Charge transport of electron-doped Mott insulators on a triangular lattice

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The charge transport of electron-doped Mott insulators on a triangular lattice is investigated within the t-J model. The conductivity spectrum shows a low-energy peak and an unusual midinfrared band, while the resistivity is characterized by a crossover from the high temperature metalliclike behavior to low temperature insulatinglike behavior, in qualitative agreement with experiments. Our results also show that the mechanism that causes this unusual charge transport is closely related to a competition between the kinetic energy and the magnetic energy in the system.

The recent discovery of superconductivity in doped cobaltates, $Na_xCoO_2 \cdot yH_2O$ with the superconducting transition temperature $T_c \sim 5$ K, has generated great interest due to the role of strong electron correlations.[1] This compound has a lamellar structure consisting of the two-dimensional (2D) CoO_2 layers separated by a thick insulating layer of Na^+ ions and H_2O molecules, where the one-half spin Co^{4+} ions are arranged on a triangular lattice. This structure is similar to cuprates superconductors in the sense that they also have a layered structure of the square lattice of the CuO_2 plane separated by insulating layers.[2] It has been well established that Cu^{2+} ions exhibit an antiferromagnetic (AF) long-range order (AFLRO) in the parent compounds of cuprate superconductors, where superconductivity occurs when the AFLRO state is suppressed by hole or electron doping.[2,3] However, $Na_xCoO_2 \cdot yH_2O$ is viewed as an electron-doped Mott insulator, where superconductivity appears with electron doping.[1] A fundamental similarity between cuprate and cobaltate superconductors has been seen in the decreases in the superconducting transition temperature for both underdoped and overdoped materials.[4] The optimal doping for superconductivity occurs at 0.3 electrons per Co above the ground-state $Na_0CoO_2 \cdot 1.3H_2O$, which is a half-filled two-electron t_{2g}- derived band, while for cuprates, the optimal doping occurs at 0.15 holes (electrons) added to the half-filled band of the parent compound.[4] On the other hand, doped Mott insulators on a triangular lattice, where the geometric spin frustration exists, are also of interest in their own right, with many unanswered fascinating questions. Historically, the undoped Mott insulator on a triangular lattice was first proposed to be a model for microscopic realization of the spin liquid due to the existence of the strong spin frustration.[5] It has been argued that this spin liquid state may play a crucial role in the mechanism for superconductivity in doped cuprates.[6] Thus the unexpected finding of superconductivity in doped cobaltates has raised the hope that it may help solve the unusual physics in doped cuprates.

Some experimental studies have revealed a non-Fermi liquid behavior in doped cobaltates.[1,7,8] Among the striking features in the normal state, a hallmark is the charge transport, where the resistivity shows a temperature linear variation in a wide range of temperatures.[1,7] Moreover, this unusual charge transport is found to be intriguingly related to the magnetic correlation. The charge transport measurement is a powerful probe for interacting electron systems,[9] and can provide very detailed knowledges about the low-energy excitations as electrons are doped to cobaltates. One of the important issues in experimental and theoretical investigations of doped cobaltates is the understanding of the low-energy excitations in these compounds, where the clue to their superconductivity possibly already lies in their nonconventional normal-state properties. Therefore it is of interest to have a detailed look at the charge transport. In this paper, we try to study this issue. We show that the resistivity of electron-doped cobaltates is characterized by a crossover from the high temperature metalliclike behavior to low temperature insulatinglike behavior, and the mechanism that causes this unusual transport is closely related to a competition between the kinetic energy and the magnetic energy in the system.

In electron-doped cobaltates, the characteristic feature is the presence of the 2D CoO_2 plane[1] as mentioned above, and it seems evident that the unusual behaviors are dominated by this plane. It has been argued that the essential physics of the doped CoO_2 plane is contained in the t-J model on a triangular lattice,[10]

$$H = -t_e \sum_{i\hat{\eta}\sigma} P C_{i\sigma}^\dagger C_{i+\hat{\eta}\sigma} P^\dagger - \mu \sum_{i\sigma} P C_{i\sigma}^\dagger C_{i\sigma} P^\dagger + J \sum_{i\hat{\eta}} \mathbf{S}_i \cdot \mathbf{S}_{i+\hat{\eta}},$$

