Pair–pair interactions as a mechanism for high-\(T_c\) superconductivity

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Abstract

The mutual interaction between Cooper pairs is proposed as a mechanism for the superconducting state. Above \(T_c\), pre-existing but fluctuating Cooper pairs give rise to the unconventional pseudogap (PG) state, well-characterized by experiment. At the critical temperature, the pair–pair interaction induces a Bose-like condensation of these preformed pairs leading to the superconducting (SC) state. Below \(T_c\), both the condensation energy and the pair–pair interaction \(\beta\) are proportional to the condensate density \(N_{sc}(T)\), while the usual Fermi-level spectral gap \(\Delta_p\) is independent of temperature. The new order parameter \(\beta(T)\), can be followed as a function of temperature, carrier concentration and disorder—i.e. the phase diagrams. The complexity of the cuprates, revealed by the large number of parameters, is a consequence of the coupling of quasiparticles to Cooper-pair excitations. The latter interpretation is strongly supported by the observed quasiparticle spectral function.

Keywords: cuprates, unconventional superconductivity, pseudogap phenomena, novel mechanisms

1. Introduction

As Occam’s razor would suggest, second order phase transitions often depend on few parameters [1]. Such is the case for the familiar magnetic, spin glass, charge-density wave, structural transitions, etc, and the more exotic Kosterlitz–Thouless case for two-dimensional systems [2]. The superconducting phase of ‘classical’ materials follows this trend wherein a weak attractive electron–electron interaction is responsible for the transition. Essentially a single energy scale, the SC gap parameter \(\Delta_0\) at zero temperature, is relevant. In the conventional theory of Bardeen, Cooper, Schrieffer (BCS) [3] pair-breaking quasiparticle excitations restore the normal-metal state at the transition, such that \(\Delta_0 = 1.76 k_B T_c\). Moreover, \(\Delta_0\) fixes the scale of the critical currents, for example in a Josephson junction, the upper critical field and the coherence length, such as the vortex core radius.

In highly-correlated electron systems such as cuprates, many physical properties of their SC state are not yet fully understood. In addition to many theoretical ideas [4–9] the experiments reveal an inherent complexity: a complex quasiparticle self-energy [10–12], coupling to a spin collective mode [10–18], anomalous specific heat [19], multiple energy scales [20–23], spatial inhomogeneities [24–29], checkerboard oscillations [30, 31], pseudogap phenomena above \(T_c\) [7, 19, 32–35], etc. The SC state of cuprates is indeed far from the desired \(\textit{lex parsimoniae}\).

The clear jump in magnitude of the physical parameters is also striking. In addition to \(T_c\), the very small coherence length, large penetration depth, large Fermi-level gap value, small lower critical-field, etc, come to mind. In the case of cuprates, the precise link of the measured Fermi-level gap to the condensate is still controversial. Consider BiSrCaCuO (2212) as a typical example. The energy gap is \(\Delta \approx 32\) meV near optimal doping \((p)\) which, if directly tied to the critical temperature using the BCS ratio, would give the erroneous value of \(T_c \sim 220\) K. Moreover, on the underdoped side of the phase diagram the energy gap increases while \(T_c\) decreases—a clear paradox. Thus, \(\Delta\) is not the scale of \(k_B T_c\).
The quasiparticle (QP) spectral function, expressing the material’s microscopic interactions, also contradicts the conventional picture \([10–12, 18, 36, 37]\). Measured by tunneling or ARPES as a function of rising temperature, the cuprate SC gap does not close at \(T_c\) as would be expected, but rather a pseudogap remains at the Fermi level. To within thermal broadening, the pseudogap has about the same value as the SC gap, with no coherence peaks, and it finally vanishes at a higher temperature \(\sim T^*\) \([37]\). An analogous PG exists in the vortex core \([38–40]\) or due to disorder \([25, 28]\), where SC coherence is also lost. Thus in the case of cuprates, the direct sign of the condensate is not the gap \(\text{per se}\), but instead the finer structure of the QP spectral function (peak-dip-hump).

