

Pseudospin S=1 description of the cuprate complexity: the charge triplets model

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Abstract. We introduce a minimal model for 2D cuprates with the on-site Hilbert space reduced to only three effective valence centers $\text{CuO}_4^{7-,6-,5-}$ (nominally $\text{Cu}^{1+,2+,3+}$) and make use of the S=1 pseudospin formalism. Despite its seeming simplicity the model is believed to capture the salient features both of the hole- and electron-doped cuprates. The pseudospin formalism elucidates a unique fermion-boson duality of the doped cuprates, does provide an unified standpoint for classification of the "myriad" of electronic phases in cuprates and the evolution of the CuO_2 planes under a nonisovalent doping, introduces the on-site mixed valence quantum superpositions and order parameters to be novel features of the cuprate physics, does provide a comprehensive description of the correlated one- and two-particle transport, coexistence of *p*- and *n*-type carriers, electron-hole asymmetry, anticorrelation of conventional spin and superconducting order parameters. Concept of the electron and hole centers, differing by a composite local boson, and electron-hole pairing are shown to explain central points of the cuprate puzzles, in particular, the HTSC itself, pseudogap, and Fermi surface reconstruction.

1. Introduction

Recently [1] we argued that an unique property of high- T_c cuprates is related with a dual nature of the Mott insulating state of the parent compounds that manifests itself in two distinct energy scales for the charge transfer (CT) reaction: $\text{Cu}^{2+} + \text{Cu}^{2+} \rightarrow \text{Cu}^{1+} + \text{Cu}^{3+}$. Indeed, the *d-d* CT gap as derived from the optical measurements in parent cuprates such as La_2CuO_4 is 1.5-2.0 eV while the true (thermal) *d-d* CT gap, or effective correlation parameter U_d , appears to be as small as 0.4-0.5 eV. It means cuprates should be addressed to be *d-d* CT unstable systems whose description implies accounting of the three many-electron valence states $\text{CuO}_4^{7-,6-,5-}$ ($\text{Cu}^{1+,2+,3+}$) on an equal footing as a well-defined charge triplet.

Below we introduce a minimal model with the on site Hilbert space reduced to only three states, three effective valence centers $\text{CuO}_4^{7-,6-,5-}$ (nominally $\text{Cu}^{1+,2+,3+}$) where the electronic and lattice degrees of freedom get strongly locked together, and make use of the S=1 pseudospin formalism we have proposed earlier [1-4]. Such a formalism constitutes a powerful method to study complex phenomena in interacting quantum systems characterized by the coexistence and competition of various ordered states [5].

2. Pseudospin formalism

In frames of the pseudospin formalism we address a mixed-valence (MV) system with three possible stable on-site valence states M^0, M^\pm , hereafter a bare center $M^0 = \text{CuO}_4^{6-}$, a hole center $M^+ = \text{CuO}_4^{5-}$, and an electron center $M^- = \text{CuO}_4^{7-}$, respectively, and neglect all other degrees



of freedom focusing on the quantum charges. Three different valence charge states M^0, M^\pm we associate with three components of the $S = 1$ pseudo-spin (isospin) triplet with $M_S = 0, \pm 1$, respectively. Such a correspondence immediately implies introduction of the unconventional on-site MV quantum superpositions $|\Psi\rangle = \sum_M c_M |1M\rangle$, or

$$|\Psi\rangle = c_{-1}|Cu^{1+}\rangle + c_0|Cu^{2+}\rangle + c_1|Cu^{3+}\rangle, \quad (1)$$

that points to many novel effects related with local Cu states. However, we should note that at variance with spinless ground states of Cu^{1+} and Cu^{3+} centers the bare Cu^{2+} center has a conventional spin $s=1/2$, in other words we arrive at the $S=1$ pseudospin model with doubly degenerate $M=0$ value. In the partition function of the classical spin model, this leads to a factor of 2 for every Cu^{2+} site.

