Strong-coupling theory of high-temperature superconductivity and colossal magnetoresistance

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We argue that the extension of the BCS theory to the strong-coupling regime describes the high-temperature superconductivity of cuprates and the colossal magnetoresistance (CMR) of ferromagnetic oxides if the phonon dressing of carriers and strong attractive correlations are taken into account. The attraction between carriers, which is prerequisite to high-temperature superconductivity, is caused by an almost unretarted electron-phonon interaction sufficient to overcome the direct Coulomb repulsion in the strong-coupling limit, where electrons become polarons and bipolarons (real-space electron or hole pairs dressed by phonons). The long-range Fröhlich electron-phonon interaction has been identified as the most essential in cuprates providing "superlight" lattice polarons and bipolarons. A number of key observations have been predicted and/or explained with polarons and bipolarons including unusual isotope effects, normal state (pseudo)gaps, upper critical fields, etc. Here some kinetic, magnetic, and more recent thermomagnetic normal state measurements are interpreted in the framework of the strong-coupling theory, including the Nernst effect and normal state diamagnetism. Remarkably, a similar strong-coupling approach offers a simple explanation of CMR in ferromagnetic oxides, while the conventional double-exchange (DEX) model, proposed half a century ago and generalised more recently to include the electron-phonon interaction, is in conflict with a number of modern experiments. Among these experiments are site-selective spectroscopies, which have shown that oxygen p-holes are current carriers rather than d-electrons in ferromagnetic manganites (and in cuprates) ruling out DEX mechanism of CMR. Also some samples of ferromagnetic manganites manifest an insulating-like optical conductivity at all temperatures contradicting the DEX notion that their ferromagnetic phase is metallic. On the other hand, the pairing of oxygen holes into heavy bipolarons in the paramagnetic phase and their magnetic pair-breaking in the ferromagnetic phase account for the first-order ferromagnetic phase transition, CMR, isotope effects, and pseudogaps in doped manganites. Here we propose an explanation of the phase coexistence and describe the shape of resistivity of manganites near the transition in the framework of the strong-coupling approach.

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I. INTRODUCTION: THE "FRÖHLICH-COULOMB" MODEL

Although high-temperature superconductivity (HTS) has not yet been targeted as 'the shame and despair of theoretical physics', - a label attributed to low-temperature superconductivity during the first half-century after its discovery - controversy of current theoretical constructions has led many researchers to say that there is no theory of HTS and no progress in understanding the phenomenon. A significant fraction of theoretical research in the field has suggested that the interaction in novel superconductors is essentially repulsive and unretarted, and it could provide high T_c without phonons. Indeed strong onsite repulsive correlations (Hubbard U) are essential in shaping the insulating state of undoped (parent) compounds. Different from conventional band-structure insulators with completely filled and empty Bloch bands, the Mott insulator arises from a potentially metallic half-filled band as a result of the Coulomb blockade of electron tunnelling to neighboring sites [1]. However, the Hubbard U model shares an inherent difficulty in determining the order when the Mott-Hubbard insulator is doped. While some groups have claimed that it describes high- T_c superconductivity at finite doping, other authors could not find any superconducting instability. Therefore it has been concluded that models of this kind are highly conflicting and confuse the issue by exaggerating the magnetism rather than clarifying it [2].

The Hubbard-U model of high temperature superconductivity or its strong-coupling "t-J" approximation are also refutable on the experimental ground. The characteristic magnetic interaction, which is allegedly responsible for the pairing in the model, is the spin exchange interaction, $J = 4t^2/U$, of the order of 0.1 eV (here t is the hopping integral). On the other hand, a simple

parameter-free estimate of the Fröhlich electron-phonon interaction (routinely neglected within the Hubbard U approach) yields the effective attraction as high as 1 eV [3]. This estimate is obtained using the familiar expression for the polaron level shift, E_p , the high-frequency, ϵ_{∞} , and the static, ϵ_0 , dielectric constants of the host insulator, measured experimentally [4],

$$E_p = \frac{1}{2\kappa} \int_{BZ} \frac{d^3q}{(2\pi)^3} \frac{4\pi e^2}{q^2},\tag{1}$$

where $\kappa^{-1} = \epsilon_0^{-1} - \epsilon_0^{-1}$ and the size of the integration region is the Brillouin zone (BZ). Since $\epsilon_\infty = 5$, $\epsilon_0 = 30$ in La₂CuO₄ and $\epsilon_\infty = 3.9$, $\epsilon_0 = 16$ in LaMnO₃ one obtains $E_p = 0.65$ eV and $E_p = 0.88$ eV in La₂CuO₄ and LaMnO₃, respectively. Hence the attraction, which is about $2E_p$, induced by the lattice deformation in cuprates and manganites is one order of magnitude larger than the exchange (magnetic) interaction. There is virtually no screening of e-ph interactions with c-axis polarized optical phonons in cuprates because the upper limit for the out-of-plane plasmon frequency (< 200 cm⁻¹)[5] is well below the characteristic phonon frequency, $\omega \approx 400$ - 1000 cm⁻¹. The screening in manganites is also very poor since the mobility of carriers is very low. As a result of poor screening the magnetic interaction remains small compared with the Fröhlich interaction at any doping. Further compelling evidence for the strong e-ph interactions has come from the isotope effects in cuprates [6] and manganites [7], recent high resolution angle resolved photoemission spectroscopies [8], and a number of earlier optical [9, 10] and neutron-scattering [11] studies. Hence any realistic approach to HTS in cuprates, other doped oxides and fullerenes, and to CMR in ferromagnetic oxides should treat the long-range Coulomb and unscreened e-ph interactions on an equal footing.

