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Low temperature thermodynamics of charged bosons in a random potential and the specific heat of $La_{2-x}Sr_xCuO_4$ below T_c .

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(January 26, 2006)

We propose a simple analytical form of the partition function for charged bosons localised in a random potential and derive the consequent thermodynamics below the superfluid transition temperature. In the low temperature limit, the specific heat C depends on the localisation length exponent ν : C is linear for $\nu \leq 1$, but for $\nu > 1$ we obtain $C \propto T^{1/\nu}$. This unusual sub-linear temperature dependence of the specific heat has recently been observed in $La_{2-x}Sr_xCuO_4$ below T_c .

74.20.-z, 74.65.+n, 74.60.Mj

I. INTRODUCTION

A charged $2e$ Bose liquid of small bipolarons as a microscopic model of the ground state of cuprates¹ explains the high value of T_c and its ‘boomerang’ doping dependence², the divergent upper critical field³, the normal state in-plane and ‘c’-axis transport, the magnetic susceptibility and the pseudogap⁴. The (bi)polaronic nature of carriers in these materials is supported by observations of the characteristic polaronic spectral function in the infrared spectra⁵, of the isotope effect on the carrier mass⁶, of flat bands in the high-resolution photoemission spectra⁷, and by a few other experiments such as the pair-distribution analysis of neutron scattering⁸.

The bipolaron model overcomes a fundamental problem with any theory of high- T_c which is the short-range Coulomb repulsion. As stressed by Emery *et al*⁹ the theories involving real-space pairs with a pure electronic (exchange) mechanism of pairing are *a priori* implausible due to the strong short-range Coulomb repulsion between two carriers. The direct repulsion is usually much stronger than any exchange interaction. The attractive potential generated by the electron-phonon interaction of the Holstein model may overcome the short-range Coulomb repulsion, but inevitably involves a huge carrier mass enhancement (otherwise the phonon frequency would be extremely high). On the other hand, one of us showed² that the Fröhlich electron-phonon interaction can provide *intrinsically* mobile intersite small bipolarons which are condensed at high T_c (of the order of 100K). This interaction, operating on a scale of the order of 1eV, compensates the Coulomb repulsion and allows the deformation potential (together with an exchange interaction of any origin) to bind two holes into an intersite mobile bipolaron in the CuO_2 plane. The mass

renormalisation appears to be smaller by several orders of magnitude than in the Holstein model (with the same attractive potential). Hence the charge $2e$ Bose liquid, being already an important reference system of quantum statistics, is now of particular physical interest. Because doped Mott insulators are disordered, the localisation of carriers by a random potential plays an important role in their low-temperature thermodynamics and transport¹.

In this paper we study the thermodynamics of charged bosons in the superfluid phase assuming that some of them are localised by disorder. The fact that in the superconducting phase the chemical potential μ is pinned at the mobility edge, $\mu = E_c$, significantly simplifies the problem. By using a plausible form of the single well partition function and a model density of localised states, we find the temperature dependence of the specific heat. Under certain conditions the predicted specific heat follows precisely the sub-linear temperature dependence observed¹⁰ in $La_{2-x}Sr_xCuO_4$ at low temperatures.

II. PARTITION FUNCTION

The picture of interacting bosons with a short-range interaction filling up all the localised single-particle states in a random potential is known in the literature^{11–13}. To calculate the partition function of localised bosons one has to take into account the repulsion between them. One cannot ignore the fact that the localisation length ξ generally varies with energy and diverges at the mobility edge. One would expect that the number of *hard core* bosons in a localised state near the mobility edge diverges in a similar way to the localisation length. Only a repulsive interaction can stop all particles condensing into the lowest localised state. Thus, as stressed by Fisher *et al*¹³, for a Bose gas (in a random field) there is no sensible non-interacting starting point about which to perturb; this is in contrast to the case of a Fermi gas.

