Normal state diamagnetism of charged bosons in cuprate superconductors

A. S. Alexandrov

Department of Physics,

Loughborough University,

Loughborough LE11 3TU, United Kingdom

Abstract

Normal state orbital diamagnetism of charged bosons quantitatively accounts for recent high-resolution magnetometery results near and above the resistive critical temperature T_c of superconducting cuprates. Our parameter-free descriptions of normal state diamagnetism, T_c, upper critical fields and specific heat anomalies unambiguously support the 3D Bose-Einstein condensation of preformed real-space pairs with zero off-diagonal order parameter above T_c at variance with phase fluctuation scenarios of cuprates.

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

arXiv:cond-mat/0511108 v2   6 Nov 2005

A possibility of real-space pairing, as opposed to the Cooper pairing, has been the subject of many discussions, particularly heated over the last 20 years after the discovery of high temperature superconductivity in cuprates [1]. The first proposal for high temperature superconductivity, made by Ogg Jr in 1946 [2], already involved real-space pairing of individual electrons into bosonic molecules with zero total spin. This idea was further developed as a natural explanation of conventional superconductivity by Schafroth and Butler and Blatt [3]. However, with one or two exceptions, the Ogg-Schafroth picture was condemned and practically forgotten because it neither accounted quantitatively for the critical behavior of conventional (i.e. low T_c) superconductors, nor did it explain the microscopic nature of attractive forces which could overcome the Coulomb repulsion between two electrons constituting a pair. The failure of the 'bosonic' picture of individual electron pairs became fully transparent when Bardeen, Cooper and Schrieffer [4] proposed that two electrons in a superconductor were indeed correlated, but on a very large distance of about 10^3 times of the average inter-electron spacing.

Highly successful for low-Tc metals and alloys the BCS theory has led many researchers to believe that novel high-temperature superconductors should also be "BCS-like". However, the Ogg-Schafroth and the BCS descriptions are actually two opposite extremes of the same electron-phonon interaction. Indeed by extending the BCS theory towards the strong interaction between electrons and ion vibrations, a charged Bose gas (CBG) of tightly bound electron pairs surrounded by lattice deformations (i.e. of *small bipolarons*) was predicted by us [5] with a further prediction that high T_c should exist in the crossover region of the electron-lattice interaction strength from the BCS-like to bipolaronic superconductivity [6]. Experimental evidence for an exceptionally strong electron-phonon interaction in novel superconductors [7, 8, 9, 10] is so overwhelming that bipolaronic CBG [11] could be a feasible alternative to BCS-like scenarios of cuprates. Nevertheless, some authors [12] have dismissed any real-space pairing, advocating a *collective* pairing (i.e *Cooper* pairs in the momentum space) at some high temperature T^* which are phase coherent below a lower temperature $T_c < T^*$. Ref.[12] has argued that the superconducting transition in cuprates is an almost two-dimensional Kosterlitz-Thouless (KT) transition, where a vortex liquid exists above T_c different from the BCS theory and its strong-coupling bipolaronic extension with a perfectly "normal" state without any off-diagonal order.

So far there has been no decisive conclusion on the origin of anomalous normal state

of cuprates. Some normal state properties have been satisfactorily interpreted within the Fermi-liquid approach, while many others have been understood with preformed real-space [11] or Cooper [12] pairs, in particular on the underdoped side of the phase diagram. Moreover preformed real-space pairs could coexist with the Fermi-liquid, which effectively hides them in the normal state kinetics. Any direct evidence in favor of either scenario is highly desirable. If real-space pairs indeed exist in superconducting cuprates, then their superconducting state should be a three-dimensional (3D) Bose-Einstein condensate (BEC) of CBG. Its critical behavior [11] is rather different from any "universal" criticality like mean-field BCS [4], 3D "XY" or KT [12] transitions.

Here I show that high-resolution magnetometry in the critical and normal regions provides unambiguous evidence for real-space pairing in cuprates.