In the past decade we have developed a "Fröhlich-Coulomb" model (FCM) [3, 12, 13] to deal with the strong long-range Coulomb and e-ph interactions in cuprates, manganites and other related compounds. The model Hamiltonian explicitly includes a long-range electron-phonon and the Coulomb interactions as well as the kinetic and deformation energies. The implicitly present large Hubbard U term prohibits double occupancy and removes the need to distinguish fermionic spins since the exchange interaction is negligible compared with the direct Coulomb and the electron-phonon interactions. The model also provides a simple explanation of CMR in ferromagnetic oxides if the exchange interaction of p-holes with d-electron spins is included in the Hamiltonian [14] (see below). Introducing spinless fermionic, $c_{\mathbf{n}}$, and phononic, $d_{\mathbf{m}\alpha}$, operators the Hamiltonian of the model is written as

$$H = -\sum_{\mathbf{n} \neq \mathbf{n}'} \left[t(\mathbf{n} - \mathbf{n}') c_{\mathbf{n}}^{\dagger} c_{\mathbf{n}'} - V_{c}(\mathbf{n} - \mathbf{n}') c_{\mathbf{n}}^{\dagger} c_{\mathbf{n}} c_{\mathbf{n}'}^{\dagger} c_{\mathbf{n}'} \right]$$

$$- \sum_{\mathbf{n} \mathbf{m}} \omega_{\alpha} g_{\alpha}(\mathbf{m} - \mathbf{n}) (\mathbf{e}_{\alpha} \cdot \mathbf{u}_{\mathbf{m} - \mathbf{n}}) c_{\mathbf{n}}^{\dagger} c_{\mathbf{n}} (d_{\mathbf{m}\alpha}^{\dagger} + d_{\mathbf{m}\alpha})$$

$$+ \sum_{\mathbf{m} \alpha} \omega_{\alpha} \left(d_{\mathbf{m}\alpha}^{\dagger} d_{\mathbf{m}\alpha} + 1/2 \right),$$

$$(2)$$

where \mathbf{e}_{α} is the polarization vector of the α th vibration coordinate, $\mathbf{u}_{\mathbf{m}-\mathbf{n}} \equiv (\mathbf{m}-\mathbf{n})/|\mathbf{m}-\mathbf{n}|$ is the unit vector in the direction from electron \mathbf{n} to ion \mathbf{m} , $g_{\alpha}(\mathbf{m}-\mathbf{n})$ is the dimensionless e-ph coupling function, and $V_c(\mathbf{n}-\mathbf{n}')$ is the inter-site Coulomb repulsion. $g_{\alpha}(\mathbf{m}-\mathbf{n})$ is proportional to the *force* acting between the electron on site \mathbf{n} and the ion on \mathbf{m} . For simplicity, we assume that all the phonon modes are non-dispersive with the frequency ω_{α} . We also use $\hbar = k_B = c = 1$.

The Hamiltonian, Eq.(2), has been solved analytically by using the " $1/\lambda$ " multi-polaron expansion technique [3] in the strong limit where the e-ph coupling constant is large, $\lambda = E_p/zt > 1$. Here the polaron level shift is $E_p = \sum_{\mathbf{n}\alpha} \omega_{\alpha} g_{\alpha}^2(\mathbf{n}) (\mathbf{e}_{\alpha} \cdot \mathbf{u}_{\mathbf{n}})^2$, and zt is a half-bandwidth in the rigid lattice. The model shows a reach phase diagram depending on the ratio of the inter-site Coulomb

repulsion V_c and the polaron level shift E_p [13]. The ground state of FCM is a polaronic Fermi liquid when the Coulomb repulsion is large, a bipolaronic high-temperature superconductor at intermediate Coulomb repulsions, and a charge-segregated insulator if the repulsion is weak. FCM predicts superlight polarons and bipolarons in cuprates with a remarkably high superconducting critical temperature. Cuprate bipolarons are relatively light because they are inter-site rather than on-site pairs due to the strong on-site repulsion, and because mainly c-axis polarized optical phonons are responsible for the in-plane mass renormalization. The relatively small mass renormalization of polaronic and bipolaronic carries in FCM has been confirmed numerically using the exact QMC [15], cluster diagonalization [16] and variational [17] simulations.

(Bi)polarons describe many properties of cuprates [3], in particular normal-state transport properties (section 2), the Nernst effect (section 3), and the normal state diamagnetism (section 4). The strong-coupling theory also provides an explanation for the phase separation and coexistence and describes the shape of resistive and magnetic transitions in manganites (section 5).

II. NORMAL STATE IN-PLANE RESISTIVITY, HALL EFFECT AND MAGNETIC SUSCEPTIBILITY OF CUPRATES IN THE BIPOLARON MODEL

The low-energy FCM electronic structure of cuprates is shown in Fig.1 [18]. Polaronic p-holes are bound into lattice inter-site singlets (A) or into singlets and triplets (B) (if spins are included in Eq.(2)) at any temperature. Above T_c a charged bipolaronic Bose-liquid is non-degenerate and below T_c phase coherence (ODLRO) of the preformed bosons sets in. The state above T_c is perfectly "normal" in the sense that the off-diagonal order parameter (i.e. the Bogoliubov-Gor'kov anomalous average $\mathcal{F}(\mathbf{r}, \mathbf{r}') = \langle \psi_{\downarrow}(\mathbf{r})\psi_{\uparrow}(\mathbf{r}') \rangle$ is zero above the resistive transition temperature T_c . Here $\psi_{\downarrow,\uparrow}(\mathbf{r})$ annihilates electrons with spin \downarrow,\uparrow at point \mathbf{r} . Triplet and singlet states are separated by the exchange energy J which explains the spin gap observed in a number of NMR and neutron scattering experiments. There are also thermally excited single polarons in the model. Their density becomes comparable with the bipolaron density at the temperature T^* which is about half of the bipolaron binding energy Δ , in accordance with the experimentally observed crossover regime at $T^* > T_c$ and the normal state pseudogaps in cuprates.

A nonlinear temperature dependence of the *in*-plane resistivity below T^* , a temperature-dependent paramagnetic susceptibility, and a peculiar maximum in the Hall ratio well above T_c have remained long-standing problems of cuprate physics. The bipolaron model provides their quantitative description [19]. Thermally excited phonons and (bi)polarons are well decoupled in the strong-coupling regime of the electron-phonon interaction [3], so the conventional Boltzmann kinetics for mobile polaronic and bipolaronic carries is applied. Here we use a 'minimum' bipolaron model Fig.1A, which includes the singlet bipolaron band and the spin 1/2 polaron band separated by T^* , and the τ -approximation in weak electric \mathbf{E} and magnetic fields, $\mathbf{B} \perp \mathbf{E}$.