Although the comprehensive scaling analysis of neutral¹³ and charged bosons¹⁴ allows us to describe the quantum Bose glass-superfluid transition, the shape of the scaling functions as well as the thermodynamics of each phase away from the transition remain unknown. A physically plausible model of neutral¹⁵ and charged bosons¹⁶ in a random potential based on a separation of localised single-particle states from delocalised states might be helpful. As discussed in Ref.¹⁴, such separation, strictly speaking, is not possible as the localised states will be subsumed into the collective mode. Nevertheless,

the effect of this mode can be understood by analysing the high density limit where the dimensionless Coulomb repulsion is small: $r_s = 4me^2/\epsilon_0(4\pi n/3)^{1/3} \leq 1$. Here m is the boson mass, n is the density and ϵ_0 the static dielectric constant of the host material (and we take $\hbar = 1$). In this limit the excitation spectrum at $T = 0$ was calculated by Foldy¹⁷, who worked at zero temperature using the Bogoliubov¹⁸ approach. The Bogoliubov method leads to the result that the elementary excitations of the system have, for small momenta, energies characteristic of plasma oscillations which pass over smoothly for large momenta to the energies characteristic of single particle excitations. They screen an external charge with a screening radius, which is temperature independent at low temperatures^{19,20}. At high density, $r_s \ll 1$, one can expect a disappearance of localised states because of the screening similar to the Mott metal-insulator transition in doped semiconductors.

It is more likely, however, that the dimensionless strength of the Coulomb repulsion is about unity or larger, $r_s \geq 1$, and it is only when r_s is very large $r_s \gg 1$ that the Wigner crystallisation of charged carriers occurs. Hence, there is a wide interval of densities where the localised states coexist with a superfluid. Being screened they depend on the interaction and the superfluid density. Nevertheless, based on the screening in the high density limit^{19,20} we expect that the density of localised states ρ_L near the mobility edge, E_c remains temperature independent at low temperatures ($k_B T \rho_L \ll 1$). If this is so, then the renormalisation of the effective single-particle energies by the collective mode does not affect the temperature dependence of any of the thermodynamic functions at low temperatures.

With all these reservations we assume that at some finite temperature T_c , bosons are condensed at $E = E_c$ so that the chemical potential $\mu = E_c$. The excitation spectrum of the delocalised superfluid has a gap of the order of the plasma frequency¹⁷ and so the Bogoliubov collective modes can be ignored in the thermodynamics of the system (in 3D their contribution is exponentially small while in 2D their energy scales as T^5 and the specific heat as T^4). Even in the case of a short range repulsion the sound modes yield an energy proportional to T^{d+1} and hence a specific heat which behaves like $C \propto T^d$ (where $d = \text{dimensionality}$)¹³. Thus, for $d \geq 2$, the contribution to thermodynamics from the delocalised bosons appears to be negligible at low temperatures compared with that from bosons localised in shallow potential wells. Hence in the following we calculate the partition function and specific heat of localised bosons only.

For simplicity, we choose

$$E_c = 0 \quad (1)$$

When two or more charged bosons are in a single localised state of energy E there may be significant Coulomb energy and we try to take this into account as follows. The localisation length ξ is thought to depend on E via

$$\xi \propto \frac{1}{(-E)^\nu} \quad (2)$$

where $\nu > 0$. The Coulomb potential energy of p charged bosons confined within a radius ξ can be expected to be

$$\text{potential energy} \sim \frac{p(p-1)e^2}{\epsilon_0 \xi}. \quad (3)$$

Thus the total energy of p bosons in a localised state of energy E is taken to be

$$w(E) = pE + p(p-1)\kappa(-E)^\nu \quad (4)$$

where $\kappa > 0$. Hence we see that the behaviour of charged bosons in localised states can be thought of as intermediate between Bose-Einstein statistics and Fermi-Dirac statistics. When $\kappa = 0$ we have an equally spaced set of levels, i.e. Bose-Einstein behaviour, whereas when $\kappa = \infty$ we have Fermi-Dirac behaviour since the only levels with finite energy are $p = 0$ and $p = 1$, thus enforcing an exclusion principle. When $0 < \kappa < \infty$ we have the intermediate ‘parastatistics’ that the level spacing $\delta w \rightarrow \infty$ as $p \rightarrow \infty$.