A number of experiments (see, for example, [13, 14, 15, 16, 17, 18] and references therein), including torque magnetometery, showed enhanced diamagnetism above T_c. Originally it was explained as the conventional fluctuation diamagnetism in quasi-2D BCS superconductors (see, for example Ref. [16]). 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 $YBa_2Cu_3O_{7-\delta}$ [14]. The dependence of magnetization (or $M/B^{1/2}$) on the magnetic field was shown to vanish at some characteristic temperature below T_c. Importantly more recent data taken in high magnetic fields (up to 30 Tesla) show that the crossing point, anticipated for low-dimensional superconductors and associated with conventional superconducting fluctuations, does not explicitly exist in magnetic fields above 5 Tesla [15, 18].

Ref.[18] has linked the enhanced normal state diamagnetism with mobile vortexes well above T_c where conventional fluctuations should be negligible. Surprisingly the same torque magnetometery [13, 15, 18] uncovered that the diamagnetic signal above T_c *increases* in magnitude with applied magnetic field, B. Such magnetic field dependence of magnetisation $M(T, B)$ is entirely inconsistent with what one expects from vortex liquid. While $-M(B)$ decreases logarithmically at temperatures well below T_c, the experimental curves clearly show that $-M(B)$ increases with the field at and above T_c , just opposite to what one could expect in a conventional vortex liquid. These significant departures from the London liquid behavior indicates that vortex liquid does not appear above the resistive phase transition (see also Ref.[13]). Also accepting the vortex scenario and fitting the magnetization data in Bi-2212
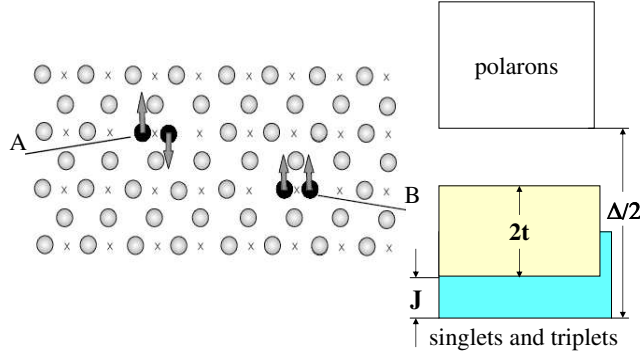
3

FIG. 1: Bipolaron picture of high temperature superconductors. A corresponds to a singlet oxygen intersite bipolaron, B is a triplet intersite bipolaron.

with the conventional logarithmic field dependence [18], one obtains surprisingly high upper critical fields $H_{c2} > 120$ Tesla and a very large Ginzburg-Landau parameter, $\kappa = \lambda_H/\xi > 450$ even at temperatures close to T_c. The in-plane low-temperature magnetic field penetration depth is $\lambda_H \approx 220$ nm in optimally doped Bi-2212 (see, for example [23]). Hence the zero temperature coherence length ξ turns out to be about the lattice constant, $\xi \lesssim 0.5$nm. Such a small coherence length is perfectly compatible with direct STM measurements of the individual vortex cores in Bi-2212 [19] and with the size of the vortex core in CBG [11]. However it rules out the "preformed Cooper pairs" [12], since the pairs are virtually not overlapped at any size of the Fermi surface.

Here I calculate the magnetization, $M(T,B)$, of anisotropic CBG on a lattice, and compare the result with diamagnetism of cuprates recently measured in Ref. [18]. A low-energy structure of cuprates in the bipolaron model is shown in Fig.1, where oxygen holes are bound into real-space intersite singlets (A) and triplets (B) separated by an exchange energy J [20], which is estimated as a few tens or hundreds Kelvin depending on doping in agreement with experimental charge and spin pseudogaps in cuprates [21]. Bipolarons are almost ideal charged bosons, because their Coulomb repulsion is strongly suppressed by a large lattice dielectric constant. When the magnetic field is applied perpendicular to

copper-oxygen plains the quasi-2D bipolaron energy spectrum is quantized as

$$E_\alpha = \omega(n + 1/2) + 2t_c[1 - \cos(K_z d)], \tag{1}$$

where α comprises $n = 0, 1, 2, ...$ and in-plane K_x and out-of-plane K_z center-of-mass quasi-momenta, $\omega = 2\hbar eB/\sqrt{m_x m_y}$, t_c and d are the hopping integral and the lattice period perpendicular to the planes. The spectrum consists of two degenerate brunches, the so-called "x" and "y" bipolarons [11], with anisotropic in-plane bipolaron masses $m_x \equiv m$ and $m_y \approx 4m$. Expanding the Bose-Einstein distribution function in powers of $exp[(\mu - E)/k_B T]$ with the negative chemical potential μ one can after summation over n readily obtain the boson density