Bipolaron and single-polaron non-equilibrium distributions are found as

$$f(\mathbf{k}) = f_0(E) + \tau \frac{\partial f_0}{\partial E} \mathbf{v} \cdot \{ \mathbf{F} + \Theta \mathbf{n} \times \mathbf{F} \}, \qquad (3)$$

where $\mathbf{v} = \partial E/\partial \mathbf{k}$, $\mathbf{F} = \vec{\nabla}(\mu - 2e\phi)$, $f_0(E) = [y^{-1}\exp(E/T) - 1]^{-1}$ and the Hall angle $\Theta = \Theta_b = 2eB\tau_b/m_b$ for bipolarons with the energy $E = k^2/(2m_b)$, and $\mathbf{F} = \vec{\nabla}(\mu/2 - e\phi)$, $f_0(E) = \{y^{-1/2}\exp[(E+T^*)/T] + 1\}^{-1}$, $E = k^2/(2m_p)$, and $\Theta = \Theta_p = eB\tau_p/m_p$ for thermally excited polarons. Here m_b and m_p are the bipolaron and polaron mass, respectively, $y = \exp(\mu/T)$, μ is the chemical potential, and $\mathbf{n} = \mathbf{B}/B$ is a unit vector in the direction of the magnetic field. Eq.(3) is

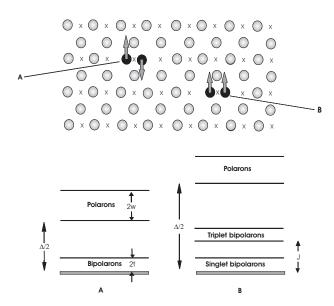


FIG. 1: Bipolaron picture of high temperature superconductors. A corresponds to the singlet intersite bipolaron. B is the triplet intersite bipolaron, which naturally includes the addition of an extra excitation band. The crosses are copper sites and the circles are oxygen sites, w is a half bandwidth of the polaron band, t is a half bandwidth of the bipolaronic band, $\Delta/2$ is the bipolaron binding energy per polaron and J is the exchange energy per bipolaron.

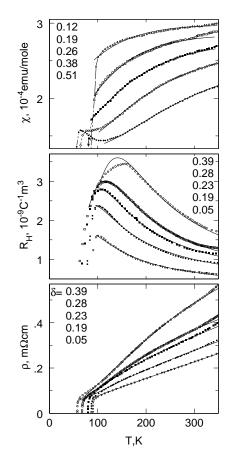


FIG. 2: Uniform magnetic susceptibility, $\chi(T)$, Hall ratio, $R_H(T)$ and resistivity, $\rho(T)$, of underdoped $YBa_2Cu_3O_{7-\delta}$ fitted by the theory; see the Table for parameters.

used to calculate the electrical resistivity and the Hall ratio as

$$\rho = \frac{m_b}{4e^2\tau_b n_b (1 + An_p/n_b)},\tag{4}$$

$$R_H = \frac{1 + 2A^2 n_p / n_b}{2e n_b (1 + A n_p / n_b)^2},\tag{5}$$

where $A = \tau_p m_b/(4\tau_b m_p)$. The atomic densities of quasi two-dimensional carriers are found as

$$n_b = \frac{m_b T}{2\pi} |\ln(1 - y)|, \tag{6}$$

$$n_p = \frac{m_p T}{\pi} \ln \left[1 + y^{1/2} \exp\left(-T^*/T\right) \right].$$
 (7)

and the chemical potential is determined by doping x using $2n_b + n_p = x - n_L$, where n_L is the number of carriers localised by disorder (here we take the lattice constant a = 1).

Polarons are not degenerate. Their number remains small compared with twice the number of bipolarons, $n_p/(2n_b) < 0.2$, in the relevant temperature range $T < T^*$, so that

$$y \approx 1 - \exp(-T_0/T),\tag{8}$$

where $T_0 = \pi(x - n_L)/m_b \approx T_c$ is about the superconducting critical temperature of the (quasi)twodimensional Bose gas. Because of this reason, the experimental T_c was taken as T_0 in our fits. Using Eqs. (7,6,5) we obtain

$$R_H(T) = R_{H0} \frac{1 + 2A^2 y^{1/2} (T/T_c) \exp(-T^*/T)}{[1 + A(T/T_c) y^{1/2} \exp(-T^*/T)]^2},$$
(9)

where $R_{H0} = [e(x - n_L)]^{-1}$. If we assume that the number of localised carriers depends only weakly on temperature in underdoped cuprates since their average ionisation energy is sufficiently large, then R_{H0} is temperature independent at $T < T^*$. As proposed in Ref.[20] the scattering rate at relatively high temperatures is due to inelastic collisions of itinerant carriers with those localised by disorder, so it is proportional to T^2 . We also have to take into account the residual scattering of polarons off optical phonons, so that $\tau^{-1} = aT^2 + b \exp(-\omega/T)$, if the temperature is low compared with the characteristic phonon energy ω . The relaxation times of each type of carriers scales with their charge e^* and mass as $\tau_{p,b} \propto m_{p,b}^{-3/2}(e^*)^{-2}$, so we estimate $A = (m_b/m_p)^{5/2} \approx 6$ if we take $m_b \approx 2m_p$. As a result the in-plane resistivity is given by

$$\rho(T) = \rho_0 \frac{(T/T_1)^2 + \exp(-\omega/T)}{[1 + A(T/T_c)y^{1/2}\exp(-T^*/T)]},$$
(10)

where $\rho_0 = bm_b/[2e^2(x-n_L)]$ and $T_1 = (b/a)^{1/2}$ are temperature independent. Finally, one can easily obtain the uniform magnetic susceptibility due to nondegenerate spin 1/2 polarons as [21]

$$\chi(T) = By^{1/2} \exp\left(-T^*/T\right) + \chi_0,\tag{11}$$

where $B = (\mu_B^2 m_p/\pi)$, and χ_0 is the magnetic susceptibility of the parent Mott insulator.

