How normal is the "normal" state of superconducting cuprates?

V. N. Zavaritsky and A. S. Alexandrov
Department of Physics, Loughborough University, Loughborough LE11 3TU, United Kingdom

High magnetic field studies of the cuprate superconductors revealed a non-BCS temperature dependence of the upper critical field $H_{c2}(T)$ determined resistively by several groups. These determinations caused some doubts on the grounds of the contrasting effect of the magnetic field on the in-plane, $\rho_{ab}$, and out-of-plane, $\rho_{c}$ resistances reported for large sample of Bi2212. Here we present careful measurements of both $\rho_{ab}(B)$ and $\rho_{c}(B)$ of tiny Bi2212 crystals in magnetic fields up to 50 Tesla. None of our measurements revealed a situation when on field increase $\rho_{c}$ reaches its maximum while $\rho_{ab}$ remains very small if not zero. The resistive $H_{c2}(T)$ estimated from $\rho_{ab}(B)$ and $\rho_{c}(B)$ are approximately the same. We also present a simple explanation of the unusual Nernst signal in superconducting cuprates as a normal state phenomenon. Our results support any theory of cuprates, which describes the state above the resistive phase transition as perfectly ‘normal’ with a zero off-diagonal order parameter.

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

A pseudogap is believed to be responsible for the non Fermi-liquid normal state of cuprate superconductors. Various microscopic models of the pseudogap proposed are mostly based on the strong electron correlations, and/or on the strong electron-phonon interaction. There is also a phenomenological scenario, where the superconducting order parameter (the Bogoliubov-Gor’kov anomalous average $F(\mathbf{r},\mathbf{r}') = \langle \psi_{\downarrow}(\mathbf{r})\psi_{\uparrow}(\mathbf{r}') \rangle$) does not disappear at the resistive $T_c$ but at much higher (pseudogap) temperature $T^*$. While the scenario was found to be inconsistent with the ‘intrinsic tunnelling’ I-V characteristics, the discovery of the Joule heating origin of the gap-like I-V nonlinearities made the objection irrelevant.

In line with this scenario several authors suggested a radical revision of the magnetic phase diagram of the cuprates with an upper critical field much higher than the resistive $H_{c2}(T)$-line. In particular, Ref. questioned the resistive determination of $H_{c2}(T)$ claiming that while $\rho_{c}$ is a measure of the inter-plane tunnelling, only the in-plane data may represent a true normal state. The main argument in favour of this conclusion came from the radically different field dependencies of $\rho_{c}$ and $\rho_{ab}$ in Fig.2 of Ref. also shown in our Fig.1 (inset B). According to these findings, magnetic field sufficient to recover normal state $\rho_{c}$, leaves in-plane superconductivity virtually unaffected. The difference suggests that Bi2212 crystals do not loose their off-diagonal order in the CuO$_2$ planes even well above $H_{c2}(T)$ determined from the c-axis data. This conclusion is based on one measurement so that it certainly deserves experimental verification, which was not possible until recently because of the lack of reliable $\rho_{ab}(B,T)$ for Bi2212.

Quite similar conclusion followed from the thermomagnetic studies of superconducting cuprates. A large Nernst signal well above $T_c$ has been attributed to a vortex motion in a number of cuprates. As a result the magnetic phase diagram of the cuprates has been revised with the upper critical field $H_{c2}(T)$ curve not ending at $T_{c0}$ but at much higher temperatures. Most surprisingly, Ref. estimated $H_{c2}$ at the zero-field transition temperature of Bi2212, $T_{c0}$, as high as 50-150Tesla.

On the other hand, any phase fluctuation scenario such as of Ref. is difficult to reconcile with the extremely sharp resistive and magnetic transitions at $T_c$ in single crystals of cuprates. Above $T_c$ the uniform magnetic susceptibility is paramagnetic and the resistivity is perfectly ‘normal’, showing only a few percent positive or negative magnetoresistance (MR). Both in-plane and out-of-plane resistive transitions remain sharp in the magnetic field in high quality samples providing a reliable determination of a genuine $H_{c2}(T)$. These and some other observations do not support any superconducting order parameter above $T_c$.