$$n_b = \frac{2eB}{\pi \hbar d} \sum_{r=1}^{\infty} I_0(2t_c r/k_B T) \frac{\exp[(\tilde{\mu} - 2t_c)r/k_B T]}{1 - \exp(-\omega r/k_B T)}, \tag{2}$$

and the magnetization, $M(T, B) = -k_B T \partial/\partial B \sum_\alpha \ln[1 - \exp(\mu - E_\alpha)/k_B T]$,

$$M(T, B) = -n_b \mu_b + \frac{2ek_B T}{\pi \hbar d} \sum_{r=1}^{\infty} I_0 \left(\frac{2t_c r}{k_B T} \right) \times \tag{3}$$

$$\frac{\exp[(\tilde{\mu} - 2t_c)r/k_B T]}{1 - \exp(-\omega r/k_B T)} \left(\frac{1}{r} - \frac{\omega \exp(-\omega r/k_B T)}{k_B T[1 - \exp(-\omega r/k_B T)]} \right),$$

where $\mu_b = \hbar e/\sqrt{m_x m_y}$, $\tilde{\mu} = \mu - \omega/2$ and $I_0(x)$ is the modified Bessel function. At low temperatures $T \to 0$ Schafroth's result [3] is recovered, $M(0, B) = -n_b \mu_b$. The magnetization of charged bosons is field-independent at low temperatures. At high temperatures, $T \gg T_c$ the chemical potential has a large magnitude, and we can keep only the terms with $r = 1$ in Eqs.(2,3) to obtain $M(T, B) = -n_b \mu_b \omega/(6k_B T)$ at $k_B T \gg k_B T_c \gg \omega$, which is the familiar Landau orbital diamagnetism of nondegenerate carriers.

The critical region $\tau = T/T_c - 1 \ll 1$ requires numerical calculations, which have been done [22] for an anisotropic 3D CBG with $t_c \gtrsim k_B T_c/2$ and $I_0(x) \approx e^x/\sqrt{2\pi x}$ in Eqs.(2,3). Notwithstanding, one can nicely map the exact results, Fig.2, with a simple analytical expression by replacing summation over all but the first Landau level for integration,

$$\frac{M(T, B)}{n_b \mu_b} = -\frac{0.22\omega}{k_B T_c} \left[\tau + \sqrt{\frac{0.37\omega}{k_B T_c} + \tau^2} \right]^{-1}. \tag{4}$$

