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Normal state Nernst effect, semiconducting-like resistivity and diamagnetism of underdoped cuprates

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Semiconducting-like low-temperature in-plane resistivity indicates that there are no remnants of superconductivity above the resistive phase transition at $T > T_c$ in underdoped cuprates. The model with the chemical potential pinned near the mobility edge inside the charge-transfer optical gap describes quantitatively the Nernst effect, thermopower, diamagnetism and the unusual low-temperature resistivity of underdoped cuprates as normal state phenomena above T_c.

PACS numbers: 74.40.+k, 72.15.Jf, 74.72.-h, 74.25.Fy

In the framework of the weak-coupling BCS theory the superconducting state is described by a nonzero Gor'kov anomalous average $\mathcal{F}(\mathbf{r},\mathbf{r}') = \langle\psi_\downarrow(\mathbf{r})\psi_\uparrow(\mathbf{r}')\rangle$, which is zero above the resistive phase transition temperature T_c. When the BCS theory is extended to the strong-coupling regime, electrons are paired into lattice bipolarons, which are real-space pairs dressed by phonons, *both* below and above T_c [1]. The state above T_c is a normal charged Bose-liquid and below T_c phase coherence of the preformed bosons sets in. In this regime $\mathcal{F}(\mathbf{r},\mathbf{r}')$ describes bosons in the Bose-Einstein condensate similar to the Bogoliubov anomalous average of the annihilation operator in the Bose-gas. As in the BCS theory the state above T_c is perfectly "normal" in the sense that the off-diagonal order parameter $\mathcal{F}(\mathbf{r},\mathbf{r}')$ is zero at $T > T_c$.

In disagreement with the weak-coupling BCS and the strong-coupling bipolaron theories a significant fraction of research in the field of superconducting cuprates claims 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 state of cuprates (see [2, 3, 4] and references therein). Refs [2, 5] 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-pairs" or "the phase fluctuation" model [6]. The model is based on the assumption that superfluid density is small compared with the normal carrier density in cuprates. These claims seriously undermine many theoretical and experimental works on superconducting cuprates, which consider the state above T_c as perfectly normal with no off-diagonal order.

However, the vortex scenario is unreconcilable with the extremely sharp resistive transitions at T_c in high-quality samples of cuprates. For example, the in-plane and out-of-plane resistivity of $Bi-2212$, where the anomalous Nernst signal has been measured [2], is perfectly normal above T_c, showing only a few percent positive or negative magnetoresistance [7]. The preformed Cooper-pairs model [6] is clearly incompatible with a great number of thermodynamic, magnetic, kinetic and optical 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 [6] that the superfluid density x is small compared with the normal-state carrier density $1-x$ is also inconsistent with the theorem [8], which proves that the number of supercarriers at $T = 0$K should be the same as the number of normal-state carriers in any clean superfluid.

Faced with these inconsistences we have recently described the unusual Nernst signal in overdoped $La_{1.8}Sr_{0.2}CuO_4$ in a different manner as the normal state phenomenon [9]. Here we extend our description to cuprates with low doping level accounting not only for their anomalous Nernst signal, but also for the thermopower, normal state diamagnetism and a semiconducting-like in-plane low-temperature resistivity as observed in recent [2, 3, 4, 5] and more earlier experiments.

In underdoped cuprates strong on-site repulsive correlations (Hubbard U) are essential in shaping the insulating state of parent compounds. The Mott-Hubbard insulator arises from a potentially metallic half-filled band as a result of the Coulomb blockade of electron tunnelling to neighboring sites [10]. The first band to be doped in cuprates is the oxygen band inside the Hubbard gap. The strong electron-phonon interaction (see for experimental facts Ref. [1]) creates oxygen hole polarons and inter-site bipolarons. Hence the chemical potential remains inside the optical charge-transfer gap, as clearly observed in the tunnelling experiments by Bozovic et al. [11]. Disorder, inevitable with doping, creates localised impurity states for holes separated by a mobility edge from their extended states like in conventional amorphous semiconductors [10, 12]. Then the chemical potential should be found at or near the mobility edge in slightly doped cuprates, if they superconduct.