Very different interpretations have been proposed: the two-energy scale gap function, implying a non-retarded pairing interaction \([22, 41, 42]\), and the coupling to a spin collective mode \([11–18, 43–46]\). Although phenomenological, the static model describes very well the particular signatures of the spectral function, the SC density of states (DOS), and the transition to the pseudogap state \([22]\). Still, the origin of these signatures remains to be clarified.

Many studies with scanning tunneling spectroscopy (STS) \([24–29, 40]\), showed real-space inhomogeneities even for pristine samples at low temperature. According to conventional wisdom, without an applied magnetic field or current, the SC order parameter should be homogeneous. The phenomenon has led to in-depth theoretical studies of the mechanisms underlying long-range SC order with particular emphasis on the effects of disorder and percolation \([47–50]\). By studying a variety of spectral data, we find that they have a distinctive shape on the mesoscopic scale; the underlying parameters then become meaningful.

To summarize, a solution of the high-\(T_c\) problem must account for:

(i) multiple energy scales and large spectral gap \(\Delta_p\),
(ii) large \(T_c\), but paradoxically: \(k_B T_c \ll \Delta_p\),
(iii) the dome-shape of the \(T – p\) phase diagram,
(iv) the SC to pseudogap transition,
(v) the unconventional QP dispersion (peak-dip-hump).

In this work a microscopic theory is presented that addresses these requirements. We assume the existence of pre-existing fluctuating pairs above \(T_c\) in the incoherent pseudogap state \([6–9, 21, 34, 51]\). Since the carrier-density and coherence length are small, a system of interacting bosons is relevant; a new pair–pair interaction, \(\beta_{ij}\), leads to a Bose-like condensation to the coherent SC state at the critical temperature (section 3).

Simple expressions for the gap equation and the condensation energy are given and the nature of the SC to PG transition is described. A new order parameter \(\beta(T)\) naturally emerges, whose properties are examined as a function of temperature, carrier concentration and disorder—are the phase diagrams. It extends the notion of ‘pair field’ that we reported in the coarse-grain approach \([42]\), and gives a direct link between the SC coherence and the parameters of the QP spectral function.

While the Bose-like condensation of preformed pairs is relatively straightforward, the interpretation of the experimental QP spectra suggests a larger number of parameters. In this work we argue that the further complexity is due to the coupling of quasiparticles to Cooper-pair excitations. This proposition is strongly supported by the variety of experimental spectra, analysed in sections 4 and 5, and other widely accepted properties of high-\(T_c\).

2. Hamiltonian for interacting pairs

In the absence of pre-formed pairs, the so-called ‘pairing’ Hamiltonian \([52]\) takes the form:

\[
H = H_0 + H_{\text{pair}} + H',
\]

where, in standard notation

\[
H_0 = \sum_{k,\sigma} e_k c_{k,\sigma}^\dagger c_{k,\sigma},
\]

\[
H_{\text{pair}} = -\sum_k \left( \Delta_k b_k^\dagger + \Delta_k^* b_k - \Delta_k \left( b_k^\dagger b_k^\dagger \right) \right)
\]

with: \(b_k = c_{-k,\uparrow} c_{k,\downarrow}\) and \(H'\) is bilinear in the fluctuation terms: \(b_k - \langle b_k \rangle\). Now, in the problem we are addressing, we assume pre-existing pairs that have the gap/energy distribution \(\Delta_i\) where \(i\) labels an energy state with the probability \(P_0(\Delta_i)\). We assume that these pairs are still described by the BCS model, so that equation (3) takes the form:

\[
H_\text{pair} = -\sum_i \sum_k \left( \Delta_k b_k^\dagger + \Delta_k^* b_k \right)
\]

after dropping the constant term. We assume that the bilinear fluctuating terms in \(H'\) that are diagonal in \(i\) only renormalize the pair amplitude. On the contrary, we keep the non-diagonal terms such that a new mutual interaction between pairs emerges. We find \(H' = H_{\text{int}}\) can be written:

\[
H_{\text{int}} = \frac{1}{2} \sum_{i,j,k,l} \beta_{ijkl}^b b_i^\dagger b_j^\dagger b_l b_k + \text{h.c.},
\]

where \(\beta_{ijkl}^b\) is the microscopic coupling. This term is neglected in the conventional approach.