We take the total energy of charged bosons in localised states to be the sum of the energies of the bosons in the individual potential wells. The partition function Z for such a system is then the product of the partition functions for each of the wells, and the system free energy $F = k_B T \ln Z$ is simply the sum of the individual free energies $k_B T \ln z(E)$. The free energy of the localised bosons in one unit cell is then given by

$$F = k_B T \int_{-\infty}^0 dE \rho_L(E) \ln z(E), \quad (5)$$

where $\rho_L(E)$ is the one-particle density of localised states per unit cell below the mobility edge.

In this paper we assume the following physically plausible density of localised states $\rho_L(E)$:

$$\rho_L(E) = \frac{n_L}{\gamma} e^{\frac{E}{\gamma}} \quad (6)$$

We shall see below that, in the most interesting case, if the width of the tail γ is large compared with $k_B T$, the specific heat is insensitive to the details of the shape of $\rho_L(E)$, depending only on the value of $\rho_L(0)$.

III. THE THERMODYNAMICS OF A SINGLE POTENTIAL WELL

Before considering the thermodynamics of charged bosons in our assumed density of localised states, we first summarise here the results we have previously obtained¹⁶ for the thermodynamics of a single potential well with only one single particle level of energy ϵ , i.e. for the case

$$\rho_L(E) = n_L \delta(E - \epsilon) \quad (7)$$

The probability for this state to be occupied by p bosons is proportional to

$$e^{-\beta\{w(\epsilon)-p\mu\}}$$

where μ is the chemical potential and $\beta \equiv 1/k_B T$. We can re-express $w - p\mu$ as

$$w - p\mu = \kappa(-\epsilon)^\nu (p - p_0)^2 - \kappa(-\epsilon)^\nu p_0^2 \quad (8)$$

where

$$p_0 = \frac{1}{2} + \frac{\mu - \epsilon}{2\kappa(-\epsilon)^\nu} \quad (9)$$

Fig 1 shows a graph of $w - p\mu$ as a function of p . The partition function $z(\epsilon)$ for such a single localised state is

$$\begin{aligned} z(\epsilon) &= \sum_{p=0}^{\infty} e^{-\beta(w-p\mu)} \\ &= e^{p_0^2 \beta \kappa(-\epsilon)^\nu} \sum_{p=0}^{\infty} e^{-\beta \kappa(-\epsilon)^\nu (p-p_0)^2}. \end{aligned} \quad (10)$$

The partition function is thus completely determined by the dimensionless parameters p_0 and $k_B T / [\kappa(-\epsilon)^\nu]$.

The corresponding mean occupancy $\langle p \rangle$ given by

$$\langle p \rangle = k_B T \frac{\partial \ln z(\epsilon)}{\partial \mu} \quad (11)$$

and specific heat capacity c given by

$$c = \beta^2 \frac{\partial^2 \ln z(\epsilon)}{\partial \beta^2}. \quad (12)$$

are shown in Figs 2 and 3 for the case $\mu = 0$. These results were calculated by simply truncating the partition function series at 100 terms.

We now attempt to understand these results in more detail, looking separately at each temperature range.

1. $k_B T \ll \kappa(-\epsilon)^\nu$

At low temperatures the partition function is dominated by the term with p closest to p_0 , i.e. the value of p giving the lowest value of $w - p\mu$, and so the mean occupancy $\langle p \rangle$ is an integer and goes up in steps as p_0 increases, as seen in Fig 2. The changeover in dominance from one term to another occurs when p_0 is a half-integer, at which point the two lowest energy states are degenerate.

So long as one term dominates the partition function, the specific heat c will be close to zero. However when p_0 is close to a half-integer we have a two level system and a corresponding Schottky anomaly in the specific heat capacity. Hence, at fixed temperature, the low temperature specific heat capacity (i.e. $k_B T \ll p_0^2 \kappa(-\epsilon)^\nu$) is periodic in p_0 .

2. $k_B T > \kappa(-\epsilon)^\nu$

We can approximate the sum by an integral

$$z(\epsilon) \approx e^{p_0^2 \beta \kappa(-\epsilon)^\nu} \int_0^\infty dp e^{-\beta \kappa(-\epsilon)^\nu (p-p_0)^2} \quad (13)$$

• $\kappa(-\epsilon)^\nu < k_B T < p_0^2 \kappa(-\epsilon)^\nu$

In this case we can approximate the lower limit of the integral as $-\infty$, i.e. the partition function can be approximated by an untruncated gaussian, and is therefore approximately symmetrical about p_0 . Hence, in this temperature range we have

$$\langle p \rangle \approx p_0 \quad (14)$$

as is clearly seen in Fig 2, and

$$c \approx \frac{1}{2} k_B \quad (15)$$

as seen in Fig 3.