δ	T_c	ρ_0	R_{H0}	$10^4 B$	$10^{4}\chi_{0}$	T^*	ω	T_1
	K	$m\Omega cm$	$\frac{10^{-9}m^3}{C}$	$\frac{emu}{mole}$	$\frac{emu}{mole}$	K	K	K
0.05	90.7	1.8	0.45			144	447	332
0.12	93.7			2.6	2.1	155		
0.19	87	3.4	0.63	4.5	1.6	180	477	454
0.23	80.6	5.7	0.74			210	525	586
0.26	78			5.4	1.5	259		
0.28	68.6	8.9	0.81			259	594	786
0.38	61.9			7.2	1.4	348		
0.39	58.1	17.8	0.96			344	747	1088
0.51	55			9.1	1.3	494		

The present model numerically fits the Hall ratio, $R_H(T)$, the in-plane resistivity, $\rho(T)$, and the magnetic susceptibility $\chi(T)$ of $YBa_2Cu_3O_{7-\delta}$ within the physically relevant range of all parameters (see Fig. 2 and the Table). The ratio of polaron and bipolaron mobilities A=7 used in all fits is close to the above estimate, and $\chi_0 \approx 1.5 \times 10^{-4} emu/mole$ is very close to the susceptibility of a slightly doped insulator [22]. The maximum of $R_H(T)$ is due to the contribution of thermally excited polarons into transport, and the temperature dependence of the in-plane resistivity below T^* is due to this contribution and the combination of the carrier-carrier and carrier-phonon scattering. The characteristic phonon frequency from the resistivity fit (Table) decreases with doping and the pseudogap T^* shows the doping behaviour as observed in other independent experiments.

Notwithstanding our explanation of the Hall ratio, the in-plane resistivity and the bulk magnetic susceptibility might be not so convincing as a direct measurement of the double charge 2e on carriers in the normal state. In 1993, we discussed the thermal conductivity of preformed bosons [23]. The contribution from carriers to the thermal transport provided by the Wiedemann-Franz law depends strongly on the elementary charge as $\sim (e^*)^{-2}$ and should be significantly suppressed if $e^* = 2e$. The Lorenz number, L, has been directly measured in $YBa_2Cu_3O_{6.95}$ by Zhang et al. [24] using the thermal Hall conductivity. Remarkably, the measured value of L just above T_c was found just the same as predicted by the bipolaron model [23], $L \approx 0.15L_e$, where L_e is the conventional Fermiliquid Lorenz number. The breakdown of the Wiedemann-Franz law has been also explained in the framework of the bipolaron model [25].

III. NORMAL-STATE NERNST EFFECT

In disagreement with the weak-coupling BCS and the strong-coupling bipolaron theories a significant fraction of research in the field of high-temperature superconductivity suggests that the superconducting transition is only a phase ordering while the superconducting order parameter $\mathcal{F}(\mathbf{r}, \mathbf{r}')$ remains nonzero above the resistive T_c . One of the key experiments supporting this viewpoint is the large Nernst signal observed in the normal (i.e. resistive) state of cuprates (see Ref. [26, 27, 28] and references therein). Some authors [26, 29] claim that numerous resistive determinations of the upper critical field, $H_{c2}(T)$ in cuprates have been misleading since the Nernst signal [26] and the diamagnetic magnetization [29] imply that $H_{c2}(T)$ remains large at T_c and above. They propose a "vortex scenario", where the long-range phase coherence is destroyed by mobile vortices, but the amplitude of the off-diagonal order parameter remains finite and the Cooper pairing with a large binding energy exists well above T_c supporting the so-called "preformed Cooper-pair" or "phase fluctuation" model [30]. The model is based on the assumption that the superfluid density is small compared with the normal carrier density in cuprates. These interpretations seriously undermine

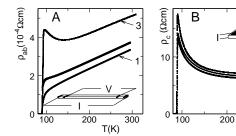


FIG. 3: In-plane (A) and out-of-plane (B) resistivity of 3 single crystals of Bi₂Sr₂CaCu₂O₈ [31] showing no signature of phase fluctuations well above the resistive transition temperature.

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many theoretical and experimental works on superconducting cuprates, which consider the state above T_c as perfectly normal with no off-diagonal order, either long or short.

We believe that the vortex (or phase fluctuation) scenario contradicts straightforward resistive and other measurements, and it is theoretically inconsistent. This scenario is impossible to reconcile with the extremely sharp resistive transitions at T_c in high-quality underdoped, optimally doped and overdoped cuprates. For example, the in-plane and out-of-plane resistivity of Bi - 2212, where the anomalous Nernst signal has been measured [26], is perfectly "normal" above T_c , Fig.3, showing only a few percent positive or negative magnetoresistance [31]. Both in-plane [32, 33, 34, 35, 36] and out-of-plane [37, 38, 39] resistive transitions of high-quality samples are sharp and remain sharp in the magnetic field providing a reliable determination of the genuine $H_{c2}(T)$. The vortex entropy [27] estimated from the Nernst signal is an order of magnitude smaller than the difference between the entropy of the superconducting state and the extrapolated entropy of the normal state obtained from the specific heat. The preformed Cooper-pair model [30] is incompatible with a great number of thermodynamic, magnetic, and kinetic measurements, which show that only holes (density x), doped into a parent insulator are carriers both in the normal and the superconducting states of cuprates. The assumption [30] that the superfluid density is small compared with the normal-state carrier density is also inconsistent with the theorem [40], which proves that the number of supercarriers at T = 0K should be the same as the number of normal-state carriers in any clean superfluid.

Recently we described the unusual Nernst signal in cuprates in a different manner as the normal state phenomenon [41]. Here we extend our description to cuprates with very low doping level accounting for their Nernst signal, the thermopower and the insulating-like in-plane low temperature resistance [26, 27, 28].

