SUPPLEMENTARY INFORMATION

Tuning the metal-insulator transition in NdNiO$_3$ heterostructures via Fermi surface instability and spin-fluctuations

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DFT calculations

Earlier calculations in the heterostructure of LaNiO$_3$ and LaAlO$_3$ show that its electronic structure is very much same as the bulk LaNiO$_3$ apart from tiny reduction of three-dimensionality [1]. Following the same strategy of Ref. [2], we carry out the density functional calculation of NNO using the Vienna *ab-initio* simulation package [3] within the GGA+$U$ of PBE parametrization [4]. Projected augmented-wave (PAW) [5, 6] pseudo-potentials are used to describe core electrons. We use $U = 3.5$ eV, which is larger than the values of $U$ used in the self-energy calculation. This is expected since the bare $U$ used in the self-energy calculation is further multiplied by various components of the susceptibility to provide the effective many-body potential in this calculation, as described below. Bulk NdNiO$_3$ belongs to the P4/mmm space group and a $A$ type antiferromagnetic spin ordering [2] is imposed in the system to obtain a stable ground state [2]. The electronic wave-function is expanded using plane waves up to a cutoff energy of 500 eV. Brillouin zone sampling is done by using a (12x12x6) Monkhorst-Pack $k$-grid.

Tight binding fitting

To accurately compute the electron-electron correlation in this system, we derive the tight-binding Hamiltonian for the three lowest energy states. As evident from the DFT band structure, the low-energy states mainly composed of two $e_g$ orbitals which constitute the electron and hole pockets, while a third hole pocket develops near 'A' point coming from the hybridized state of $t_{2g}$ of Ni atoms with the $p$-orbitals of O atoms. To restrict our methodology within the essential three bands model, we model the third band as an effective state of all these orbitals. Therefore, in the three band tight-binding model, the non-interacting Hamiltonian can be written as

$$
\hat{H} = \hat{\Psi}^\dagger \begin{pmatrix}
\xi_{1,\mathbf{k}} & \xi_{13,\mathbf{k}} \\
\xi_{12,\mathbf{k}} & \xi_{23,\mathbf{k}} \\
h.c. & \xi_{3,\mathbf{k}}
\end{pmatrix} \hat{\Psi}.
$$

(1)

The symbol tilde over a quantity in this paper denotes it to be a matrix. Here the non-interacting dispersions for intra- and inter-orbital hoppings are derived by considering the three-dimensional crystal symmetry of the tetragonal lattice. If we define $\xi_{1,\mathbf{k}}$, $\xi_{2,\mathbf{k}}$, and $\xi_{3,\mathbf{k}}$ as dispersion for $d_{x^2-y^2}$, $d_{z^2}$, and effective $d_{xy}$ orbitals, respectively, we obtain

$$
\begin{align*}
\xi_{1,\mathbf{k}} &= -2t_1(c_x + c_y) - 4t_2c_xc_y + \delta_1 - \mu, \\
\xi_{2,\mathbf{k}} &= -2t_4(c_x + c_y) - 4t_5c_xc_y - 2t_7c_z + \delta_2 - \mu, \\
\xi_{3,\mathbf{k}} &= -2t_9(c_x + c_y) - 2t_7c_z + \delta_3 - \mu,
\end{align*}
$$

(2)

and

$$
\begin{align*}
\xi_{12,\mathbf{k}} &= -2t_5(c_x - c_y), \\
\xi_{13,\mathbf{k}} &= -2t_9(c_x + c_y), \\
\xi_{23,\mathbf{k}} &= -2t_{10}c_xc_y.
\end{align*}
$$

(3)