In the absence of the pair–pair interaction \(H_{\text{int}} = 0\), then \(H_{\text{PG}} = H_0 + H_{\text{pair}}\) describes a pseudogap state of uncondensed pairs having a binding energy compatible with \(\sim T^* \gg T_c\). In general, \(\Delta_i\) can be complex when taking into account phase fluctuations. In our model, however, we consider only the real energy distribution. The justification is that clear experimental evidence for phase fluctuation effects is lacking: the Kosterlitz–Thouless, transition, for example, is not pertinent at these energy scales.

The fluctuating amplitudes \(\Delta_i\) (now real), appearing in equation (4), are distributed according to:

\[
P_0(\Delta_i) = \frac{\sigma_0^2}{(\Delta_k - \Delta_{0,k})^2 + \sigma_0^2},
\]

where \(\Delta_{0,k}\) and \(\sigma_0\) are the average pairing energy and width, respectively. These two parameters will remain central to our model throughout this work.
In the non superconducting state, $H_{SC}$, can be diagonalized for each state $\Delta^\pm$ independently using the canonical BCS–Bogoliubov transformation, leading to the spectrum:

$$E'_k = \sqrt{\frac{\epsilon_k}{2} + \left(\Delta^+\right)^2};$$

the final PG state being a simple superposition. The corresponding spectral function is:

$$A(\epsilon_k, E) = -\frac{1}{\pi} \text{Im} \int_0^\infty d\Delta^+ P_0(\Delta^+) \frac{1}{E - E'_k + i \Gamma},$$

where $\Gamma$ is the Dynes QP broadening parameter [53]. As shown in [42], the above spectral function and corresponding DOSs leads to a smeared gap of width $\Delta_0$ at the Fermi level characterized by vanishing coherence peaks (similar to figure 4, spectrum 5).

The interaction between Cooper pairs, $\beta^{ij}_{k,k'}$, responsible for the superconducting state, is a key ingredient of the model. In close analogy with the original BCS coupling $V_{k,k'}$, where two fermions interact only within an energy window $\hbar\omega_D$ with respect to the Fermi level, we assume that the pre-formed pairs interact in the energy window $\pm \epsilon_0$ with respect to $\Delta_0$. This property is satisfied by choosing the form:

$$\beta^{ij}_{k,k'} = g_k g_{k'} P_0(\Delta^+_k) P_0(\Delta^+_k')$$

where the factor $g_k$ preserves the $d$-wave symmetry, and $P_0(\Delta^+_k)$ is the pair distribution. With this potential the equalizing of the $\Delta^+_k$ in the final SC state is favored while retaining the memory of the initial state. It has the useful property of being separable, which allows for the decoupling of the equations.

In the mean-field approximation, at $T = 0$, the operator $b^+_k$ is replaced by its quantum average: $\langle b^+_k \rangle = \Delta^+_k/(2 E'_k)$, so that the effective interaction reduces to:

$$H_{int} = \sum_{i,k} 2 \beta_k P_0(\Delta^+_k) b^+_k + \text{h.c.},$$

where

$$\beta_k = g_k \sum_{j,k'} g_{k'} P_0(\Delta^+_k) \Delta^+_k \frac{4 \pi}{8 E'_k}$$

which defines $\beta_k$ in terms of all other pairs $j \neq i$ in the system.

We assume that in the PG state, $\beta_0$ is negligible due to the fluctuations of $\Delta_0$, see equation (10). On the contrary, upon pair condensation all the bosons must be in the same quantum state, at $T = 0$, so that $\Delta^+_k \rightarrow \Delta_k$, which no longer depends on $j$. One has:

$$\beta_k = N_p \times g_k \sum_k g_{k'} P_0(\Delta^+_k) \frac{4 \pi}{8 E'_k}$$

which becomes large due to the factor $N_p$, the number of condensed pairs. Moreover, equation (11) is a new self-consistent equation which depends on the exact $\Delta_k$ and the corresponding QP excitations:

$$E_k = \sqrt{\frac{\epsilon_k}{2} + \left(\Delta_k^\pm\right)^2}.$$  

To obtain the SC gap equation, we first assume that all pairs join the condensate (a condition we relax in section 3):
\(\Delta_p \approx 38\) meV. The three characteristic energies: \(\Delta_p, \varepsilon_c,\) and \(\Delta_0 = \Delta_p + \varepsilon_c,\) are illustrated in the phase diagram figure 1.