• $k_B T > \kappa(-\epsilon)^\nu$ and $k_B T \gg p_0^2 \kappa(-\epsilon)^\nu$

In this case we can approximate the partition function as being an integral over half a Gaussian. We then obtain

$$\langle p \rangle \approx \sqrt{\frac{k_B T}{\pi \kappa(-\epsilon)^\nu}} \quad (16)$$

and the specific heat is again given by $c \approx k_B/2$; both of these results are seen in Figs 2 and 3.

IV. THE NUMBER OF BOSONS IN LOCALISED STATES

Having established the partition function $z(E)$ for a single potential well containing one single particle level of energy E we now use equations 5 and 6 to derive the thermodynamics of localised bosons.

The average number N_L of bosons in localised states in each unit cell is

$$N_L = \int_{-\infty}^0 dE \langle p \rangle \rho_L(E) \quad (17)$$

$\langle p \rangle$ can be approximated as

$$\langle p \rangle \approx \begin{cases} p_0 & \text{if } k_B T < p_0^2 \kappa(-E)^\nu \\ \sqrt{\frac{k_B T}{\pi \kappa(-E)^\nu}} & \text{otherwise} \end{cases} \quad (18)$$

Hence a reasonable approximation for $\langle p \rangle$ at all temperatures is

$$\langle p \rangle \approx \frac{1}{2} + \frac{1}{2\kappa(-E)^{\nu-1}} + \sqrt{\frac{k_B T}{\pi\kappa(-E)^\nu}} \quad (19)$$

With this approximation we obtain

$$N_L = n_L \left\{ \frac{1}{2} + \frac{\Gamma(2-\nu)}{2\kappa\gamma^{\nu-1}} + \sqrt{\frac{k_B T}{\pi\kappa\gamma^\nu}} \Gamma(1 - \frac{\nu}{2}) \right\} \quad (20)$$

Hence if $\nu > 2$ then N_L becomes infinite and so the formation of a superfluid is excluded.

V. THE SPECIFIC HEAT OF LOCALISED BOSONS

We have shown above that the contribution to the specific heat c from a single localised state is determined solely by the values of two dimensionless quantities: p_o and $k_B T / [\kappa(-E)^\nu]$. If ν is temperature independent, c can instead be considered as a function of a different pair of quantities: $-E/[k_B T]$ and $\tau = \kappa(k_B T)^{\nu-1}$. The integrated specific heat C of the localised bosons is then

$$C = \int_{-\infty}^0 dE \rho_L(E) c\left(\frac{-E}{k_B T}, \tau\right) \quad (21)$$

A. Specific heat for $\nu = 1$

In this case $\tau = \kappa$ and is therefore temperature independent. Thus we obtain:

$$C = k_B T \int_{-\infty}^0 d\left(\frac{-E}{k_B T}\right) \rho_L(E) c\left(\frac{-E}{k_B T}, \kappa\right) \quad (22)$$

If $\rho_L(E)$ is constant in the region $-E < \sim k_B T / \kappa$ then equation 22 yields $C \propto T$; for ρ_L given by equation 6 this result applies if $\gamma \gg k_B T / \kappa$.

B. Specific heat for $\nu > 1$

For $\nu > 1$ the integral in equation 21 can be thought of as an integral over three regions:

$$1. -E/[k_B T] < \tau^{-1/\nu} \quad \{\text{i.e. } p_o > \frac{1}{2}(1 + \tau^{-1/\nu})\}$$

