Parameter-free expression for superconducting T_c in cuprates

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A parameter-free expression for the superconducting critical temperature of layered cuprates is derived which allows us to express T_c in terms of experimentally measured parameters. It yields T_c values observed in about 30 lanthanum, yttrium and mercury-based samples for different levels of doping. This remarkable agreement with the experiment as well as the unusual critical behaviour and the normal-state gap indicate that many cuprates are close to the Bose-Einstein condensation regime.

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An ultimate goal of the theory of superconductivity is to provide an expression for T_c as a function of some well-defined parameters characterizing the material [1]. The BCS theory provides a 'material' aspect in an estimate of T_c since the characteristic phonon frequency and the electron-phonon coupling constant can be measured [2] while the Coulomb pseudopotential is determined in the first principle (LDA) band-structure calculations. There is no general restrictions on the BCS value of T_c if the dielectric function formalism is properly applied [3,4]. On the other hand, there is a growing evidence for a non-Fermi-liquid normal state and for a non-BCS superconducting state of doped cuprates (see, for example [5]). In particular, the correlation of T_c with the carrier density and with their effective mass [6,7], the carrier specific heat near the transition and its magnetic field dependence [8], the divergent upper critical field [9–11] are reminicent of a charged Bose liquid [5,12,13].

In this letter we derive a parameter-free expression for T_c of a charged Bose-liquid on a quasi-2D lattice. It yields T_c values observed in many cuprates for any level of doping. The main point of our letter is largely independent of the microscopical interpretation of charged bosons, which might be lattice and/or spin bipolarons [5], or any other preformed pairs.

In the framework of this rather general model T_c is calculated from the density sum rule as the Bose-Einstein condensation temperature of 2e charged bosons on a lattice. Just before the discovery [14] we estimated T_c as high as $\simeq 100K$ by applying our expression for the bipolaron effective mass [15]. That was tested by Uemura [7,16] with the conclusion that cuprates are neither BCS nor BEC superfluids but they are in a crossover region from one to another. The experimental T_c has been found about 3 or more times below the BEC temperature.

We now calculate the critical temperature of a charged Bose-liquid taking carefully into account the microscopic band structure of bosons in layered cuprates. It appears that due to a multi-band structure bosons have internal quantum number that might give a two-fold degeneracy as derived by one of us for a particular case of a 'peroxy' bipolaron [17]. We arrive at the parameter-free expression for T_c, which involves the in-plane, λ_{ab} and out-of-plane, λ_c magnetic field penetration depths and the normal state Hall coefficient R_H just above the transition. It describes the experimental data for a few dozen different samples clearly indicating that many cuprates are in the BEC regime.

Bound pairs in cuprates are intersite pairs [5] because of a strong on-site repulsion. In the case of the electron-phonon coupling this is confirmed by the numerical simulations of ionic perovskite lattices [18], where two types of pairs were found, i.e a 'peroxy' bipolaron and an 'in-plane' one. The energy spectrum of the 'peroxy bipolaron (a bound state of the in-plane and apical holes) is

a doublet due to two (x and y) oxygen orbitals elongated along the CuO_2 planes [17]. The energy band minima are found at the Brillouin zone boundary, $(\pm\pi, 0)$ and $(0, \pm\pi)$ owing to the opposite sign of the $pp\sigma$ and $pp\pi$ oxygen hopping integrals. Near these points an effective mass approximation is applied with the following result for the x and y bipolaron bands:

$$E_{\mathbf{k}}^{x,y} = \frac{\hbar^2 k_{x,y}^2}{2m_x} + \frac{\hbar^2 k_{y,x}^2}{2m_y} + t_c[1 - cos(k_z d)], \qquad (1)$$

where d is the interplane distance and $t_c/2$ is the interplane hopping integral. The wave vectors corresponding to the energy minima belong to the stars with two prongs. Their group has only 1D representations. It means that the spectrum is degenerate with respect to the number of prongs of the star. The spectrum Eq.(1) belongs to the star with two prongs, and, hence it is a two-fold degenerate.