Remarkably Eq.(4) predicts almost field-independent diamagnetism well below T_c, $|\tau| \gg \omega/k_B T_c$, a linear field dependence $M(T_c, B) \sim B$ well above T_c, $\tau \gg \omega/k_B T_c$, and an unusual square root behavior at $T = T_c$, $M(T_c, B) \sim B^{1/2}$. Here T_c is the familiar Bose-Einstein condensation temperature $k_B T_c = 3.31\hbar^2 (n_b/2)^{2/3}/(m_x m_y m_c)^{1/3}$, with $m_c = \hbar^2/|t_c|d^2$.
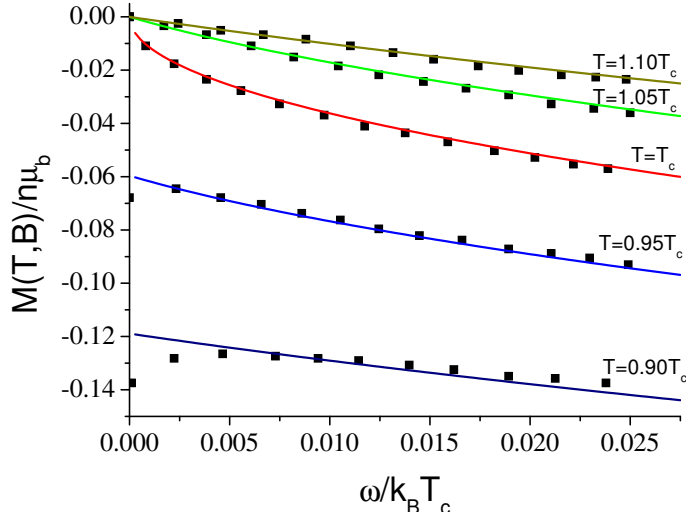
FIG. 2: Exact numerical magnetization (symbols) compared with the analytical approximation, Eq.(4) (lines) .
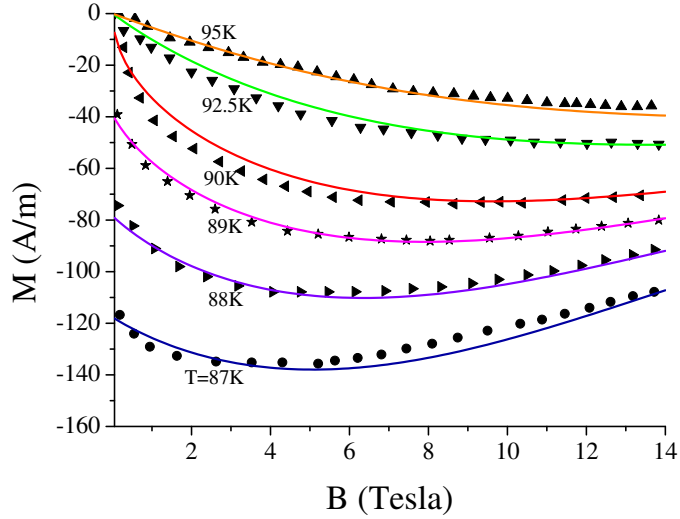

FIG. 3: Diamagnetism of optimally doped Bi-2212 (symbols)[18] compared with magnetization of CBG, Eq.(6), near and above T_c (lines).

Comparing with experimental data one has to take into account a temperature and field depletion of singlets due to their thermal excitations into spin-split triplets and single polaron states, Fig.1B. If the spin gap is small compared with the charge pseudogap, $J < \Delta/2$, triplets mainly contribute to temperature and field dependencies of the singlet bipolaron

6

density near T_c,

$$n_b(T, B) = n_b(T_c, 0)[1 - \alpha\tau - (B/B^*)^2],　\qquad (5)$$

where $\alpha = 3(2n_c t)^{-1}[J(e^{J/k_B T_c} - 1)^{-1} - k_B T_c \ln(1 - e^{-J/k_B T_c})]$, $\mu_B B^* = (2k_B T_c n_c t)^{1/2} \sinh(J/2k_B T_c)$, $\mu_B \approx 0.93 \times 10^{-23}$ Am^2 is the Bohr magneton, n_c is the atomic density of singlets at $T = T_c$ in zero field ($n_c \lesssim 0.1$ in optimally doped cuprates), and $2t$ is the triplet bandwidth, which is taken much larger than $k_B T_c$. A triplet contribution to diamagnetism remains negligible compared with the singlet diamagnetism if $\tau \ll J/k_B T_c$. Then Eq.(4) mapping numerical magnetization in the critical region is modified as

$$M(T, B) = -\frac{0.22 n_b(T_c, 0)\mu_b B}{B_0(1 + 2\alpha/3)} \times \qquad (6)$$

$$\left[\tau + \frac{(B/B^*)^2}{1 + 2\alpha/3} + \sqrt{\frac{0.37 B/B_0}{(1 + 2\alpha/3)^2} + \left(\tau + \frac{(B/B^*)^2}{1 + 2\alpha/3}\right)^2} \right]^{-1},$$

