Generalized Statistics and High $T_c$ Superconductivity

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Abstract

Introducing the generalized, non-extensive statistics proposed by Tsallis [1], into the standard s-wave pairing BCS theory of superconductivity in 2D yields a reasonable description of many of the main properties of high temperature superconductors, provided some allowance is made for non-phonon mediated interactions.

The discovery of superconductivity in the copper oxides in 1986 [2] and the subsequent race for even higher critical temperatures (125K by 1993 [3, 4]) raised hopes for the application of superconducting phenomenon at operating temperatures approaching room temperature. The inability of the Bardeen-Cooper-Schrieffer (BCS) model [5] to describe superconductivity in these materials satisfactorily, appears to indicate that we are dealing with a completely different class of superconductors. Various theoretical models have been proposed to explain this phenomenon, ranging from d-wave superconductivity [6, 7] to models incorporating non-phononic coupling mechanisms [8, 9, 10, 11]. Different degrees of success have been achieved in explaining specific aspects of these high-$T_c$ materials, but no inclusive model exists.

A number of characteristics must be incorporated in any such model. Certainly the main common denominator in all high-$T_c$ materials is the large anisotropy in the crystal structure, resulting in conduction electron states in the CuO$_2$ planes being very nearly two-dimensional in character. It is therefore reasonable to assume that this is essential for the high critical

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temperatures and that arguably the 'ideal' high-\(T_c\) superconductor is purely two-dimensional in character. It is, however, well known that in BCS, a pure 2D model presents problems as the phase loses its coherence due to fluctuations \[12\]. This calls for at least a quasi-2D model in all realistic cases. Although the results of measurements on the superconducting gap are somewhat varied, there seems to be convergence towards a ratio of \(\frac{2\Delta_0}{k_B T_c}\) equal to somewhere between 6 and 8 which is not consistent with the universal value of \(\sim 3.5\) obtained for normal BCS superconductors. This is particularly so in the case of tunneling measurements \[13, 14, 15, 16\], where ratios as high as 8.9 have been measured in \(\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4\) \[17\], but appears to contradict optical measurements\[18, 19, 20\] which seem to yield results closer to the BCS ratio for weak coupled superconductors. In spite of the apparent BCS behaviour of the tunneling currents, the violation of universality is difficult to understand. Thirdly, experimental data suggests that the electronic contribution to the specific heat in cuprates does not exhibit the exponential form of normal weak coupled superconductors. It is rather of the linear form \(\gamma T\), where \(\gamma\) is of the order of a few mJ/mol-K\(^2\). Magnetic properties of the high-\(T_c\) superconductors also differ appreciably from their normal counterparts, most notably in their very high upper critical fields. These fields are extrapolated to be of the order of tens or hundreds of Teslas and are too large to be attained with other present technologies. Furthermore it should also be noted that all high-\(T_c\) superconductors are type II superconductors. One of the strongest arguments for the phonon coupling scheme of the BCS model is the existence of an isotope effect. In cuprate materials, where there is a strong doping dependence of the isotope effect, this requires some additional consideration. In optimally doped materials the effect is often strongly suppressed (\(T_c\) may scale as a power \(\alpha = 0.1\) of the mass), whereas in overdoped or underdoped materials it might be more prevalent, with the scaling in some cases even exceeding \(\alpha = 0.5\) \[21\].

Recently in statistical mechanics there has been interest in a generalization of the Boltzmann-Gibbs (BG) statistics to a non-extensive form proposed by Tsallis\[1\] in which the former is recovered in an appropriate limit. This formalism has had considerable success in providing an appropriate mathematical framework for dealing with physical systems with long-range interactions. This is markedly so in the so-called stellar "polytropes" \[22\] where the usual Boltzmann distribution functions yield unphysical results. A variety of other applications have also been considered along with the generalization of many well-known theorems and principles, see for example the references in \[23\].

In this paper we propose and motivate the possibility that the cuprate oxide materials have an inherent underlying non-BG like character responsible for their high-critical temperatures and other unusual properties. We suggest that the s-wave BCS model is essentially correct in employing an effective weak coupling Hamiltonian that includes the kinetic energy of free
electrons along with a constant attractive potential between electrons of equal momentum and opposite spin \[.\] The BCS ansatz in a purely 2D form is used for the ground state, with a generalized form of the Fermi-distribution function \[24\] appearing at finite temperatures. We show elsewhere that such a 2D model is justified as the fluctuation-dissipation theorem is no longer valid in the Tsallis formalism \[25\]. The assumption that the coupling mechanism is purely phonon mediated, needs to be modified to obtain reasonable fits to the experimental data, particularly in the case of very high temperature superconducting materials.