Thermomagnetic effects appear in conductors subjected to a longitudinal temperature gradient $\nabla_x T$ in x direction and a perpendicular magnetic field in z direction. The transverse Nernst-Ettingshausen effect [42] (here the Nernst effect) is the appearance of a transverse electric field E_y in the third direction. When bipolarons are formed in the strong-coupling regime, the chemical potential is negative, Eq.(8). It is found in the impurity band just below the mobility edge at $T > T_c$. Carriers, localised below the mobility edge contribute to the longitudinal transport together with the itinerant carriers in extended states above the mobility edge. Importantly the contribution of localised carriers of any statistics to the transverse transport is normally small [43] since a microscopic Hall voltage will only develop at junctions in the intersections of the percolation paths, and it is expected that these are few for the case of hopping conduction among disorder-localised states [44]. Even if this contribution is not negligible, it adds to the contribution of itinerant carriers to produce a large Nernst signal, $e_y(T, B) \equiv -E_y/\nabla_x T$, while it reduces the thermopower S and the Hall angle Θ . This unusual "symmetry breaking" is completely at variance with ordinary metals where the familiar "Sondheimer" cancelation [45] makes e_y much smaller than S tan Θ because of the

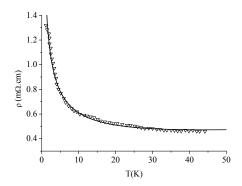


FIG. 4: Normal state in-plane resistivity of underdoped $La_{1.94}$ $Sr_{0.06}CuO_4$ (triangles [27]) as revealed in the field B = 12 Tesla and compared with the bipolaron theory, Eq.(17) (solid line).

electron-hole symmetry near the Fermi level. Such behaviour originates in the "sign" (or "p-n") anomaly of the Hall conductivity of localised carriers. The sign of their Hall effect is often *opposite* to that of the thermopower as observed in many amorphous semiconductors [43] and described theoretically [46].

The Nernst signal is expressed in terms of the kinetic coefficients σ_{ij} and α_{ij} as

$$e_y = \frac{\sigma_{xx}\alpha_{yx} - \sigma_{yx}\alpha_{xx}}{\sigma_{xx}^2 + \sigma_{xy}^2},\tag{12}$$

where the current density is given by $j_i = \sigma_{ij} E_j + \alpha_{ij} \nabla_j T$. When the chemical potential μ is at the mobility edge, the localised carriers contribute to the transport, so σ_{ij} and α_{ij} in Eq.(12) can be expressed as $\sigma_{ij}^{ext} + \sigma_{ij}^l$ and $\alpha_{ij}^{ext} + \alpha^l ij$, respectively. Since the Hall mobility of carriers localised below μ , σ_{yx}^l , has the sign opposite to that of carries in the extended states above μ , σ_{yx}^{ext} , the sign of the off-diagonal Peltier conductivity α_{yx}^l should be the same as the sign of α_{yx}^{ext} . Then neglecting the magneto-orbital effects in the resistivity (since $\Theta \ll 1$ [26]) we obtain

$$S \tan \Theta \equiv \frac{\sigma_{yx}\alpha_{xx}}{\sigma_{xx}^2 + \sigma_{xy}^2} \approx \rho(\alpha_{xx}^{ext} - |\alpha_{xx}^l|)(\Theta^{ext} - |\Theta^l|)$$
(13)

and

$$e_y \approx \rho(\alpha_{yx}^{ext} + |\alpha_{yx}^l|) - S \tan \Theta,$$
 (14)

where $\Theta^{ext} \equiv \sigma_{yx}^{ext}/\sigma_{xx}$, $\Theta^l \equiv \sigma_{yx}^l/\sigma_{xx}$, and $\rho = 1/\sigma_{xx}$ is the resistivity.

Clearly the model, Eqs.(13,14) can account for a low value of $S \tan \Theta$ compared with a large value of e_y in some underdoped cuprates [26, 28] due to the sign anomaly. Even in the case when localised bosons contribute little to the conductivity their contribution to the thermopower $S = \rho(\alpha_{xx}^{ext} - |\alpha_{xx}^l|)$ could almost cancel the opposite sign contribution of itinerant carriers [41]. Indeed the longitudinal conductivity of itinerant two-dimensional bosons, $\sigma^{ext} \propto \int_0 dE E df(E)/dE$ diverges logarithmically when μ in the Bose-Einstein distribution function $f(E) = [\exp((E - \mu)/T) - 1]^{-1}$ goes to zero and the relaxation time τ is a constant. At the same time $\alpha_{xx}^{ext} \propto \int_0 dE E(E - \mu) df(E)/dE$ remains finite, and it could have the magnitude comparable with α_{xx}^l . Statistics of bipolarons gradually changes from Bose to Fermi statistics with lowering energy across the mobility edge because of the Coulomb repulsion of bosons in localised states [47]. Hence one can use the same expansion near the mobility edge as in ordinary amorphous semiconductors to obtain the familiar textbook result $S = S_0 T$ with a constant S_0 at low temperatures [48]. The model becomes particularly simple, if we

neglect the localised carrier contribution to ρ , Θ and α_{xy} , and take into account that $\alpha_{xy}^{ext} \propto B/\rho^2$ and $\Theta^{ext} \propto B/\rho$ in accordance with the Boltzmann theory. Then Eqs.(13,14) yield

$$S \tan \Theta \propto T/\rho \tag{15}$$

and

$$e_y(T,B) \propto (1 - T/T_1)/\rho. \tag{16}$$

According to our earlier suggestion [49] the insulating-like low-temperature dependence of $\rho(T)$ in underdoped cuprates originates from the elastic scattering of nondegenerate itinerant carriers off charged impurities. As in section 2 we assume here that the carrier density is temperature independent at low temperatures in agreement with the temperature-independent Hall effect [50]. The relaxation time of nondegenerate carriers depends on temperature as $\tau \propto T^{-1/2}$ for scattering off short-range deep potential wells, and as $T^{1/2}$ for very shallow wells [49]. Combining both scattering rates we obtain

$$\rho = \rho_0 [(T/T_2)^{1/2} + (T_2/T)^{1/2}]. \tag{17}$$

Eq.(17) with $\rho_0 = 0.236$ m Ω ·cm and $T_2 = 44.6$ K fits extremely well the experimental insulating-like normal state resistivity of underdoped La_{1.94} Sr_{0.06}CuO₄ in the whole low-temperature range from 2K up to 50K, Fig.4, as revealed in the field B = 12 Tesla [27, 28]. Another high quality fit can be obtained combining the Brooks-Herring formula for the 3D scattering off charged impurities, as proposed in Ref.[51] for almost undoped LSCO, or the Coulomb scattering in 2D ($\tau \propto T$) and a temperature independent scattering rate off neutral impurities with the carrier exchange [52] similar to the scattering of slow electrons by hydrogen atoms. Importantly our expressions (15,16) for $S \tan \Theta$ and e_y do not depend on the particular scattering mechanism. Taking into account the excellent fit of Eq.(17) to the experiment, they can be parameterized as

$$S \tan \Theta = e_0 \frac{(T/T_2)^{3/2}}{1 + T/T_2},\tag{18}$$

and

$$e_y(T,B) = e_0 \frac{(T_1 - T)(T/T_2)^{1/2}}{T_2 + T},$$
(19)

where T_1 and e_0 are temperature independent.

