Hund-Heisenberg model in superconducting infinite-layer nickelates

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We theoretically investigate the unconventional superconductivity in the newly discovered infinite-layer nickelates Nd1−xSr2xNiO4 based on a two-band model. By analyzing the transport experiments, we propose that the doped holes dominantly enter the Ni dxy orbital and form a conducting band. Via the onsite Hund coupling, the doped holes as carriers are coupled to the Ni localized holes in the dz2−y2 orbital band. We demonstrate that this two-band model could be further reduced to a Hund-Heisenberg model. Using the reduced model, we show the non-Fermi liquid liquid state above the critical Tc could stem from the carriers coupled to the spin fluctuations of the localized holes. In the superconducting phase, the short-range spin fluctuations mediate the carriers into Cooper pairs and establish dz2−y2-wave superconductivity. We further predict that the doped holes ferromagnetically coupled with the local magnetic moments remain itinerant even at very low temperature, and thus the pseudogap hardly emerges in nickelates. Our work provides a new superconductivity mechanism for strongly correlated multi-orbital systems and paves a distinct way to exploring new superconductors in transition or rare-earth metal oxides.

Introduction—Although the origin of superconductivity (SC) in cuprate and heavy fermion materials remain highly controversial theoretically, new unconventional superconductors have proliferated experimentally over the recent decades [1–3]. The quasi-two-dimensional (2D) iron pnictides have triggered a new boom of the SC in recent decades [1–3]. The quasi-2D Ni-O plane is geometrically analog to the Cu-O plane in cuprates. The Ni localized holes in the dz2−y2 orbital are the key carriers and thermodynamic properties [6]. Moreover, it is debated whether the doped hole forms a spin singlet or a triplet doublon with the original hole on a Ni ion [26–30]. Several microscopic models have been proposed, such as the t-J model, the metallic gas coupled to a 2D Hubbard model and the spin freezing model [26, 22, 23, 27–30]. More surprisingly, absence of superconductivity was recently claimed in the bulk nickelates and the film prepared on various oxide substrates different from SrTiO3, and it was suggested that the absence possibly results from the hydrogen intercalation [31–33]. Own to these confusions, more insights into the microscopic mechanism in nickelates is imperative.

In this letter, we investigate the nickelate SC based on the analysis of the transport experiments. Considering the positive Hall coefficient and the suppressed self-doping effects at low temperature, we suggest that the doped holes go to the Ni dxy orbital and establish a conducting band [22, 24]. The onsite Hund interaction couples the conducting band with the localized dz2−y2 orbital together. The correlation between the sparse carriers and the kinetic energy of the localized holes could then be ignored. Thus, the two-band model is simplified into a Hund-Heisenberg model. We show both the non-Fermi liquid in normal state and the superconductivity is determined by the spin fluctuations of the localized holes. This SC mechanism could be realized in multi-orbital strongly correlated systems with both Hund and
Microscopic Hamiltonian—We first analyze the electronic properties in normal state based on the transport experiments [6]. In the parent compounds, both the resistivity and Hall effect measurements show Kondo transport with a logarithmic temperature dependence from tens to around several kelvin. The Kondo effects were attributed to the hybridization between the Nd 5d states and the Nd 4f or Ni 3d states as in rare-earth heavy fermion compounds although the \textit{ab initio} study suggests the hybridization between the Ni 3d state and the Nd 5d states is negligible, and the 4f electron spin fluctuation should be weak due to the large magnetic moment and the energy far away from Fermi energy level [22, 23, 30]. In Nd$_{1-x}$Sr$_x$NiO$_2$, above 60 K, the negative Hall coefficient indicates that the Nd 5d electrons dominate the transport and thermodynamic properties. With the decreasing of temperature, the self-doping effect is reduced as in semiconductors and the Hall coefficient also changes its sign from negative to positive. This means that the doped holes take over the dominant role in the transport and thermodynamics at low temperature. It is still controversial whether the doped hole forms a high spin triplet or a low spin singlet doublon with the original hole on the $d_{x^2-y^2}$ orbital [24, 30]. In fact, a Ni$^{2+}$ ion with $d^8$ configuration often has a high spin $S = 1$ in common Nickel oxides as the result of Hund coupling.