(1)

where $t_e > 0$, the summation is over all sites i, and for each i, over its nearest-neighbor $\hat{\eta}$, $C_{i\sigma}^\dagger$ ($C_{i\sigma}$) is the electron creation (annihilation) operator, $\mathbf{S}_i = C_i^\dagger \sigma C_i / 2$ is the spin operator with $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ as the Pauli matrices, μ is the chemical potential, and the projection operator P removes zero occupancy, i.e., $n_{i\uparrow} + n_{i\downarrow} \geq 1$ with $n_{i\sigma} = C_{i\sigma}^\dagger C_{i\sigma}$. In the past 15 years, some useful methods have been proposed to treat the no double occupancy local constraint in hole-doped cuprates.[11] In particular, a fermion-spin theory based on the partial charge-spin separation has been developed to study the

physical properties of doped cuprates,[12] where the no-double-occupancy local constraint can be treated properly in analytical calculations. Within this theory, the charge transport of the underdoped cuprates has been discussed,[12,13] and the results are consistent with the experiments.[9,14] To apply this theory in electron-doped cobaltates, the *t-J* model (1) can be rewritten in terms of a particle-hole transformation $C_{i\sigma} \rightarrow f^\dagger_{i-\sigma}$ as[10]

$$H = -t\sum_{i\hat{\eta}\sigma} f^\dagger_{i\sigma} f_{i+\hat{\eta}\sigma} + \mu \sum_{i\sigma} f^\dagger_{i\sigma} f_{i\sigma} + J\sum_{i\hat{\eta}} \mathbf{S}_i \cdot \mathbf{S}_{i+\hat{\eta}}, \quad (2)$$

supplemented by a local constraint $\sum_\sigma f^\dagger_{i\sigma} f_{i\sigma} \leq 1$ to remove double occupancy, where as a matter of convenience, we have set $t=-t_e$, $f^\dagger_{i\sigma}$ $(f_{i\sigma})$ as the hole creation (annihilation) operator, and $\mathbf{S}_i = f^\dagger_i \sigma f_i / 2$ as the spin operator in the hole representation. Now we follow the partial charge-spin separation fermion-spin theory,[12] and decouple hole operators $f_{i\uparrow}$ and $f_{i\downarrow}$ as

$$f_{i\uparrow} = a^\dagger_{i\uparrow} S^-_i, \quad f_{i\downarrow} = a^\dagger_{i\downarrow} S^+_i, \quad (3)$$

where the spinful fermion operator $a_{i\sigma} = e^{-i\Phi_{i\sigma}} a_i$ describes the charge degree of freedom together with the phase part of the spin degree of freedom (dressed fermion), while the spin operator S_i describes the amplitude part of the spin degree of freedom (dressed spinon), then the no-double-occupancy local constraint, $\sum_\sigma f^\dagger_{i\sigma} f_{i\sigma} = S^+_i a_{i\uparrow} a^\dagger_{i\uparrow} S^-_i + S^-_i a_{i\downarrow} a^\dagger_{i\downarrow} S^+_i = a_i a^\dagger_i (S^+_i S^-_i + S^-_i S^+_i) = 1 - a^\dagger_i a_i \leq 1$, is satisfied in analytical calculations, and the double dressed fermion occupancy, $a^\dagger_{i\sigma} a^\dagger_{i-\sigma} = e^{i\Phi_{i\sigma}} a^\dagger_i a^\dagger_i e^{i\Phi_{i-\sigma}} = 0$ and $a_{i\sigma} a_{i-\sigma} = e^{-i\Phi_{i\sigma}} a_i a_i e^{-i\Phi_{i-\sigma}} = 0$, are ruled out automatically. It has been shown that these dressed fermion and spinon are gauge invariant, and in this sense, they are real and can be interpreted as the physical excitations. Since the phase factor $e^{-i\Phi_{i\sigma}}$ is separated from the bare spinon operator, it therefore describes a spinon cloud.[12] In this case, the dressed fermion $a_{i\sigma}$ is a spinless fermion a_i incorporated a spin cloud $e^{-i\Phi_{i\sigma}}$ (magnetic flux), and is a magnetic dressing. In other words, the gauge invariant dressed fermion carries some spin messages, i.e., it shares some effects of the spin configuration rearrangements due to the presence of the electron itself.[15] Although in common sense $a_{i\sigma}$ is not a real spinful fermion, it behaves like a spinful fermion. In this partial charge-spin separation fermion-spin representation, the low-energy behavior of the *t-J* model (2) can be expressed as[12]

