

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

A. S. Alexandrov

*Department of Physics, Loughborough University,  
Loughborough, United Kingdom*

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  $T_c$ .

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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  $T = 0K$  should be the same as the number of normal-state carriers in any clean superfluid.

Faced with these inconsistencies 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  $\Theta$ . This unusual "symmetry breaking" is at variance with ordinary metals where the familiar "Sondheimer" cancellation [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  $\sigma_{ij}$  and  $\alpha_{ij}$  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  $\mu$  is at the mobility edge, the localised carriers contribute to the transport, so  $\sigma_{ij}$  and  $\alpha_{ij}$  in Eq.(1) can be expressed as  $\sigma_{ij}^{ext} + \sigma_{ij}^l$  and  $\alpha_{ij}^{ext} + \alpha_{ij}^l$ , respectively [9]. Since the Hall mobility of carriers localised below  $\mu$ ,  $\sigma_{yx}^l$ , has the sign opposite to that of carries in the extended states above  $\mu$ ,  $\sigma_{yx}^{ext}$ , the sign of the off-diagonal Peltier conductivity  $\alpha_{yx}^l$  should be the same as the sign of  $\alpha_{yx}^{ext}$ . 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,  $\sigma_{xx}^{ext} \propto \int_0^\infty dE E df(E)/dE$  diverges logarithmically when  $\mu$  in the Bose-Einstein distribution function  $f(E) = [\exp((E - \mu)/T) - 1]^{-1}$  goes to zero

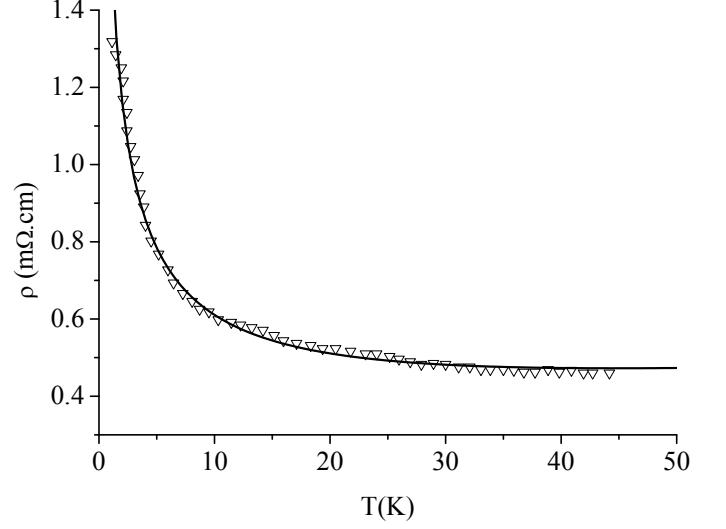


FIG. 1: Normal state in-plane resistivity of underdoped  $\text{La}_{1.94}\text{Sr}_{0.06}\text{CuO}_4$  (triangles [3]) as revealed in the field  $B = 12$  Tesla and compared with the bipolaron theory, Eq.(6) (solid line).

and the relaxation time  $\tau$  is a constant (here and further we take  $\hbar = c = k_B = 1$ ). At the same time  $\alpha_{xx}^{ext} \propto \int_0^\infty dE E(E - \mu) df(E)/dE$  remains finite, and it could have a magnitude comparable with  $\alpha_{xx}^l$ . 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  $\rho$ ,  $\Theta$  and  $\alpha_{xy}$ , 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. The relaxation time of *non-degenerate* carriers depends on temperature as  $\tau \propto T^{-1/2}$  for scattering by short-range deep potential wells, and as  $T^{1/2}$  for scattering by very shallow wells as discussed in Ref. [17]. Combining both scattering rates yields