The relevant question is, of course, why one would resort to changing the entropic measure in a high-\(T_c\) superconductor, and if one does, why the Tsallis formulation should be appropriate. A very compelling argument can be found from a consideration of the electronic specific heat. The more exotic coupling methods mentioned above, certainly may have the effect of removing problems indirectly related to the BCS-Hamiltonian such as the lattice instabilities predicted by Migdal \[26\]. The experimental evidence of an energy gap, however, requires an energy spectrum for the total energy of excitations, of the form

\[
E_k^2 = \varepsilon_k^2 + \Delta^2, \quad (1)
\]

where \(\varepsilon\) is the excitation energy and \(\Delta\) the energy gap, independent of any proposed model. Assuming a Fermi distribution, the electronic specific heat capacity \((C)\) at temperatures \(kT \ll \Delta\), can shown to have the following form \[27\]

\[
C \sim e^{-\frac{\Delta}{kT}} \quad (2)
\]

One is therefore obliged to contend with an exponential form of the heat capacity in any model that relies on the Fermi distribution function used in BG statistical mechanics. One way to circumvent this is to introduce a different distribution function.

It should be remembered that the most basic interaction in an electron gas is that of the Coulomb repulsion. This interaction is of infinite range and may also introduce correlation effects. One example of this is the condensation of a true electron gas, of low enough density, into a Wigner lattice \[28\]. Very early attempts at explaining superconductivity, quite intuitively, but unsuccesfully used the Coulomb potential as a starting point \[29, 30\]. More recently models like those of Hubbard \[31\], describing contributions due to repulsion between electrons sharing an atomic orbital, have been applied to superconductivity. The condensation of electrons into its superfluid state is ultimately the result of an effective, attractive interaction. The extent of the effect of Coulomb repulsion might not be obvious and in spite of the fact that we do not explicitly take it into account in the effective interaction, we argue that it is precisely the effects of this long-range interaction which may render the system more suitable for description by generalized statistics.
The generalized entropy postulated by Tsallis \cite{1} takes the form:

\[ S_q = k \frac{\sum_{i=1}^{\infty} \{p_i - p_i^q\}}{q - 1} \quad (q \in \mathbb{R}) \tag{3} \]

where \( w \) is the total number of microstates in the system and \( p \) is the associated probabilities. It has been shown that this entropy obeys the usual properties of concavity, equiprobability, positivity and irreversibility and Shannon additivity and preserves the Legendre transformation structure of thermodynamics \cite{32}. It is also straightforward to verify that the usual BG entropy is recovered in the limit \( q \to 1 \). Associated with equation (3) is the generalized Fermi distribution given by \cite{24}

\[ f_q = \frac{1}{[1 + \beta(q - 1)e_k]^{1/q} + 1} \tag{4} \]

Once again the usual Fermi distribution of BG statistics is recovered in the limit \( q \to 1 \). It is this generalized distribution, we will henceforth assume, that excited independent quasi-particles obey at finite temperature. In 2D the BCS gap equation for weak coupled superconductors at finite temperature \cite{5} is given as

\[ \Delta_p = -\sum_k g_{kp} \Delta_k \frac{2E_k}{[1 - f_{k\uparrow} - f_{k\downarrow}] (1 - f_{k\uparrow} - f_{k\downarrow})} \tag{5} \]

where \( g \) is the coupling constant, \( E_k \) is the quasi-particle energy given by equation (1) and \( f_{k\sigma} \) is the Fermi distribution function. Replacing \( f_{k\sigma} \) by equation (4) and the sum by an integral over the density of states we obtain in 2D a generalized form for the gap equation:

\[ \frac{1}{N(0)g} = \int \frac{d\varepsilon}{\sqrt{\varepsilon^2 + \Delta^2}} \frac{[1 + \beta(q - 1)\varepsilon^2 + \Delta^2]^{1/q} - 1}{[1 + \beta(q - 1)\varepsilon^2 + \Delta^2]^{1/q} + 1} \tag{6} \]

where \( N(0) \) represents the density of states. For \( q=1 \) in 3D, one recovers the gap equation in the standard BCS theory and, of course, a description of normal superconductors in the weak coupling limit. As required, equation (6) is independent of the distribution function and at zero temperature reduces to

\[ \frac{1}{N(0)g} = \int \frac{d\varepsilon}{(\Delta_0^2 + \varepsilon^2)^{1/2}} \tag{7} \]

where \( \Delta_0 \) is the zero temperature gap.