Naturally carriers, localised below the mobility edge, contribute to the normal-state longitudinal transport together with the itinerant carriers in extended states. On the other hand, the contribution of localised carriers of any statistics to the *transverse* transport is usually small as in many amorphous semiconductors [12]. Importantly, if the localised-carrier 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 at variance with ordinary metals where the familiar "Sondheimer" cancelation [13] makes e_y much smaller than $S\tan\Theta$ because of the electron-hole symmetry near the Fermi level. Such behavior 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 [12] and described theoretically [14].

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

$$e_y = \frac{\sigma_{xx}\alpha_{yx} - \sigma_{yx}\alpha_{xx}}{\sigma_{xx}^2 + \sigma_{xy}^2}, \quad (1)$$

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.(1) can be expressed as $\sigma_{ij}^{ext} + \sigma_{ij}^l$ and $\alpha_{ij}^{ext} + \alpha^l ij$, respectively [9]. 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 $\Theta \ll 1$ [2]) we obtain

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

and

$$e_y \approx \rho(\alpha_{yx}^{ext} + |\alpha_{yx}^l|) - S\tan\Theta, \quad (3)$$

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.(2,3) can account for a low value of $S\tan\Theta$ compared with a large value of e_y in underdoped cuprates [2, 4] because of the sign anomaly. Even in the case when localised carriers 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. Indeed, if the carriers are bosons, their longitudinal conductivity in two-dimensions, $\sigma^{ext} \propto \int_0 dEEdf(E)/dE$ diverges logarithmically when μ in the Bose-Einstein distribution function $f(E) = [\exp((E-\mu)/T) - 1]^{-1}$ goes to zero

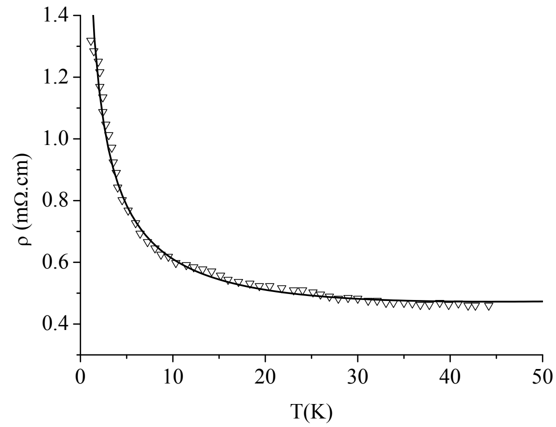

FIG. 1: Normal state in-plane resistivity of underdoped $La_{1.94}$ $Sr_{0.06}CuO_4$ (triangles [3]) as revealed in the field $B = 12$ Tesla and compared with the bipolaron theory, Eq.(6) (solid line).

and the relaxation time τ is a constant (here and further we take $\hbar = c = k_B = 1$). At the same time $\alpha_{xx}^{ext} \propto \int_0 dEE(E-\mu)df(E)/dE$ remains finite, and it could have a magnitude comparable with α_{xx}^l. Statistics of bipolarons effectively changes from Bose to Fermi-like statistics with lowering energy below the mobility edge because of the Coulomb repulsion of bosons in localised states [15]. 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 [16].

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 the Boltzmann theory. Then Eqs.(2,3) yield

$$S\tan\Theta \propto T/\rho \quad (4)$$

and

$$e_y(T,B) \propto (1 - T/T_1)/\rho. \quad (5)$$

According to our earlier suggestion [17] the semiconducting-like dependence of $\rho(T)$ in underdoped cuprates ([3, 4] and references therein) at low temperatures originates from the elastic scattering of non-degenerate itinerant carriers by charged impurities. Different from scenarios based on any kind of metal-insulator transitions, our suggestion is fully compatible with almost temperature-independent carrier density as measured by the temperature-independent Hall voltage at very low temperatures [18]. The relaxation time of *non-degenerate* carriers in two dimensions depends on temperature as $\tau \propto T^{-1/2}$ for scattering by short-range potential wells, and as $T^{1/2}$ for charged impurities. Combining both scattering rates yields