In spite of all simplifications, the model describes remarkably well both $S \tan \Theta$ and e_y measured in La_{1.94} Sr_{0.06}CuO₄ with a *single* fitting parameter, $T_1 = 50 \text{K}$ using the experimental $\rho(T)$. The constant $e_0 = 2.95 \ \mu\text{V/K}$ scales the magnitudes of $S \tan \Theta$ and e_y . The magnetic field B = 12 Tesla destroys the superconducting state of the low-doped La_{1.94} Sr_{0.06}CuO₄ down to 2K, Fig.4, so any residual superconducting order above 2K is clearly ruled out, while the Nernst signal, Fig.5, is remarkably large. The coexistence of the large Nernst signal and a nonmetallic resistivity is in sharp disagreement with the vortex scenario, but in agreement with our model. Taking into account the field dependence of the conductivity of localised carriers, the phonon-drug effect, and their contribution to the transverse magnetotransport can well describe the magnetic field dependence of the Nernst signal [41] and improve the fit in Fig.5 at the expense of the increasing number of fitting parameters.

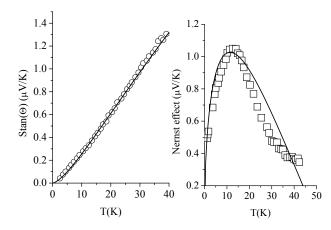


FIG. 5: $S \tan \Theta$ (circles [28]) and the Nernst effect e_y (squares [27]) of underdoped La_{1.94} Sr_{0.06}CuO₄ at B = 12 Tesla compared with the bipolaron theory, Eqs. (18,19) (solid lines).

IV. NORMAL STATE DIAMAGNETISM IN CUPRATES

A number of experiments (see, for example, [29, 53, 54, 55, 56, 57] and references therein), including torque magnetometries, showed enhanced diamagnetism above T_c , which has been explained as the fluctuation diamagnetism in quasi-2D superconducting cuprates (see, for example Ref. [56]). The data taken at relatively low magnetic fields (typically below 5 Tesla) revealed a crossing point in the magnetization M(T, B) of most anisotropic cuprates (e.g. Bi - 2212), or in $M(T, B)/B^{1/2}$ of less anisotropic YBCO [54]. The dependence of magnetization (or $M/B^{1/2}$) on the magnetic field has been shown to vanish at some characteristic temperature below T_c . However the data taken in high magnetic fields (up to 30 Tesla) have shown that the crossing point, anticipated for low-dimensional superconductors and associated with superconducting fluctuations, does not explicitly exist in magnetic fields above 5 Tesla [55].

Most surprisingly the torque magnetometery [53, 55] uncovered a diamagnetic signal somewhat above T_c which increases in magnitude with applied magnetic field. It has been linked with the Nernst signal and mobile vortexes in the normal state of cuprates [29]. However, apart from the inconsistences mentioned above, the vortex scenario of the normal-state diamagnetism is internally inconsistent. Accepting the vortex scenario and fitting the magnetization data in Bi-2212 with the conventional logarithmic field dependence [29], one obtains surprisingly high upper critical fields $H_{c2} > 120$ Tesla and a very large Ginzburg-Landau parameter, $\kappa = \lambda/\xi > 450$ even at temperatures close to T_c . The in-plane low-temperature magnetic field penetration depth is $\lambda = 200$ nm in optimally doped Bi-2212 (see, for example [58]). Hence the zero temperature coherence length ξ turns out to be about the lattice constant, $\xi = 0.45$ nm, or even smaller. Such a small coherence length rules out the "preformed Cooper pairs" [30], since the pairs are virtually not overlapped at any size of the Fermi surface in Bi-2212. Moreover the magnetic field dependence of M(T,B)at and above T_c is entirely inconsistent with what one expects from a vortex liquid. While -M(B)decreases logarithmically at temperatures well below T_c , the experimental curves [29, 53, 55] clearly show that -M(B) increases with the field at and above T_c , just opposite to what one could expect in the vortex liquid. This significant departure from the London liquid behavior clearly indicates that the vortex liquid does not appear above the resistive phase transition [53].

Some time ago we explained the anomalous diamagnetism in cuprates as the Landau normal-state diamagnetism of preformed bosons [59]. The same model predicted the unusual upper critical field

[60] observed in many superconducting cuprates [32, 33, 34, 35, 36, 37, 61]. Here we extend the model to high magnetic fields taking into account the magnetic pair-breaking of singlet bipolarons and the anisotropy of the energy spectrum.

When the strong magnetic field is applied perpendicular to the copper-oxygen plains the quasi-2D bipolaron energy spectrum is quantized as

$$E_{\alpha} = \omega(n + 1/2) + 2t_c[1 - \cos(k_z d)], \tag{20}$$

where $\omega = 2eB/m_b$, n = 0, 1, 2, ..., and t_c , k_z , d are the hopping integral, the momentum and the lattice period perpendicular to the planes. Quantum numbers α also include the momentum along one of the in-plane directions. Expanding the Bose-Einstein distribution function in powers of $exp[(\mu - E_{\alpha})/T]$ with the negative μ one can readily obtain (after summation over n) the boson density

$$n_b = \frac{eB}{\pi d} \sum_{k=1}^{\infty} I_0(2t_c k/T) \frac{\exp[(\tilde{\mu} - 2t_c)k/T]}{1 - \exp(-\omega k/T)},$$
(21)

and the magnetization

$$M(T,B) = -n_b \mu_b + \frac{eT}{\pi d} \sum_{k=1}^{\infty} I_0(2t_c k/T) \frac{\exp[(\tilde{\mu} - 2t_c)k/T]}{1 - \exp(-\omega k/T)}$$

$$\times \left(\frac{1}{k} - \frac{\omega \exp(-\omega k/T)}{T[1 - \exp(-\omega k/T)]}\right), \tag{22}$$

where $\mu_b = e/m_b$, $\tilde{\mu} = \mu - \omega/2$ and $I_0(x)$ is the modified Bessel function. At low temperatures $T \to 0$ Schafroth's result [62] is recovered, $M(0,B) = -n_b\mu_b$. The magnetization of charged bosons is field-independent at low temperatures. At high temperatures, $T \gg T_c$ the chemical potential has a large magnitude, so we can keep only terms with k = 1 in Eqs.(21,22) to obtain

$$M(T,B) = -n_b \mu_b + \frac{T n_b}{B} \left(1 - \frac{\omega \exp(-\omega/T)}{T[1 - \exp(-\omega/T)]} \right). \tag{23}$$