In this region $k_B T > \kappa(-E)^\nu$ and so

$$c \approx \frac{1}{2} k_B \quad (23)$$

$$2. \tau^{-1/\nu} < -E/[k_B T] < \tau^{-1/(\nu-1)}$$

$$\{\text{i.e. } \frac{1}{2}(1 + \tau^{-1/\nu}) > p_o > 1\}$$

This region only exists if $\tau < 1$. c is dominated by Schottky anomalies, each centred on a half-integer value of p_o . The last full Schottky anomaly in the series is at $p_o = \frac{3}{2}$, so the total number s of Schottky anomalies is

$$s = \frac{1}{2} \tau^{-1/\nu} - \frac{1}{2} \quad (24)$$

If we ignore all except the lowest two energy levels then for each anomaly

$$\int_{below}^{above} dE c\left(\frac{-E}{k_B T}, \tau\right) = \frac{\pi^2}{3(\nu-1)} k_B^2 T \quad (25)$$

$$3. -E/[k_B T] > \tau^{-1/(\nu-1)} \quad \{\text{i.e. } p_o < 1\}$$

Here the $p = 1$ level is the lowest energy level, and the $p = 0$ level is the next lowest. If higher levels are neglected then c is solely a function of $-E/[k_B T]$ and is non-negligible only for $0.1 < -E/[k_B T] < 10$. Hence this region can be neglected entirely if $\tau < (\frac{1}{10})^{\nu-1}$. If, on the other hand, $\tau > 10^{\nu-1}$ the integrated specific heat for this region will be that of half a Schottky anomaly:

$$\int_{region 3} dE c\left(\frac{-E}{k_B T}, \tau\right) \approx \frac{1}{6} \pi^2 k_B^2 T \quad (26)$$

If $\kappa\gamma^{\nu-1} \gg 1$ then $e^{E/\gamma} \approx 1$ throughout regions (1) and (2). If $\gamma \gg k_B T$ then $e^{E/\gamma} \approx 1$ throughout region (3). We take both of these conditions to hold so that equation 21 can be re-written as

$$\frac{C}{T} \approx \frac{n_L k_B}{\gamma} \int_0^\infty d\left(\frac{-E}{k_B T}\right) c\left(\frac{-E}{k_B T}, \tau\right) \quad (27)$$

i.e. if ν is fixed then C/T is solely a function of τ .

Hence we can distinguish two cases for which an approximate analytical formula for C can be derived:

$$1. \tau < \left(\frac{1}{10}\right)^{\nu-1}$$

In this case region (3) can be neglected. From equations 23, 24 and 25 we then obtain

$$\frac{C}{T} \approx \left[\frac{\pi^2}{6(\nu-1)} + \frac{1}{2} \right] \frac{n_L k_B^2}{\tau^{1/\nu} \gamma} \quad (28)$$

This implies

$$C \propto T^{1/\nu} \quad (29)$$

i.e. the specific heat has a power law dependence on temperature in which the power is less than unity.

2. $\tau > 10^{\nu-1}$

In this case the specific heat C is dominated by region (3). From equation 26 we then obtain

$$\frac{C}{T} \approx \frac{\pi^2 n_L k_B^2}{6\gamma} \quad (30)$$

i.e. C/T is temperature independent.

Fig. 4 compares these two formulae (for the case $\nu = 1.5$) with the result of calculating C from equation 27 by numerical integration (in which c is calculated from the first 100 terms of the partition function).

C. Specific heat for $\nu < 1$

Now p_o increases as $-E$ increases. Again we can distinguish three regions

1. $-E/[k_B T] < \tau^{-1/\nu}$ {i.e. $p_o < \frac{1}{2}(1 + \tau^{-1/\nu})$ }

Here $k_B T > \kappa(-E)^\nu$ and so once again $c \approx k_B/2$.

2. $\tau^{-1/\nu} < -E/[k_B T] < \tau^{1/(1-\nu)}$

{i.e. $\frac{1}{2}(1 + \tau^{-1/\nu}) < p_o < 1$ }

Here the partition function is dominated by the two levels corresponding to $p = 1$ and $p = 0$. If $\tau < 1$ this region does not exist.