where $B_0 = k_B T_c/2\mu_b$. Using the magnetic field in-plane penetration depth, $\lambda_H^{-2} \approx 21$ $(\mu m)^{-2}$ of optimally doped Bi-2212 [23] and of CBG [11], $\lambda_H^{-2} = 2n_b e^2(m_x + m_y)/(\mu_0 m_x m_y)$, we estimate the bipolaron mass as $m \approx 7.5 m_e$ in agreement with the analytical and numerical QMC results [11], and $n_b(T_c, 0)\mu_b = \hbar\mu_0(m_x m_y)^{1/2}/2e\lambda_H^2(m_x + m_y) \approx 2100$A/m. The BEC temperature corresponds to the temperature were the in-plane resistivity starts to drop with temperature lowering, which is about $T_c = 90$K in optimally doped Bi-2212, so that $B_0 = 524$ Tesla. This choice of $T_c = 90$K is also justified by low-field magnetization [18], which has an exponent close to $1/2$, $M(90K, B) \sim B^{1/2}$ just for this temperature. The remaining two parameters in Eq.(6) are found using the experimental field dependence of $M(T, B)$ at any fixed temperature near T_c. Fitting $M(T, B)$ at $T = 89$ K, Fig.3, yields $\alpha = 0.62$ and $B^* = 56$ Tesla, which according to Eq.(5) corresponds to the singlet-triplet exchange energy $J \approx 20$K. Quite remarkably all other experimental curves in the critical region are well described by Eq.(6) without any fitting parameters, Fig.3.

I conclude that the normal state diamagnetism observed in many cuprates [13, 14, 15, 16, 17, 18] provides unambiguous evidence for charged real-space bosons. The experimental data, Fig.3, clearly contradict BCS (with or without conventional fluctuations) and KT scenarios of the phase transition in cuprates. If we define a critical exponent as $\delta = \ln B/\ln|M(T, B)|$ for $B \to 0$, the T dependence of $\delta(T)$ in CBG is dramatically different
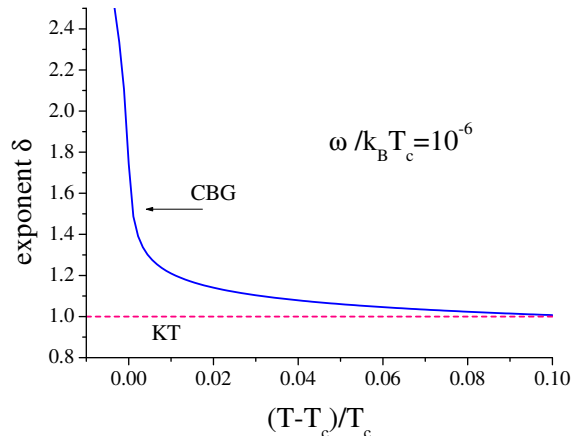
FIG. 4: Critical exponents of the low-field magnetization in CBG and in KT transition.

from KT and other "universal" critical exponents, but it is very close to the experimental [18] $\delta(T)$, Fig.4.

Another strong argument in favor of 3D BEC in cuprates has been drawn using parameter-free fitting of experimental T_c with BEC T_c in more than 30 underdoped, optimally and overdoped samples [24]. Whereas the KT critical temperature expressed through the in-plane penetration depth [12, 25] $k_B T_{KT} \approx 0.9 d\hbar^2/(16\pi e^2 \lambda_H^2)$ appears several times higher than the experimental values in many cases. There are also quite a few samples with about the same λ_H and the same d, but with very different values of T_c, in disagreement with the KT transition. The large Nernst signal, allegedly supporting vortex liquid in the normal state of cuprates [26], has been explained as perfectly normal state phenomenon owing to a partial localization of charge carriers in a random potential inevitable in cuprates [27]. CBG upper critical field and the specific heat in the magnetic field have been found in striking consensus with experimental data [28] following our prediction [29]. More recently the d-wave symmetry and real-space modulations of the order parameter have been also explained with CBG in underdoped [11] and overdoped [30] cuprates.

[1] J.G. Bednorz and K.A. Müller, Z. Phys. B**64**, 189 (1986).

[2] R.A. Ogg Jr., Phys. Rev. **69**, 243 (1946).

[3] M.R. Schafroth, Phys. Rev. **100**, 463 (1955); J.M. Blatt and S.T. Butler, Phys. Rev. **100**, 476 (1955).

[4] J. Bardeen, L.N. Cooper, and J.R. Schrieffer, Phys. Rev **108**, 1175 (1957).

[5] A. Alexandrov and J. Ranninger, Phys. Rev. B**23**, 1796 (1981), ibid **24**, 1164 (1981).

