, $d_{x^2-y^2}$, and $d_{xz}/d_{yz}$ orbital characters are presented.

FIG. S2. Discretization of the Brillouin zone so as to decompose the Drude peak into two band components (Ni $d_{x^2-y^2}$ and an uncorrelated hybridized band) that cross the Fermi level. In the Fermi surface of LaNiO$_2$ computed with DFT+DMFT, each band character is well separated spatially in the Brillouin zone indicated by the black squares.
FIG. S3. Decomposition of the Drude peak in the infinite-layer LaNiO$_2$ at a broad temperature range. The Drude peak computed with DFT+DMFT is decomposed into each band component that crosses the Fermi level, that are Ni $d_{x^2-y^2}$ and an uncorrelated hybridized band.
FIG. S4. (a) The optical conductivity of the paramagnetic metallic phase of V$_2$O$_3$ computed within the DFT+DMFT method. The data is adopted from Ref. [S65]. (b) The integrated optical spectral weight $K(\Omega) = \int_0^\Omega \sigma_1(\omega) d\omega$ as a function of cutoff frequency $\Omega$. It decreases upon heating, which is a key characteristic of a Mott system.

FIG. S5. DMFT self-energy in the infinite-layer LaNiO$_2$. The imaginary part of self-energy for Ni $d_{x^2-y^2}$ orbital is presented on the real axis for several temperatures.

FIG. S6. Low-energy physical quantities for Ni 3$d$ orbitals in the infinite-layer LaNiO$_2$. (a) Quasiparticle weight $Z$ as a function of temperature. (b) Quasiparticle scattering rate $1/\tau_{qp} = -Z \text{Im} \Sigma(\theta^+) + \text{Im} \Sigma''(\theta^+) - \Gamma$ as a function of temperature. (c) Quasiparticle scattering rate $\Gamma = 1/\tau_{qp}$ divided by $k_B T$ as a function of temperature. Error bars presented in (a), (b), and (c) originate from the statistical errors in CTQMC simulations. The dash-dotted lines in (a) and (b) are guides for the eye by fitting $Z$ and $1/\tau_{qp}$ to linear ($aT + b$) and quadratic ($aT^2 + b$) functions, respectively. The dash-dotted lines in (c) are guides for the eye and have a form of $aT + b/T$ where the coefficients $a$ and $b$ are obtained from the fitting in (b).
FIG. S7. DMFT Fermi surfaces of the infinite-layer La$_{1-x}$Sr$_x$NiO$_2$ as a function of doping ratio $x$. In this figure, Im$\Sigma(\omega)$ is set to be zero to obtain the quasiparticle DMFT Fermi surfaces.

FIG. S8. Hund’s rule correlation in the infinite-layer LaNiO$_2$. (a) The DMFT valence histogram of the Ni-3$d$ shell for LaNiO$_2$ is provided for Hund’s coupling $J_H =$ (left) 1, (middle) 0.3, and (right) 0 eV. The 1024 possible atomic configurations are sorted by the number of 3$d$ electrons of the individual configuration. The probability of the atomic configuration of (N = 8, $e_g$ S = 1), where the spin triplet state is realized in Ni $e_g$ orbitals, decreases as $J_H$ becomes smaller. (b) Quasiparticle weight $Z$ as a function of Hund’s coupling $J_H$. (c) Electronic coherence scale $-\text{Im}\Sigma(i0^+)$ as a function of Hund’s coupling $J_H$. The dash-dotted line in (c) is a guide for the eye. Error bars originate from the statistical errors in CTQMC simulations.
FIG. S9. Coherence-incoherence crossover in LaNiO$_2$. (a) Quasiparticle scattering rate $\Gamma = -\text{ZIm}(\delta^+)$ of Ni $d_{x^2-y^2}$ divided by $k_BT$ as a function of temperature. Error bars originate from the statistical errors in CTQMC simulations. The red dash-dotted line is a guide for the eye by fitting $\Gamma/k_BT$ to a linear function. $\Gamma/k_BT \approx 1$ at the coherent temperature $T_{coh} \sim 450\,\text{K}$. Above $T_{coh}$, $\Gamma/k_BT$ shows deviation from linearity, indicating that a quasiparticle is no longer well-defined. The coherence-incoherence crossover is clearly shown in (b), where Fermi surfaces are plotted at temperature below and above $T_{coh}$.

FIG. S10. Two-band model. (a) DFT (blue solid line) and Wannier (red dot) band dispersions of LaNiO$_2$. (b) Isosurface plots for (left) Ni-centered $d_{x^2-y^2}$-like and (right) Ni-centered $d_{z^2}$-like with extended La-centered $d_{z^2}$-like Wannier orbitals. These two orbitals describe the DFT band dispersion well near the Fermi level in (a).