Let us, as a starting point, choose as cutoff to the gap equation the Debye frequency \( \hbar \omega_D \). The analytical solution to equation (6) is

\[ \Delta_0 = \left( \hbar \omega_D + \sqrt{\hbar^2 \omega_D^2 + \Delta_0^2} \right) e^{\frac{1}{\hbar \omega_D}}. \tag{8} \]
Using experimental values for the gap and Debye frequency, one can solve for \( N(0)g \) at zero temperature. The problem then reduces to finding that \( q \) in the gap equation, which yields a vanishing gap at the critical temperature. With \( q \) thus fixed, the temperature dependence of the gap can be determined.

Consider again the gap in equation (5) at \( T_c \), independent of the particular choice of statistics. A change in the integration variable \( \varepsilon \rightarrow \epsilon k_B T_c \) may always be made such that the integration limits are from 0 to \( \frac{\theta_D}{T_c} \). Integrating by parts yields:

\[
\frac{1}{N(0)g} = (1 - 2f(\frac{\theta_D}{T_c})) \ln \frac{\theta_D}{T_c} - \int_0^{\frac{\theta_D}{T_c}} (\epsilon - 2F(\epsilon)) \ln \epsilon \ d\epsilon \quad (9)
\]

where \( F(\epsilon) \) is the indefinite integral of \( f(\epsilon) \). Taking the integral on the right to the left hand side and dividing by \( 1 - 2f(\frac{\theta_D}{T_c}) \) yields some number \( \varphi \) which depends, of course, on the choice of \( f(\epsilon) \). Exponentiating both sides yields:

\[
T_c = \theta_D e^{\varphi} \quad (10)
\]

which clearly demonstrates that the isotope effect is preserved in the BCS formulation irrespective of the particular form of statistical mechanics employed.

It is at this stage appropriate to note that despite reasonable physical arguments in the case of normal superconductors, the choice of the Debye frequency as a cutoff is mathematically quite arbitrary. The temperature dependent solution to the gap equation in fact converges to a well defined value for any cutoff of sufficient magnitude. It is only when the cutoff is of a magnitude of the order of the gap itself, that deviations occur. In normal superconductors the Debye frequency is greater than the gap by \( \sim 10^2 \) and thus adequate. This is, however, certainly not the case in high-\( T_c \) superconductors where the gap at zero temperature may even be larger than the Debye frequency, e.g. in \( \text{TL}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10} \) \( \text{[3]} \) (see \( \text{[3]} \) for Debye frequencies of other high \( T_c \) superconductors). Abandoning the Debye frequency as a cutoff is tantamount to acknowledging that the electron-phonon interaction is not the only interaction involved in forming the condensed state. Changing the cutoff will, of course, influence the isotope effect. It might be argued that the observed suppression in the isotope effect may be a consequence of this.

It is interesting to note that the convergence of the gap at greater cutoffs, is accompanied by a simultaneous convergence of \( q \). Consider a transformation of the gap in equation (8) via the substitution \( \varepsilon = \frac{k_B T_c \epsilon'}{2} \). Let us also define the cutoff in terms of multiples of the energy gap, \( e.g. \ n\Delta_0 \). Then for a given value of the gap to critical temperature ratio, \( \frac{2\Delta_0}{k_B T_c} = m \), one can show that the gap equation reduces to a form dependent only on the ratio...
Thus, if for a specific ratio of $\Delta_0$ to $T_c$, the temperature dependent gap is convergent independent of cutoff, then $q$ must have a uniquely defined value. It therefore seems appropriate to define the cutoff in terms of the convergence of $q$ as this can be specified independent of the critical temperature pertaining to a specific material.

\[ 1 = \int_0^{mn} \frac{d\epsilon'}{\epsilon'} \left\{ \frac{[1 + (q - 1)\epsilon']}{[1 + (q - 1)\epsilon']/2} \frac{1}{\sqrt[q-1]{1 + \frac{1}{2}}} - 1 \right\} \]

(11)

Figure 1: The ratio $\frac{2\Delta_0}{k_BT_c}$ vs. $q$ is given by the solid curve. For comparison the dashed curve representing $\frac{2\Delta_0}{qk_BT_c}$ vs. $q$ is included.

A relevant question now is whether a generalization of the BCS universality condition of $\frac{2\Delta_0}{k_BT_c} \sim 3.52$ exists for the generalized statistics of Tsallis. Clearly the generalization must be $q$ dependent because of the dependence of $T_c$ on $q$ and reduce to the BCS universality condition for $q=1$. In Fig.1
a graph of $\frac{2\Delta_0}{qk_B T_c}$ versus $q$ is given. This ratio does not deviate appreciably from 3.5 which suggests the following generalization of the universality condition

$$\frac{2\Delta_0}{qk_B T_c} \sim 3.52$$  \hspace{1cm} (12)

A more detailed analysis might lead to replacing $q$ by some function of $q$ which should be $\sim q$.

