Coherent *ab* and *c* Transport Theory of High-T_c Cuprates

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We propose a microscopic theory of the normal state transport in copper oxides based on the bipolaron theory which describes qualitatively 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 in underdoped, optimally doped, and not very heavily overdoped $La_{2-x}Sr_xCuO_4$ for the temperature range from T_c up to 600 K. A free parameter relation between the anisotropy and the spin susceptibility is derived, $\rho_c(T,x)/\rho_{ab}(T,x) \sim x/T^{1/2}\chi_s(T,x)$, which agrees quantitatively with the experiment. The normal state gap is explained and its doping and temperature dependence clarified. [S0031-9007(96)01809-1]

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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 the high-T_c cuprates are recognized 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, $x = 0.15$, $\sigma_{ab} \simeq 2 \times 10^5\ \Omega^{-1}\,\mathrm{m}^{-1}$, and $\sigma_c \simeq 10^3\ \Omega^{-1}\,\mathrm{m}^{-1}$ [4], one obtains

$$\frac{E_F\tau}{\hbar} = \frac{\pi\hbar d\sigma_{ab}}{e^2} \simeq 1.5 \qquad (1)$$

and the ratio of the mean-free path on the *c* direction l_c to the interplane distance $d \simeq 6$ Å as small as $l_c/d < 0.01$, where E_F is the Fermi energy and τ is the transport relaxation time. This estimate as well as the semiconductorlike 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 and thermodynamics measurements [1] led us [6] to the conclusion that the so-called spin gap observed previously in the magnetic susceptibility [7] (in the spin channel) could belong to the charge carriers, which are intersite bipolarons. In particular, the explanation of the NMR linewidth and the linear in-plane resistivity as well as of the Hall effect was proposed [8,9]. The comprehensive analysis by Batlogg and co-workers [10] revealed transport features incompatible with the spin-charge separation and the doping dependence of the gap, as well as the semiconducting doping dependence of the resistivity. It appears that high-T_c oxides are doped semiconductors rather than metals, irrespective to the level of doping [11].

In this Letter the normal 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 [6].

The small polarons are paired *above* T_c in strongly correlated Mott-Hubbard insulators with the electron-phonon coupling constant above an intermediate value $\lambda \geq 0.5$ [12]. 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 [6]. Singlets tunnel along the plane with an effective mass m^{**}_{ab} of the order of a single-polaron mass m^{*}_{ab} as shown by one of us [11]. 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, $m^{**}_{c} \gg m^{**}_{ab} \sim m^{*}_{ab}$. In that way we explain a large transport anisotropy ($>10^3$) of copper oxides at low temperatures when polarons are frozen out. 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. At the same time the polaron contribution to the *ab* transport is small at any temperature due to their low density compared with the bipolaron one. As a result we have a mixture of the *nondegenerate* quasi-two-dimensional spinless bosons and the thermally excited fermions, which are capable to propagate between the planes. Only polarons contribute to the spin susceptibility which leads to a fundamental relation between the anisotropy and the magnetic susceptibility.

Quite generally the in-plane bipolaron, σ_{ab}, *c*-axis polaron, σ_c conductivities, and the uniform spin susceptibility χ_s are expressed as ($\hbar = k_B = 1$)

$$\sigma_{ab}(T,x) = -4\int_0^\infty dE\,\sigma_b(E)\,\frac{\partial f_b}{\partial E}, \qquad (2)$$

$$\sigma_c(T,x) = -2\int_0^\infty dE\,\sigma_{pc}(E)\,\frac{\partial f_p}{\partial E}, \qquad (3)$$

