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# Hall effect and resistivity in underdoped cuprates

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The behaviour of the Hall ratio  $R_H(T)$  as a function of temperature is one of the most intriguing normal state properties of cuprate superconductors. One feature of all the data is a maximum of  $R_H(T)$  in the normal state that broadens and shifts to temperatures well above  $T_c$  with decreasing doping. We show that a model of preformed pairs-bipolarons provides a selfconsistent quantitative description of  $R_H(T)$  together with in-plane resistivity and uniform magnetic susceptibility for a wide range of doping.

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The theory of high temperature superconductivity remains the biggest challenge in condensed matter physics today. One way of thinking is that this phenomenon is of purely electronic origin and phonons are irrelevant [1, 2, 3, 4]. Other authors (see, for example [5, 6, 7, 8, 9, 10]) explore an alternative view, namely that the extension of the BCS theory towards the strong interaction between electrons and ion vibrations describes the phenomenon. High temperature superconductivity could exist in the crossover region of the electron-phonon interaction strength from the BCS to bipolaronic superconductivity as was argued before the discovery [11]. In the strong coupling regime,  $\lambda \gtrsim 1$ , pairing takes place in real space (i.e. *individual pairing*) due to a polaron collapse of the Fermi energy [12]. At first sight, polaronic carriers have a mass too large to be mobile; however it has been shown that the inclusion of the on-site Coulomb repulsion leads to the favoured binding of intersite oxygen holes [13, 14]. The intersite bipolarons can then tunnel with an effective mass of about 10 electron masses [8, 13, 14, 15].

The possibility of real-space pairing, as opposed to the Cooper pairing, has been the subject of much discussion. Experimental [16, 17, 18, 19, 20, 21] and theoretical [22, 23, 24, 25] evidence for an exceptionally strong electron-phonon interaction in all novel superconductors is now so overwhelming that even some advocates of non-phononic mechanisms [26] accept this fact. Nevertheless, the same authors [2, 26] dismiss any real-space pairing, suggesting a *collective* pairing (i.e the *Cooper* pairs in the momentum space) at some temperature  $T^* > T_c$  but without phase coherence. The existence of noncoherent Cooper pairs might be a plausible idea for the crossover region of the coupling strength, as was proposed still earlier by Dzhumanov [10]. However, apart from this, Refs.[2, 26] argue that the phase coherence and superconducting critical temperature  $T_c$  are determined by the superfluid density, which is proportional to doping  $x$ , rather than to the density of normal state holes, which is  $(1+x)$  in their scenario. On the experimental side, the scenario is not compatible with a great number

of thermodynamic, magnetic, and kinetic measurements, which show that only holes *doped* into a parent insulator are carriers *in both* the normal and superconducting state of *underdoped* cuprates. On theoretical grounds, this preformed *Cooper*-pair (or phase-fluctuation) scenario contradicts a theorem [27], which proves that the number of supercarriers (at  $T = 0$ ) and normal-state carriers should be the same in any *clean* translation-invariant superfluid. A periodic crystal-field potential does not affect this conclusion because the coherence length is larger than the lattice constant in cuprates. Objections against real-space pairing also contradict a parameter-free estimate of the renormalised Fermi energy  $\epsilon_F$ [28], that yields  $\epsilon_F$  less than the normal state charge pseudogap  $\Delta/2$  in underdoped cuprates. The condition for real-space pairing,  $\epsilon_F \lesssim \pi\Delta$ , is well satisfied if one admits that the bipolaron binding energy,  $\Delta$ , is twice the pseudogap.

Bipolarons in cuprates could be formed by the Fröhlich interaction of holes with optical phonons [13], and by molecular phonons (Jahn-Teller interactions [9]) because of molecular-like crystal structure of these materials. Mott and Alexandrov proposed a simple model [23] of the cuprates based on bipolarons. In this model, all the holes (polarons) are bound into small intersite bipolarons at any temperature. Above  $T_c$  this Bose gas is non-degenerate and below  $T_c$  phase coherence of the preformed bosons sets in, followed by superfluidity of the charged carriers. Of course, there are also thermally excited single polarons in the model. There is much evidence for the crossover regime at  $T^* \simeq \Delta/2$  and normal state charge and spin pseudogaps in the cuprates [29]. Many experimental observations have been satisfactorily explained using this particular approach including the in-plane [30, 31] and out-of-plane resistivity [32, 33], magnetic susceptibility [32, 34], tunneling spectroscopy [35], isotope effect [16, 36], upper critical field and specific heat anomaly [37]. ARPES measurements indicate the presence of a pseudogap as well. They also indicate an angular dependent narrow peak and a featureless background. In the polaronic model, the ARPES spectrum can be numerically explained if one considers a charge

transfer Mott-insulator and the single polaron spectral function [38].

