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Interplay of charge and spin correlations in nickel perovskites

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Analyzing the motion of low-spin ($s = 1/2$) holes in a high-spin ($S = 1$) background, we derive a sort of generalized t-J Hamiltonian for the NiO_2 planes of Sr-doped nickelates. In addition to the rather complex carrier-spin and spin-spin couplings we take into account the coupling of the doped holes to in-plane oxygen breathing modes by a Holstein-type interaction term. Because of strong magnetic confinement effects the holes are nearly entirely prelocalized and the electron-phonon coupling becomes much more effective in forming polarons than in the isostructural cuprates. In the light of recent experiments on $La_{2-x}Sr_xNiO_4$ we discuss how the variety of the observed transport and charge/spin-ordering phenomena can be qualitatively understood in terms of our model Hamiltonian.

In contrast to the superconducting cuprates, e.g. $La_{2-x}Sr_xCuO_4$, the layered transition metal oxide $La_{2-x}Sr_xNiO_4$ becomes metallic only near $x = 1$. In either case, however, the measurements of the electrical transport and the magnetic susceptibility, as well as the investigations of the lattice and magnetic structures revealed a rich variety of phenomena indicating the close interconnection between the charge- and spin-ordering and the transport properties [1, 2].

The studies on doped $La_{2-x}Sr_xNiO_4$ indicate that the additional holes in the NiO_2 planes have their own magnetic moment, i.e., they carry a spin $s = 1/2$ and couple to the Ni^{2+} (d^8) ions with spin $S = 1$ (the high-spin state, HSS) in a way that a low-spin state (LSS) with total spin $J = 1/2$ is formed [3]. Excluding the hole doubly-occupied sites, we shall define our model in the tensor product space of local hole states $\{|i,\sigma\rangle,\ |i,0\rangle\}$ and local $S = 1$ states $\{|i,S,m\rangle\}$. Here, $|i,\sigma\rangle$ means an eigenfunction of spin operators $\vec{s}_i^{\,2}$, s_i^z of a hole at the site i, $|i,0\rangle$ corresponds to no extra hole at i, and $|i,S,m\rangle$ denotes the eigenfunction of $\vec{S}_i^{\,2}$, S_i^z of the HSS with spin projection $m = 1, 0, -1$ at the site i.

In the case of nonmaximal total spin J, we shall construct our Hamiltonian in the subspace of the tensor product space defined by the local basis vectors $\{|i,S,m\rangle|i,0\rangle\}$ and $\{|i,J = 1/2, M = \pm 1/2\rangle\}$, i.e.,

$$|i,\tfrac{1}{2},M\rangle = \tfrac{1}{\sqrt{3}}\left\{-[\tfrac{3}{2}-M]^{1/2}\,|i,S,M-\tfrac{1}{2}\rangle|i,\uparrow\rangle + [\tfrac{3}{2}+M]^{1/2}\,|i,S,M+\tfrac{1}{2}\rangle|i,\downarrow\rangle\right\}. \quad (1)$$

In Ref. [3], the hole-transport Hamiltonian given by all processes conserving the total-spin z-component was expressed by means of operators creating HSS from the state of closed Ni-shells $|0\rangle_S$, namely $B_{im}|0\rangle_S = |i,S,m\rangle$, and by fermionic operators creating and annihilating the LSS. In contrast to [3], we take into account the 'interior structure' of the LSS (1) by the Clebsch-Gordon coefficients. This way the hole-transport Hamiltonian can be written as

$$\mathcal{H}_t = -t\sum_{\langle i,j\rangle}\left(X_{i\uparrow}X_{j\uparrow}^{\dagger} + X_{i\downarrow}X_{j\downarrow}^{\dagger}\right) \quad (2)$$

with

$$X_{i\uparrow} = -\sqrt{\tfrac{1}{3}}B_{i,0}^{\dagger}B_{i,0}h_{i\uparrow} + \sqrt{\tfrac{2}{3}}B_{i,0}^{\dagger}B_{i,1}h_{i\downarrow} - \sqrt{\tfrac{2}{3}}B_{i,-1}^{\dagger}B_{i,-1}h_{i\uparrow} + \sqrt{\tfrac{1}{3}}B_{i,-1}^{\dagger}B_{i,0}h_{i\downarrow}, \quad (3)$$

and the time-reversal transformed expression for $X_{i\downarrow}$, where the fermionic operators $h_{i\sigma}$ are defined by $h_{i\sigma}|i,\sigma\rangle = |i,0\rangle$, $h_{i\sigma}^{\dagger}|i,0\rangle = |i,\sigma\rangle$.