$$H = -t\sum_{i\hat{\eta}} (a_{i\uparrow} S^+_i a^\dagger_{i+\hat{\eta}\uparrow} S^-_{i+\hat{\eta}} + a_{i\downarrow} S^-_i a^\dagger_{i+\hat{\eta}\downarrow} S^+_{i+\hat{\eta}}) - \mu \sum_{i\sigma} a^\dagger_{i\sigma} a_{i\sigma}$$

$$+ J_{\text{eff}} \sum_{i\hat{\eta}} \mathbf{S}_i \cdot \mathbf{S}_{i+\hat{\eta}}, \quad (4)$$

with $J_{\text{eff}} = (1-\delta)^2 J$. $\delta = \langle a^\dagger_{i\sigma} a_{i\sigma} \rangle = \langle a^\dagger_i a_i \rangle$ is the electron-doping concentration. At the zero doping, the *t-J* model is reduced to the Heisenberg model. Many authors[16] have shown unambiguously that as in a square lattice, there is indeed AFLRO in the ground state of the AF Heisenberg model on a triangular lattice. However, this AFLRO is destroyed more rapidly with increasing doping than that on a square lattice, due to the strong geometry frustration. Therefore there is no AFLRO away from the zero doping, i.e., $\langle S^z_i \rangle = 0$. On the

other hand, in the electron underdoped regime where superconductivity occurs, the weak ferromagnetism can be induced,[17,10] since the effective magnetic coupling in the present case is $J_{\text{eff}} - 2\phi|t|$ with the dressed fermion mean-field (MF) particle-hole parameter $\phi = \langle a^\dagger_{i\sigma} a_{i+\hat{\eta}\sigma} \rangle$.

Since the no-double-occupancy local constraint has been treated properly within the partial charge-spin separation fermion-spin theory, this leads to the disappearing of the extra gauge degree of freedom related to the local constraint.[12] In this case, the charge fluctuation couples only to dressed fermions.[12,13] Based on this theory, the charge transport of the hole-doped Mott insulator on a square lattice has been discussed,[12,13] and the results are consistent with experiments.[9,14] Following their discussions, the optical conductivity of electron-doped Mott insulators on a triangular lattice can be obtained as

$$\sigma(\omega) = (Z\chi te)^2 \frac{1}{2N} \sum_{k\sigma} \gamma^2_{sk} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} A^{(a)}_\sigma(\mathbf{k}, \omega' + \omega) A^{(a)}_\sigma$$

$$\times (\mathbf{k}, \omega') \frac{n_F(\omega' + \omega) - n_F(\omega')}{\omega}, \quad (5)$$

where Z is the number of the nearest-neighbor sites, the spinon correlation function $\chi = \langle S^+_i S^-_{i+\hat{\eta}} \rangle$, $\gamma^2_{sk} = \{[\sin k_x + \sin \times (k_x/2)\cos(\sqrt{3}k_y/2)]^2 + 3[\sin(\sqrt{3}k_y/2)\cos(k_x/2)]^2\}/9$, $n_F(\omega)$ is the fermion distribution function, and the dressed fermion spectral function $A^{(a)}_\sigma(k, \omega)$ is obtained as $A^{(a)}_\sigma(k, \omega) = -2\,\text{Im}\,g_\sigma(k, \omega)$. The full dressed fermion Green's function $g^{-1}_\sigma(k, \omega) = g^{(0)-1}_\sigma(k, \omega) - \Sigma^{(a)}(k, \omega)$ with the MF dressed fermion Green's function $g^{(0)-1}_\sigma(k, \omega) = \omega - \xi_k$, and the second-order dressed fermion self-energy from the dressed spinon pair bubble,[12,13]

$$\Sigma^{(a)}(k, \omega) = \frac{1}{2}(Zt)^2 \frac{1}{N^2} \sum_{pq} \frac{B_{q+p} B_q}{4\omega_{q+p}\omega_q} (\gamma_{q+p+k} + \gamma_{q-k})^2$$

$$\times \left(\frac{F^{(1)}(k,p,q)}{\omega + \omega_{q+p} - \omega_q - \xi_{p+k}} + \frac{F^{(2)}(k,p,q)}{\omega - \omega_{q+p} + \omega_q - \xi_{p+k}} \right.$$

$$\left. + \frac{F^{(3)}(k,p,q)}{\omega + \omega_{q+p} + \omega_q - \xi_{p+k}} + \frac{F^{(4)}(k,p,q)}{\omega - \omega_{q+p} - \omega_q - \xi_{p+k}} \right), \quad (6)$$