The $S = 1$ spin algebra includes eight independent nontrivial pseudo-spin operators, three dipole and five quadrupole operators:

$$\hat{S}_z; \hat{S}_\pm = \mp \frac{1}{\sqrt{2}}(S_x \pm iS_y); \hat{S}_z^2; \hat{T}_\pm = \{S_z, S_\pm\}; \hat{S}_\pm^2. \quad (2)$$

The two fermion-like pseudospin raising/lowering operators S_\pm and T_\pm change the pseudo-spin projection by ± 1 , with slightly different properties

$$\begin{aligned} \langle 0|\hat{S}_\pm|\mp 1\rangle &= \langle \pm 1|\hat{S}_\pm|0\rangle = \mp 1, \\ \langle 0|\hat{T}_\pm|\mp 1\rangle &= -\langle \pm 1|\hat{T}_\pm|0\rangle = +1. \end{aligned} \quad (3)$$

In lieu of \hat{S}_\pm and \hat{T}_\pm operators one may use two novel operators \hat{P}_\pm and \hat{N}_\pm :

$$\hat{P}_\pm = \frac{1}{2}(\hat{S}_\pm + \hat{T}_\pm); \hat{N}_\pm = \frac{1}{2}(\hat{S}_\pm - \hat{T}_\pm), \quad (4)$$

which do realize transformations $Cu^{2+} \leftrightarrow Cu^{3+}$ and $Cu^{1+} \leftrightarrow Cu^{2+}$, respectively. The boson-like pseudospin raising/lowering operators \hat{S}_\pm^2 do change the pseudo-spin projection by ± 2 and define a local nematic order parameter $\langle S_\pm^2 \rangle = \frac{1}{2}(\langle S_x^2 - S_y^2 \rangle \pm i\langle \{S_x, S_y\} \rangle)$. This on-site off-diagonal order parameter is nonzero only for the on-site $M^-(Cu^{1+})-M^+(Cu^{3+})$ superpositions. It is worth noting that the \hat{S}_+^2 (\hat{S}_-^2) operator creates an on-site hole (electron) pair, or composite boson, with a kinematic constraint $(\hat{S}_\pm^2)^2 = 0$, that underlines its "hard-core" nature. Obviously, the pseudospin nematic average $\langle S_\pm^2 \rangle$ can be addressed to be a local complex superconducting order parameter. Both $\hat{S}_+(\hat{S}_-)$ and $\hat{T}_+(\hat{T}_-)$ can be anyhow related with a conventional $s=1/2$ spin single particle creation (annihilation) operators, however, these are not standard fermionic ones, as well as \hat{S}_+^2 (\hat{S}_-^2) operators are not standard bosonic ones.

3. Effective S=1 pseudospin Hamiltonian

Effective S=1 pseudospin Hamiltonian which does commute with the z-component of the total pseudospin $\sum_i S_{iz}$ thus conserving the total charge of the system can be written to be a sum of potential and kinetic energies:

$$\hat{H} = \hat{H}_{ch} + \hat{H}_{tr}, \quad (5)$$

where

$$\hat{H}_{ch} = \sum_i (\Delta_i S_{iz}^2 - (\mu - h_i) S_{iz}) + \sum_{i<j} V_{ij} S_{iz} S_{jz}, \quad (6)$$

and $\hat{H}_{tr} = \hat{H}_{tr}^{(1)} + \hat{H}_{tr}^{(2)}$ being a sum of one-particle and two-particle transfer contributions. In terms of \hat{S}_{\pm} and \hat{T}_{\pm} operators $\hat{H}_{tr}^{(1)}$ and $\hat{H}_{tr}^{(2)}$ read as follows:

$$\begin{aligned} \hat{H}_{tr}^{(1)} = & \sum_{i<j} t_{ij}(S_{i+}S_{j-} + S_{i-}S_{j+}) + \sum_{i<j} t'_{ij}(T_{i+}T_{j-} + T_{i-}T_{j+}) \\ & + \frac{1}{2} \sum_{i<j} t''_{ij}(S_{i+}T_{j-} + S_{i-}T_{j+} + T_{i+}S_{j-} + T_{i-}S_{j+}); \end{aligned} \quad (7)$$

$$\hat{H}_{tr}^{(2)} = \sum_{i<j} t_{ij}^b(S_{i+}^2S_{j-}^2 + S_{i-}^2S_{j+}^2), \quad (8)$$