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$$\chi_s(T,x) = -2\mu_B^2 \int_0^\infty dE\, N_p(E)\, \frac{\partial f_p}{\partial E}, \qquad (4)$$

where $f_b = [y^{-1}\exp(E/T) - 1]^{-1}$ and $f_p = [y^{-1/2} \times \exp(E/T + \Delta/2T) + 1]^{-1}$ 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 [6]. Therefore polarons are not degenerate at any temperatures. Above T_c bipolarons are also not degenerate, so that $f_b \simeq y\exp(-E/T)$, and $f_p \simeq y^{1/2} \times \exp(-E/T - \Delta/2T)$. If the scattering mechanism is the same for polarons and bipolarons the ratio of the differential bipolaron $\sigma_b(E)$ and polaron $\sigma_{pc}(E)$ conductivities [13] is independent of the energy and doping

$$\frac{\sigma_b(E)}{\sigma_{pc}(E)} \equiv A = \text{const.} \qquad (5)$$

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 [11] with a quasi-one-dimensional polaron density of states as observed with the high resolution angle-resolved photoemission spectroscopy (ARPES) [14],

$$N_p(E) \simeq \frac{1}{4\pi da}\left(\frac{m^*}{2E}\right)^{1/2}, \qquad (6)$$

where a is the in-plane lattice constant. Then the c-axis resistivity as well as the spin susceptibility are expressed as

$$\frac{\rho_c(T,x)}{\rho_{ab}(T,x)} = 2Ay^{1/2}\exp\left(\frac{\Delta}{2T}\right) \qquad (7)$$

and

$$\chi_s(T,x) = \frac{\mu_B^2}{2da}\left(\frac{ym^*}{2\pi T}\right)^{1/2}\exp\left(-\frac{\Delta}{2T}\right). \qquad (8)$$

The chemical potential, $y = 2\pi n_b(T,x)/Tm_{ab}^{**}$, is calculated by taking into account the Anderson localization of bipolarons in a random potential [9]. The bipolaron density $n_b(T,x)$ per cm^2 appears to be linear in temperature and doping, $n_b(T,x) \simeq xT/\gamma a^2$, in a wide temperature range below the bipolaron ($\sim 1/m_{ab}^{**}a^2$) and the impurity (γ) bandwidth. Then the temperature and doping dependence of the Hall effect $R_H \sim 1/2en_b(T,x)$ as well as the linear *ab* resistivity are well described [9]. As a result we find the temperature independent $y \sim x$ [15] and

$$\frac{\rho_c(T,x)}{\rho_{ab}(T,x)} = C\,\frac{x}{T^{1/2}\chi_s(T,x)}, \qquad (9)$$

$$\chi_s(T,x) = C'\left(\frac{x}{T}\right)^{1/2}\exp\left(-\frac{\Delta(T,x)}{2T}\right), \qquad (10)$$

where $C = 2A\mu_B^2(2\pi m^*)^{1/2}/\gamma m_{ab}^{**}a\Omega$ and $C' = \mu_B^2 \times (m^*/\gamma m_{ab}^{**})^{1/2}/\Omega$ are assumed to be constants, and Ω a unit cell volume. We expect a strong dependence of the binding energy, $\Delta \equiv \Delta(T,x)$, on doping because of the screening. 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, $q_s = [16\pi e^2 n_b(T,x)/\epsilon_0 T]^{1/2}$, and the static dielectric constant $\epsilon_0 \simeq 30$ one obtains the value of $q_s \simeq 3\ \text{Å}^{-1}$ at room temperature which is about 3 times larger than the reciprocal lattice vector q_d. Hence the polaron (Franck-Condon) level shift E_p is suppressed by the screening being proportional to q_d^2/q_s^2 for large q_s [12]. Consequently, the normal state gap depends on the bipolaron density as $\Delta(T,x) \sim T/n_b(T,x)$, which yields the temperature independent gap,

$$\Delta = \Delta_0/x, \qquad (11)$$

where Δ_0 is doping independent, if $n_b(T,x) \sim xT$. This estimate is applied in a restricted range of doping near the optimum value, $x = 0.15$. At small doping the screening radius is larger than the lattice constant. As a result, we do not expect any doping dependence of Δ below $x = 0.05$. On the other hand, at large doping the screening weakens the polaron orthogonality (phonon overlap) blocking of the tunneling. The (bi)polaron band widens and becomes comparable with E_p. Then the binding energy Δ, which is the difference between the minimum of the polaronic and bipolaronic bands [see Fig. 2(a) below], becomes smaller than both E_p and the bandwidth. As a result, the doping suppression of Δ is expected to be more significant than Eq. (11) suggests for the overdoped samples.

