Hall Effect and Resistivity of High-T_c Oxides in the Bipolaron Model

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We discuss the Hall effect and resistivity above T_c, using a variant of the bipolaron theory which takes into account Anderson localization of the bosons by disorder. The model supposes that $R_H = 1/2en_bc$, where n_b is the number of delocalized carriers. Temperature and doping dependences of ρ, R_H, $\cot\Theta_H$, and the "spin" gap in $YBa_2Cu_3O_{7-\delta}$ are explained.

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The purpose of this paper is to apply the bipolaron model of high T_c, with consideration of the localization of the carriers by disorder, to the Hall effect and resistivity of Y-Ba-Cu-O (YBCO). It is recognized that other theories which are basically different have successfully explained many features of the Hall "constant" and resistivity; our preference for the bipolaron model and some criticism of others is contained in our recent papers [1–3]. Mott has shown some time ago [4] that a T-dependent Hall coefficient can be explained if the carriers are spin bipolarons . More recently he suggested [1] that, since inelastic neutron scattering does not reveal any moments, the spin polarons must transform into highly correlated metal disks. If so, we do not think the predicted Hall behavior should occur. In general both lattice vibrations and spin fluctuations give rise to polarons and bipolarons, which can be small or large, depending on the bandwidth and the strength of the interaction. It is important for application to high-T_c superconductivity that the bipolaron bounds more easily in two dimensions than in 3D, and that the mean value of the pair radius is a few angstroms [5–7]. Thus the ground state of high-T_c oxides can be seen as a charged Bose liquid of $2e$ spin-lattice bipolarons [2]. Our assumption is then that above T_c a material such as YBCO contains a nondegenerate gas of singlet bipolarons. We have suggested elsewhere that above 200 K there are also triplets with a slightly lower mass due to the lower binding energy. The main new point is to assume that a proportion of bipolarons are in Anderson localized states, and as pointed out by one of us [1] Coulomb repulsion limits the number of bosons in each localized state, so that the distribution function will show a mobility edge E_c. Because of low dimensionality of high-T_c copper oxides (2D rather then 3D) any random potential leads to localization no matter how weak it is. Moreover one-particle density of extended states above E_c is constant for practically all energies within the bipolaronic band of the width $2w$:

$$N(\epsilon) = \frac{1}{2w}, \qquad E_c < \epsilon < E_c + 2w. \tag{1}$$

The main part of the electron-electron (Hubbard U) and electron-phonon correlation energy is included in the binding energy of bipolarons and in their bandwidth renormalization. The rest, including the boson-boson repulsion, may be treated for extended states as perturbation resulting in the canonical Boltzmann kinetics or in the Bogoliubov excitations in the superconducting state. In the normal state the corrections due to the interaction to the single-particle spectrum are small if the gas parameter (for a short-range repulsion) or the random phase approximation parameter r_s for Coulomb forces is not very large. In that case the density of extended (free) bosons is given by

$$n_b(T) = \frac{T}{2w} \ln\left(\frac{1 - ye^{-2w/T}}{1 - y}\right), \tag{2}$$

where $y = \exp[(\mu + E_c)/T]$, $\mu < E_c$ is the chemical potential, and k_B is taken to be unity. To calculate the density of localized bosons $n_L(T)$ one should take into account the repulsion between them. One cannot ignore the fact that the localization length ξ generally varies with energy and diverges at the mobility edge. In the case of *charged* bosons their number in a single potential well is determined by the competition between their long-range Coulomb repulsion $\simeq 4e^2/\xi$ and the binding energy $E_c - \epsilon$. If the localization length diverges with the critical exponent $\nu < 1$ [8], $\xi \sim (E_c - \epsilon)^{-\nu}$, one can apply a "single-well–single-particle" approximation assuming that one can place only one boson in each potential well. The gross features of the temperature behavior of $n_L(T)$ are not influenced by this approximation if the number of bosons in a potential well is finite [8]. Within this approximation localized *charged* bosons obey the Fermi-Dirac statistics:

$$n_L(T) = \int_{-\infty}^{E_c} \frac{N_L(\epsilon)d\epsilon}{\exp(\frac{\epsilon-\mu}{T}) + 1}, \tag{3}$$

where the density of localized states $N_L(\epsilon)$ may be approximated in many cases by the exponential tail:

$$N_L(\epsilon) = \frac{n_L}{\gamma} \exp\left(\frac{\epsilon - E_c}{\gamma}\right), \tag{4}$$