Like many other properties, the Hall ratio in high- $T_c$  cuprates shows a non-Fermi-liquid behaviour [39, 40, 41]. A Fermi-liquid approach may describe  $R_H(T)$  for  $T \gg T^*$  only if vertex corrections are included. However, the advocates of this approach [42] admit that it is inappropriate for  $T < T^*$ . On the other hand, the bipolaron model described an enhanced magnitude, doping and  $1/T$  temperature dependence of the Hall ratio [13, 30], but the maximum of  $R_H(T)$  in underdoped cuprates well above  $T_c$  [39, 40, 41] was not addressed. Also, a nonlinear temperature dependence of the *in*-plane resistivity below  $T^*$  remains one of the unsolved problems. In this paper, we give an explanation of these long-standing problems from the standpoint of the bipolaron model.

Thermally excited phonons and (bi)polarons are well decoupled in the strong-coupling regime of electron-phonon interaction [5], so that the conventional Boltzmann kinetics for renormalised carries is applied. Here we use a ‘minimum’ bipolaron model, which includes a singlet bipolaron band and a spin 1/2 polaron band separated by  $T^*$ , and the  $\tau$ -approximation [43] in an electric field  $\mathbf{E} = -\vec{\nabla}\phi$  and in a weak magnetic field  $\mathbf{B} \perp \mathbf{E}$ . The bipolaron and single-polaron non-equilibrium distributions are found as

$$f(\mathbf{k}) = f_0(E) + \tau \frac{\partial f_0}{\partial E} \mathbf{v} \cdot \{\mathbf{F} + \Theta \mathbf{n} \times \mathbf{F}\}, \quad (1)$$

where  $\mathbf{v} = \partial E / \partial \mathbf{k}$ ,  $\mathbf{F} = \vec{\nabla}(\mu - 2e\phi)$  and  $f_0(E) = [y^{-1} \exp(E/T) - 1]^{-1}$  for bipolarons with the energy  $E = k^2/(2m_b)$ , and  $\mathbf{F} = \vec{\nabla}(\mu/2 - e\phi)$  and  $f_0(E) = \{y^{-1/2} \exp[(E + T^*)/T] + 1\}^{-1}$ ,  $E = k^2/(2m_p)$  for thermally excited polarons. Here  $m_b \approx 2m_p$  and  $m_p$  are the bipolaron and polaron masses of *quasitwo*-dimensional carriers,  $y = \exp(\mu/T)$ ,  $\mu$  is the chemical potential,  $\hbar = c = k_B = 1$ , and  $\mathbf{n} = \mathbf{B}/B$  is a unit vector in the direction of the magnetic field. Eq.(1) is used to calculate the electrical resistivity and the Hall ratio as

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

$$R_H = \frac{1 + 2A^2 n_p / n_b}{2e n_b (1 + A n_p / n_b)^2}, \quad (3)$$

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

$$n_b = \frac{m_b T}{2\pi} |\ln(1 - y)|, \quad (4)$$

$$n_p = \frac{m_p T}{\pi} \ln \left[ 1 + y^{1/2} \exp(-T^*/T) \right]. \quad (5)$$

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

$T \lesssim T^*$ , so that

$$y \approx 1 - \exp(-T_0/T), \quad (6)$$

where  $T_0 = \pi(x - n_L)/m_b \approx T_c$  is about the superconducting critical temperature of the (quasi)two-dimensional Bose gas [5]. Because of this reason, experimental  $T_c$  was taken as  $T_0$  for our fits.

Then using Eqs.(2,3) we obtain

$$R_H(T) = R_{H0} \frac{1 + 2A^2 y^{1/2} (T/T_c) \exp(-T^*/T)}{[1 + A(T/T_c) y^{1/2} \exp(-T^*/T)]^2}, \quad (7)$$

where  $R_{H0} = [e(x - n_L)]^{-1}$ . In the following, we assume that the number of localised carriers depends only weakly on temperature in underdoped cuprates because their average ionisation energy is sufficiently large, so that  $R_{H0}$  is temperature independent if  $T \lesssim T^*$ . As proposed in Ref.[30] the scattering rate ( $\propto T^2$ ) is due to inelastic collisions of itinerant carriers with those localised by disorder. Here we also take into account the scattering off optical phonons [43], so that  $\tau^{-1} = aT^2 + b \exp(-\omega/T)$ , if the temperature is low compared with the characteristic phonon energy  $\omega$ . 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 that  $A = (m_b/m_p)^{5/2} \approx 6$  in the Born approximation for any short-range scattering potential. As a result we obtain the in-plane resistivity as