Here $c_i = \cos k_i$ for $i = x, y, z$. $t_i$ are various nearest and next-nearest neighbor hoppings, $\delta_i$ are the onsite potentials for different orbitals, and $\mu$ is the system’s chemical potential. The corresponding tight-binding parameters are obtained by fitting the eigenvalues of the above Hamiltonian to the corresponding DFT band structure as shown in Fig. S1. Hence we obtain $t_1=0.41$, $t_2=-0.12$, $\delta_1=0$, $t_4=0.05$, $t_5=0.35$, $\delta_2=-0.1$, $t_6=0.23$, $t_7=0.15$, $\delta_3=-1.8$, $t_8=0.2$, $t_9=0.1$, $t_{10}=0.1$, all in eV. All the parameters remain same for the case of NNO/LAO, except the onsite term for the $d_{xy}$ is reduced to -1.5 eV, and the hopping between $d_{xy}$ to the two $e_g$ orbitals is reduced to 0.15 eV. The chemical potential is estimated for each samples to keep the number of electrons same after including the self-energy effects. After projecting the orbital character on the corresponding band structure, we find that the dominant orbital contribution to both electron and hole pockets comes from the $d_{x^2-y^2}$ orbital as in cuprates (without the electron pocket). The corresponding Fermi surface is shown in the paper, see Fig. 6(h). For NNO/LAO case, the electron-pocket becomes smaller while the hole pocket increases, and an additional hole-pocket appears around the ‘A’ point, see Fig. 6(i) of the paper.

More details of the MRDF method

The transition metal oxides reside in the metal-insulator boundary which indicates that the strength of the Coulomb interaction is neither sufficiently large to localize all electrons, nor weak enough to define a quasiparticle peak at all energy and momenta. In this spirit it is appropriate to apply an intermediate coupling theory, [9–11] which allows the coexistence of itinerant and localized electrons in different momentum and energy regions of the spectra.
FIG. 1. DFT band structure for NNO. Superimposed are the tight-binding bands calculated from the Hamiltonian in Eq. (1). The color coating of the tight-binding line gives the relative weight of \(d_{z^2}\) (blue) to \(d_{x^2-y^2}\) (red) orbital character while light green color gives the weight of the hybridized \(d_{xy}\)-orbital.

The single-particle Green's function is defined as 
\[
\tilde{G}_0(k, i\omega_n) = \left(i\omega_n \tilde{1} - \tilde{H}\right)^{-1},
\]
where \(i\omega_n\) is the Matsubara frequency for the fermions, and \(H\) is the non-interacting tight-binding Hamiltonian. The explicit form of \(G\) is then obtained as
\[
G_{mn}(k, i\omega_n) = \sum_{\nu} \phi_{k,m}^\nu \phi_{k,n}^{\nu\dagger} i\omega_n - \xi_{k}^{\nu}.
\]
(4)

Here \(k\) and \(\omega\) are the quasiparticle momentum and frequency, and \(q\) and \(\omega_p\) are the bosonic excitation momentum and frequency, respectively. \(\phi_{k,m}^\nu\) is the eigenstate for the \(\nu\)-th tight-binding band \((\xi_{k}^{\nu})\), projected onto the \(m\)-th orbital. The non-interacting density fluctuation susceptibility is
\[
\chi_{0, mn}(q, \omega_p) = -\frac{1}{\Omega_{BZ}\beta} \sum_{k,n} G_{mn}(k, i\omega_n)G_{st}(k + q, i\omega_n + \omega_p),
\]
(5)
where \(\beta = 1/k_B T\), and \(k_B\) is the Boltzmann constant and \(T\) is temperature. \(\Omega_{BZ}\) is the electronic phase space volume. \(f_k^\nu\) and \(n_p\) are the fermion and boson occupation numbers, respectively. After performing the Matsubara summation over the fermionic frequency \(\omega_n\) and taking analytical continuation to the real frequency as \(\omega_n \rightarrow \omega + i\delta\), we get
\[
\chi_{0, mn}(q, \omega_p) = -\frac{1}{\Omega_{BZ}} \sum_{k,v,v'} \phi_{k+q,v}^{\nu\dagger} \phi_{k+q,v'}^{\nu} \phi_{k,n}^{\nu\dagger} \phi_{k,m}^{\nu} \frac{f_{k+q}^{\nu} - f_{k'}^{\nu}}{\omega_p + i\delta - \xi_{k}^{\nu} + \xi_{k+q}^{\nu}}.
\]
(6)
FIG. 2. Comparison of self-energy and band renormalization for NNO/NGO and NNO/LAO. For the same value of the interaction parameters, we find that the band renormalization and imaginary part of the self-energy are smaller in NNO/LAO than the other one.