Figure 2: The normalized electronic specific heats $\frac{C}{C_{\text{max}}}$ vs. $\frac{T}{T_c}$ for various choices of $\frac{2\Delta_0}{k_B T_c}$, $q$ and cutoffs where in each case $C_{\text{max}}$ corresponds to the maximum value of $C$. The corresponding values of the normalized gaps are shown in the inset.

The electronic specific heat capacity may be expressed as

$$C = T \frac{dS}{dT}$$  \hspace{1cm} (13)

where the entropy is given by
Figure 3: Comparison of the temperature dependence of experimental values of $\Delta$ from Ref [13] for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ with $T_c = 85K$ and $\frac{2\Delta_0}{k_BT_c} = 6.2$ with the theoretical results for $q=1.71$

\[ S_q = 2k_B\left[\frac{\int d\varepsilon\{f(E) - f(E)^q\}}{q - 1} + \frac{\int d\varepsilon\{(1 - f(E)) - (1 - f(E))^q\}}{q - 1}\right] \quad (14) \]

and we use $N(0) = \frac{m}{2\pi\hbar^2}$ for the density of states in 2D and the bare electron mass $m_e$ in all cases. The effect of the cutoff on the specific heat is shown in Figure 2 for LSCO ($T_c = 36K$). The linear nature of the specific heat can be seen over most of the superconducting region with $\hbar\omega_D = 390K$ as the cutoff. Clearly the situation deteriorates as the critical temperature is reached. Increasing the cutoff to $70\Delta_0$ yields a more linear form of the specific heat near $T_c$ with little change in the shape of the gap (see inset in Fig. 2). Changing the cutoff from $\hbar\omega_D$ to $70\Delta_0$ increases the $\gamma$ from 5.5 mJ/mol-K$^2$ to 10.7 mJ/mol-K$^2$ with a slight change in $q$ (from 1.752 to 1.679). Note that in spite of the fact that the slopes of the specific heat may be altered
by using an effective mass \( (\gamma \to \frac{m^*}{m}\gamma) \), these values are not in disagreement with many of the experimental results which seem to lie between 3 and 12 mJ/mol-K\(^2\) for most cuprates \[34\]. For comparison the ratios of \( \frac{2\Delta_0}{k_BT_c} = 4.0 \) and 8.0, the corresponding results for the specific heats and the gaps are shown. Note the specific heat is much more linear when larger cutoffs are used and that the nonlinearity around \( T_c \) disappears almost completely for \( \frac{2\Delta_0}{k_BT_c} = 8.0 \). In this case \( \gamma = 20.0 \) J/mol-K\(^2\).

In Fig. 3 the experimental data obtained by Briceno and Zettl \[13\] for the temperature dependence of the gap of Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_8\) with \( T_c = 85 \) K and \( \frac{2\Delta_0}{k_BT_c} = 6.2 \) are compared with the theoretical results with \( q=1.717 \). In spite of a linear specific heat \( (\gamma = 13.9 \) mJ/mol-K\(^2\) using \( m_e \)) which is quite consistent with experiment \[34\], the shape of the gap differs slightly from that obtained experimentally.

Due to the type II nature of the cuprate oxides, a full analysis of the critical magnetic field would require including contributions to the free energy resulting from the flux lattice as well as the mutual energy of the surface current and flux line currents. We therefore present only a qualitative analysis of the thermodynamic critical field, \( B_c \), and make no attempt at solving either the lower- \( (B_{c1}) \) or the upper- \( (B_{c2}) \) critical fields.

In 2D, the thermodynamic critical field is given by

\[
\frac{B_{c2}^2}{\mu_0} = F_n - F_s
\]  

where \( F_n \) and \( F_s \) are the free energies in the normal- and superconducting states respectively and \( \mu_0 \) is the permeability of free space. The free energy in the superconducting state is given by

\[
F_s = N(0)(\hbar\omega)^2 \left[1 + \left(\frac{\Delta_0}{\hbar\omega}\right)^2\right]^{\frac{1}{2}} - 1 - 2N(0) \int d\varepsilon \left(\frac{2\varepsilon^2 + \Delta^2}{E}\right) f_q(\beta E)
\]  

\[
- \frac{4kT}{q-1} N(0) \int d\varepsilon \left[(1 - f_q(\beta E))^q - (1 - f_q(\beta E))\right].
\]