where $B_k = \lambda[(2\epsilon\chi^z + \chi)\gamma_k - (\epsilon\chi + 2\chi^z)]$, $\lambda = 2ZJ_{\text{eff}}$, $\epsilon = 1 + 2t\phi/J_{\text{eff}}$, $\gamma_k = [\cos k_x + 2\cos(k_x/2)\cos(\sqrt{3}k_y/2)]/3$, and

$$F^{(1)}(k,p,q) = n_F(\xi_{p+k})[n_B(\omega_q) - n_B(\omega_{q+p})]$$

$$+ n_B(\omega_{q+p})[1 + n_B(\omega_q)], \quad (7a)$$

$$F^{(2)}(k,p,q) = n_F(\xi_{p+k})[n_B(\omega_{q+p}) - n_B(\omega_q)]$$

$$+ n_B(\omega_q)[1 + n_B(\omega_{q+p})], \quad (7b)$$

$$F^{(3)}(k,p,q) = n_F(\xi_{p+k})[1 + n_B(\omega_{q+p}) + n_B(\omega_q)]$$
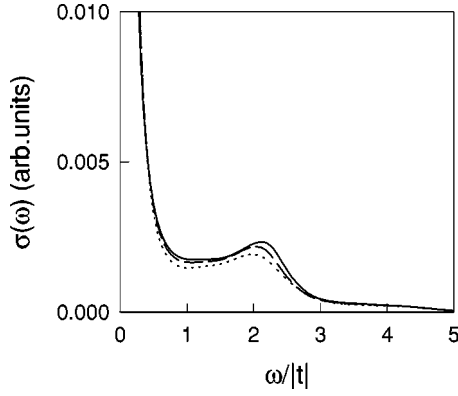
$$+ n_B(\omega_q)n_B(\omega_{q+p}), \quad (7c)$$

FIG. 1. The conductivity of electron-doped Mott insulators on a triangular lattice as a function of frequency at electron doping δ =0.40 (solid line), δ=0.35 (dashed line), and δ=0.30 (dotted line) for $t/J=-2.5$ with temperature $T=0$.

$$F^{(4)}(k,p,q) = [1 + n_B(\omega_q)][1 + n_B(\omega_{q+p})]$$
$$- n_F(\xi_{p+k})[1 + n_B(\omega_{q+p}) + n_B(\omega_q)], \quad (7d)$$

where $n_B(\omega)$ is the Bose distribution function, and the MF dressed fermion and spinon excitation spectra are given by

$$\xi_k = Zt\chi\gamma_k - \mu, \tag{8a}$$

$$\omega_k^2 = A_1\gamma_k^2 + A_2\gamma_k + A_3, \tag{8b}$$

respectively, where

$$A_1 = \alpha\epsilon\lambda^2\left(\frac{1}{2}\chi + \epsilon\chi^z\right), \tag{9a}$$

$$A_2 = \epsilon\lambda^2\left[\frac{1}{Z}(1-Z)\alpha\left(\frac{1}{2}\epsilon\chi + \chi^z\right) - \alpha\left(C^z + \frac{1}{2}C\right)\right.$$
$$\left. - \frac{1}{2Z}(1-\alpha)\right], \tag{9b}$$

$$A_3 = \lambda^2\left[\alpha\left(C^z + \frac{1}{2}\epsilon^2 C\right) + \frac{1}{4Z}(1-\alpha)(1+\epsilon^2)\right.$$
$$\left. - \alpha\epsilon\frac{1}{Z}\left(\frac{1}{2}\chi + \epsilon\chi^z\right)\right], \tag{9c}$$

with spinon correlation functions $\chi^z = \langle S_i^z S_{i+\hat{\eta}}^z\rangle$, $C = (1/Z^2)\Sigma_{\hat{\eta}\hat{\eta}'}\langle S_{i+\hat{\eta}}^+ S_{i+\hat{\eta}'}^-\rangle$, and $C^z = (1/Z^2)\Sigma_{\hat{\eta}\hat{\eta}'}\langle S_{i+\hat{\eta}}^z S_{i+\hat{\eta}'}^z\rangle$. In order to satisfy the sum rule for the correlation function $\langle S_i^+ S_i^-\rangle = 1/2$ in the absence of AFLRO, a decoupling parameter α has been introduced in the MF calculation, which can be regarded as the vertex correction.[18,19] All these mean-field order parameters, the decoupling parameter, and the chemical potential are determined self-consistently.[18]