The interaction \(\beta\) has a dominant effect on the energy changes involved in the transition. To illustrate, consider the BCS expression for the self-consistent gap:

\[
\Delta_p = 2E_0 e^{-1/(\hbar \nu_0)},
\]

where \(E_0\) is the cut-off energy, \(N_0\) is the Fermi-level DOS and \(V_p\) is the total pair potential. Using \(V_p = V_0 + V_{\text{int}} = V_0(1 - \beta),\) where \(\beta\) is the relative pair–pair interaction, we assume \(\Delta_0\) and \(\varepsilon_c\) are of the form:

\[
\Delta_0 = \Delta_p(1 + \beta) \quad \text{and} \quad \varepsilon_c = B \Delta_p.
\]

As illustrated in figure 2, the spectral gap \(\Delta_p\) decreases sharply as a function of \(\beta,\) becoming vanishingly small as \(\beta \to 1.\) \(\Delta_0\) begins equal to \(\Delta_p,\) for \(\beta = 0,\) and then follows a similar trend as \(\Delta_p,\) albeit with \(\Delta_0 > \Delta_p.\) The main observation is that \(\varepsilon_c\) is nearly dome-shaped—a general property of high-\(T_c\) and indeed other types of superconductors. Here, the pair–pair interaction \(\beta\) causes the rapid decrease in \(\Delta_p\) and the dome-shape of the condensation energy.

The physical origin of the pair–pair interaction within the condensate can now be addressed. We consider that it is a non-retarded interaction mediated directly by the quasiparticles. Consequently it is long-range, implying that \(\beta\) is proportional to the number of condensed pairs \(N_{\text{oc}}(T).\) It can therefore be written:

\[
\beta(T) = \beta_0 N_{\text{oc}}(T).
\]

The effect of this interaction on the condensation mechanism is investigated in the following section 3 and on the quasiparticle spectrum in section 4.

### 3. Pair condensation

Consider the phase diagram of figure 1 in the context of uncondensed pairs above \(T_c.\) Qualitatively, the average pair energy \(\Delta_p\) follows the critical curve shown and vanishes at \(\sim T^*.\) In particular, the SC critical point, denoted by \(C,\) is situated on the plateau of the critical curve where \(\Delta_p,\) is about equal to its zero-temperature value. The fact that \(\Delta_p,\) remains constant in the SC to PG transition can be directly seen as a function of temperature [36, 37], disorder [25, 28] and in the vortex core [38, 39].

In the BCS gap equation, the reduction of the gap with rising temperature below \(T_c\) is associated with quasiparticle excitations given by the factor:

\[
1 - 2f(E_k, T) = \tanh \left( \frac{E_k}{2k_B T} \right),
\]

where \(f(E_k, T)\) is the Fermi–Dirac function and \(E_k\) the QP energy. In the case of cuprates, as the temperature approaches \(T_c\) from below, the smallest value of this function is \(\tanh(\Delta_p/(2k_B T_c)) \sim .96\) and is \(\sim 1\) otherwise. Clearly, pair-breaking linked to thermally excited quasiparticles is not relevant.

Assuming 2D free-electrons for a single conducting plane, we note that the pair density per unit area is about: \(N_p/\Lambda \approx m\Delta_p/(2\pi \hbar^2).\) Then, consider the typical number of pairs within the disk of area \(S = \pi \xi^2,\) with \(\sim \xi\) being a pair diameter:

\[
N_p \approx \frac{m\Delta_p \xi^2}{2\hbar^2} \sim 1 - 2.
\]

Clearly, the combination of small \(\xi\) and small pair overlap strongly supports an interacting boson model.