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

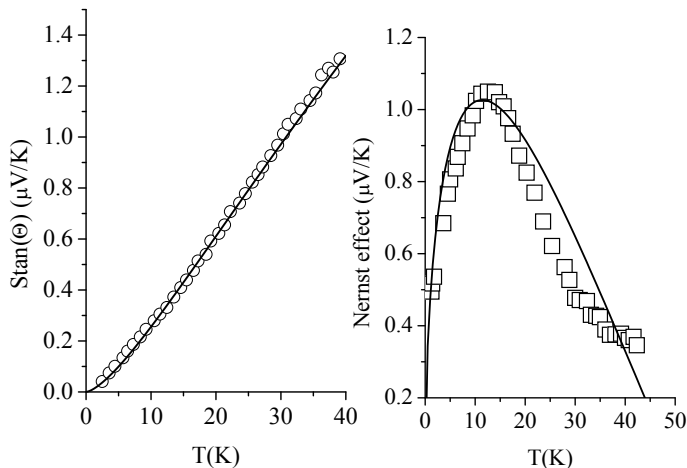


FIG. 2:  $S \tan \Theta$  (circles [4]) and the Nernst effect  $e_y$  (squares [3]) of underdoped  $\text{La}_{1.94}\text{Sr}_{0.06}\text{CuO}_4$  at  $B = 12$  Tesla compared with the bipolaron theory, Eqs.(7,8) (solid lines).

Eq.(6) with  $\rho_0 = 0.236 \text{ m}\Omega\text{-cm}$  and  $T_2 = 44.6\text{K}$  fits well the experimental semiconducting-like normal state resistivity of underdoped  $\text{La}_{1.94}\text{Sr}_{0.06}\text{CuO}_4$  in the whole low-temperature range from 2K up to 50K, Fig.1, as revealed in the field  $B = 12$  Tesla [3, 4]. Another high quality fit can be obtained combining the Brooks-Herring formula for the 3D scattering off screened charged impurities, as proposed in Ref.[18] for almost undoped *LSCO*, or the Coulomb scattering in 2D ( $\tau \propto T$ ) and a temperature independent scattering rate off neutral impurities with the carrier exchange [19] similar to the scattering of slow electrons by hydrogen atoms in three dimensions. Hence the scale  $T_2$ , which determines the crossover toward an insulating behavior, depends on the relative strength of two scattering mechanisms. Importantly the expressions (4,5) for  $S \tan \Theta$  and  $e_y$  do not depend on particular scattering mechanisms, but only on the experimental  $\rho(T)$ . Taking into account the excellent fit of Eq.(6) to the experiment, these expressions can be parameterized as

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

and

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

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  $\text{La}_{1.94}\text{Sr}_{0.06}\text{CuO}_4$  with a *single* fitting parameter,  $T_1 = 50\text{K}$  using the experimental  $\rho(T)$ . The constant  $e_0 = 2.95 \mu\text{V/K}$  scales the magnitudes of  $S \tan \Theta$  and  $e_y$ . The magnetic field  $B = 12$  Tesla destroys the superconducting state of the low-doped  $\text{La}_{1.94}\text{Sr}_{0.06}\text{CuO}_4$  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-drag 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 vortices above  $T_c$  [5], is enhanced diamagnetism. A number of experiments (see, for example, [5, 20, 21, 22, 23, 24] 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. [23]). 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* [21]. 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 [22]. Most surprisingly the torque magnetometry [20, 22] 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  $\lambda \approx 200 \text{ nm}$  in optimally doped *Bi-2212* (see, for example [25]). Hence the zero temperature coherence length  $\xi$  turns out to be about the lattice constant,  $\xi = 0.45\text{nm}$ , 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, 20, 22] 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 [20].

Some time ago [26] 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 planes the quasi-2D bipolaron energy spectrum is quantized,  $E = \omega(n+1/2) + 2t_c[1 - \cos(k_z d)]$ , where  $\omega = 2eB/m_b$ ,  $n = 0, 1, 2, \dots$ , 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 [27]. 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  $M(0, B) \approx 2000 \text{ A/m}$  with the bipolaron density  $n_b = 10^{21} \text{ cm}^{-3}$ . 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, 22].

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. Also the singlet-triplet exchange energy of inter-site bipolarons depends on the field for the same reason. As a result the number of singlet bipolarons and thermally excited triplet pairs and single polarons depend on the field and on the temperature. 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, and the normal state magnetization of singlet bipolarons fits experimental  $M(T, B)$  curves [5] in the whole normal state and critical regions [28].