The experimental conditions are such that $T \gg \omega$ when T is of the order of T_c or higher, so that

$$M(T,B) = -n_b \mu_b \frac{\omega}{6T},\tag{24}$$

which is the Landau orbital diamagnetism of nondegenerate carriers. The bipolaron in-plane mass in cuprates is about $m_b \approx 10 m_e$ [3]. Using this mass yields $M(0,B) \approx 2000$ A/m with the bipolaron density $n_b = 10^{21}$ cm⁻³. Then the magnitude and the field/temperature dependence of M(T,B) near and above T_c are about the same as experimentally observed in Refs [29, 55]. The pseudogap temperature T^* depends on the magnetic field predominantly because of the magnetic-field splitting of the single-polaron band in Fig.1. As a result the bipolaron density depends on the field (as well as on temperature) near T_c as

$$n_b(T,B) = n_b(T_c,0) \left[1 + (T_c - T)/\tilde{T}_0 - (B/B_0)^{\beta} \right], \tag{25}$$

where \tilde{T}_0 and B_0 are constants depending on T^* , $\beta = 2$ if the polaron spectrum is spin-degenerate, and $\beta = 1$ if the spin degeneracy is removed by the crystal field already in the absence of the external field.

Theoretical temperature and field dependencies of M(T,B), Eq.(22) agree qualitatively with the experimental curves in Bi-2212 [29, 55], if the depletion of the bipolaron density, Eq.(25) is taken into account. The depletion of n_b accounts for the absence of the crossing point in M(T,B) at high magnetic fields. Nevertheless a quantitative fit to experimental M(T,B) curves using \tilde{T}_0 and B_0 as the fitting parameters is premature. The experimental diamagnetic magnetization has been extracted from the total magnetization assuming that the normal state paramagnetic contribution remains temperature-independent at all temperatures [29, 55]. This assumption is inconsistent with a great number of NMR and the Knight shift measurements, and even with the preformed Cooperpair model itself. The Pauli spin-susceptibility has been found temperature-dependent in these experiments revealing a normal-state pseudogap, contrary to the assumption. Hence the experimental diamagnetic M(T,B) [29, 55] has to be corrected by taking into account the temperature dependence of the spin paramagnetism at relatively low temperatures.

V. PHASE COEXISTENCE AND RESISTIVITY NEAR THE FERROMAGNETIC TRANSITION IN MANGANITES

Ferromagnetic oxides, in particular manganese perovskites, show a huge magnetoresistance near the ferromagnetic transition. The resistivity change is so large that it could not compare with any other forms of magnetoresistance. The effect observed in these materials was therefore named 'colossal' magnetoresistance (CMR) to distinguish it from the giant magnetoresistance observed in magnetic multilayers. The discovery raised expectations of a new generation of magnetic devices, and launched a frenetic scientific race to understand the cause of the effect. Significant progress has been made in understanding their properties, but new questions have arisen. The ferromagnetic metal-insulator transition in manganites has long been thought as the consequence of the so-called double exchange mechanism (DEX), which results in a varying bandwidth of electrons in the Mn³⁺ d-shell as a function of temperature [63]. More recently it has been noticed [64] that the effective spin-exchange interaction of the double-exchange model cannot account for CMR alone. In fact there is strong experimental evidence for exceptionally strong e-ph interactions in doped manganites from the optical data (see section 2), the giant isotope effect [7], the Arrhenius behaviour of the drift and Hall mobilities [65] in the paramagnetic phase above the Curie temperature, T_m , etc. Therefore Ref. [64] and some subsequent theoretical studies combined DEX with the Jahn-Teller e-ph interaction in d-orbitals arriving at the conclusion that the low-temperature ferromagnetic phase is a spin-polarised metal, while the paramagnetic phase is a polaronic insulator.

However, some low-temperature optical [66], electron-energy-loss (EELS) [67], photoemission[68] and thermoelectric [69] measurements showed that the ferromagnetic phase of manganites is not a conventional metal. In particular, broad incoherent spectral features and a pseudo-gap in the excitation spectrum were observed. EELS confirmed that manganites were charge-transfer doped insulators having p-holes as current carriers rather than d Mn^{3+} electrons. Photoemission and x-ray absorption spectroscopies of $La_{1-x}Sr_xMnO_3$ also showed that the itinerant holes doped into LaMnO₃ are of oxygen p-character. CMR has been observed in the ferromagnetic pyrochlore manganite $Tl_2Mn_2O_7$ [70], which has neither the mixed valence for DEX magnetic interaction nor the Jahn-Teller cations such as Mn^{3+} .