We thus retain the preformed-pair hypothesis above \(T_c\) and propose that their condensation follows Bose statistics. To proceed, one needs the excited states of the system which are critical. A simple model for the energy needed to remove a pair from the condensate is a mini-gap \(\delta,\) separating the condensate energy

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Figure 2. Influence of a pair–pair interaction, \(\beta.\)

\(\Delta_p\) and \(\varepsilon_c\) are of the form:

\[\Delta_0 = \Delta_p(1 + \beta)\quad \text{and} \quad \varepsilon_c = \beta \Delta_p.\]

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\(\beta(T) = \beta_0 N_{\text{oc}}(T).\) The effect of this interaction on the condensation mechanism is investigated in the following section 3 and on the quasiparticle spectrum in section 4.
from the excited states. We thus write:

$$N_{oc}(T) = 1 - A \int_{\Delta_0 + \delta}^{\infty} d\Delta P_0(\Delta) f_\beta(\Delta - \Delta_p, T)$$

(21)

to calculate the occupation number. The free-energy at the transition can likewise be written [54]:

$$F(T_c) = A \int_{\Delta_p + \delta}^{\infty} d\Delta P_0(\Delta) g(\Delta - \Delta_p, T_c).$$

(22)

where $g(\Delta, T) = -k_B T \ln(1 - \exp(-\Delta/(k_B T)))$.

In the case of the $d$-wave symmetry, the in-plane wave vector $\vec{k} = (k, \theta)$ implies the further integration over $\theta$ so that equation (21) becomes:

$$N_{oc}(T) = 1 - A \int_0^{\pi} d\theta \int_{\Delta_0 \cos \theta + \delta}^{\infty} d\Delta
\times P_0(\Delta) f_\beta(\Delta - \Delta_p \cos \theta, T)$$

(23)

which is the equation used in the problem.

The interpretation of the mini-gap $\delta$ will be discussed later. In practise, we use the experimental value of $T_c$ to determine the normalization $A$ of the pair distribution. For an optimally-doped BiSrCaCuO(2212), implementing equation (23) with $\delta = 2.1$ meV and $T_c = 95$ K leads to $F(T_c) \approx 0.84 k_B T_c$, which is a satisfactory order of magnitude.

The above figure 3 summarizes just how well the Bose condensation model fits the data. First, precise fits to the QP tunneling spectra from Renner et al. [37], as a function of $T$, allow one to determine the parameters $\Delta_0$ and $\sigma_0$, as well as the QP peak heights (PHs). As we showed in our previous work [22, 42], the attenuation of the DOS PH, analogous to the effect shown in the spectra of figure 4, is due to the gradual decrease in the interaction $\beta(T)$, as expected from equation (19). The condensation energy $\varepsilon_c(T)$ is also proportional $N_{oc}(T)$, equation (17), due to its dependence on $\beta(T)$.

In the upper right panel, we plot both the data points from Renner et al. [37] and the best fit of $N_{oc}(T)$ using equation (23), which is excellent except for a slight jump near $T_c$. Unfortunately, similar STS experiments as a function of temperature are rare. In the lower right panel of figure 3 we plot the critical current $I_c R_N$ obtained by Miyakawa et al. using a SIS break-junction setup [55]. We emphasize that the Josephson $I_c R_N$ is directly sensitive to the SC condensate. Here it decreases monotonically with $T$ whereas the gap value $\Delta_p$ remains constant in this temperature range, in agreement with our model. In both experiments, there is a shift in $T_c$ comparing the local vanishing of $N_{oc}(T)$ and the reported bulk value. Still, the quantity $\beta(T) \propto N_{oc}(T)$ has the required properties for an order parameter.