According to the first principle calculation, the energy of the triplet state is around 1 eV lower than that of the singlet, and the top of the Ni $d_{xy}$ or/and $d_{x^2-y^2}$ orbital band is also close to the Fermi energy [23, 34, 35]. Moreover, the holes doped on the $d_{xy}$ or/and $d_{x^2-y^2}$ orbitals could itinerate freely, agreeing well with the positive Hall coefficient at low temperature. The delocalization of the doped holes on $d_{xy}$ or/and $d_{x^2-y^2}$ orbitals is attributed to the fact that the doped hole concentration is dilute, and under the short range antiferromagnetic (AF) correlation background, a hole can hop freely to its next nearest neighbor sites without energy cost as long as these sites are not occupied by another doped hole. In contrast, the doped holes on the already half-filled $d_{x^2-y^2}$ orbitals tend to be localized at low temperature, otherwise the hopping disturbs the magnetic configurations of short-range AF correlations as in cuprates [3].

Based on the aforementioned analysis, we confine our study to the Ni-O planes and assume that the holes doped on the $d_{xy}$ or/and $d_{x^2-y^2}$ orbitals form a conducting band, coexisting with the localized $d_{x^2-y^2}$ orbital band. $c_i = (c_{i\uparrow}, c_{i\downarrow})^T$ is introduced as the annihilation operator of the bare carrier particles on the $d_{xy}$ or/and $d_{x^2-y^2}$ orbitals of the $i$th Ni site, and $d_i = (d_{i\uparrow}, d_{i\downarrow})^T$ is the real space annihilation operator of the localized holes on the Ni $d_{x^2-y^2}$ orbitals. The Hamiltonian is written as

$$H = H_c + H_d + U_c \sum_i n_{ci\uparrow} n_{ci\downarrow} + U_d \sum_i n_{di\uparrow} n_{di\downarrow}$$

with

$$H_c = \varepsilon_{d0} \sum_i c_i^\dagger c_i - \sum_{i,j} t_{cij} c_i^\dagger c_j$$

and

$$H_d = \varepsilon_{d0} \sum_i d_i^\dagger d_i - \sum_{i,j} t_{dij} c_i^\dagger c_j,$$
safely ignore the correlation between the carriers. Furthermore, we take the \( t_{ij} \) hopping as the perturbation and then the Hubbard model for the \( d_{x^2-y^2} \) orbital band is reduced to a Heisenberg model. Finally, we arrive at a Hund-Heisenberg model,

\[
H = H_c - J_H \sum_i \mathbf{S}_i \cdot \mathbf{S}_j + J_H \sum_{\langle i,j \rangle} \mathbf{S}_{di} \cdot \mathbf{S}_{dj} \tag{4}
\]

where \( \mathbf{S}_i = c_i^\dagger \mathbf{\sigma} c_i / 2 \) and \( \mathbf{S}_{di} = d_i^\dagger \mathbf{\sigma} d_i / 2 \) with the Pauli vector \( \mathbf{\sigma} \). \( J_H \) is the Heisenberg interaction between the \( d_{x^2-y^2} \) orbital holes on the Ni square lattice. Here, we have ignored the kinetic energy of the localized holes, and the carrier chemical potential \( \varepsilon_{c0} \) has been replaced by \( \varepsilon_c \) in \( H_c \) to include the energy renormalization from the Coulomb interaction of the holes on the \( d_{x^2-y^2} \) orbitals.