$$\rho(T) = \rho_0 \frac{(T/T_1)^2 + \exp(-\omega/T)}{[1 + A(T/T_c) y^{1/2} \exp(-T^*/T)]}, \quad (8)$$

where  $\rho_0 = b m_b / [2e^2(x - n_L)]$  and  $T_1 = (b/a)^{1/2}$  are temperature independent. Finally, we easily obtain the uniform magnetic susceptibility due to nondegenerate spin 1/2 polarons as [32]

$$\chi(T) = B y^{1/2} \exp(-T^*/T) + \chi_0, \quad (9)$$

where  $B = (\mu_B^2 m_p / \pi)$ , and  $\chi_0$  is the magnetic susceptibility of the parent Mott insulator.

The present model fits the Hall ratio,  $R_H(T)$ , the in-plane resistivity,  $\rho(T)$ , and the magnetic susceptibility  $\chi(T)$  of underdoped  $YBa_2Cu_3O_{7-\delta}$  with a selfconsistent set of parameters (see Fig. 1-2 and the Table). The ratio of polaron and bipolaron mobilities  $A = 7$  used in all fits is close to the above estimate, and  $\chi_0 \approx 1.5 \times 10^{-4} \text{ emu/mole}$  is very close to the susceptibility of a slightly doped insulator [44]. The comprehensive analysis by Mihailovic et al. [29] yields  $T^*$  in the range from 200K to 1000K depending on doping, and  $\omega$  should be about 500K or so from the optical data by Timusk et al. [19].

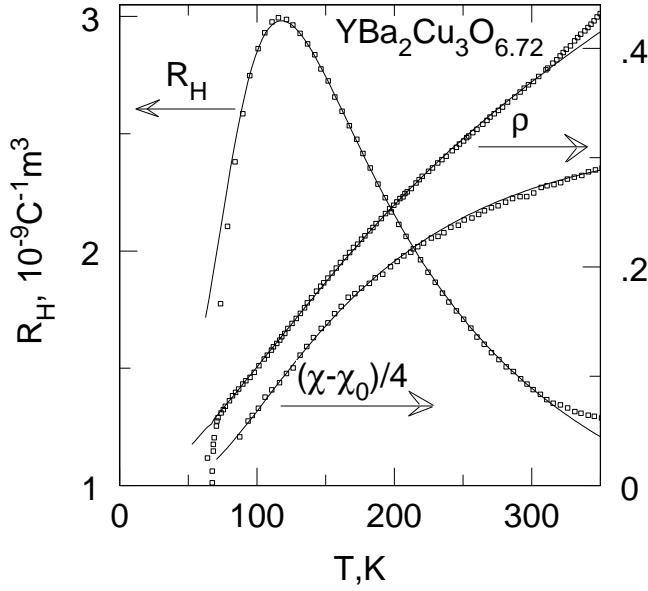


FIG. 1: The Hall ratio and resistivity ( $\rho$ ,  $m\Omega cm$ ) of underdoped  $YBa_2Cu_3O_{6.72}$  fitted by the present theory; also shown the fit of rescaled susceptibility ( $\chi$ ,  $10^{-4} emu/mole$ ) for similarly doped sample,  $\delta = 0.26$ . (See Table for the parameters.)

$\delta$	$T_c$ K	$\rho_0$ $m\Omega cm$	$R_{H0}$ $\frac{10^{-9} m^3}{C}$	$10^4 B$ $\frac{emu}{mole}$	$10^4 \chi_0$ $\frac{emu}{mole}$	$T^*$ K	$\omega$ K	$T_1$ 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		

As shown in Fig.1 and Fig.2, the model describes remarkably well the experimental data with the parameters in this range (Table), in particular the unusual maximum of the Hall ratio, well above  $T_c$  (Fig.1), and the non-linear temperature dependence of the in-plane resistivity. 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. It is also quite remarkable that the characteristic phonon frequency from the resistivity fit (Table) decreases with doping as observed in the neutron scattering experiments and the pseudogap  $T^*$  shows the doping behaviour as observed in other experiments [29]. The temperature dependences of  $R_H(T)$ ,  $\rho(T)$  and  $\chi(T)$  in underdoped  $La_{2-x}Sr_xCuO_4$  and in other underdoped cuprates are very similar to  $YBa_2Cu_3O_{7-\delta}$ , when the temperature