with γ of the order of a binding energy in a single ran-

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dom potential well and n_L the total number of localized states per unit cell. The integral in Eq. (3) is equal to the total number of occupied localized states and varies linearly with temperature over a wide range $T < (\gamma, 2w)$ because the chemical potential is pinned in this temperature region near the mobility edge E_c, which one can choose as an energy origin, $\mu \simeq E_c = 0$. The pinning of μ follows from conservation of the total number of bosons per cell, $n = n_b(T) + n_L(T)$, which gives for the chemical potential

$$\frac{T}{2w} \ln \frac{1}{1-y} - \frac{n_L T}{\gamma} \ln\left(\frac{1+y}{y}\right) = n - n_L. \tag{5}$$

If $T \ll (\gamma, 2w)$, the solution of this equation is $y \simeq 1$ with an exception of a very narrow region of "compensation" $n - n_L \ll T/2w$, where y is approximately 0.6 for $\gamma = 2w$. The density n_b depends on y logarithmically, therefore its temperature dependence remains practically linear up to $T \simeq \gamma$:

$$n_b(T) = n - n_L + n_L b T, \tag{6}$$

with $b = \gamma^{-1} \ln(\frac{1+y}{y})$ and thus temperature independent. Solving the Boltzmann equation with a weak magnetic field H for extended bosons, scattered by acoustical phonons and by unscreened random potential in the relaxation time approximation [9], or using a variational approach for the inelastic boson-boson scattering one obtains the canonical expressions for the Hall ratio R_H, resistivity, ρ and $\cot\Theta_H$:

$$R_H = \frac{v_0 \langle \tau^2 \rangle}{2e n_b(T) \langle \tau \rangle^2}, \tag{7}$$

$$\rho = \frac{v_0 m}{4e^2 n_b(T) \langle \tau \rangle}, \tag{8}$$

$$\cot\Theta_H = \frac{\langle \tau \rangle}{\omega_c \langle \tau^2 \rangle}, \tag{9}$$

where $m = \pi\hbar^2/wa^2$ is the in-plane boson mass, a is the in-plane lattice constant, $\omega_c = 2eH/mc$, τ is the transport relaxation time, v_0 is the volume of an elementary cell (0.167 nm^3 for $YBa_2Cu_3O_{7-\delta}$), and $\langle \cdots \rangle$ stands for an average with the energy times the energy derivative of the Bose-Einstein distribution function. The transport relaxation rate due to 2D boson-phonon scattering has been shown to be energy independent and linear in temperature [9]:

$$\frac{1}{\tau_{b-ac}} = m C_{ac} T, \tag{10}$$

where the constant C_{ac} is proportional to a deformation potential. In the case of Bose-Einstein statistics and $T \ll 2w$ an umklapp scattering can be neglected so the scattering between extended bosons does not contribute to the resistivity. However, the inelastic scattering of an extended boson by localized bosons gives a contribution because momentum is not conserved in two-particle collisions in the presence of the impurity potential. In a "single-well–single-particle approximation" the role of the Pauli exclusion principle is played by the dynamical repulsion between bosons. That is why the boson-boson "transport relaxation rate" has the same temperature dependence as the fermion-fermion scattering and identical to that calculated by Xing and Liu in the case of localized fermions [10]. This transport relaxation rate is proportional to T^2 because only localized bosons within the energy shell of the order of T near the mobility edge contribute to the scattering and because the number of the final states is proportional to temperature:

$$\frac{1}{\tau_{b-b}} = \frac{\alpha e^2 b n_L}{m} T^2, \tag{11}$$

with α as a constant determined in Ref. [10]. Unoccupied potential wells with the density $bn_L T$ also contribute to the scattering, giving rise to the energy independent elastic relaxation rate, which is linear in temperature:

$$\frac{1}{\tau_{b-im}} = m C_{im} n_L T \tag{12}$$

with C_{im} as a constant. Substitution of Eqs. (10)–(12) into Eqs. (7)–(9) yields

$$R_H = \frac{v_0}{2e(n - n_L + b n_L T)}, \tag{13}$$

$$\rho = (m^2 C v_0/4e^2) \frac{T + \sigma_b T^2}{n - n_L + b n_L T}, \tag{14}$$

where $C = C_{ac} + n_L C_{im}$ and $\sigma_b = \alpha e^2 b n_L / m^2 C$ is the relative boson-boson scattering cross section, and