We have performed a numerical calculation for the optical conductivity in Eq. (5), and the results at electron doping δ =0.40 (solid line), δ=0.35 (dashed line), and δ=0.30 (dotted line) for parameter $t/J=-2.5$ in temperature $T=0$ are shown in Fig. 1, where the charge e has been set as the unit. The conductivity spectrum shows a low-energy peak appearing at
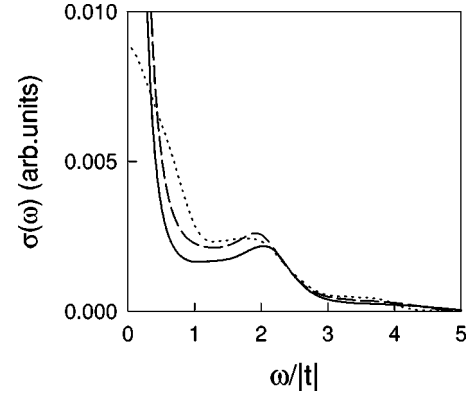

FIG. 2. The conductivity of electron-doped Mott insulators on a triangular lattice as a function of frequency at electron doping δ =0.35 for $t/J=-2.5$ with temperature $T=0$ (solid line), $T=0.3J$ (dashed line), and $T=0.7J$ (dotted line).

$\omega \sim 0$, which decays rapidly, and a broad midinfrared band. This midinfrared band is electron-doping dependent, and the component increases with increasing electron-doping for $1|t| < \omega < 3|t|$, and is nearly independent of electron doping for $\omega > 3|t|$. This reflects an increase in mobile carrier density, and indicates that the spectral weight of the midinfrared side band is taken from the Drude absorption, then the spectral weight from both low-energy peak and midinfrared band represent the actual free-carrier density. For a better understanding of the optical properties of electron-doped Mott insulators on a triangular lattice, we have studied the conductivity at different temperatures, and the results at electron doping δ=0.35 for $t/J=-2.5$ with temperature $T=0$ (solid line), $T=0.3J$ (dashed line), and $T=0.7J$ (dotted line) are plotted in Fig. 2. It is shown that the conductivity spectrum is temperature dependent for $\omega < 3|t|$, and almost temperature independent for $\omega > 3|t|$. The low-energy peak broadens and decreases in height with increasing temperatures, and there is a tendency towards Drude-like behavior. At the same time, the midinfrared band is severely suppressed with increasing temperatures, and vanishes at high temperatures, in qualitative agreement with the hole-doped Mott insulators on a square lattice.[9,12,13,20] As in doped cuprates, the charge transport is governed by the dressed fermion scattering, therefore δ-dressed fermions are responsible for the conductivity, i.e., the optical conductivity in electron-doped cobaltates is carried by δ electrons. Since strong electron correlation is common for both hole-doped cuprates and electron-doped cobaltates, these similar behaviors observed from the optical conductivity are expected.

Now we turn to discuss the resistivity, which can be obtained in terms of conductivity as $\rho(T)=1/\lim_{\omega\to 0}\sigma(\omega)$, and the results for $t/J=-2.5$ at electron doping δ=0.36 (solid line), δ=0.35 (dashed line), and δ=0.34 (dotted line) are plotted in Fig. 3, in comparison with the experimental data[1] taken from $Na_xCoO_2 \cdot yH_2O$ (inset). Our present results show obviously that $\rho(T)$ is characterized by a crossover from the high temperature metalliclike behavior $(d\rho(T)/dT>0)$ to low temperature insulatinglike behavior $(d\rho(T)/dT<0)$, but the metalliclike linear temperature dependence dominates over a wide temperature range, which is qualitatively consistent with experiments.[1,7]
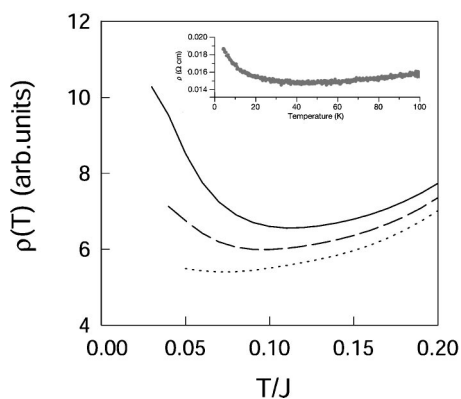
FIG. 3. The resistivity of electron-doped Mott insulators on a triangular lattice as a function of temperature for $t/J = -2.5$ at electron doping $\delta = 0.36$ (solid line), $\delta = 0.35$ (dashed line), and $\delta = 0.34$ (dotted line). Inset: the experimental result on $Na_{0.35}CoO_2 \cdot 1.3H_2O$ taken from Ref. 1.