While the X-ray absorption experiments confirm an important role of apical holes in some cuprates [19] there are multilayer compounds with inner layers without apex oxygens. The in-plane p-band hybridised with copper might have a higher energy than other bands and the first to be doped. In those cases the in-plane boson (a bound state of two oxygen holes in the CuO_2 plane) is the ground state with the energy minimum, which might be found at the Γ point of the Brillouin zone. One can apply the method of invariants to derive its spectrum. The space group of $La_{2-x}Sr_xCuO_4$ in the tetragonal phase is D_{4h}^{17}. The wave functions at the Γ point transform as 1D $A_{1g,u}$, $A_{2g,u}$, $B_{1g,u}$, $B_{2g,u}$ or 2D E_g and E_u representations of the point group D_{4h}. For the case of E_g (basis functions xz and yz) and E_u (basis functions x and y) the spectrum is expressed as the eigenvalues of the 2x2 matrix. This matrix can be written as a linear combination of Pauli matrices, τ_i. Taking into account that τ_0 transforms as A_{1g} (basis functions $k_x^2 + k_y^2$, k_z^2), τ_x transforms as B_{1g} (basis functions $k_x^2 - k_y^2$), τ_y transforms as B_{2g} (basis functions $k_x k_y$), τ_z transforms as A_{2g} (basis functions s_z) we write the matrix of the Hamiltonian as $H = \left[A(k_x^2 + k_y^2) + Bk_z^2\right]\tau_0 + C(k_x^2 - k_y^2)\tau_x + Dk_x k_y \tau_y$. The eigenvalues of this Hamiltonian are $E_{\mathbf{k}}^{1,2} = A(k_x^2 + k_y^2) + Bk_z^2) \pm \sqrt{C^2(k_x^2 - k_y^2)^2 + D^2 k_x^2 k_y^2}$. Here A, B, C, and D are fenomenological constants that parametrize the effective mass tensor. As a result the in-plane spectrum is degenerate in Γ point as well if it belongs to a 2D representation.

It should be pointed out that a low temperature phase has lower symmetry (space group D_{2h}^{18}). It means that the degeneracy of the spectrum might be removed below the structural phase transition. Nevertheless, if the level splitting is less or of the order of T_c we can apply a theory with a two-fold degenerate spectrum. On the other hand, if the degeneracy is removed the theoretical T_c would be higher by $2^{2/3}$ (see below).

Substituting the spectrum, Eq.(1) into the density sum rule,

$$\sum_{\mathbf{k},i=(x,y)} \left[\exp(E_{\mathbf{k}}^i/k_BT_c) - 1\right]^{-1} = n_B \qquad (2)$$

one readily obtains T_c as

$$k_BT_c = f\left(\frac{t_c}{k_BT_c}\right) \times \frac{3.31\hbar^2(n_B/2)^{2/3}}{(m_xm_ym_c)^{1/3}}, \qquad (3)$$

where the coefficient $f \simeq 1$ is shown in Fig.1 as a function of the anisotropy, t_c/k_BT_c, and $m_c = \hbar^2/|t_c|d^2$. This expression is rather ambiguous so far because the effective mass tensor as well as the boson density n_B are unknown and doping dependent due to the screening of the interaction by free carriers and their localisation by disorder. Fortunately, one can express the band-structure parameters through the in-plane, $\lambda_{ab} = [m_xm_y/8\pi n_Be^2(m_x + m_y)]^{1/2}$ and out-of-plane penetration depth, $\lambda_c = [m_c/16\pi n_Be^2]^{1/2}$ (we take $c = 1$). The boson density is expressed through the in-plane Hall constant (above the transition) as [17]:

$$R_H = \frac{1}{2en_B} \times \frac{4m_xm_y}{(m_x + m_y)^2}, \qquad (4)$$

which leads to

$$T_c = 1.64f \times \left(\frac{eR_H}{\lambda_{ab}^4\lambda_c^2}\right)^{1/3} \qquad (5)$$

with T_c measured in Kelvin, eR_H in cm^3 and λ in cm. The coefficient f is about unity in a very wide range of the anisotropy $t_c/k_BT_c \geq 0.01$, Fig.1. As a result we arrive at a parameter-free expression, which unambiguously can tell us how far cuprates are from the BEC regime:

$$T_c \simeq T_c(3D) = 1.64\left(\frac{eR_H}{\lambda_{ab}^4\lambda_c^2}\right)^{1/3}. \qquad (6)$$

It does not contain the mass tensor explicitly. Hence, any other dispersion law would lead to about the same result for T_c if the spectrum is two-fold degenerate. As an example, taking for simplicity $C = D = 0$ in the in-plane bound state dispersion, one arrives at $T_c \simeq 3.31(n_B/2)^{2/3}/(m^2m_c)^{1/3}$, $R_H = 1/2en_B$, and $\lambda_{ab} = [m_{ab}/16\pi n_Be^2]^{1/2}$, $\lambda_c = [m_c/16\pi n_Be^2]^{1/2}$, and with Eq.(6) as well. Here $m_{ab} = \hbar^2/2A$ and $m_c = \hbar^2/2B$.