The overall behavior of $N_{oc}(T)$ from underdoped to overdoped BiSrCaCuO(2212) is illustrated in the main panel of figure 3. The curves are quite different from the conventional BCS order parameter having a short plateau towards low-$T$ (spanning ~10 K) then descending rapidly to intersect the $T$-axis with a finite slope. Starting from the fitted estimate of the mini-gap, $\delta = 2.1$ meV, we scaled $\delta$ with $T_c$ using $3.85 \delta = k_B T_c$. The values of the parameters $\Delta_0$ and $\sigma_0$ of the pair distribution were obtained from precise fits to the experimental spectra on BiSrCaCuO(2212), the subject of the following section.

4. Quasiparticle Spectrum

The QP spectral function and DOS, obtained by ARPES and tunneling respectively, contain valuable information on the
Figure 4. Left panel: fits to a series of 5 QP spectra from McElroy et al [28] using equation (29) (dots: experiment, solid line: theory). The spectra are ordered according to the ‘strength’ of their SC features. Inset: experimental spectra on the occupied side before removing background slope. Right panel: values of important parameters from the spectra. $\Delta_D$, $\sigma_0$, and $E_{disp}$ are all moving upwards, following $\Delta_p$. The pair–pair interaction $\beta \propto N_c$ moves oppositely towards lower energy, parallel to the peak heights (PH). $\sigma$ varies from 2 to 16 meV from spectrum 1 to 5 (not shown).

SC state at the microscopic scale. The SC spectral function at zero temperature is:

$$A(\epsilon_k, E) = -\frac{1}{\pi} \text{Im} \frac{1}{E - E_k + i \Gamma},$$

(24)

where $E_k$ is the QP energy. The BCS–Bogoliubov coherence factors are omitted since, as described by Schrieffer [56], they are redundant if $\Delta_k(\pm \epsilon_k)$ is symmetric with respect to the Fermi level.

The QP DOS is obtained directly from the spectral function using: $N_k(E) = 2 \sum\limits_{\epsilon_k} A(\epsilon_k, E)$ where, for a $d$-wave pairing symmetry, $\Delta_k(\theta) = \Delta_k \cos(2\theta)$ leading to the typical $V$-shaped DOS [40, 57, 58]. However, as we have shown repeatedly [22, 41, 42], such a $d$-wave DOS, with weak logarithmic singularities at the coherence peaks and no dip-hump structures, fails to match the experimental spectra.

We have shown that the total Hamiltonian (1) with the interaction term (9) not only expresses the ground-state, with all $\Delta'_k$ within the condensate $\Delta_c = \Delta_k$, but also the pair excited states, with the corresponding distribution, $P_0(\Delta'_k)$. The latter excited states lower the condensate occupation number $N_{oc}(T)$, equation (23), and eventually the SC coherence is lost at $T_c$. How does this relatively simple Bose-like transition affect the QP spectral function?

In our model, the pair–pair interaction is directly mediated by the quasiparticles. An excited pair, i.e. $\Delta'_k > \Delta_k$, can be coupled to a quasiparticle of the same energy and conversely, an excited QP of energy $E'_k$ is linked to such a ‘virtual’ pair. We propose that this energy-conserving coupling is manifested directly in the pair–pair interaction term of the Hamiltonian, equation (9). Indeed, replacing $\Delta'_k$ by $E'_k$ we have:

$$H_{int} = \sum\limits_{i,k} 2 \beta_k P_0(E'_k) b_i^+ b_i + \text{h.c.}$$

(25)

In the final SC state, this leads to a non-retarded energy-dependant gap:

$$\Delta_k(E_k) = \Delta_{0,k} - 2 \beta_k P_0(E_k).$$

(26)

Note that the QP dispersion, while having the same functional form:

$$E_k = \sqrt{\epsilon_k^2 + \Delta_k(E_k)^2}$$

(27)

is in fact greatly modified due to this energy dependence and must be used for calculating the corresponding DOS. With surprisingly few parameters, this QP dispersion with gap function equations (26) and (27) accurately matches the wide peaks and characteristic dips as measured in the spectra, see figure 4.