with a charge density constraint:

$$\frac{1}{2N} \sum_i \langle S_{iz} \rangle = \Delta n, \quad (9)$$

where Δn is the deviation from a half-filling ($N_{M^+} = N_{M^-}$). Hamiltonian \hat{H}_{ch} corresponds to a classical spin-1 Ising model with a single-ion anisotropy in the presence of a longitudinal magnetic field. The first single-site term describes the effects of a bare pseudo-spin splitting, or the local energy of $M^{0,\pm}$ centers and relates with the on-site density-density interactions. The second term may be related to a pseudo-magnetic field $\mathbf{h}_i \parallel Z$, in particular, a real electric field which acts as a chemical potential (μ is the hole chemical potential, and h_i is a (random) site energy). The third term in \hat{H}_{ch} describes the effects of the short- and long-range inter-site density-density interactions including screened Coulomb and covalent couplings.

Hamiltonian $\hat{H}_{tr}^{(1)}$ describes the one-particle inter-site hopping and represents an obvious extension of the conventional Hubbard model which assumes that the electronic orbital is infinitely rigid irrespective of occupation number, and has much in common with so-called dynamic Hubbard models [6] that describe a correlated hopping. Indeed, conventional Hubbard model implies all the single particle transfer (7) is governed only by the SS -term in (7) while the TT and ST terms describe a non-Hubbard correlated density-dependent single-particle hopping. In terms of \hat{P}_{\pm} and \hat{N}_{\pm} operators the Hamiltonian $\hat{H}_{tr}^{(1)}$ transforms as follows:

$$\begin{aligned} \hat{H}_{tr}^{(1)} = & \sum_{i<j} t_{ij}^p(P_{i+}P_{j-} + P_{i-}P_{j+}) + \sum_{i<j} t_{ij}^n(N_{i+}N_{j-} + N_{i-}N_{j+}) \\ & + \frac{1}{2} \sum_{i<j} t_{ij}^{pn}(P_{i+}N_{j-} + P_{i-}N_{j+} + N_{i+}P_{j-} + N_{i-}P_{j+}), \end{aligned} \quad (10)$$

where

$$t_{ij}^{p,n} = t_{ij} + t'_{ij} \pm t''_{ij}; \quad t_{ij}^{pn} = t_{ij} - t'_{ij}. \quad (11)$$

All the three terms in (10) suppose a clear physical interpretation. The first PP -type term describes one-particle hopping processes: $Cu^{3+} + Cu^{2+} \leftrightarrow Cu^{2+} + Cu^{3+}$, that is a rather conventional motion of the hole M^+ (Cu^{3+}) centers in the lattice formed by M^0 (Cu^{2+})-centers (p -type carriers, respectively) or the motion of the M^0 (Cu^{2+})-centers in the lattice formed by hole M^+ (Cu^{3+}) centers (n -type carriers, respectively). The second NN -type term describes one-particle hopping processes: $Cu^{1+} + Cu^{2+} \leftrightarrow Cu^{2+} + Cu^{1+}$, that is a rather conventional motion of the electron M^- (Cu^{1+}) centers in the lattice formed by M^0 (Cu^{2+})-centers (n -type carriers) or the motion of the M^0 (Cu^{2+})-centers in the lattice formed by electron M^- (Cu^{1+}) centers (p -type carriers). These hopping processes are typical ones for heavily underdoped or heavily overdoped cuprates. It is worth noting that the ST -type contribution of the one-particle transfer differs in sign for the PP and NN transfer thus breaking the electron-hole

symmetry. The third PN (NP) term in (10) defines a very different one-particle hopping process: $Cu^{2+} + Cu^{2+} \leftrightarrow Cu^{3+} + Cu^{1+}$, $Cu^{1+} + Cu^{3+}$, that is the *local disproportionation/recombination*, or the *EH-pair creation/annihilation*. Interestingly, the term can be related with a local pairing as the Cu^{1+} center can be addressed to be electron pair (= composite electron boson) localized on the Cu^{3+} center or *vice versa* the Cu^{3+} center can be addressed to be hole pair (= composite hole boson) localized on the Cu^{1+} center.

