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Coherent ‘ab’ and ‘c’ transport theory of high- T_c cuprates

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We propose a microscopic theory of the ‘c’-axis and in-plane transport of copper oxides based on the bipolaron theory and the Boltzmann kinetics. The fundamental relationship between the anisotropy and the spin susceptibility is derived, $\rho_c(T, x)/\rho_{ab}(T, x) \sim x/\sqrt{T}\chi_s(T, x)$. The temperature (T) and doping (x) dependence of the in-plane, ρ_{ab} and out-of-plane, ρ_c resistivity and the spin susceptibility, χ_s are found in a remarkable agreement with the experimental data in underdoped, optimally and overdoped $La_{2-x}Sr_xCuO_4$ for the entire temperature regime from T_c up to 800K. The normal state gap is explained and its doping and temperature dependence is clarified.

The absolute value and qualitatively different temperature dependence of the in-plane and c -axis resistivity [1] as well as the normal state gap observed with NMR, neutron scattering, thermodynamic and kinetic measurements in copper based high- T_c oxides are recognised now as the key to our understanding of the high- T_c phenomenon [2,3]. By the use of the room-temperature values of the ab and c -axis conductivities of the prototypical cuprate $La_{2-x}Sr_xCuO_4$ at the optimal doping, $\sigma_{ab} \simeq 2 \times 10^5 \Omega^{-1}m^{-1}$ and $\sigma_c \simeq 10^3 \Omega^{-1}m^{-1}$, $x = 0.15$, [4] one obtains

$$\frac{E_F \tau}{\hbar} = \frac{\pi \hbar d \sigma_{ab}}{e^2} \simeq 1 \quad (1)$$

and the ratio of the mean-free path l_c to the inter-plane distance d as

$$\frac{l_c}{d} = \frac{2\pi\hbar\sigma_c}{e^2k_F^2d} < 0.01 \quad (2)$$

where E_F and $\hbar k_F$ are the Fermi energy and $2D$ Fermi momentum, respectively, and τ is the transport relaxation time. This estimate as well as the semiconductor-like behavior of $\rho_c(T)$ contrasting with the linear $\rho_{ab}(T)$ do not agree with any Fermi-liquid description [2]. Another challenging problem is a three-dimensional coherent superconducting state of these quasi-two dimensional conductors which is hardly compatible with several phenomenological models [1,5], based on the assumption that the c-axis transport is *incoherent*. To meet this challenge some authors [2] alleged the spin-charge separation abandoning the Fermi-liquid and Boltzmann approach.

At the same time the results of the kinetic [6,7,8,9] and the heat capacity [10] measurements led us [11] to the conclusion that the so-called spin-gap observed previously in the magnetic susceptibility [12,13,14] (in the spin channel) belongs in fact to the charge carriers, which are inter-site bipolarons. In particular, the quantitative explanations of the temperature dependence of the NMR line-width and the linear in-plane resistivity [15] as well as of the Hall effect [16] were proposed. Several authors [7,4,5] attributed the ‘semiconducting’ temperature dependence $\rho_c(T)$ to the ‘normal state gap’ in the density of states. The comprehensive analysis by Batlogg and co-workers [17] revealed transport features incompatible with the spin-charge separation and the doping dependence of the normal state gap as well the semiconducting-like doping dependence of resistivity. It became clear that high- T_c oxides are doped semiconductors rather than metals, irrespective to the level of doping [18].

In this letter the *coherent* theory of $\rho_c(T, x)$, $\chi_s(T, x)$ and of the normal state gap, $\Delta(T, x)$ is developed based on the bipolaron theory of high-temperature superconductivity by Alexandrov and Mott [11].