Resolution of these issues, which affect fundamental conclusions about the nature of superconductivity in highly anisotropic layered cuprates, requires further careful experiments and transparent interpretations. Here we present systematic measurements of both in-plane and out-of-plane MRs of small Bi2212 single crystals subjected to magnetic fields, $B \leq 50$ Tesla, $B \perp (ab)$. Our measurements reproduced neither the unusual field dependence of $\rho_{ab}$ nor the contrasting effect of the field as in Ref., which are most probably an experimental artefact. On the contrary, they show that the resistive upper critical fields estimated from the in-plane and out-of-plane data are nearly identical. We also present a simple explanation of the unusual Nernst signal in cuprates as a normal state phenomenon, thus supporting any microscopic theory of cuprates with a zero off-diagonal order parameter above resistive $T_c$.

Reliable measurements of the resistivity tensor require defect-free samples. This is of prime importance for the in-plane MR because even unit-cell scale defects will result in a significant out-of-plane contribution owing to the extreme anisotropy of Bi2212. Because of this reason much attention has been paid to the sample preparation. We studied $\rho_{c}$ and $\rho_{ab}$ of the same high quality, optically and slightly underdoped Bi2212 crystals, $T_{c0} \approx 87-92K$. Differently from Ref. small crystals were pre-
pared in order to reduce eddy currents and the forces acting on the sample during the pulse. We measured $\rho_c$ on samples with in-plane dimensions from $\sim 30 \times 30 \mu m^2$ to $\sim 80 \times 80 \mu m^2$ while $\rho_{ab}$ was studied on a longer crystals, from $\sim 300 \times 11 \mu m^2$ to $\sim 780 \times 22 \mu m^2$. Metallic type of zero-field $\rho_{ab}(T)$ and the sign of its normal state MR [14] indicate vanishing out-of-plane contribution. All samples selected for $\rho_c$ and $\rho_{ab}$ measurements were cut from the same parent crystals of $1-3 \mu m$ thickness. The absence of hysteresis in the $\rho(B)$ data obtained on the rising and falling sides of the pulse and the consistency of $\rho(B)$ taken at the same temperature in pulses of different $B_{max}$ exclude any measurable heating effects. Ohmic response is confirmed by a consistency of the dc $\rho(B)$ measured at identical conditions with different currents, 10-1000 A/cm$^2$ for $\rho_{ab}$ and 0.1-20 A/cm$^2$ for $\rho_c$.

Fig.1 shows the typical $\rho_c(B)$ and $\rho_{ab}(B)$ taken below $T_{c0}$ of a Bi2212 single crystal. The low-field portions of the curves correspond to the resistance driven by vortex dynamics. Here a non-linear $\rho(B)$ dependence is followed by a regime, where a linear dependence fits the experimental observations rather well, Fig.1. It is natural to attribute the high field portions of the curves in Fig.1 (assumed to be above $H_{c2}$) to a normal state. Then, the c-axis high-field MR appears to be negative and quasi-linear in B in a wide temperature range both above and below $T_{c0}$. Contrary to $\rho_c(B)$, the normal state in-plane MR is positive (see [14] and references therein for an explanation). The reasonable concordance of $H_{c2}(T)$ estimates from $\rho_c(B)$ and $\rho_{ab}(B)$ (Inset A to Fig.1) favours our association of the resistive $H_{c2}$ with the upper critical field especially given the apparently different mechanisms responsible for $\rho_{ab}$ and $\rho_c$ [14].

Our conclusion is based on the results obtained during few hundred measurements performed on three pairs of crystals. None of those revealed a situation when on field increase $\rho_c$ reaches its maximum while $\rho_{ab}$ remains very small if not zero as reported in Ref. [5] (see inset B in Fig.1). Since the authors of Ref. [5] measured $\rho_{ab}(B)$ by means of contacts situated on the same face of the crystal, their curve could not represent the true $\rho_{ab}$. Moreover, neither the current redistribution (discussed in [16] for homogeneous medium) nor imperfections of their huge crystals were accounted for in Ref. [5].