\( F_n \) is obtained by setting \( \Delta = 0 \) and using the value of \( q \) determined from solution of the corresponding gap equation. This implies that in our generalized BCS model the normal state is also described by the generalized statistics of Tsallis since \( B_c \) must vanish at \( T_C \). Figure 4 shows the thermodynamic field obtained for a superconductor with \( \frac{2\Delta_0}{k_BT_c} = 6.0 \) and \( T_c = 36K \). The inset shows the behaviour of the free energies. They differ from the weak coupled normal superconductor case where the free energy is always negative except at \( T = 0 \) where it vanishes. The difference, however, remains a positive definite quantity and no problems arise in calculating \( B_c \). The zero temperature result of figure 4 agrees exceptionally well with an extrapolation based on experimental results due to \[35\]. They find, for
a La$_{1.846}$Sr$_{0.154}$CuO$_4$ crystal with $T_c = 35$K that $B_c = 0.251$ T, while our result is 0.234 T.

The Josephson quasi-particle current was first treated in detail by Shapiro et al. [36] (the other terms are considered elsewhere [37]). The quasi-particle current between two different superconductors is given by the integral:

$$I_{qp} = \frac{1}{eR_N} \int_{-\infty}^{\infty} [f(\omega) - f(\omega + eV)][|\omega + eV| |\omega| + V] \left[ \frac{\theta(|\omega| - \Delta_1)\theta(|\omega + eV| - \Delta_2)}{(\omega^2 - \Delta_1^2)^{1/2}[(\omega + eV)^2 - \Delta_2^2]^{1/2}} \right] d\omega$$

where $V$ is the applied voltage, $e$ the electron charge, $R_N$ the junction resistance and $\Delta_1$, $\Delta_2$ the energy gap on either side of the tunneling junction. In Figure 5 we show the result of evaluating equation (18) at 5K for a junction with identical superconductors having $\frac{2\Delta}{k_B T_c} = 6.0$ and using the generalized distribution function with $q = 1.679$. Of interest is the fact that $I_{qp}$ is not negligible below an applied voltage of $2\Delta/e$. The equivalent calculation using BG statistics yields a vanishing contribution in this region. Included in the
Figure 5: $\frac{dI}{dV}$ vs. $eV$ (inset) obtained from the quasi-particle contribution to the Josephson tunneling current in the region $eV < 2\Delta$, as shown in the main figure.

Figure is the derivative $\frac{dI}{dV}$. Although we do not obtain all of the structure observed experimentally in LSCO, the existence of a $I_{qp}$ contribution raises the possibility that interference due to other admixtures, perhaps d-wave or Andreev bound states, will produce the sought after structure. Analysis of the other contributions using generalized statistics does not appreciably alter the results in the $eV < 2\Delta$ region. The use of generalized statistics, in the BCS equations, however, allows the experimentally observed discontinuity in the quasi-particle tunneling current to be interpreted as being coincident with the energy gap at low T, whilst consistently predicting the gap to vanish at the correct critical temperature.

In conclusion we have shown that a simple 2D s-wave BCS pairing model which incorporates generalized statistics can provide an adequate description of many of the main features of high temperature superconductivity. Non-phonon mediated interactions, however, appear to play a role as the critical


temperature increases.

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List of Figures

1. The ratio $\frac{2\Delta}{k_B T_c}$ vs. $q$ is given by the solid curve. For comparison the dashed curve representing $\frac{\Delta}{k_B T_c}$ vs. $q$ is included. ................................................. 6

2. The normalized electronic specific heats $\frac{C}{C_{max}}$ vs. $\frac{T}{T_{C}}$ for various choices of $\frac{2\Delta}{k_B T_c}$, $q$ and cutoffs where in each case $C_{max}$ corresponds to the maximum value of $C$. The corresponding values of the normalized gaps are shown in the inset. ........ 7

3. Comparison of the temperature dependence of experimental values of $\Delta$ from Ref [13] for Bi$_2$Sr$_2$CaCu$_2$O$_8$ with $T_c = 85K$ and $\frac{2\Delta}{k_B T_c} = 6.2$ with the theoretical results for $q=1.71$ .... 8

4. The critical magnetic field $B_c$ vs. $T$ obtained from free energies given in the inset .................................................. 10

5. $\frac{dI}{dV}$ vs. $\frac{eV}{2\Delta}$ (inset) obtained from the quasi-particle contribution to the Josephson tunneling current in the region $eV < 2\Delta$, as shown in the main figure. .................................................. 11