One can describe all qualitative features of the c-axis resistivity and the magnetic susceptibility of $La_{2-x}Sr_xCuO_4$ by the use of Eqs. (9) and (10) and Eq. (11) as the comparison of Figs. 1(a)–1(b) and Figs. 1(c)–1(d) shows. The linear temperature dependence of the *ab* resistivity has been explained within the same approach [8,9]. Thus the c-axis resistivity is now understood as well. The anisotropy is described *quantitatively* by Eq. (9) with the *experimental* value of $\chi_s(T,x)$ [see Fig. 1(d)], allowing small sample variation of the constant C within less than 15% as shown in Fig. 1(c). A temperature independent anisotropy of a heavily overdoped sample, $x = 0.3$ in Fig. 1(c), is explained by the fact that polarons contribute to the *ab* transport when the bipolaron binding energy Δ is below 100 K as shown in Fig. 2(b). The normal state gap $\Delta(T,x)$ can be found from Eq. (8) by the use of the experimental values of $\chi_s(T,x)$, Fig. 1(d). With the temperature independent $y(x)$ in agreement with a flat temperature dependence of the thermoelectric power of $La_{2-x}Sr_xCuO_4$ one obtains

$$\Delta(T,x) = 2T\,\ln\frac{B_\infty}{T^{1/2}\chi_s(T,x)}, \qquad (12)$$

where $B_\infty = \mu_B^2[m^*a^2y(x)/2\pi]^{1/2}/\Omega$ shows only weak (if any) dependence on doping, $B_\infty \simeq 5.46 \times 10^{-6}\,\text{emu}\,\text{K}^{1/2}/\text{g}$. The simultaneous increase with doping of $y \sim x$ [15] and the decrease of the polaron mass m^* due to screening make B_∞ practically doping independent. The values and doping dependence of $\Delta(T,x)$,