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 normal state orbital diamagnetism of bipolarons.

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- [1] A.S. Alexandrov, *Theory of Superconductivity* (IoP Publishing, Bristol and Philadelphia, 2003).
  - [2] Z.A. Xu, N.P. Ong, Y. Wang, T. Kakeshita, and S. Uchida, *Nature (London)* **406**, 486 (2000); N.P. Ong and Y. Wang, *Physica C* **408**, 11 (2004).
  - [3] C. Capan, K. Behnia, J. Hinderer, A.G.M. Jansen, W. Lang, C. Marcenat, C. Marin, and J. Flouquet, *Phys. Rev. Lett.* **88**, 056601 (2002).
  - [4] C. Capan and K. Behnia, *cond-mat/0501288*.
  - [5] Y. Wang, L. Li, M.J. Naughton, G.D. Gu, S. Uchida, and N.P. Ong, *cond-mat/0503190*.
  - [6] V.J. Emery and S.A. Kivelson, *Nature (London)*, **374**, 434 (1995).
  - [7] V.N. Zavaritsky and A.S. Alexandrov, *Phys. Rev. B* **71**, 012502 (2005).
  - [8] A.J. Leggett, *Physica Fennica* **8** 125 (1973); *J. Stat. Phys.* **93**, 927 (1998).
  - [9] A.S. Alexandrov and V.N. Zavaritsky, *Phys. Rev. Lett.* **93**, 217002 (2004).
  - [10] N.F. Mott, *Metal-Insulator Transitions, 2nd ed* (Taylor&Francis, London, 1990).
  - [11] I. Bozovic, G. Logvenov, M. A. J. Verhoeven, P. Caputo, E. Goldobin, and T. H. Geballe, *Nature (London)* **422**, 873 (2003).
  - [12] S.R. Elliot, *Physics of amorphous materials*, pp. 222-225 (Longman, New York, 1983).
  - [13] E.H. Sondheimer, *Proc. Roy. Soc.* **193**, 484 (1948).
  - [14] L. Friedman, *J. Non-Cryst. Sol.* **6**, 329 (1971).
  - [15] A.S. Alexandrov and R.T. Giles, *J. Phys.: Cond. Matt.* **9**, 9921 (1997).
  - [16] M. Cutler and N.F. Mott, *Phys. Rev.* **181**, 1336 (1969).
  - [17] A.S. Alexandrov, *Phys. Lett. A* **236**, 132 (1997).
  - [18] C.Y. Chen, E.C. Branlund, C.S. Bae, K. Yang, M.A. Kastner, A. Cassanho, and R.J. Birgeneau, *Phys. Rev. B* **51**, 3671 (1995).
  - [19] C. Erginsoy, *Phys. Rev.* **79**, 1013 (1950).
  - [20] C. Bergemann, A.W. Tyler, A.P. Mackenzie, J. R. Cooper, S.R. Julian, and D.E. Farrell, *Phys. Rev. B* **57**, 14387 (1998).
  - [21] A. Junod, J.-Y. Genouda, G. Triscone, and T. Schneider, *Physica C* **294**, 115 (1998).
  - [22] M. J. Naughton, *Phys. Rev. B* **61**, 1605 (2000).
  - [23] J. Hofer, T. Schneider, J.M. Singer, M. Willemin, H. Keller, T. Sasagawa, K. Kishio, K. Conder, and J. Karpinski, *Phys. Rev. B* **62**, 631 (2000).
  - [24] I. Iguchi, A. Sugimoto, and H. Sato, *J. Low Temp. Phys.* **131**, 451 (2003).
  - [25] J.L. Tallon, J.R. Cooper, S.H. Naqib, and J.W. Loram, *cond-mat/0410568*.
  - [26] C.J. Dent, A.S. Alexandrov, and V.V. Kabanov, *Physica*

- C**341-348**, 153 (2000).
- [27] M.R. Schafroth, Phys. Rev. **100**, 463 (1955).
- [28] A.S. Alexandrov, cond-mat/0511108.