The resistive upper critical field, which is about the same from in- and out-of-plane data for Bi2212, shows a non-BCS temperature dependence, Fig.1. These results are supported by the independent studies of $\rho_c$ and $\rho_{ab}$ in a single-layer cuprate Bi2201 with the similar anisotropy. If we apply the routine procedure for resistive $H_{c2}(T)$ evaluation [7], the very similar values of $H_{c2}(T)$ are obtained from $\rho_{ab}$ and $\rho_c$ measured on the same crystals [17] and films [18] (see dashed and solid lines in Fig.2). Remarkably, $H_{c2}(T)$ obtained are compatible with the Bose-Einstein condensation field of preformed charged bosons [19], and also with some other models [16, 20].

Finally we address the origin of the large Nernst voltage measured above $T_{c0}$ in superconducting cuprates (see
It is expressed in terms of the kinetic coefficients $\sigma_{ij}$ and $\alpha_{ij}$, as

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

where the current density per spin is given by $j_i = \sigma_{ij} E_j + \alpha_{ij} \nabla_j T$. Carriers in doped semiconductors and disordered metals occupy states localised by disorder and itinerant Bloch-like states. Both types of carriers contribute to the transport properties, if the chemical potential $\mu$ (or the Fermi level) is close to the energy, where the lowest itinerant state appears (i.e. to the mobility edge). Superconducting cuprates are among such poor conductors and their superconductivity appears as a result of doping, which inevitably creates disorder. Indeed, there is strong experimental evidence for the coexistence of itinerant and localised carriers in cuprates in a wide range of doping.

When the chemical potential is near the mobility edge, and the effective mass approximation is applied, there is no Nernst signal from itinerant carriers alone, because of a so-called Sondheimer cancellation \cite{24}. However, when the localised carriers contribute to the longitudinal transport, $\sigma_{xx}$ and $\alpha_{xx}$ in Eq.(1) should be replaced by $\sigma_{xx} + \sigma_l$ and $\alpha_{xx} + \alpha_l$, respectively. Since the Hall mobility of localised carriers is often much smaller than their drift mobility \cite{22}, there is no need to add their contributions to the transverse kinetic coefficients. One can also neglect field orbital effects because the Hall angle remains very small for the experimentally accessible fields in poor conductors, $\Theta_H < 1\ [4, 8], $ so that

$$ e_y(T, B) = \frac{\sigma_{xx} \alpha_{yx} - \sigma_{yx} \alpha_{xx}}{(\sigma_{xx} + \sigma_l)^2}. \quad (2) $$

The conductivity of itinerant carriers $\sigma_{xx}$ in the superconducting cuprates dominates over that of localised carriers \cite{22}, $\sigma_{xx} \gg \sigma_l$, which simplifies Eq.(2) as

$$ e_y = \frac{k_B}{e} r \theta_0, \quad (3) $$

where $\rho = 1/[(2s+1)\sigma_{xx}]$ is the resistivity, $s$ is the carrier spin, and $r$ is a constant,

$$ r = \frac{e|\alpha_l|}{k_B \sigma_l} + \frac{\int_0^\infty dE E (E - \mu) \partial f(E)/\partial E}{k_B T \int_0^\infty dE E \partial f(E)/\partial E}. \quad (4) $$

Here $N(E)$ is the density of states (DOS) near the band edge ($E = 0$), and $\mu$ is taken with respect to the edge. The ratio $e|\alpha_l|/k_B \sigma_l$ is a number of the order of one. For example, $e|\alpha_l|/k_B \sigma_l \approx 2.4$, if $\mu = 0$ and the conductivity index $\nu = 1/2$. Calculating the integrals in Eq.(4) yields $r \approx 14.3$ for fermions ($s = 1/2$), and $r \approx 2.4$ for bosons ($s = 0$) with the two-dimensional DOS, $N(E) = constant$.