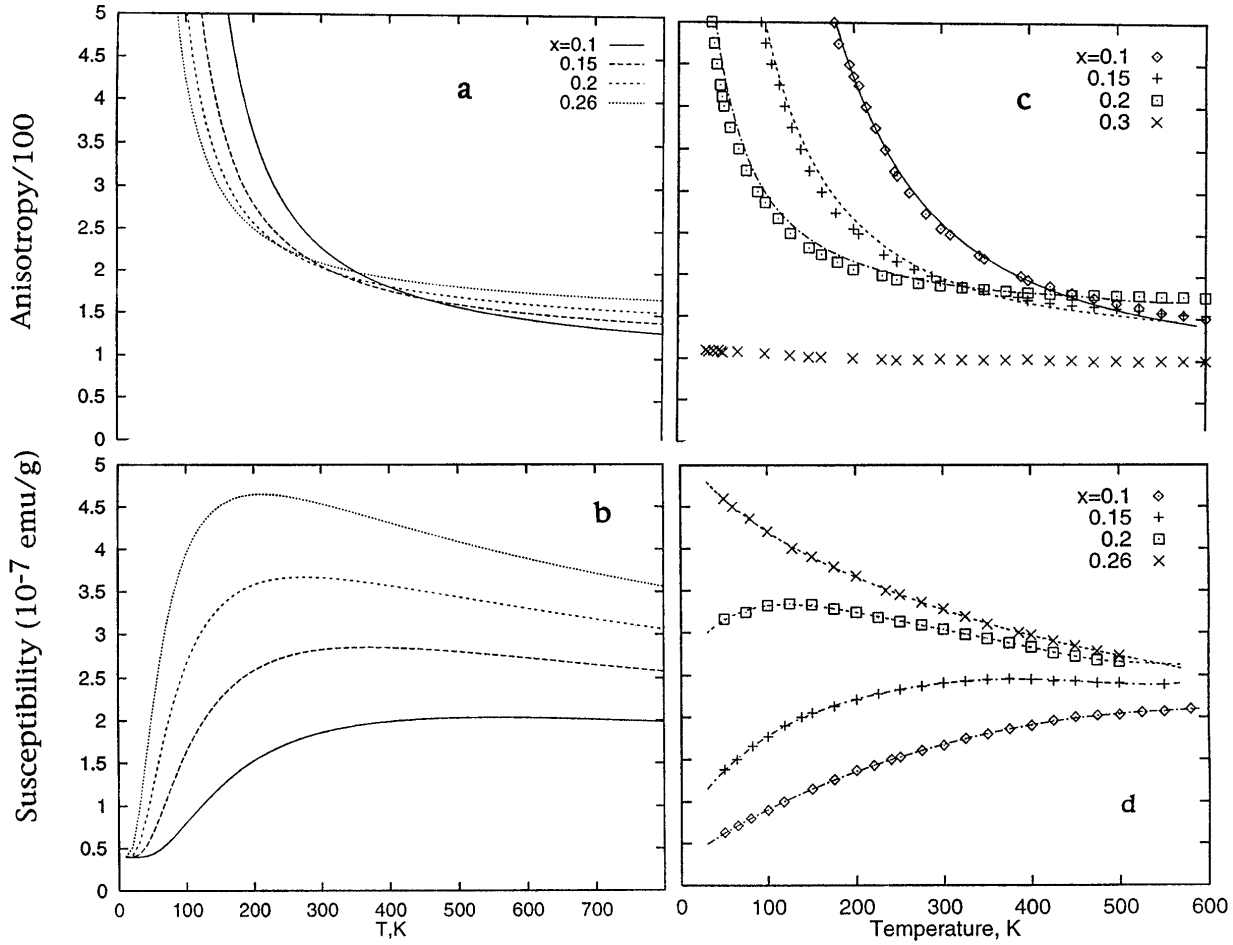

FIG. 1. Theoretical anisotropy (a) and magnetic susceptibility (b) $\chi = \chi_s(T,x) + \chi_0$ with $\chi_0 = 0.4 \times 10^{-7}$ emu/g and with the temperature independent gap, Eq. (11) ($\Delta_0 = 55$ K). The anisotropy ratio (c) [4] compared with the theory (lines) by the use of Eq. (9) and the experimental $\chi(T,x)$ [7] (d). The lines in (d) are used to calculate the anisotropy (c) and the normal state gap in Fig. 2(b).

determined with Eq. (12), Fig. 2(b), are about the same as Batlogg's normal-state-gap temperature $T^*(x)$ [10]. The normal state gap $\Delta(T,x)$, Fig. 2(b), is the "spin gap" in the uniform spin susceptibility. For the finite wave vector $q \sim (\pi,\pi)$ the inverse of which is of the order of the size of the intersite bipolaron, the bipolaron formation affects $\chi_s(q)$ less than $\chi_s(0)$. Therefore the intersite bipolaron picture alone might be insufficient to resolve all problems related to $1/T_1T$.

The proposed kinetics of high-T_c cuprates is derived from the generic Hamiltonian, which includes the bare hopping integral, the electron-phonon interaction, and the *direct* Coulomb repulsion. It can be diagonalized *exactly* in the limit $\lambda \simeq E_p/E_F \to \infty$ [6,12]. The ground state bipolaron configuration is found by the use of the lattice minimization technique [16] fully taking into account the direct Coulomb repulsion. Then applying $1/\lambda$ perturbation theory the bipolaron effective mass tensor is readily derived [11]. The in-plane oxygen-oxygen pairs appear to be the ground state in perovskite structures. Therefore the in-plane bipolaron tunneling is essentially one particle, and the in-plane effective mass is of the order of the small polaron mass about $10m_e$. On the other hand, the *c*-axis tunneling is only possible via a Josephson-like hopping. In that case one derives [12,17]

$$\frac{1}{m_c^{**}} \simeq 4t^2d^2\left(\frac{2\pi}{\omega\Delta}\right)^{1/2} \exp\left[-\frac{\Delta}{\omega}\left(1 + \ln\frac{2g^2\omega}{\Delta}\right)\right]. \tag{13}$$