The RPA Hamiltonian for the multiband system is

$$H_{\text{int}} = \sum_{k_1-k_4} \left[ U_c \sum_m c_{k_1,m\sigma}^\dagger c_{k_2,m\sigma} c_{k_3,m\bar{\sigma}}^\dagger c_{k_4,m\bar{\sigma}} + \sum_{m<n,\sigma} \left( V_{c_k,m\sigma} c_{k_2,m\sigma} c_{k_3,n\bar{\sigma}}^\dagger c_{k_4,n\bar{\sigma}} + (V-J_H)c_{k_1,m\sigma} c_{k_2,m\sigma} c_{k_3,n\bar{\sigma}}^\dagger c_{k_4,n\bar{\sigma}} \right) \right] + \sum_{m<n,\sigma} \left( J_H c_{k_1,m\sigma}^\dagger c_{k_2,m\sigma} c_{k_3,n\bar{\sigma}}^\dagger c_{k_4,n\bar{\sigma}} + J' c_{k_1,m\sigma}^\dagger c_{k_2,m\sigma} c_{k_3,n\bar{\sigma}} c_{k_4,n\bar{\sigma}} + \text{h.c.} \right).$$

The nonzero components of the matrices $\tilde{U}_c$ and $\tilde{U}_s$ are given as

$$\tilde{U}_{mm}^{s,c} = U, \quad \tilde{U}_{mm}^{s,c} = \frac{1}{2} J_H, \quad \tilde{U}_{mn}^{s,c} = \frac{1}{4} J_H + V, \quad \tilde{U}_{mn}^{s,c} = J',$n

Of course, it is implicit that all the interaction parameters are orbital dependent. Within the RPA, spin and charge channels become decoupled. The collective many-body corrections of the density-fluctuation spectrum can be written in matrix representation: $\tilde{\chi}_{s/c} = \tilde{\chi}_0[1 \mp \tilde{U}_{s/c}\tilde{\chi}_0]^{-1}$, for spin and charge densities, respectively. $\tilde{\chi}_0$ matrix consists of components $\chi_{0,mn}^{s,c}$, with the same basis in which the interactions $\tilde{U}_{s/c}$ are defined above.

Finally, the density fluctuation exchange potentials for the electronic state are computed as

$$V_{mn,i}(q_1,\omega_p) = \frac{\eta_i}{2} \left[ \tilde{U}_{i}(q_1,\omega_p) \tilde{U}_{i}^\dagger \right]_{mn}^{s,c},$$

where $i$ stands for spin and charge components, $\eta = 3, 1$ for the spin and charge channels, respectively. The feedback effect of the electronic interaction on the electronic spectrum is then calculated via self-energy calculation within the
FIG. 3. The Fermi surface maps of 20 uc NdNiO$_3$ thin films grown on NdGaO$_3$, measured at 200 K sample temperature and as a function of photon energies ranging from 60 eV to 115 eV with 5 eV steps.

MRDF method[9–11]

$$\Sigma_{mn,i}(\mathbf{k}, \omega) = \frac{1}{\Omega_{BZ}} \sum_{\mathbf{q}_{st,\nu}} \int_{-\infty}^{\infty} d\omega_p \Gamma_{mn,\nu}^{st}(\mathbf{q}, \omega_p)\tilde{\Gamma}_{mn,\nu}^{st}(\mathbf{k}, \mathbf{q}) \left[ \frac{1 - f_{\mathbf{k}-\mathbf{q} + \mathbf{n}_p} - f_{\mathbf{k}-\mathbf{q} + \mathbf{n}_p}}{\omega + i\delta - \xi_{\mathbf{k}-\mathbf{q} + \mathbf{n}_p} + \omega_p} + \frac{f_{\mathbf{k}-\mathbf{q} + \mathbf{n}_p} + f_{\mathbf{k}-\mathbf{q} + \mathbf{n}_p}}{\omega + i\delta - \xi_{\mathbf{k}-\mathbf{q} + \mathbf{n}_p} + \omega_p} \right],$$  