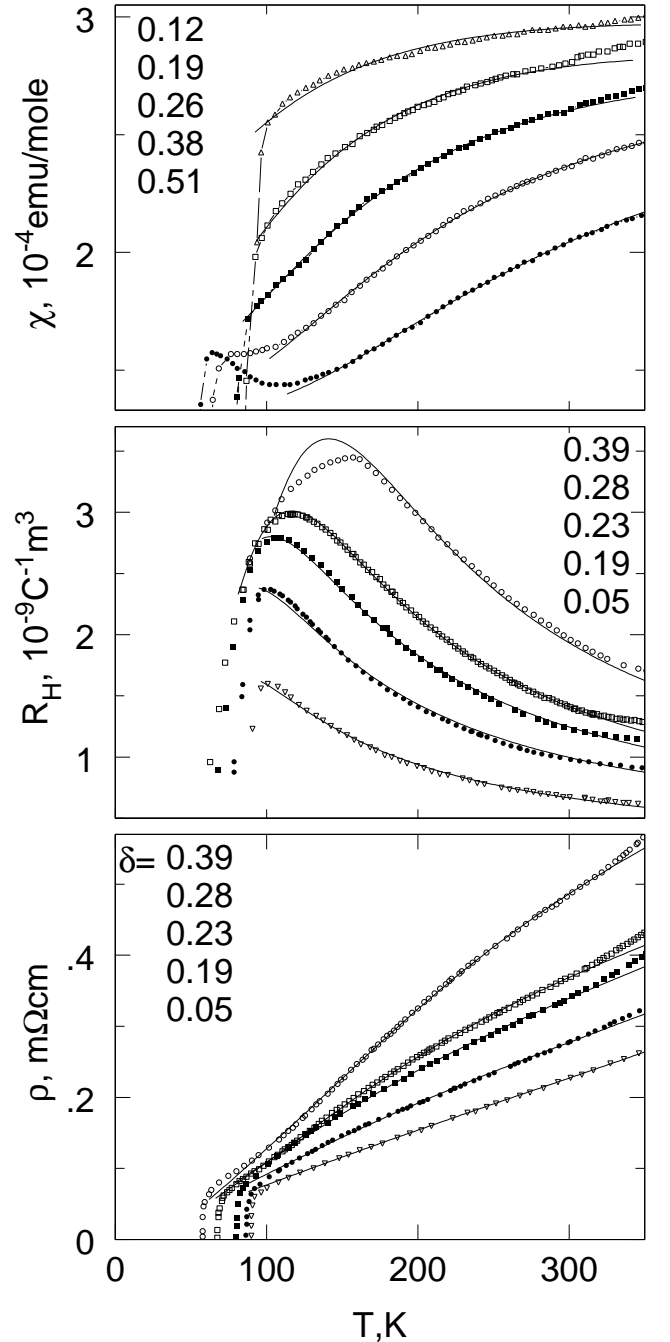


FIG. 2: Uniform magnetic susceptibility,  $\chi(T)$ , Hall ratio,  $R_H(T)$  and resistivity,  $\rho(T)$ , of underdoped  $YBa_2Cu_3O_{7-\delta}$  fitted by the theory; see Table for the parameters.

is rescaled by the pseudogap. It should be noted that adding a triplet bipolaron band [17, 23] could improve the fit further (in particular, above room temperature) however increasing the number of fitting parameters.

To conclude: we applied the multi-polaron approach based on the extension of the BCS theory to the strong-coupling regime [5] to describe peculiar normal state kinetics of underdoped cuprates. The low energy physics in

this regime is that of small bipolarons and thermally excited polarons. Using this approach, we have explained the temperature dependence of the Hall ratio, the in-plane resistivity and the bulk magnetic susceptibility of underdoped cuprates. A direct measurement of the double elementary charge  $2e$  on carriers in the normal state could be decisive. In 1993, Mott and Alexandrov [45] discussed the thermal conductivity  $\kappa$ ; the contribution from the carriers provided by the Wiedemann-Franz ratio depends strongly on the elementary charge as  $\sim (e^*)^{-2}$  and should be significantly suppressed in the case of  $e^* = 2e$ . Recently, a new way to determine the Lorenz number has been applied by Zhang et al. [46], based on the thermal Hall conductivity. As a result, the Lorenz number has been directly measured in  $YBa_2Cu_3O_{6.95}$ . Remark-

ably, the measured value of  $L$  just above  $T_c$  is the same as predicted by the bipolaron model,  $L \approx 0.15L_e$  ( $L_e$  is the conventional Lorenz number). A breakdown of the Wiedemann-Franz law has been also explained in the framework of the bipolaron model [47].

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