$$\rho = \rho_0[(T/T_2)^{1/2} + (T_2/T)^{1/2}]. \quad (6)$$

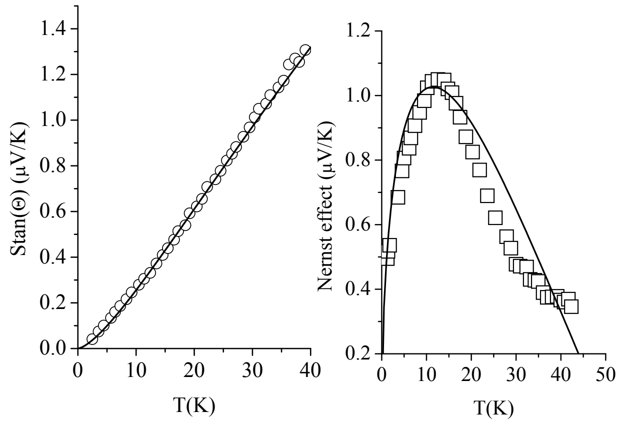

FIG. 2: $S\tan\Theta$ (circles [4]) and the Nernst effect e_y (squares [3]) of underdoped $La_{1.94}$ $Sr_{0.06}CuO_4$ at $B = 12$ Tesla compared with the bipolaron theory, Eqs.(7,8) (solid lines).

Eq.(6) with $\rho_0 = 0.236$ mΩ·cm and $T_2 = 44.6$K fits well the experimental semiconducting-like normal state resistivity of underdoped $La_{1.94}$ $Sr_{0.06}CuO_4$ in the whole low-temperature range from 2K up to 50K, Fig.1, as revealed in the field $B = 12$ Tesla [3, 4]. Importantly the expressions (4,5) for $S\tan\Theta$ and e_y do not depend on a particular scattering mechanism, but only on the experimental temperature dependence of $\rho(T)$. Taking into account the excellent fit of Eq.(6) to the experiment, they can be parameterized as

$$S\tan\Theta = e_0\frac{(T/T_2)^{3/2}}{1+T/T_2}, \tag{7}$$

and

$$e_y(T,B) = e_0\frac{(T_1-T)(T/T_2)^{1/2}}{T_2+T}, \tag{8}$$

where T_1 and e_0 are temperature independent.

In spite of many simplifications, the model describes remarkably well both $S\tan\Theta$ and e_y measured in $La_{1.94}$ $Sr_{0.06}CuO_4$ with a *single* fitting parameter, $T_1 = 50$K using the experimental $\rho(T)$. The constant $e_0 = 2.95$ μV/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 $La_{1.94}$ $Sr_{0.06}CuO_4$ down to 2K, Fig.1, so any residual superconducting order above 2K is clearly ruled out. At the same time the Nernst signal, Fig.2, is remarkably large. The coexistence of the large Nernst signal and a nonmetallic resistivity is in sharp disagreement with the vortex scenario, but is in agreement with our model. Taking into account the field dependence of the conductivity of localised carriers, their contribution to the transverse magnetotransport and the phonon-drug effect (at elevated temperatures) can well describe the magnetic field dependence of the Nernst signal [9] and improve the fit in Fig.2 but at the expense of the increasing number of fitting parameters.

Another experimental observation, which has been linked with the Nernst signal and mobile vortexes above T_c [5], is enhanced diamagnetism. A number of experiments (see, for example, [5, 19, 20, 21, 22, 23] and references therein), including torque magnetometries, showed enhanced diamagnetism near and above T_c, which has been explained as fluctuation diamagnetism in quasi-2D superconducting cuprates (see, for example Ref. [22]). 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 $YBCO$ [20]. 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 in agreement with conventional fluctuations. 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 [21]. Most surprisingly the torque magnetometery [19, 21] uncovered a diamagnetic signal somewhat above T_c which *increases* in magnitude with applied magnetic field.