The Nernst signal, Eq.(3), is positive, and its maximum value $e_{yx}^{max} \approx (k_B/e)r\Theta$ is about 5 to 10 $\mu V/K$ with $\Theta = 10^{-2}$ and $\sigma_l \approx \sigma_{xx}$, as observed \cite{8, 34}. Actually, the magnetic and temperature dependencies of the unusual Nernst effect in cuprates are described by Eq.(3) quantitatively, if $\sigma_l$ obeys the Mott’s law,

$$ \sigma_l = \sigma_0 \exp\left[-(T_0/T)^x\right], \quad (5) $$

where $\sigma_0$ is about a constant. The exponent $x$ depends on the type of localised wavefunctions and variation of DOS, $N_l$ below the mobility edge \cite{24, 27, 28}. In two dimensions one has $x = 1/3$ and $T_0 \approx 8\alpha^2/(k_B N_l)$, where $N_l$ is at the Fermi level.

In sufficiently strong magnetic field\cite{20} the radius of the ‘impurity’ wave function $\alpha^{-1}$ is about the magnetic length, $\alpha \approx (eB)^{1/2}$. If the relaxation time of itinerant carriers is due to the particle-particle collisions, the Hall angle depends on temperature as $\Theta_H \propto 1/T^2$, and the resistivity is linear, since the density of itinerant carriers is linear in temperature, both for fermionic and/or bosonic carriers \cite{34}. Hence, the model explains the temperature dependence of the normal-state Hall angle and resistivity in cuprates at sufficiently high temperatures. Then using Eq.(3) and Eq.(5) the Nernst signal is given by

$$ e_y/B\rho = a(T) \exp\left[-b(B/T)^{1/3}\right], \quad (6) $$

where $a(T) \propto T^{-2}$ and $b = 2(e/(k_B N_l))^{1/3}$ is a constant. The phonon drag effect should be taken into account at low temperatures in any realistic model. Then $a(T)$ in Eq.(6) is found to be enhanced by this effect as $a(T) \propto T^{-6}$ \cite{21}. The theoretical field dependence of $e_y/(B\rho)$, Eq.(6), is in excellent quantitative agreement with the experiment, as shown in Fig.3 for $b = 7.32 (K/\text{Tesla})^{1/3}$. The corresponding temperature dependence of $a(T)$ follows closely $T^{-6}$, inset to Fig.3. The density of impurity states $N_l = 8e/(b^3k_B)$ is about

\[\text{FIG. 3: Eq.(6) fits the experimental signal (symbols) in La}_{1.8}\text{Sr}_{0.2}\text{CuO}_4 with } b = 7.32(K/\text{Tesla})^{1/3}.\text{ Inset shows } a(T) \text{ obtained from the fit (dots) together with } a \propto T^{-6} (\text{line}).\]
$4 \times 10^{13}$ cm$^{-2}$(eV)$^{-1}$, which corresponds to the number of impurities $N_{im} \lesssim 10^{21}$ cm$^{-3}$, as it should be.

If carriers are fermions, then the product $S\tan \Theta_H$ of the thermopower $S$ and of the Hall angle should be larger or of the same order as $e_y$, because their ratio is proportional to $\sigma_{xx}/\sigma_l \gg 1$ in our model. Although it is the case in many cuprates, a noticeable suppression of $S\tan \Theta_H$, as compared with $e_y$, was reported to occur close to $T_c$ in strongly underdoped LSCO and in a number of Bi2201 crystals. These observation could be generally understood if we take into account that underdoped cuprates are strongly correlated systems, so that a substantial part of carriers is (most probably) preformed bosonic pairs. The second term in Eq.(4) vanishes for (quasi)two dimensional itinerant bosons, because the denominator diverges logarithmically suppressed. It can be almost cancelled by the opposite sign contribution of the localised carriers, even if $\sigma_{xx} \gtrsim \sigma_l$. When it happens, the Nernst signal is given by $e_y = \rho \alpha_{xy}$, where $\alpha_{xy} \propto \tau^2$. Differently from that of fermions, the relaxation time of bosons is enhanced critically near the Bose-Einstein condensation temperature, $T_c(B)$, $\tau \propto (T - T_c(B))^{-1/2}$, as in atomic gases. Providing $S\tan \Theta_H \ll e_y$, this critical enhancement of the relaxation time describes well the temperature dependence of $e_y$ in Bi2201 and in strongly underdoped LSCO close to $T_c(B)$.