[6] A.S. Alexandrov, Russ. J. Phys. Chem. **57**, 167 (1983).

[7] G.M. Zhao and D.E. Morris Phys. Rev. B **51**, R16487 (1995); G.-M. Zhao, M. B. Hunt, H. Keller, and K. A. Müller, Nature (London) **385**, 236 (1997); R. Khasanov, D. G. Eshchenko, H. Luetkens, E. Morenzoni, T. Prokscha, A. Suter, N. Garifianov, M. Mali, J. Roos, K. Conder, and H. Keller Phys. Rev. Lett. **92**, 057602 (2004).

[8] A. Lanzara, P.V. Bogdanov, X.J. Zhou, S.A. Kellar, D.L. Feng, E.D. Lu, T. Yoshida, H. Eisaki, A. Fujimori,K. Kishio, J.I. Shimoyana, T. Noda, S. Uchida, Z. Hussain and Z.X. Shen, Nature (London) **412**, 510 (2001).

[9] D. Mihailovic, C.M. Foster, K. Voss, and A.J. Heeger, Phys. Rev. B**42**, 7989 (1990).

[10] T.R. Sendyka, W. Dmowski, T. Egami, N. Seiji, H. Yamauchi, and S. Tanaka, Phys. Rev. B**51**, 6747 (1995).

[11] A.S. Alexandrov, *Theory of Superconductivity: From Weak to Strong Coupling* (IoP Publishing, Bristol and Philadelphia, 2003), and references therein.

[12] V.J. Emery and S.A. Kivelson, Nature (London), **374**, 434 (1995).

[13] C. Bergemann, A.W. Tyler, A.P. Mackenzie, J. R. Cooper, S.R. Julian, and D.E. Farrell, Phys. Rev. B **57**, 14387 (1998).

[14] A. Junod, J-Y. Genouda, G. Trisconea, and T. Schneider, Physica C **294**, 115 (1998).

[15] M. J. Naughton, Phys. Rev. B**61**, 1605 (2000).

[16] 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).

[17] I. Iguchi, A. Sugimoto, and H. Sato, J. Low Temp. Phys. **131**, 451 (2003).

[18] Y. Wang, L. Li, M.J. Naughton, G.D. Gu, S. Uchida, and N.P. Ong, cond-mat/0503190; L. Li, Y. Wang, M.J. Naughton, S. Ono, Y. Ando, and N.P. Ong, cond-mat/0507617; Y. Wang, L. Li and N.P. Ong, cond-mat/0510470.

[19] B.W. Hoogenboom, M. Kugler, B. Revaz, I. Maggio-Aprile, O. Fischer, and Ch. Renner, Phys. Rev. B **62**, 9179 (2000).

[20] A.S. Alexandrov and N.F. Mott, J. Supercond (US), **7**, 599 (1994).

[21] D. Mihailovic, V.V. Kabanov, K. Zagar, and J. Demsar, Phys. Rev. B**60**, R6995 (1999) and references therein.

[22] C.J. Dent, A.S. Alexandrov, and V.V. Kabanov, Physica C**341-348**, 153 (2000).

[23] J.L. Tallon, J.W. Loram, J.R. Cooper, C. Panagopoulos, and C. Bernhard, Phys. Rev. B **68**, 180501(R) (2003).

[24] A.S. Alexandrov and V.V. Kabanov Phys. Rev. B **59**, 13628 (1999).

[25] Y.J. Uemura et al., Phys. Rev. Lett. **62**, 2317 (1989).

[26] Z.A. Xu, N.P. Ong, Y. Wang, T. Kakeshita, and S. Uchida, Nature (London) **406**, 486 (2000).

[27] A.S. Alexandrov and V.N. Zavaritsky, Phys. Rev. Lett. **93**, 217002 (2004).

[28] V.N. Zavaritsky, V.V. Kabanov and A.S. Alexandrov, Europhys. Lett. **60**, 127 (2002).

[29] A.S. Alexandrov, Phys. Rev. B**48**, 10571 (1993) and references therein.

[30] A.F. Andreev, Pis'ma Zh. Eksp. Teor. Fiz. **79**, 100 (2004).