Here we argue that such behaviors are incompatible with the vortex scenario but can be understood with bipolarons. Accepting the vortex scenario and fitting the magnetization data for $Bi-2212$ with the conventional logarithmic field dependence [5], one obtains surprisingly high upper critical fields $H_{c2} > 120$ Tesla even at temperatures close to T_c, and a very large Ginzburg-Landau parameter, $\kappa = \lambda/\xi > 450$. The in-plane low-temperature magnetic field penetration depth is $\lambda = 200$ nm in optimally doped $Bi-2212$ (see, for example [24]). 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" [6], 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 of a vortex liquid. While $-M(B)$ decreases logarithmically at temperatures well below T_c, the experimental curves [5, 19, 21] clearly show that $-M(B)$ increases with the field at and above T_c , just the opposite of what one could expect in a vortex liquid. This significant departure from the London liquid behavior clearly indicates that the vortex liquid does not appear above the resistive phase transition [19].

Some time ago [25] we proposed that anomalous diamagnetism $M(T,B)$ in cuprates could be the Landau normal-state diamagnetism of preformed bosons. When the strong magnetic field is applied perpendicular to the copper-oxygen plains the quasi-2D bipolaron energy spectrum is quantized, $E = \omega(n+1/2)+2t_c[1-\cos(k_zd)]$, 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. Differentiating the thermodynamic potential one can readily obtain $M(0,B) = -n_b\mu_b$ at low temperatures, $T \ll T_c$, which is the familiar Schafroth's result [26]. Here n_b is the bipolaron density, $\mu_b = e/m_b$ is the "bipolaron" Bohr magneton, and m_b is the bipolaron in-plane mass. The magnetization of charged bosons is field-independent at low temperatures. At high temperatures, $T \gtrsim T_c$ the bipolaron gas is almost classical. The experimental conditions are such that $T \gg \omega$, when T is of the order of T_c or higher, so $M(T,B) \approx -n_b\mu_b\omega/6T$. It is the familiar Landau orbital diamagnetism of non-degenerate carriers. The bipolaron in-plane mass in cuprates is about $m_b \approx 10m_e$ as follows from a number of independent experiments and numerical (QMC) simulations [1]. 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 $M(T,B)$ at and above T_c are about the same as experimentally observed in Refs [5, 21].

The pseudogap temperature T^*, which is half of the bipolaron binding energy in the model, depends on the magnetic field because of spin-splitting of the single-polaron band by the magnetic-field. As a result the number of bipolarons and thermally excited polarons depend on the field and on the temperature. It is easy to show that when the depletion of the bipolaron density with temperature and magnetic field is taken into account, the crossing point in $M(T,B)$ disappears at high magnetic fields as observed. Nevertheless, a quantitative fit to experimental $M(T,B)$ curves 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 [5, 21]. This assumption is inconsistent with a great number of NMR and the Knight shift measurements, and even with the preformed Cooper-pairs model itself where the Pauli spin-susceptibility has been found temperature-dependent. Hence the experimental curves for diamagnetic $M(T,B)$ [5, 21] have to be corrected by taking into account the temperature and field dependencies of the spin paramagnetism at relatively low temperatures.

In summary, we have described the normal state Nernst effect, the thermopower, the diamagnetism and the semiconducting-like in-plane resistivity of underdoped cuprates at low temperatures as the normal-state properties of non-degenerate oxygen holes doped into the Mott-Hubbard charge-transfer insulator with the chemical potential close to the mobility edge. The familiar "sign" (or "$p-n$") anomaly of the Hall conductivity of localised carriers accounts for a small value of $S\tan\Theta_H$ compared with a large value of e_y. The semiconducting-like temperature dependence of the in-plane resistivity at low temperatures originates from the elastic scattering of non-degenerate itinerant carriers by charged impurities, rather than from any localisation. The enhanced diamagnetism at $T > T_c$ is the Landau orbital diamagnetism of non-degenerate carriers above the mobility edge.

I thank V.V. Kabanov and V.N. Zavaritsky for valuable discussions. The work was supported by EPSRC (UK) (grant EP/C518365/1).

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