We compare the theoretical expressions, Eq.(5,6) with the experimental T_c of more than 30 different cuprates, for which both λ_{ab} and λ_c are measured along with R_H (see Table 1 and Fig.2). The Hall coefficient has a strong temperature dependence above T_c. Therefore, we use the experimental Hall 'constant' $R_H \equiv R_H(T_c+0)$ just above the transition as prescribed by Eqs.(5,6). In a few cases (mercury compounds), where $R_H(T_c + 0)$ is unknown, we have taken the inverse chemical density of carriers (divided by e) as R_H. Almost for all samples the theoretical T_c coincides with the experimental one within the experimental error bar for the penetration depth (about $\pm 10\%$). There are a few Zn doped YBCO samples, Fig.2, with the experimental critical temperature higher than the theoretical one. We believe that the degeneracy of the boson spectrum is removed by the random potential of Zn, so for these samples the theoretical T_c is actually higher than Eq.(6) suggests. Multiplying the theoretical T_c in Table 1 by $2^{2/3}$ for three Zn-doped samples we obtain T_c=73 K, 57 K, 41 K in good agreement with the experimental values T_c=68 K, 55 K and 46 K, respectively.

One can argue that cuprates belong to a 2D 'XY' universality class with the Kosterlitz-Thouless critical temperature T_{KT} of preformed bosons [20,21] or Cooper pairs [22] due to a large anisotropy. Would it be the case one could hardly discriminate Cooper pairs with respect to local pairs in cuprates by the use of their T_c values. The Kosterlitz-Thouless temperature is expressed trough the in-plane penetration depth alone as [22]

$$k_BT_{KT} \simeq \frac{0.9d\hbar^2}{16\pi e^2\lambda_{ab}^2}. \qquad (7)$$

It appears significantly higher than the experimental value in most cases (see Table 1). What is more crucial, however, is the fact that cuprates have the specific heat [8] of a 3D charged Bose gas [12].

The boson-boson interaction might be rather strong leading to self-energy effects and to some renormalisation of the effective mass tensor. It is important, that Eq.(6) does not contain the mass and, hence, is not affected by the interaction. Nevertheless, it is interesting to evaluate the effective mass tensor in terms of the penetration depth and the Hall constant. The in-plane and out-of-plane boson masses are presented in Table 2 for a few samples of La$_{2-x}$Sr$_x$Cu$_4$ and YBa$_2$Cu$_3$O$_{7-x}$. The in-plane boson dispersion with $C = D = 0$ has been applied. Theoretical estimates of the in-plane mass ($\simeq 10m_e$ i.e. about $5m_e$ per hole [17,23,24,18,25]) fit well our empirical values, Table 2. There is an intersting opposite tendency in the doping dependence of the effective mass of La$_{2-x}$Sr$_x$CuO$_4$ and YBa$_2$Cu$_3$O$_{7-x}$. While the mass increases with doping in La$_{2-x}$Sr$_x$CuO$_4$, it slightly decreases in YBa$_2$Cu$_3$O$_{7-x}$. We believe that it is a result of an interplay between an interaction responsible for the mass enhancement and disorder. We notice, however, that the absolute value of the effective mass in terms of the free electron mass, Table 2, does not describe the actual band mass renormalisation because the unrenormalised (bare) band mass remains unknown.

Many thermodynamic, magnetic and kinetic properties of cuprates were understood with charged bosons on a lattice [5,26]. We admit, however, that one experimental fact might be sufficient to destroy any theory.

In particular, the single-particle spectral function seen by ARPES [27] was interpreted by several authors as a Fermi liquid feature of the normal state incompatible with bipolarons. Most (but not all) of these measurements produced a large Fermi surface. This should evolve with doping as $(1 - x)$ in a clear contradiction with low frequency kinetics and thermodynamics, which show an evolution proportional to x (x is the number of holes introduced by doping). Recently it has been established, however, that there is a normal state gap in ARPES and SIN tunnelling, existing well above T_c irrespective to the doping level [27–29]. 'The 'Fermi surface' showed missing segments just near the points [28] where we expect the Bose-Einstein condensation [30]. A plausible explanation is that there are two liquids in cuprates, the normal Fermi liquid and the charged Bose-liquid (this mixture has been theoretically discussed a long time ago [31]). However, it is difficult to see how this scenario could explain the doping dependence of dc and ac conductivity as well as of the magnetic susceptibility and carrier specific heat. On the other hand, the single-particle spectral function of a pure 'bosonic' system has been recently derived by one of us [30]. It describes the spectral features of tunnelling and photoemission in cuprates. Any single-particle spectral weight at the chemical potential appears in our model due to states localised by disorder inside the normal state gap [30]. The model is thus compatible with the doping evolution of thermodynamic and kinetic properties.

In conclusion, we have shown that the experimental critical temperate of superconducting cuprates is not very different from the Bose-Einstein condensation temperature of two-fold degenerate charged bosons on a lattice. Our empirical expression for T_c describes the experimental data remarkably well with no parameters to fit. This possibility originates in two different energy scales in cuprates: a strong attractive interaction and a small bandwidth. Their difference allows us to 'integrate out' the interaction and express T_c via static response functions.

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Figure Captures

Fig.1. Correction coefficient to the 3D Bose-Einstein condensation temperature as a function of the anisotropy.