(10)

where the subscript $i$ stands for spin, charge and phonon contributions. The vertex correction $\Gamma_{mn,\nu}^{st}(\mathbf{k}, \mathbf{q})$ encodes both the angular and dynamical parts of the vertex, which are combined to obtain $\Gamma_{mn,\nu}^{st}(\mathbf{k}, \mathbf{q}) = \phi_{\mathbf{k}-\mathbf{q} + \mathbf{n}_p}^{\nu} \phi_{\mathbf{k}-\mathbf{q} + \mathbf{n}_p}^{\nu} (1 - \partial \Sigma_{mn}(\mathbf{k}-\mathbf{q}, \omega) / \partial \omega_0)$. Full self-consistency requires the bare Green’s function $G_0$ in Eq. (4) to be replaced with the self-energy dressed $G^{-1}(\mathbf{k}, \omega) = G_0^{-1}(\mathbf{k}, \omega) - \Sigma(\mathbf{k}, \omega)$, where the total self-energy tensor is $\Sigma(\mathbf{k}, \omega) = \Sigma_1(\mathbf{k}, \omega) + \Sigma_2(\mathbf{k}, \omega) + \Sigma_3(\mathbf{k}, \omega)$. The resulting self-energy dressed quasiparticle dispersions $\xi^{\nu}(\mathbf{k}) = Z_k^{\nu} \epsilon^{\nu}(\mathbf{k})$ are used in Eqs. (4)-(10), which keep all the formalism unchanged with respect to the momentum resolved orbital selective quasiparticle renormalization factor $Z_k$.

Origin of stronger correlation in NNO/NGO system:

Eq. 10 indicates that the dominant contributions to the self-energy come from the large density of states (embedded in the Green’s function $G$), and the susceptibilities term $\chi$ for a given set of the Coulomb interaction terms $U$. To effectively explain the origin of the large renormalization effect in NNO/NGO than NNO/LAO, we keep all the band parameters and the Coulomb interaction $U$ same in both systems, and change only the onsite potential by about 300 meV and the inter-orbital hoppings between $e_g$ and $t_{2g}$ orbitals. As seen from the band structure plot in Fig. S1, as we go from NNO/LAO to NNO/NGO, we approach toward the van-Hove singularity (VHS) of $d_{x^2-y^2}$ orbital on the $k_z = 0$ plane and of $d_{z^2}$ orbital on the $k_z = \pi$ plane. That means the system approaches towards large density of states instability. A similar increment in the susceptibility is also visible for the NNO/NGO system. In this many-body case, the spin fluctuation is clearly more important than the charge counterparts. By comparing the integrated spin susceptibilities for the two systems, we see a considerable changes. In the low-energy region, we see that the
FIG. 4. (a) Typical RHEED intensity oscillations of NNO/LAO. (b,c) RHEED patterns of LAO substrate as well as NNO films grown on LAO substrate, respectively.

susceptibility is stronger in NNO/NGO, and concentrated around the wavevector $q \sim (1/4, 1/4, 1/4 \pm \delta)$ instead of being spread out over a large momentum span as in NNO/LAO. This means NNO/NGO is prone to a magnetic instability. Similarly, the integrated spin susceptibility over the whole energy scale shows that while it is very much similar in the $k_z = 0$ plane, on the $k_z = \pi$ plane, it is larger for the NNO/NGO system. This result indicates that the dynamical fluctuation, and thus the renormalization effect is larger in NNO/NGO than in NNO/LAO.

This observation is also reflected in the numerical values of $\Sigma$ and corresponding renormalization effect as shown in Fig. S2. In the top panels of Fig. S2, we show the same energy dependent self-energy for three orbitals and two systems at five representative high-symmetry momenta values. The general conclusion drawn at all these momenta by comparing the solid (NNO/NGO) and dashed (NNO/LAO) lines that the self-energies are larger in intensity for the NNO/NGO case, and the corresponding slope in $\Sigma'$ (determining the strength of mass enhancement at the Fermi level) is also higher in this system. The overall $k$-profiles of the $Z$ also reflect the same that the average renormalization increases in NNO/NGO than in NNO/LAO in all orbitals except in the $d_{xy}$ one. This is because, the $d_{xy}$ only crosses the Fermi level for the latter system which thereby obtain stronger renormalization effect.

Additional experimental details:

In order to show the evolution of the electron and hole pockets, we present, in Fig. S3, the Fermi surface maps of 20 uc NdNiO$_3$ thin films grown on NdGaO$_3$, measured at 200 K sample temperature and as a function of photon energies ranging from 60 eV to 115 eV with 5 eV steps. In Fig. S4, we show typical RHEED oscillations during the growth of NNO thin films on LAO substrate. The corresponding RHEED pattern is shown in Figs. S(b,c) for LAO substrate and NNO/LAO system, respectively, which show the well-ordered, high-quality single crystalline surface of the deposited thin films.

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