Hamiltonian $\hat{H}_{tr}^{(2)}$ describes the two-particle (local composite boson) inter-site hopping, that is the motion of the electron (hole) center in the lattice formed by the hole (electron) centers, or the exchange reaction: $Cu^{3+} + Cu^{1+} \leftrightarrow Cu^{1+} + Cu^{3+}$. In other words, t_{ij}^b is the transfer integral for the local composite boson.

Conventional spin degree of freedom can be build in our effective Hamiltonian, if we take into account Heisenberg spin exchange Cu^{2+} - Cu^{2+} coupling as follows

$$\hat{H}_{ex} = \sum_{i>j} (1 - \hat{S}_{iz}^2)(1 - \hat{S}_{jz}^2) I_{ij} (\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j), \quad (12)$$

where $(1 - \hat{S}_{iz}^2)$ is a projection operator which picks out the $s=1/2$ Cu^{2+} center, I_{ij} is an exchange integral. Obviously, the spin exchange provides an energy gain to the parent antiferromagnetic insulating (AFMI) phase with $\langle \hat{S}_{iz}^2 \rangle = 0$, while local superconducting order parameter is maximal given $\langle \hat{S}_{iz}^2 \rangle = 1$. In other words, the superconductivity and magnetism are nonsymbiotic phenomena with competing order parameters giving rise to an inter-twinning, glassiness, and other forms of electronic heterogeneities.

Effective pseudospin Hamiltonian (5) is significantly complicated as compared with a typical $S=1$ spin Hamiltonian with uniaxial single-ion and exchange anisotropies on a square lattice:

$$\hat{H} = \sum_{i>j} J_{ij} (S_{ix}S_{jx} + S_{iy}S_{jy} + \lambda S_{iz}S_{jz}) + \sum_i D S_{iz}^2 - \sum_i h S_{iz}, \quad (13)$$

investigated rather extensively in recent years (see, e.g., Refs. [7] and references therein). Correspondence with our pseudospin Hamiltonian points to $D = \Delta$, $J_{ij} = -t_{ij}$, $\lambda J_{ij} = V_{ij}$ ($t_{ij}^a = t_{ij}^b = 0$; $t_{ij}^c = 0$). The Hamiltonian (13) is invariant under the transformation $J, \lambda \rightarrow -J, -\lambda$ and a shift of the Brillouin zone $\mathbf{k} \rightarrow \mathbf{k} + (\pi, \pi)$. The spectrum of the spin Hamiltonian (13) in the absence of external magnetic field changes drastically as Δ varies from very small to very large positive or negative values. A strong "easy-plane" anisotropy for large positive $\Delta > 0$ favors a singlet phase where all the spins are in the $S_z = 0$ ground state. This quadrupole ($Q_{zz} = -\frac{2}{3}$) phase has no magnetic order, and is aptly referred to as a quantum paramagnetic phase (QPM), which is separated from the "ordered" state by a quantum critical point (QCP) at some $\Delta = \Delta_1$. A strong "easy-axis" anisotropy for large negative $\Delta \leq \Delta_2$, favors a spin ordering along Z , the "easy axis", with the on-site $S_z = \pm 1$ (Z -phase). The order parameter will be "Ising-like" and long-range (staggered) diagonal order will persist at finite temperature, up to a critical line $T_c(\Delta)$. For intermediate values $\Delta_1 > \Delta > \Delta_2$ the Hamiltonian will have $O(2)$ symmetry and the system is in a gapless XY phase. At $T=0$ the $O(2)$ symmetry will be spontaneously broken and the system will exhibit spin order in some direction. Although there will be no ordered phase at finite temperature one expects a finite temperature Kosterlitz-Thouless transition. At finite effective field h_z but $\lambda=1$ the XY phase transforms into a canted antiferromagnetic XY - Z_{FM} phase, the spins acquire a uniform longitudinal component which increases with field and saturates at the fully polarized (FP) state (all $S_z = 1$, Z_{FM} phase) above the saturation field h_s . However, at $D > 0$ and $\lambda > 1$ the phase diagram contains an extended spin supersolid or biconical phase XY - Z_{FIM} with ferrimagnetic z -order that does exist over a range of magnetic fields [7].