We expect that small polarons are paired *above* T_c in strongly correlated Mott-Hubbard insulators with the electron-phonon coupling constant above an intermediate value $\lambda \geq 0.5$ [19]. The ground and low-energy states are well described by the mixture of the intersite in-

plane singlet pairs (small bipolarons) and thermally excited polarons. Above T_c , which is the condensation temperature of the charged Bose-gas, all carriers are nondegenerate. Singlets tunnel along the plane with an effective mass m^{**} of the order of a single-polaron mass m^* as shown by one of us [18]. However, their c -axis tunneling can be only Josephson-like involving simultaneous hopping of two holes. Therefore the singlet c -axis mass is strongly enhanced, $m_c^{**} \gg m^{**} \sim m^*$. In that way we explain a large transport anisotropy of copper oxides at low temperatures when polarons are frozen out which is difficult to understand from the band-structure calculations alone. The crucial point of our theory is that polarons dominate in the c -axis transport at intermediate and high temperatures because they are much lighter in the c -direction than bipolarons (see below). Along the planes they propagate with about the same effective mass as singlets. Therefore their contribution to the ab transport is small at any temperature due to their low density compared with the density of bipolarons. As a result we have a mixture of the *nondegenerate* quasi-two dimensional spinless bosons and the thermally excited fermions, which are capable of propagating along the c -axis. Because only polarons contribute to the spin susceptibility there is a fundamental relationship between the anisotropy and the magnetic susceptibility.

Applying the Boltzmann theory we obtain the following kinetic coefficients ($\hbar = k_B = 1$)

$$\sigma_{ab}(T, x) = - \int_0^\infty dE \sigma_b(E) \frac{\partial f_b}{\partial E}, \quad (3)$$

$$\sigma_c(T, x) = - \int_0^\infty dE \sigma_{pc}(E) \frac{\partial f_p}{\partial E}, \quad (4)$$

and

$$\chi_s(T, x) = -2\mu_B^2 \int_0^\infty dE N_p(E) \frac{\partial f_p}{\partial E}. \quad (5)$$

where $f_b = [y^{-1} \exp(E/T) - 1]^{-1}$ and $f_p = [y^{-1/2} \exp(E/T + \Delta/2T) + 1]^{-1}$ are the bipolaron and polaron distribution functions, respectively, with $y = \exp[\mu(T, x)/T]$, $\mu(T, x)$ the chemical potential, and μ_B the Bohr magneton. The bipolaron binding energy Δ is expected to be of the order of a few hundred K [11]. Therefore polarons are not degenerate at any temperatures. Above T_c bipolarons are also not degenerate, so that

$$f_b \simeq y \exp\left(-\frac{E}{T}\right), \quad (6)$$

and

$$f_p \simeq \sqrt{y} \exp\left(-\frac{E + \Delta/2}{T}\right). \quad (7)$$

If the scattering mechanism is the same for polarons and bipolarons the ratio of the differential conductivities is independent of the energy and doping

$$\frac{\sigma_b(E)}{\sigma_{pc}(E)} \equiv A = \text{constant}. \quad (8)$$

There is a large difference in the values of the $pp\sigma$ and $pp\pi$ hopping integrals between different oxygen sites. Therefore we expect a highly anisotropic polaron energy spectrum [18] with a quasi one -dimensional polaron density of states as observed with the high resolution *ARPES* [20]

$$N_p(E) \simeq \frac{1}{2\pi} \sqrt{\frac{m^*}{2E}}. \quad (9)$$

Then the c -axis resistivity as well as the spin susceptibility is expressed as

$$\frac{\rho_c(T, x)}{\rho_{ab}(T, x)} = A \sqrt{y} \exp\left(\frac{\Delta}{2T}\right) \quad (10)$$

and

$$\chi_s(T, x) = \mu_B^2 \sqrt{\frac{ym^*}{2\pi T}} \exp\left(-\frac{\Delta}{2T}\right). \quad (11)$$

The chemical potential, $y = 2\pi n_b(T, x)/Tm^{**}$, is calculated by taking into account the Anderson localisation of bipolarons in a random potential. Within a ‘single well-single particle’ approximation [16] the bipolaron density $n_b(T, x)$ appears to be linear in temperature and doping, $n_b(T, x) \sim n_L T$, with the total number of impurity levels n_L proportional to the doping x . That fits very well the temperature and doping dependence of the Hall effect $R_H = 1/2en_b(T, x)$ as well as the linear ab -resistivity in a wide temperature and doping range as shown in ref. [16]. As a result we find the temperature independent $y \sim x$ and

$$\frac{\rho_c(T, x)}{\rho_{ab}(T, x)} = constant \times \frac{x}{T^{1/2}\chi_s(T, x)}, \quad (12)$$

$$\chi_s(T, x) = constant' \times \sqrt{\frac{x}{T}} \exp\left(-\frac{\Delta(T, x)}{2T}\right). \quad (13)$$