This model is formally similar to the Kondo-Heisenberg model on a 2D square lattice \([36, 37]\). In a similar way, spin fluctuations of the localized \( d_{x^2-y^2} \) states not only act as the pairing ‘glue’ of the \( d \)-wave SC, but also results in non-Fermi liquid in normal state \([37]\). The difference is the conducting carrier ferromagnetically coupled to the localized magnetic moments rather than antiferromagnetically. Moreover, the onsite Hund ferromagnetic coupling is in favor of the delocalization of the doped holes even at very low temperature, and thus it is difficult to form a pseudogap. In the following, we study this model in normal and superconducting state, respectively.

**Normal state**—The bare doped holes are assumed to compose a dilute Fermi gas without interaction between them except in the superconducting phase. At low temperature, the transport and thermodynamic properties are determined by the imaginary part of the quasiparticle self-energy due to the interaction with the renormalized spin fluctuation \( \chi_d(q, \omega) \) of the localized \( d_{x^2-y^2} \) holes. Within the Born approximation, the imaginary part of the momentum-integral self-energy reads \([37]\)

\[
\text{Im} \Sigma_c(\omega) \sim J_H^2 \rho_0^q \int_{-\omega_c}^{\omega_c} dv \left[ n_B(v) + n_F(\omega + v) \right] \text{Im} \chi_d(v)
\]

where \( n_B \) and \( n_F \) are the Bose and Fermi functions, the magnetic fluctuations \( \chi_d(v) = \int d^2q \chi_d(q, v)/4\pi^2 \) with the upper cutoff frequency \( \omega_c \). \( \rho_0^q \) is the density of states of the carriers, a constant value for an ideal 2D Fermi gas with quadratic dispersion. It is worth noting that the neglect of the higher order self-energy corrections is based on the fact that the carriers is weakly magnetised by the local magnetic moments or \( |\langle \mathbf{S}_c \rangle| \ll 1/2 \). Therefore, the Hund coupling in the effective Hamiltonian only gives perturbation correction to the carrier self-energy despite the large Hund coupling constant.

Given that the momentum-integral spin-fluctuation spectra of localized holes take the similar form of the cuprates, e.g. \( \text{Im} \chi_d(v) \sim \tanh(v/2T) \), one has the marginal Fermi liquid-like self-energy \([37]\)

\[
\text{Im} \Sigma_c(\omega, T) \sim \pi \rho_0^q J_H^2 \left[ 1 + \frac{\omega}{2T} \tanh \left( \frac{\omega}{2T} \right) \right] \sim \max \left( |\omega|, T \right).
\]

Then the linear temperature dependence of electrical resistivity as well as some other anomalous transport properties could be explained.

In cuprates, since the doped holes are antiferromagnetically coupled to the localized holes, the hopping disturbs the original magnetic configuration, and thus the doped holes tend to be localized at low doping or at low temperature. On the contrary, in nickelates, the doped holes, which are ferromagnetically coupled to localized holes, could hop over the Ni-O Plane without affecting the magnetic background so that they are not easy to be trapped around the local magnetic moments even at low temperature. Therefore, we propose that it is almost impossible to observe a pseudogap in nickelates.

**Superconductivity**—In normal state, the carriers are scattered by the short-range spin fluctuation as the metallic gas by phonons. In the superconducting state, the carrier pairing is mediated by the spin fluctuations as the BCS mechanism. It is worthy of note that we assume that the conducting carriers only partially screen the local moments without formation of localized triplets or singlets, and then the Heisenberg interaction between the screened moments and their surroundings could survive. Thus, the carriers on site \( i \) and \( j \) could interact with each other by exchanging the spin fluctuations. The interaction Hamiltonian can be written in the coordinate representation as \([37]\)

\[
H_{sc} = J_H^2 \chi_d(\langle i, j \rangle, \omega) \sum_{\langle i, j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \tag{7}
\]

where the nearest neighbor \( \chi_d(\langle i, j \rangle, \omega) \) is assumed to be space independent in homogenous systems. Combining with \( H_c \), a \( t-J \)-like model is reached. Interestingly, despite the formal similarity with the conventional \( t-J \) model \([38]\), here the spin-like operator \( \mathbf{S}_c \) is associated with the carriers on the Ni sites rather than the local moments. Therefore, it is distinct from the conventional \( t-J \) model. In consideration of the short AF spin correlation length at low temperature, we only take the nearest-neighbor magnetic coupling into account in Eq. \((7)\). After transforming to the momentum space, on the square lattice of the Ni-O planes, the pairing interaction is written as