4. Pseudospin description of the cuprate physics

The pseudospin Hamiltonian, Eq.(5) differs from its simplified version (13) in several points. This concerns the charge density constraint (9), a significantly more complicated form of the "transversal" (XY) term, existence of the conventional spin $s=1/2$ for $M = 0$ pseudospin states. The pseudospin parameters, in particular Δ , V_{ij} , h in the effective Hamiltonian (5) can be closely linked to each other. At variance with typical spin systems the pseudospin system appears to be strongly anisotropic one with an enhanced role of frustrative effects of in-plane next-nearest neighbor couplings, inter-plane coupling, and different non-Heisenberg biquadratic interactions. Despite the difference we can translate many results of the spin $S=1$ algebra to our pseudospin system. Turning to a classification of the possible homogeneous phases of the charge states of the model cuprates and its phase diagram we introduce *monovalent* (Cu^{1+} , Cu^{2+} , Cu^{3+}), *bivalent* MV-2 ($\text{Cu}^{1+,2+}$, $\text{Cu}^{2+,3+}$, $\text{Cu}^{1+,3+}$), and *trivalent* MV-3 ($\text{Cu}^{1+,2+,3+}$) phases in accordance with character of the on-site superpositions (1). Then, in accordance with the above nomenclature of spin phases and the charge triplet $-S=1$ pseudospin correspondence we arrive at a parent monovalent (Cu^{2+}) phase as an analogue of the QPM phase, the XY^{13} , XY^{123} , $XY-Z_{FM}^{13}$, $XY-Z_{FM}^{23}$, $XY-Z_{FM}^{12}$, $XY-Z_{FM}^{123}$, $XY-Z_{AFM}^{13}$, $XY-Z_{AFM}^{123}$, $XY-Z_{FIM}^{13}$, $XY-Z_{FIM}^{123}$, Z_{FM}^1 , and Z_{FM}^3 phases as mono-, bi-, and trivalent analogues of respective spin phases. All the metallic phases with XY^{13} and XY^{123} components do admit in principle the pseudospin nematic order $\langle S_{\pm}^2 \rangle \neq 0$ related with the high- T_c superconductivity (HTSC). In all the trivalent phases the superconducting order competes with a spin ordering. Moreover, in $XY-Z_{FIM}^{123}$ phase we deal with a competition of superconducting, spin, and charge orders. It is worth noting that the $XY-Z$ nomenclature does strictly reflect an interplay of kinetic (XY -terms) and potential (Z -term) energies, or itineracy and localization.

For the undoped model cuprate with $\sum_i \langle S_{iz} \rangle = 0$ (half filling) given rather large positive $\Delta > \Delta_1$ we arrive at insulating monovalent *quantum paramagnetic* M^0 (Cu^{2+})-phase, a typical one for Mott-Hubbard insulators. In parent cuprates, such as La_2CuO_4 , the Cu^{2+} ions form an antiferromagnetically (AF) coupled square lattice of $s = 1/2$ spins, which could possibly realize the resonant valence bond (RVB) liquid of singlet spin pairs. In the RVB state the large energy gain of the singlet pair state, resonating between the many spatial pairing configurations, drives strong quantum fluctuations strong enough to suppress long range AF order. However, by lowering the Δ below Δ_1 the undoped cuprate can be turned first into metallic and superconducting XY^{123} phase, and given $\Delta < \Delta_2$ into a fully disproportionated MV-2 system of electron M^- and hole M^+ centers (M^{\pm} -phase) with $\langle S_{iz}^2 \rangle = 1$, or *electron-hole Bose liquid* (EHBL) [1-4,8]. There is no single particle transport: $\langle S_{\pm} \rangle = \langle T_{\pm} \rangle = 0$, while the bosonic one may exist, and, in common, $\langle S_{\pm}^2 \rangle \neq 0$. Given the $\Delta \rightarrow -\infty$ condition the EHBL phase is equivalent to the lattice hard-core (hc) Bose system with an inter-site repulsion. Indeed, one may address the electron M^- center to be a system of a local composite boson (e^2) localized on the hole M^+ center: $M^- = M^+ + e^2$. For such a system, the pseudo-spin Hamiltonian (5) can be mapped onto the Hamiltonian of hc Bose gas on a lattice (see Refs. [9,10] and references therein)