Fig.2. Theoretical critical temperature compared with the experiment (the theory is exact for samples on the straight line) for $LaSrCuO$ compounds (squares) for Zn substituted $YBa_2(Cu_{1-x}Zn_x)_3O_7$ (circles) for $YBa_2Cu_3O_{7-\delta}$ (triangles) and for $HgBa_2CuO_{4+\delta}$ (diamonds). Experimental data for the London penetration depth are taken from T. Xiang $et\ al$, Int. J. Mod. Phys. B **12**, 1007 (1998) and B. Janossy $et\ al$, Physica C, **181**, 51 (1991) in $YBa_2Cu_3O_{7-\delta}$ and $YBa_2(Cu_{1-x}Zn_x)_3O_7$; from V.G. Grebennik $et\ al$, 'Hyperfine Interactions', **61**, 1093 (1990) and C. Panagopoulos (private communication) in underdoped and overdoped $La_{2-x}Sr_xCuO_4$, respectively, and from J. Hofer $et\ al$, Physica C, **297**, 103 (1998) in $HgBa_2CuO_{4+\delta}$. The Hall coefficient above T_c is taken from A. Carrington $et\ al$, Phys. Rev. B **48**, 13051 (1993) and J. R. Cooper (private communication) ($YBa_2Cu_3O_{7-\delta}$ and $YBa_2(Cu_{1-x}Zn_x)_3O_7$) and from H.Y. Hwang $et\ al$, Phys. Rev. Lett. **72**, 2636 (1994) ($La_{2-x}Sr_xCuO_4$).

TABLE I. Experimental data on T_c(K), ab and c penetration depth(nm), Hall coefficient ($10^{-3}(cm^3/C)$), and calculated values of T_c applying Eqs.(6,5,7) respectively (K)for $La_{2-x}Sr_xCuO_4$ (La), $YBaCuO(x\%Zn)$ (Zn), $YBa_2Cu_3O_{7-x}$ (Y) and $HgBa_2CuO_{4+x}$ (Hg) compounds

Compound	T_c	λ_{ab}	λ_c	R_H,	T_c	T_c	T_{KT}
$La(0.2)$	36.2	200	2540	0.8	38	41	93
$La(0.22)$	27.5	198	2620	0.62	35	36	95
$La(0.24)$	20.0	205	2590	0.55	32	32	88
$La(0.15)$	37.0	240	3220	1.7	33	39	65
$La(0.1)$	30.0	320	4160	4.0	25	31	36
$La(0.25)$	24.0	280	3640	0.52	17	19	47
$Zn(0)$	92.5	140	1260	1.2	111	114	172
$Zn(2)$	68.2	260	1420	1.2	45	46	50
$Zn(3)$	55.0	300	1550	1.2	35	36	38
$Zn(5)$	46.4	370	1640	1.2	26	26	30
$Y(0.3)$	66.0	210	4530	1.75	31	51	77
$Y(0.43)$	56.0	290	7170	1.45	14	28	40
$Y(0.08)$	91.5	186	1240	1.7	87	88	98
$Y(0.12)$	87.9	186	1565	1.8	75	82	97
$Y(0.16)$	83.7	177	1557	1.9	83	89	108
$Y(0.21)$	73.4	216	2559	2.1	47	59	73
$Y(0.23)$	67.9	215	2630	2.3	46	58	73
$Y(0.26)$	63.8	202	2740	2.0	48	60	83
$Y(0.3)$	60.0	210	2880	1.75	43	54	77
$Y(0.35)$	58.0	204	3890	1.6	35	50	82
$Y(0.4)$	56.0	229	4320	1.5	28	42	65
$Hg(0.049)$	70.0	216	16200	9.2	23	60	115
$Hg(0.055)$	78.2	161	10300	8.2	43	92	206
$Hg(0.055)$	78.5	200	12600	8.2	28	69	134
$Hg(0.066)$	88.5	153	7040	6.85	56	105	229
$Hg(0.096)$	95.6	145	3920	4.7	79	120	254
$Hg(0.097)$	95.3	165	4390	4.66	61	99	197
$Hg(0.1)$	94.1	158	4220	4.5	66	105	216
$Hg(0.101)$	93.4	156	3980	4.48	70	107	220
$Hg(0.101)$	92.5	139	3480	4.4	88	127	277
$Hg(0.105)$	90.9	156	3920	4.3	69	106	220
$Hg(0.108)$	89.1	177	3980	4.2	58	90	171

TABLE II. Mass enhancement with respect to the free electron mass

Compound	m_{ab}	m_c
$La(0.2)$	22.1	3558
$La(0.15)$	15.0	2698
$La(0.1)$	11.3	1909
$Y(0.0)$	7.2	584
$Y(0.12)$	8.3	600
$Y(0.3)$	10.6	1994