\[
V_{k,k'} = -2g \left[ \cos k_x \cos k'_x + \cos k_y \cos k'_y \right], \tag{8}
\]

where \( g = 3J_H^2 \chi_d(\langle i, j \rangle, \omega)/4 \), approximately assumed to be a constant within the spin fluctuation cutoff energy \( \hbar \omega_c \), otherwise, 0. The pairing interaction could
be further decoupled into a \( d \)-wave (\( \cos k_x - \cos k_y \)) and an extended \( s \)-wave (\( \cos k_x + \cos k_y \)) components. Since the AF susceptibility in momentum space peaks around AF wave vector \( \mathbf{q} \), only the \( d \)-wave pairing channel is favored \([23, 34]\) and the attractive pairing interaction dominantly mediates the carriers on the nearest-neighbor unit cells \([35, 36]\). Consequently, the superconducting phase emerges in \( d_{x^2−y^2} \)-wave pairing channel. The SC transition temperature \( T_c \sim \omega_c e^{-1/\lambda} \) with \( \lambda = g \delta \Omega_c/2 \) for weak coupling \( d \)-wave superconductors. Since the AF correlation in nickelates is weaker than that in cuprates, the spin fluctuation cutoff energy \( \hbar \omega_c \) should be lower than that in cuprates. Moreover, the Hund coupling \( J_h \) is smaller than the magnetic coupling \( J_K \) between the O carriers and Cu local moments in cuprates. Therefore, the lower critical temperature \( T_c \) in nickelates could be understood.

Discussion and conclusion— Actually, we could not exclude the possibility that the doped holes go to the Ni \( d_{3z^2−r^2} \) orbitals although the onsite Coulomb repulsion pushes the \( d_{x^2−y^2} \) lower Hubbard band away from the Fermi level \([3, 5, 11, 14, 15]\). Nevertheless, if the doped holes forms onsite spin singlet with the original localized hole, the already weak AF coupling is further suppressed and hence the superconductivity as well. The critical temperature should also be sensitive to the doping level. In addition, pseudogap should emerge at low temperature as in cuprates. On the contrary, the doped holes on the Ni-O planes can be further checked by neutron scattering and more transport experiments could be conducted to verify our assumption. Moreover, if the doped holes enter the Ni \( d_{xy} \) or/and \( d_{3z^2−r^2} \) orbital then the doping does not suppress the AF fluctuations. This also could be judged by the neutron scattering measurements. In addition, to experimentally determine if the doped holes enter the \( d_{x^2−y^2} \) or/and \( d_{3z^2−r^2} \) orbital, one way is to apply Ni L-edge polarized x-ray absorption near edge structure (XANES) on the single-crystals to study the distribution of holes in the Ni 3d orbitals \([37]\).

In conclusion, we have proposed a Hund-Heisenberg model to investigate the unconventional SC in the infinite-layer nickelates superconductor. By analyzing the transport experiments, we suggest that the doped holes enter the Ni \( d_{xy} \) or/and \( d_{3z^2−r^2} \) orbitals, and form a conducting band. The doped holes interact with the localized holes on \( d_{x^2−y^2} \) orbital through the onsite Hund coupling. We show that the non-Fermi liquid state in normal phase results from the carrier gas interacting with the spin fluctuations of the localized holes. In the superconducting phase, it is still the short-range spin fluctuations that mediates the carriers into Cooper pairs and leads to \( d \)-wave superconductivity. We expect experiments to check our predictions that the doped holes slightly enhance the spin fluctuations and a pseudogap hardly forms in nickelates. We have provided a new SC mechanism for multi-orbital strongly correlated systems and it should aid in searching or synthesizing new superconductors in transition or rare-earth metal oxides.

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