These and other observations [71], in particular the fact that some samples of ferromagnetic manganites manifest an insulating-like optical conductivity at all temperatures [72], clearly rule out DEX as the mechanism of CMR. They led us to a novel theory of ferromagnetic/paramagnetic phase transition and CMR based on the so-called current-carrier density collapse (CCDC) [14]. In CCDC p-holes are bound into heavy bipolarons above T_m due to the Fröhlich e-ph interaction, Eq.(2). The resistivity peak and CMR are the result of the magnetic pair-breaking below T_m , Fig.6,

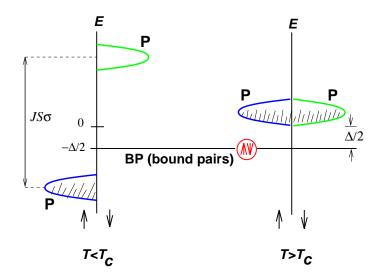


FIG. 6: Bipolaron model of CMR: pairs (BP) are localised on impurity levels in the paramagnetic phase, where the only current carriers are single thermally excited polarons (here $T_c = T_m$ is the Curie temperature). If the exchange interaction $JS\sigma$ between p-hole polarons and ordered manganese spins exceeds the pair binding energy Δ , the pairs break at $T < T_c$ because the spin-up polaron sub-band sinks abruptly below the bipolaron level. The ferromagnetic state is a polaronic conductor.

caused by the p-d spin-exchange interaction, J_{pd} , which described as

$$H_{pd} = -(2N)^{-1} \sum_{\mathbf{n}, \mathbf{m}} J_{pd} \hat{S}_{\mathbf{m}}^{z} (c_{\mathbf{n}\uparrow}^{\dagger} c_{\mathbf{n}\uparrow} - c_{\mathbf{n}\downarrow}^{\dagger} c_{\mathbf{n}\downarrow}). \tag{26}$$

Here $\hat{S}_{\mathbf{m}}^{z}$ is the z-component of Mn^{3+} spin on site \mathbf{m} , and N is the total number of sites.

Different from cuprates hole bipolarons are much heavier in manganites because the e-ph Fröhlich interaction is stronger and the band structure is less anisotropic. They are readily localised by disorder, so only thermally excited single extended polarons conduct in the paramagnetic phase. With temperature lowering single polarons polarize manganese spins at T_m via J_{pd} , and the spin polarization of manganese ions breaks the bipolaronic singlets creating a spin-polarized *polaronic* conductor. CCDC explained the resistivity peak and CMR in the experimental range of external magnetic fields [14, 73]. More recently, the theory has been further confirmed experimentally. In particular, the oxygen isotope effect has been observed in the low-temperature resistivity of La_{0.75}Ca_{0.25}MnO₃ and Nd_{0.7}Sr_{0.3}MnO₃ and explained by CCDC with polaronic carriers in the ferromagnetic phase [74]. The current-carrier density collapse has been directly observed using the Hall data in La_{0.67}Ca_{0.33}MnO₃ and $La_{0.67}Sr_{0.33}MnO_3$ [75]. And the first order phase transition at T_m , predicted by the theory [14], has been firmly established in the specific heat measurements [76]. On the other hand, the resistivity and the magnetization of some samples of La_{0.7}Ca_{0.3}Mn_{1-x}Ti_xO₃ showed a more gradual (secondorder like) transition [77]. Also the coexistence of ferromagnetic and paramagnetic phases near the Curie temperature observed in tunneling [78] and other experiments has not yet been addressed in the framework of CCDC. Here we argue that the diagonal disorder, which is inevitable with doping, explains both the phase coexistence and the resistivity/magnetization shape near the transition. The mean-field equations [14] describing the single polaron density n, p-hole polaron m and manganese σ magnetizations, and the chemical potential $\mu = T \ln y$ can be easily generalized by taking into account the random distribution of the bipolaron binding energy $\delta = \Delta/(2J_{pd})$ across the sample as

$$n_i = 6y \cosh(\sigma_i/t),$$

 $m_i = n_i \tanh(\sigma_i/t),$

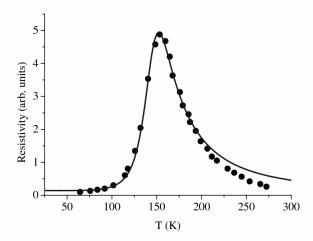


FIG. 7: CCDC model (Eq.(29), solid line) fits the experimental resistivity near ferromagnetic transition in $La_{0.7}Ca_{0.3}Mn_{1.95}Ti_{0.05}O_3$ [77] (dots), when the phase coexistence caused by disorder is taken into account.

$$\sigma_i = B_2(m_i/2t),$$

 $y^2 = \frac{x - n_i}{18} \exp(-2\delta_i/t),$ (27)

where $t = T/J_{pd}$ is the reduced temperature, B_S is the Brillouin function, x is the number of delocalised holes at zero temperature in p-orbital states, which are 3-fold degenerate. The subscript imeans different parts of the sample with different δ_i because of disorder. While averaging these transparent equations over a random distribution of δ_i is rather cumbersome, one can apply a simplified approach using the fact that the phase transition in a homogeneous system is of the first order in a wide range of δ [14]. Taking $\sigma_i \approx \Theta(T_{mi} - T)$ and $n_i \approx x\Theta(T_{mi} - T) + \sqrt{2x} \exp(-\Delta/(2T))\Theta(T - T_{mi})$ and averaging both quantities with the Gaussian distribution of random T_{mi} s around the experimental T_m we obtain the averaged manganese magnetization

$$\sigma(T) = \frac{1}{2} erfc\left(\frac{T - T_m}{\Gamma}\right) \tag{28}$$

and the resistivity, $\rho \propto 1/n$, near the transition

$$1/\rho(T) \propto erfc\left(\frac{T - T_m}{\Gamma}\right) + (2/x)^{1/2}e^{-\Delta/2T}erfc\left(\frac{T_m - T}{\Gamma}\right).$$
 (29)

Here Δ is the average bipolaron binding energy, $\Theta(y)=1$ for y>0 and zero for y<0, and $erfc(y)=(2/\pi^{1/2})\int_y^\infty dy \exp(-y^2)$. CCDC with disorder, Eq.(29) fits nicely the experimental resistivity [77] near the transition with physically reasonable parameters $\Gamma=28\mathrm{K},~\Delta=1600\mathrm{K},~T_m=102\mathrm{K},$ and x=0.1, Fig.7. A random distribution of transition temperatures with the width Γ across the sample caused by the randomness of the bipolaron binding energy is responsible for the phase coexistence near the transition [78].

In summary, the strong-coupling bipolaron extension of the BCS theory accounts for the kinetic properties of superconducting cuprates including the temperature-dependent spin susceptibility, the nonlinear in-plane resistivity, the maximum in the Hall effect, the normal-state Nernst signal and

the diamagnetism near and above T_c . CMR and ferromagnetism of ferromagnetic oxides can be well explained by the current-carrier density collapse in the framework of the same theory including the exchange magnetic interaction of p-holes with the manganese spins and disorder effects.

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