To conclude, we have shown that the understanding of the reliable experimental data does not require radical revision of the magnetic phase diagram of cuprates. Our studies of $\rho_{ab}(B)$ and $\rho_{c}(B)$ on the same Bi2212 crystals as well as the normal state model of the Nernst signal in cuprates support any microscopic theory, which describes the state above the resistive and magnetic phase transition as perfectly ‘normal’ with $F(r, r') = 0$. The carriers could be normal-state fermions, as in any BCS-like theory of cuprates, or normal-state charged bosons, as in the bipolaron theory, or a mixture of both. We believe that the resistive determinations provide the genuine $H_{c2}(T)$, and the anomalous Nernst effect in high-$T_c$ cuprates is a normal state phenomenon.

This work was supported by the Leverhulme Trust (grant F/00261/H).

[1] for a review see T. Timusk and B. Statt, Rep. Prog. Phys. 62, 61 (1999).
[2] A.S. Alexandrov, Theory of superconductivity (IOP Publishing, Bristol-Philadelphia, 2003).
[3] V.J. Emery, S.A. Kivelson, Nature, 374, 434 (1995).
[4] V.N. Zavaritsky, Physica C 404, 440 (2004) and references therein.
[5] N.Morozov, L.Krusin-Elbaum, T.Shibauchi, L.N.Bulaevskii, M.P.Maley, Yu.I.Latyshev, T.Yamashita, Phys.Rev. Lett. 84,1784 (2000).
[6] Y.Wang, S.Ono, Y.Onose, G.Gu, Y.Ando, Y.Tokura, S.Uchida, N.P.Ong, Science 299, 86 (2003).
[7] A.S. Alexandrov, V.N. Zavaritsky, W.Y. Liang, P.L. Nevsyky, Phys. Rev. Lett. 76 983 (1996).
[8] V.N. Zavaritsky, M. Springford, JETP Lett. 68, 448 (1998); V.N.Zavaritsky, JETP Lett. 71, 80 (2000).
[9] Y.Wang, N.P.Ong, Z.A.Xu, T.Kakeshita, S.Uchida, D.A.Bonn, R.Liang, W.N.Hardy, Phys. Rev. Lett. 88, 257003 (2002).
[10] A.P. Mackenzie, S.R.Julian, G.G.Lonzarich, A.Carrington, S.D.Hughes, R.S.Liu, D.C.Sinclair, Phys. Rev. Lett. 71, 1238 (1993).
[11] M.S. Ososky, R.J. Soulen, S.A. Wolf, J.M. Broto, H. Rakoto, J.C. Ousset, G.Coffe, S.Askenzay, P.Pari, I. Bozovic, J.N. Eckstein, G.F. Virshup, Phys. Rev. Lett. 71, 2315 (1993); ibid 72, 3292 (1994).
[12] D.D. Lawrie, J.P. Franck, J.R. Beamish, E.B. Molz, W.M. Chen, M.J. Graff, J. Low Temp. Phys. 107, 491 (1997).
[13] J.W. Loram, K.A. Mirza, J.R. Cooper, W.Y. Liang, J.M. Wade, J. Superconductivity, 7, 243 (1994).
[14] V.N. Zavaritsky, J.Vanacen, V.V.Moshchalkov, A.S. Alexandrov, Physica C 404, 444 (2004).
[15] A.S. Alexandrov, Phys. Rev. B 48, 10571 (1993).