Here $t(d) = T(d)e^{-g^2}$ is the interplane polaron hopping integral and ω the optical phonon frequency. The ratio of the *c*-axis singlet mass to that of the polaron one is given by

$$\frac{m_c^{**}}{m_c^*} \simeq \frac{1}{2}\left(\frac{\omega\Delta}{2\pi T^2(d)}\right)^{1/2} e^{3g^2} \gg 1\,, \tag{14}$$

for $\Delta \simeq 2E_p = 2g^2\omega$ which justifies the proposed kinetics described by Eqs. (2) and (3). The isotope effect on both T_c [18] and on the Néel temperature T_N [19] favors

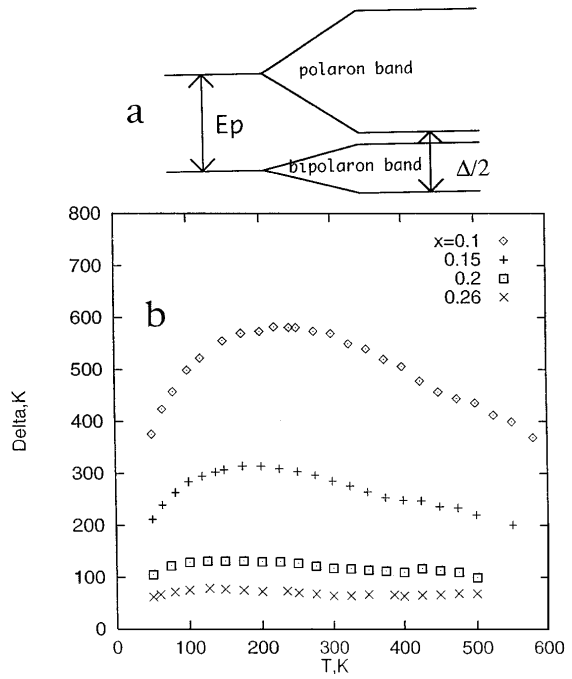

FIG. 2. The theoretical normal state gap as a function of temperature and doping (b) defined according to (a).

the electron-phonon coupling as the origin of the polaron and bipolaron formation in cuprates. The pair-distribution analysis of neutron scattering [20] also suggests that the spin gap is consistent with the formation of a bipolaronic local singlet state. However, our transport theory may be applicable beyond a pure electron-phonon model of preformed pairs.

In conclusion, we have developed a microscopic transport theory of copper based high T_c oxides, which describes the doping and temperature dependence of the *ab* and *c*-axis resistivity and the magnetic susceptibility as well as the normal state gap. A free parameter expression for the anisotropy is derived which fits well the experimental data. No question arises with the three-dimensional superconductivity. 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 [21]. At very low temperatures polarons are frozen out, so we expect the temperature independent anisotropy ρ_c/ρ_{ab} when T is low. In a magnetic field the normal state gap becomes smaller due to the spin splitting of the polaron level, Fig. 2(a), so a negative *c*-axis magnetoresistance is expected. Both features have been recently observed [22]. The unusual logarithmic divergence of resistivity [22] is explained by one of us [23] as the result of the resonance Wigner scattering of nondegenerate carriers by the attractive random potential.

This paper is inspired by Mott's original idea that doped Mott insulators are nondegenerate semiconductors in the normal state in which randomness plays an essential role. We highly appreciate enlightening discussions with Y. Ando, P. P. Edwards, J. Cooper, R. Haydock, N. Hussey, D. Khmelnitskii, W. Y. Liang, J. Loram, A. Mackenzie, J. Tallon, and K. R. A. Ziebeck, and the Royal Society financial support of one of us (V. V. K.).

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[13] Equations (2) and (3) are derived with the Kubo formalism, which is somewhat more general then the Boltzmann theory. If the Boltzmann theory and the τ approximation are applied, the differential conductivity along the $i = (a, b, c)$ direction is determined as $\sigma_i(E) = e^2 \sum_{\mathbf{k}} (\partial E_{\mathbf{k}}/\partial k_i)^2 \tau_{\mathbf{k}} \delta(E - E_{\mathbf{k}})$ for the energy spectrum $E_{\mathbf{k}}$ on a lattice. The relation Eq. (5) holds independently of the dimensionality of the spectra because $\tau_{\mathbf{k}} \sim N(E)^{-1}$.
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