An explanation for this unusual charge transport can be found from a competition between the kinetic energy and magnetic energy, since the present t-J model (4) is characterized by the competition between the kinetic energy (δt) and magnetic energy (J), with the magnetic energy J favoring the magnetic order for spins, while the kinetic energy δt favors the delocalization of electrons and tends to destroy the magnetic order. Although both dressed fermions and spinons contribute to the charge transport in the partial charge-spin separation fermion-spin theory, the dressed fermion scattering dominates the charge transport.[12,13] The dressed fermion scattering rate is obtained from the full dressed fermion Green's function (then the dressed fermion self-energy and spectral function) by considering the interaction between the dressed fermion and spinon. In this case, the crossover from the high temperature metalliclike behavior to low temperature insulatinglike behavior in the resistivity of electron-doped cobaltates is closely related to this competition. In lower temperatures, when the dressed fermion kinetic energy is much smaller than the magnetic energy, then the magnetic fluctuation is strong enough to severely reduce the dressed fermion scattering and thus, is responsible for the insulatinglike behavior in the resistivity. With increasing temperatures, the dressed fermion kinetic energy is increased, while the dressed spinon magnetic energy is decreased. In the region where the dressed fermion kinetic energy is much larger than the dressed spinon magnetic energy at higher temperatures, the dressed fermion scattering would give rise to the metalliclike resistivity.

We emphasize that this competition between the kinetic energy and magnetic energy exists in all doped Mott insulators. However, in the doped two-leg ladder Mott insulators, the charged carrier's motion is also suppressed by interference effects between the two legs;[21] therefore, in the doped two-leg ladder Mott insulators, both competition between the kinetic energy and magnetic energy and the interference effects between the two legs cause the unusual charge transport. This is why even in the hole-doped two-leg ladder systems, the metalliclike behavior state appears in a much

higher doping concentration. On the other hand, hole-doped Mott insulators (opposite sign of hopping t) on a triangular lattice have been discussed by many authors,[22,23] where the conventional quasiparticle picture may be broken by the effects of geometric spin fluctuation.[22] These and related differences[23] between hole- and electron-doped Mott insulators on a triangular lattice reflect that there is no particle-hole symmetry, and the sign of t is important.[10]

In summary, we have studied the charge transport of electron-doped cobaltates within the t-J model based on the partial charge-spin separation fermion-spin theory. It is shown that the conductivity spectrum shows a low-energy peak and a broad midinfrared band, while the resistivity is characterized by a crossover from the high temperature metalliclike to low temperature insulatinglike behavior. Our results also show that the mechanism that causes this unusual charge transport is closely related to the competition between the kinetic energy and magnetic energy in the system.

Recent experimental studies[24] have shown that the superconducting transition temperature in $Na_xCoO_2 \cdot yH_2O$ is proportional to the electron-doping concentration in the underdoped regime, and satisfies the Uemura relation for hole-doped cuprates. This and other experiments have shown that there is a remarkable resemblance in superconducting-state properties between the electron-doped cobaltate $Na_xCoO_2 \cdot yH_2O$ and hole-doped cuprates,[24,4,1] manifesting the idea that two systems have a similar underlying superconducting mechanism. Within the partial charge-spin separation fermion-spin theory, the mechanism of superconductivity in hole-doped cuprates[25] has been discussed, where dressed holons interact occurring directly through the kinetic energy by exchanging the dressed spinon excitations, leading to a net attractive force between the dressed holons, then the electron Cooper pairs originating from the dressed holon pairing state are due to the charge-spin recombination, and their condensation reveals the superconducting ground state. In this case, the electron superconducting transition temperature is determined by the dressed holon pair transition temperature, and is proportional to the hole-doping concentration in the underdoped regime, in agreement with the experiments. Since the strong electron correlation is common for both hole-doped cuprates and electron-doped cobaltates, then it is possible that superconductivity in electron-doped cobaltates is also driven by kinetic energy, as in hole-doped cuprates. Following the discussions in Ref. 25, we have studied this issue,[26] and found that the superconducting transition temperature in electron-doped cobaltates is suppressed to a lower temperature due to the strong frustration. These and related theoretical results will be presented elsewhere.

After submitting this paper, we became aware of recent optical measurements[27] on $Na_{0.7}CoO_2$ supporting our theoretical results.

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