We expect a strong dependence of the binding energy on the doping due to screening, $\Delta = \Delta(T, x)$. Bipolarons are heavy nondegenerate particles which screen very well the electron-phonon interaction. In fact, by the use of the classical expression for the inverse screening radius

$$q_s = \sqrt{\frac{16\pi e^2 n_b(T, x)}{\epsilon_0 T}} \quad (14)$$

and the static dielectric constant of *LSCO*, $\epsilon_0 \simeq 30$ one obtains the value of q_s about 3\AA^{-1} at room temperature with $n_b = 10^{21} \text{cm}^{-3}$ which is about three times larger than the reciprocal lattice vector q_d . The polaron (Franck-Condon) level shift E_p is suppressed by the screening as $(q_d/q_s)^2$ at large q_s [19]. Consequently, the normal state gap, $\Delta \simeq 2E_p$, depends on the doping and temperature as

$$\Delta(T, x) \sim \frac{T}{n_b(T, x)}. \quad (15)$$

Taking $n_b(T, x) \sim Tx$ we arrive with the temperature independent gap,

$$\Delta = \frac{\Delta_0}{x}, \quad (16)$$

where Δ_0 is doping independent. By the use of Eq.(12), Eq.(13) and Eq.(16) one can describe all qualitative features of the *c*-axis resistivity and the magnetic susceptibility of *LSCO* without any fitting parameters as the comparison of Fig.1 and Fig.2 shows. The linear temperature dependence of the *ab* resistivity was explained within the same approach [15,16]. Thus the *c*-axis resistivity is now understood as well. For a quantitative fit one has to solve the Bethe-Salpeter equation for $\Delta(T, x)$ with the realistic interaction between polarons taking into account the polarisation of the bipolaronic liquid partly localised by disorder. At temperatures close to the transition the Bose statistics becomes important. At

large temperatures the finite polaron and bipolaron bandwidth (about $1000K$) plays some role. Therefore it is not surprising that the experimental dependence of $\chi_s(T, x)$ shifts from the theoretical one, Fig.1b. At the same time the anisotropy is quantitatively described by Eq.(12) with the *experimental* values of $\chi_s(T, x)$, Fig.2a allowing small sample variations of *constant* in Eq.(12) within less than 15%. A smaller anisotropy of a heavily overdoped sample ($x = 0.3$) in Fig.2a is due to the fact that polarons contribute to the *ab* transport when the binding energy is below $100K$. Alternatively, the normal state gap $\Delta(T, x)$ can be determined by the use of Eq.(11) and the experimental values of $\chi_s(T, x)$, Fig.2b. With the temperature independent $y(x)$ (in the agreement with a flat temperature dependence of the thermoelectric power of *LSCO*) one obtains

$$\Delta(T, x) = 2T \ln \frac{B_\infty}{\sqrt{T}\chi_s(T, x)}, \quad (17)$$

where $B_\infty = \lim_{T \rightarrow \infty} \sqrt{T}\chi_s(T, x)$ for $T \rightarrow \infty$ is independent of the doping, $B_\infty \simeq 5.46 \times 10^{-6} EmuK^{1/2}/g$. The values of $\Delta(T, x)$, determined with Eq.(17), Fig.3, are about the same as Batlogg's normal- state- gap temperature $T^*(x)$ [17]. Therefore we conclude that T^* is the bipolaron binding energy. A temperature dependence of $\Delta(T, x)$ of the underdoped sample in Fig.3 is explained by the temperature dependence of the screening radius. From the Hall effect [17] one can observe that the bipolaron density is approximately constant below $200K$ for $x = 0.1$ in the agreement with our previous calculations [16]. Then, according to Eq.(15) the normal state gap is proportional to the temperature, $\Delta \sim T$ at low temperatures.