$$\cot\Theta_H = \frac{c m^2 C}{2eH} \left(T + \sigma_b T^2\right). \tag{15}$$

These formulas contain rich information about the number of bosons, localized states, and the relative strength of different scattering channels. The spin-fluctuation contribution to the relaxation rate is shown to be linear in temperature [1], so it can change only the value of the constant C. There are two fitting parameters, n and n_L, if no significant variation of m^2C, b, σ_b is expected with doping. Because of the chains the number of in-plane carriers is not fixed by the chemical formula at least in "1:2:3" YBCO. On the experimental side we have in-plane kinetic data for $YBa_2Cu_3O_{7-\delta}$ in a wide range of doping and temperatures [11,12]. The theoretical fit with parameters $n - n_L$, bn_L, m^2C, and σ_b is shown in Figs. 1–3. These parameters are presented in Table I. One can see from Table I that the number of localized states increases with doping (if the width γ remains constant). The number of extended bosons also increases. The scattering cross sections and their relative contribu-

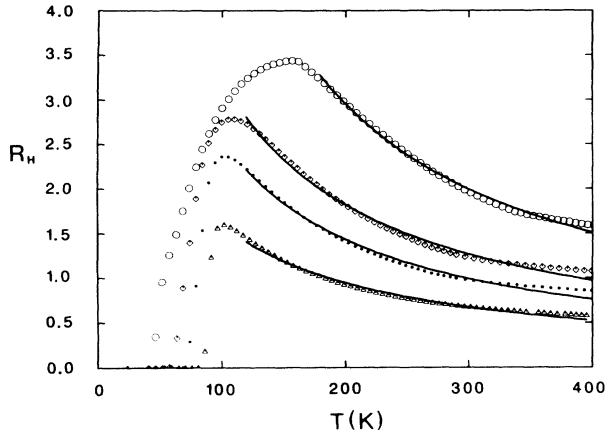

FIG. 1. Hall coefficient (in units of 10^{-9} $C^{-1}m^3$) for $YBa_2Cu_3O_{7-\delta}$ [11] compared with the theory [Eq. (13), solid lines] for different δ: 0.05 ($\triangle$), 0.19 ($\bullet$), 0.23 ($\diamond$), and 0.39 ($\circ$). Parameters of the model are presented in Table I.

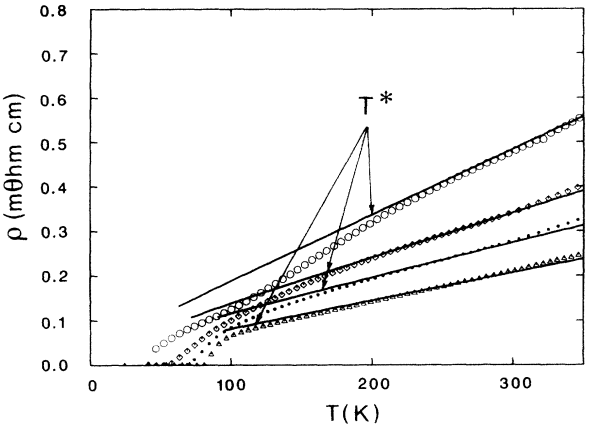

FIG. 2. Resistivity for $YBa_2Cu_3O_{7-\delta}$ [11] compared with the theory, Eq. (14). For parameters see Table I.

tion slightly depend on the doping. Underdoped samples ($\delta > 0.2$) are practically "compensated" in the sense that the total number of bosons is very close to the number of localized states. This should be the case if every additional oxygen ion produces a single localized state. From the value of σ_b (see Table I) one can see that boson-phonon and boson-boson scattering rates are comparable near T_c. At higher temperatures the latter dominates. *The residual resistivity is zero in our model.*