3. $-E/[k_B T] > \tau^{1/(1-\nu)}$ {i.e. $p_o < 1$ }

Here c is dominated by an infinite series of Schottky anomalies centred on values of E corresponding to half-integer values of p_o :

$$-E = [(2p_o - 1)\kappa]^{\frac{1}{1-\nu}} \quad (31)$$

We consider the low temperature limit, i.e. $\tau \gg 1$ (for $\nu < 1$). Let

$$\chi = \frac{-E}{\gamma} \quad (32)$$

Equation 21 becomes

$$C = n_L \int_0^\infty e^{-\chi} c d\chi \quad (33)$$

In the low temperature limit $k_B T \ll \kappa \gamma^\nu$, region 1 makes a negligible contribution to the integral in equation 33. Provided $k_B T \ll (1 - \nu)\gamma$ we can make the approximation that, when integrating through each Schottky anomaly, $e^{-\chi}$ can be taken to be a constant depending on the anomaly; hence the contribution from each anomaly is proportional to the result given in equation 25 (and equation 26). Thus, in the low temperature limit, we have $C \propto T$.

If $\gamma^{1-\nu} \gg \kappa$ then many Schottky anomalies contribute significantly to the integral in equation 33. In that case we can neglect region 2 and transform the sum over Schottky anomalies into an integral with respect to p_o , obtaining:

$$\begin{aligned} C &\approx \frac{\pi^2 n_L k_B^2 T}{3(\nu - 1)\gamma} \int_0^\infty e^{-\chi} dp_o \\ &\approx \frac{\pi^2 n_L k_B^2 T}{6\kappa \gamma^\nu} \int_0^\infty \chi^{-\nu} e^{-\chi} d\chi \\ &\approx \frac{\pi^2 k_B^2 n_L \Gamma(1 - \nu)}{6\kappa \gamma^\nu} T \end{aligned} \quad (34)$$

In the high temperature limit, on the other hand, we have $\tau \ll 1$ and $k_B T \gg \kappa \gamma^\nu$; in that case only region 1 is significant and we obtain:

$$C \approx \frac{1}{2} n_L k_B \quad (35)$$

VI. CONCLUSION

We have used a reasonable scaling of Coulomb energy with localisation length to calculate the low temperature dependence of the specific heat of a charged superfluid in a random potential. The result strongly depends on the exponent ν of the localisation length. While the specific heat is linear for $\nu \leq 1$, it is proportional to $T^{1/\nu}$ for $\nu > 1$.

We believe that our findings are relevant for those doped high- T_c cuprates having many properties of a charged Bose-liquid¹. Because of the high level of doping of these Mott-Hubbard insulators in the superconducting region one can expect a continuous density of states $\rho_L(E)$ similar to that studied by us; our prediction about the power law dependence of C (for $\nu > 1$) requires, of the density of states, only that $\rho_L(E_c) \neq 0$.

Superconducting $La_{2-x}Sr_xCuO_4$ has been observed¹⁰ to have a low temperature specific heat proportional to T^α with α being in the range $0.33 < \alpha < 0.78$ dependent on doping. This fits with our prediction for charged bosons partly localised by disorder at low temperature and $\nu > 1$. The doping dependence of $\alpha = 1/\nu$ could be due to a gradual change of the localisation length exponent ν caused by a difference in the random potential profiles for different x .

Values of α as low as 0.33 are observed¹⁰ experimentally while our model excludes the possibility of superconductivity for $\alpha = 1/\nu \leq 0.5$. However we believe that the approximation for Coulomb potential energy in equations 3-4 is likely to be inadequate at large ν .

Enlightening discussions with N. Hussey, V. Kabanov, F. Kusmartsev, J. Samson, and K.R.A. Ziebeck are highly appreciated.

FIG. 1. Graph of $(w - p\mu)/[\kappa(-E)^\nu]$ against p .

FIG. 2. The mean occupancy $\langle p \rangle$ of a single level as a function of p_0 and $\log_{10} \{k_B T / [\kappa(-\epsilon)^\nu]\}$.

FIG. 3. The specific heat capacity c of a single level as a function of p_0 and $\log_{10} \{k_B T / [\kappa(-\epsilon)^\nu]\}$.

FIG. 4. Specific heat capacity C divided by temperature T for the case $\nu = 1.5$. The continuous curve is obtained from numerical integration of the integral in equation 27, c being evaluated by direct summation of the first 100 terms of the partition function. The two dashed lines are the predictions from equations 28 and 30.

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