The proposed kinetics of high- T_c cuprates is derived from the generic Hamiltonian

$$H = \sum_{\langle ij \rangle} T(\mathbf{m} - \mathbf{n})c_i^\dagger c_j + \sum_{\mathbf{q}} \omega(\mathbf{q})(d_{\mathbf{q}}^\dagger d_{\mathbf{q}} + 1/2) + \sum_{\mathbf{q}, i} \omega(\mathbf{q})n_i[u_i(\mathbf{q})d_{\mathbf{q}} + H.c.] + \sum_{ij} V_{ij}n_i n_j, \quad (18)$$

where $T(\mathbf{m})$ is the bare hopping integral, $i = (\mathbf{m}, s)$, $j = (\mathbf{n}, s)$ ($\mathbf{m}, \mathbf{n}, s$ stands for sites and spin, respectively), $n_i = c_i^\dagger c_i$, $u_i(\mathbf{q}) = g \exp(i\mathbf{q} \cdot \mathbf{m})/\sqrt{N}$, $\omega(\mathbf{q})$ are the coupling constant and the phonon frequency, and V_{ij} is the *direct* (density-density) Coulomb repulsion. It can be diagonalised *exactly* if $T(\mathbf{m}) = 0$ (or $\lambda = \infty$). The ground state bipolaron configuration

is found by the use of the lattice minimisation technique [21] fully taking into account the direct Coulomb repulsion. Then applying $1/\lambda$ perturbation expansion the bipolaron effective mass tensor is readily derived. In perovskite structures the in-plane oxygen-oxygen pairs appears to be the ground state. Therefore the in-plane bipolaron tunneling is essentially one-particle and the in-plane effective mass appears to be of the order of the small polaron mass about $10m_e$ [18]. On the other hand, the c -axis tunneling of the bipolaron is only possible via a Josephson-like hopping. In that case one derives [22,19]

$$\frac{1}{m_c^{**}} \simeq 4t^2 d^2 \sqrt{\frac{2\pi}{\omega\Delta}} \exp\left[-\frac{\Delta}{\omega} \left(1 + \ln \frac{2g^2\omega}{\Delta}\right)\right]. \quad (19)$$

Here $t = T(d)e^{-g^2}$ is the inter-plane polaron hopping integral. Then the ratio of the c -axis singlet mass to that of the polaron one is given by

$$\frac{m_c^{**}}{m_c^*} \simeq \frac{1}{2} \sqrt{\frac{\omega\Delta}{2\pi T^2(d)}} \exp(3g^2) \gg 1, \quad (20)$$

which justifies the proposed kinetics described by Eqs(3-5). The isotope effect on both T_c [23] and on the Néel temperature T_N [24] favors the electron-phonon coupling as the origin of the polaron and bipolaron formation in *LSCO*. The pair-distribution analysis of neutron scattering [25] also suggests that the ‘spin-gap’ is consistent with the formation of a bipolaronic local singlet state.

In conclusion, we have developed a coherent transport theory of copper based high- T_c oxides, which describes the doping and temperature dependence of the resistivity and the magnetic susceptibility of *LSCO* as well as the normal state gap. Our theory is selfconsistent in the sense that the mean-free path for bipolarons remains larger than the lattice constant. The large resistivity values are due to a strongly enhanced effective mass and a small polaron density rather than to a short mean-free path. Thus the Boltzmann kinetics is applied. No question arises with the three-dimensional superconductivity either. The Bose-Einstein condensation of bipolarons explains the high value of T_c because its dependence on a huge c -axis singlet mass is only logarithmic [11]. At very low temperatures polarons are frozen out, so we expect the temperature independent anisotropy ρ_c/ρ_{ab} when T is low. In the

magnetic field the normal state gap becomes smaller due to the spin splitting of the polaron level, so a negative c -axis magnetoresistance is expected. Both features have been recently observed [26].

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FIGURES

FIG. 1. Theoretical anisotropy (*a*) and the magnetic susceptibility (*b*) with $\Delta_0 = 55K$.

FIG. 2. Experimental anisotropy [4] (*a*) compared with the theoretical one by the use of Eq.(12) with the experimental [9] $\chi(T, x) = \chi_s(T, x) + 0.4 \times 10^{-7} emu/g$ (*b*).

FIG. 3. Normal state gap as a function of temperature and doping.