It should be mentioned that the dissociation of a boson on two single nondegenerate polarons at a sufficiently high temperature has no influence on the temperature dependence of R_H and ρ if the effective mass of a boson is close to the effective mass of two single polarons. Any *nondegenerate* carriers on a two-dimensional lattice have the same temperature and doping dependences of their kinetic properties, Eqs. (13)–(15). This is also the case for triplet bipolarons. However, if triplets are lighter then singlets the slope of resistivity (proportional to m^2C) diminishes with temperature because singlets are thermally excited into triplets. The characteristic small deviation from linearity (see Fig. 2) should appear at temperature T^* where a "spin" gap in NMR and in neutron scattering [13,14] appears. Earlier we have explained with triplet bipolarons the temperature dependent Korringa ratio [9] and the change in slope of resistivity at $T = T^*$ [15], which have been measured in $YBa_2Cu_4O_8$ by Machi *et al.* [16] and by Bucher *et al.* [17] correspondingly. Recently Ito *et al.* [12] have observed a correlation in the slope of resistivity with the temperature dependent Korringa ratio in $YBa_2Cu_3O_{7-\delta}$. The temperature dependence of the normal state near-infrared absorption in $YBa_2Cu_3O_{7-\delta}$ also reflects the spin gap [18]. All these observations support our explanation [9,15] of the spin gap as a singlet-triplet bipolaron exchange energy. There is a direct connection of the proposed normal state kinetics with high-T_c phenomena. If T_c is a temperature of the Bose-Einstein condensation of charged bosons then a simple estimation with 3D corrections to $N(\epsilon)$ taken into account [9] yields

$$T_c = \frac{2w(n - n_L)}{L} \tag{16}$$

for

$$n - n_L \gg \frac{2t_\perp}{w} \tag{17}$$

with L the logarithm of the ratio of out-of-plane to in-plane mass, and

$$T_c \simeq 4t_\perp \tag{18}$$

in the opposite limit $n - n_L < 2t_\perp/w$. If one takes a resonable value of the in-plane bipolaronic half-bandwidth $w \simeq 450$ K (for estimations see Ref. [19]) in Eq. (16) or

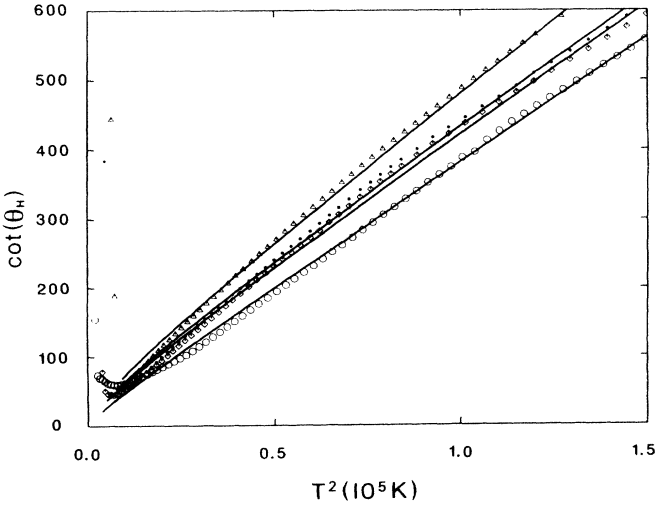

FIG. 3. $\cot\Theta_H$ [11] compared with the theory, Eq. (15).

TABLE I. Microscopic parameters of the model determined from the experimental data [11].

δ	$n - n_L$	$bn_L(10^{-3}\ \mathrm{K}^{-1})$	$m^2C\left(10^{-18}\frac{\mathrm{kg}}{\mathrm{sec\,K}}\right)$	$\sigma_b(10^{-2}\ \mathrm{K}^{-1})$
0.05	0.103	2.22	0.81	1.0
0.19	0.041	1.61	0.69	1.1
0.23	0.035	1.25	0.62	1.2
0.28	0.023	1.11	0.46	1.6
0.39	0.007	0.85	0.46	1.6

the value of the out-of-plane half-bandwidth $2t_\perp \simeq 45$ K in Eq. (18) one can explain the high value of $T_c \simeq 90$ K. The basic phenomenon that allows the high T_c is that the polaronic narrowing of the band, which eliminates the small exponential factor in the BCS or McMillan's formula, as has been already discussed by one of us [20]. On the other side the well-known shortcomings of the Schafroth model like a huge value of $T_c \simeq 10\,000$ K [21] are not shared by bipolaronic model of superconductivity. Their enhanced effective mass $> 10m_e$ and the low enough concentration $n \simeq 10^{21}$ cm^{-3} push T_c in the range of 100 K. These and other "Bose-liquid" features lead us to a firm conclusion that a charged Bose liquid is a simple but far-reaching model of low-frequency kinetics and thermodynamics of of high-T_c superconductors [1–3] in both the normal and the superconducting states [22]. In this Letter we have shown that this model gives a quantitative explanation of the longitudinal σ_{xx} and transverse σ_{xy} conductivities of copper based high-T_c oxides if one takes into account the Anderson localization.

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