$$H = \sum_{i>j} t_{ij}^b (B_i^\dagger B_j + B_j^\dagger B_i) + \sum_{i>j} V_{ij} N_i N_j - \mu \sum_i N_i, \quad (14)$$

where $N_i = B_i^\dagger B_i$, \hat{B}^\dagger (\hat{B}) (\hat{S}_{\pm}^2) are the Pauli creation (annihilation) operators which are Bose-like commuting for different sites, but $B_i^2 = (B_i^\dagger)^2 = 0$, $[B_i, B_i^\dagger] = 1 - 2N_i$. The EHBL model exhibits many fascinating quantum phases and phase transitions. Early investigations [9] point to the $T = 0$ charge order (CO= Z_{AFM}^{13}), Bose superfluid (BS= $XY-Z_{FM}^{13}$) and mixed (BS+CO= $XY-Z_{FIM}^{13}$) supersolid uniform phases with an Ising-type melting transition (CO-NO= $Z_{AFM}^{13}-Z_{FM}^{13}$) and Kosterlitz-Thouless-type (BS-NO= $XY-Z_{FM}^{13}-Z_{FM}^{13}$) phase transitions to a non-ordered normal fluid (NO= Z_{FM}^{13}) in 2D systems. At half-filling ($n_B = 0.5$, $\Delta n = 0$) given

$t^b > V_{nn}$, $V_{nnn}=0$ the EHBL system obviously prefers a superconducting BS= XY^{13} phase while at $t^b < V_{nn}$, $V_{nnn}=0$ it prefers an insulating checkerboard charge order CO= Z_{AFM}^{13} . It is worth noting that the QMC calculations [11] show that under doping away from half filling, the checkerboard solid undergoes phase separation: the superfluid (BS) and solid (CO) phases coexist but not as a single thermodynamic BS+CO phase.

The EHBL model truly reproduces many important aspects of the cuprate physics [1], in particular, the pseudogap phenomenon as a result of the EH coupling. At the same time it cannot explain a number of well-known properties, in particular, manifestation of the Cu^{2+} valence states in doped cuprates over wide doping range [12] and suppression of the superconductivity for overdoped cuprates. Such a behaviour cannot be derived from the EHBL scenario and points to realization of the more complicated "boson-fermion" dual $XY-Z_{FM}^{123}$ phase with coexisting spin and pseudospin (charge) orders in a wide doping range from parent to overdoped compounds including all the superconducting phase. The suppression of the superconductivity for the hole overdoped cuprates can be explained as a transition from the trivalent superconducting (M^{123}) phase to a bivalent nonsuperconducting M^{23} phase. Indeed, the $M^- = Cu^{1+}$ centers could be energetically gainless under hole doping particularly for overscreened EH coupling. Some properties of nonsuperconducting phases M^{23} and M^{12} , or $XY-Z_{FM}^{23}$ and $XY-Z_{FM}^{12}$, can be understood if we address limiting insulating phases M^+ or M^- (Z_{FM}^1 or Z_{FM}^3) with precisely M^+ or M^- -centers on each of the lattice sites. In frames of the pseudospin formalism these phases correspond to fully polarized ferromagnetic states with $S_z^{tot} = \pm N$, where N is the number of Cu sites. Interestingly, in frames of the pseudospin formalism the "heavily overdoped" $XY-Z_{FM}^{23}$ and $XY-Z_{FM}^{12}$ phases with $x \approx 1$ can be represented as ferromagnets where the charge constraint is realized through the occurrence of $(1-x)N$ non-interacting pseudospin magnons ($\Delta S_z = \pm 1$), that is Cu^{2+} centers, obeying Fermi statistics due to $s=1/2$ conventional spin. These heavily overdoped cuprates could be addressed to be conventional Fermi liquids with a large Fermi surface.

5. Conclusion

The $S=1$ pseudospin formalism is shown to provide a conceptual framework for an in-depth understanding and a novel starting point for analytical and computational studies of high- T_c superconductivity and other puzzles in cuprates.

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