To discuss the spin dependence of the charge transport we use a slightly modified treatment of the spin-charge decoupling proposed for the t-J model just recently [4]. In this representation, the operators $h_{i\sigma}$ defined above are expressed in terms of holon (e_i) and (pseudo) spin-1/2 ($\tilde{s}_i$) operators $h_{i\uparrow} = e_i\left(\tilde{s}_i^{+}\tilde{s}_i^{-} + e^{i\varphi}\tilde{s}_i^{-}\right)/\sqrt{2}$ and $h_{i\downarrow} = e_i\left(\tilde{s}_i^{+} + e^{i\varphi}\tilde{s}_i^{-}\tilde{s}_i^{+}\right)/\sqrt{2}$, where the arbitrary phase factor φ does not influence the matrix elements of the Hamiltonian. Consequently, we have for the hole

spin operators $s_i^+ = h_{i\uparrow}^\dagger h_{i\downarrow} = e_i^\dagger e_i \tilde{s}_i^+$, and the site-occupation operator of LSS is given by $n_i = e_i^\dagger e_i$. Then the total spin operator may be cast into the form

$$\vec{J}_i = (1 - n_i)\vec{S}_i + n_i(\vec{S}_i + \vec{\tilde{s}}_i)\,. \tag{4}$$

The spin correlations are determined by antiferromagnetic (AF) exchange interactions

$$\mathcal{H}_{ex} = \sum_{\langle i,j \rangle} \mathcal{J}(n_i, n_j)\vec{J}_i\vec{J}_j\,, \tag{5}$$

where the operators $\vec{J}_i$ are given by (4) and the arguments n_i, n_j of $\mathcal{J}$ indicate the dependence of the exchange on the electronic configuration of the nearest neighbour (NN) sites $\langle i, j \rangle$. We have to keep in mind that $\vec{s}_i$, $\vec{S}_i$ couple one another to form a $J = 1/2$ state, what can be enforced by adding an effective on-site interaction $\mathcal{J}_i n_i \vec{\tilde{s}}_i \vec{S}_i$ which is much stronger than all the inter-site interactions.

The influence of the magnetic correlations on the charge transport will be demonstrated for low doping $x \ll 1$ in which case the holes are moving in the AF background of Ni^{2+} spins. Using the linear spin wave approximation (LSWA) and the representation of $h_{i\sigma}$, $h_{j\sigma}$ in terms of decoupled spin- and charge variables, $\mathcal{H}_t$ given by (2), (3) assumes the form of a spin-dependent transport Hamiltonian for holons. The effective charge-transfer constant is obtained by taking the average of $\mathcal{H}_t$ over the spin degrees of freedom. The average over the spin $S = 1$ background in LSWA leads to

$$\mathcal{H}_t = -\frac{\sqrt{2}}{3} t \sum_{\langle i,j \rangle} e_i e_j^\dagger (\langle \delta S_i^z \rangle + \tfrac{1}{2}\langle S_i^+ S_j^- \rangle)(f_{ji} + g_{ji}), \tag{6}$$

where

$$f_{ji} = \vec{\tilde{s}}_i \vec{\tilde{s}}_j + \frac{1}{4}\,, \tag{7}$$

$$g_{ji} = \frac{e^{i\varphi}}{2}\left[\left(\frac{1}{2} - \tilde{s}_i^z\right)\tilde{s}_j^- + \tilde{s}_i^-\left(\frac{1}{2} + \tilde{s}_j^z\right)\right] \frac{e^{-i\varphi}}{2}\left[\left(\frac{1}{2} + \tilde{s}_i^z\right)\tilde{s}_j^+ + \tilde{s}_i^+\left(\frac{1}{2} - \tilde{s}_j^z\right)\right], \tag{8}$$

and $\langle \delta S_i^z \rangle$ is equal to the reduction of the local $|S_i^z|$ from the classical value S in the AF magnon ground state. Both the expectation values in (6) (being zero in the classical AF Néel ground state) are given by the zero-point spin fluctuations and cause a reduction of the bare hopping constant t. A similar effect is induced also by the remaining spin factor, as the mean value $\langle f_{ji} + g_{ji} \rangle = \langle 2\vec{\tilde{s}}_i \vec{\tilde{s}}_j + \frac{1}{2} \rangle$ and the spins $\vec{\tilde{s}}_i$, $\vec{\tilde{s}}_j$ ought to be AF correlated owing to their coupling to the $S = 1$ spins.

The latter arguments concerning the reduction of the hole hopping rate may be qualitatively also applied to the AF long-range order (LRO) for $x \to 1$, to the ferrimagnetic LRO at $x = 1/2$ as well as to the more general commensurate LRO (for $x = 1/3$) or incommensurate ordering of LSS and HSS with a large correlation length, provided that the spins are mostly AF correlated.

Electron diffraction studies and x-ray scattering technique revealed [5] that the observed variety of magnetic structures in the nickelates is closely related to the lattice structure modulation which indicates the ordering of local lattice deformations connected with the formation of quasi-localized polarons. These findings may be understood in the light of the above model considerations if the interaction of holes with the lattice is taken into account.

In fact, the spin correlations suppressing the charge transport facilitate the polaron self-trapping by electron-phonon coupling [6]. The Holstein-type interaction of holes with an in-plane (breathing) mode has the form

$$\mathcal{H}_{h-p} = -\sum_{\langle i,j \rangle} A u_{ij}(e_i^+ e_i - e_j^+ e_j)\,, \tag{9}$$

where u_{ij} means the displacement of the oxygen in the bond between NN ions i, j. Consequently, the deformation of the bonds leading to the hole localization and energy lowering is given by the charge difference between the NN sites. Thus we expect the polaron ordering given by the energy optimization of the distribution of HSS and LSS. Certain analogy with the usual phase separation exists: in the latter case the optimization of bond distribution is given by the exchange energy, while for the nickelates, the energy gain connected with the localized polaron formation seems to be most important.

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