The derivation of the DOS is given a full treatment in [41, 42]. We recall that, neglecting $\Gamma$-broadening, it depends on the following derivative:

$$N_k(E, \theta) = \frac{N_0}{2 \pi} \int_0^\infty d\epsilon_k \delta(E_k - E) = \frac{N_0}{2 \pi} \left[ \frac{\partial \epsilon_k}{\partial E_k} \right]_{E_k = E},$$

(28)

where $N_k(E, \theta)$ is the partial DOS in the $\theta$ direction. The final DOS therefore involves the energy-derivative of the gap function itself—a rather unusual property. A full treatment,
This unique DOS function is used to fit all the spectra in this work, figures 4 and 5, where the spectral shape is determined by the distribution \( P_0(E_k) \), giving \( \Delta_0 \) and \( \sigma_0 \), and the value of \( \beta \) (see table 1).

Above \( T_c \), there is the additional effect of thermal pair excitations above \( \Delta_p \). Also, pair excitations arise due to disorder, which we take into account by the broadening parameter \( \sigma \) of the pair potential: \( \Delta_p \rightarrow \Delta_p - i \sigma \) in equation (29). Its value is important when \( N_{bc}(T) \ll 1 \) or at low-doping (as shown in figure 6). Otherwise, in all fits, \( \Gamma \sim 1.5 \text{ meV} \), is small.

In the detailed experiments by McElroy et al [28] full STS mappings of BiSrCaCuO(2212) have been performed to study the local SC characteristics and, with atomic resolution, the effects of the local oxygen dopants. Three different samples with nominal average gaps ranging from near optimal to increasingly under-doped (45, 55 and 65 meV) are of particular interest. The DOS variation due to atomic oxygen dopants is quite significant: a resonance near \( \sim 96 \text{ meV} \) is seen on the occupied side of the spectrum and leads to a strong variation of the SC spectral shape.

Two other points in these experiments are significant for the present model. First, local spectra from one sample can be identified with spectra from another sample at locations having common characteristics (gap width, PH, dip position). Although the samples show topological variations, this suggests that on the mesoscopic scale, the SC gap function has consistent properties independent of the sample and its parameters are physically meaningful. Secondly, the optimally-doped sample is much more homogeneous than the other two—suggesting that a percolation effect must be taken into account for very underdoped samples.

The spectra in figure 4 are sequenced according to the ‘strength’ of their superconducting characteristics: in

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**Table 1. Summary of main parameters**

| Parameter          | \( \Delta_p \) | \( \beta \) | \( \Delta_0 \) | \( \sigma_0 \) | \( \sigma \) | \( \delta \) | \( \Gamma \) |
|--------------------|----------------|-------------|----------------|----------------|---------------|--------------|----------|
| SC gap equation    | self-consistent SC gap | pair–pair gap | pair distribution average | pair distribution average | pair broadening | excitation mini-gap | lifetime broadening |
| Condensate         | \( \delta \) | \( \Gamma \) | \( \Delta_0 \) | \( \sigma_0 \) | \( \sigma \) | \( \delta \) | \( \Gamma \) |
| QP spectrum        | \( \sigma \) | \( \delta \) | \( \Gamma \) | \( \Delta_0 \) | \( \sigma_0 \) | \( \sigma \) | \( \delta \) | \( \Gamma \) |

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**Figure 5.** Upper panel: QP DOS using equation (29) to fit a series of 5 spectra A–E from various authors (A: [28], B: [39], C: [55], D: [28], E: [29]) ordered according to gap width: A (overdoped), C (optimal), E (underdoped). (Data only shown in A and E.) Lower panel: plot of the corresponding gap-functions \( \Delta_k(E_k) \) A–E using fitted parameters: gap width \( \Delta_p \), minimum amplitude \( \Delta_0 \), and distribution width, \( \sigma_0 \). These parameters are plotted in the \( T - \rho \) diagram, figure 6.

**Figure 6.** Energy–doping (\( \rho \)) plot summarizing the dependence of the parameters in the present model. Points A–E correspond to the spectra of figure 5. The self-consistent gap \( \Delta_p \) is linear; \( \Delta_0 \), \( \sigma_0 \) and \( E_{\text{dip}} \) are decreasing with \( \rho \) but are slightly convex. At optimal doping \( \varepsilon_c \), the pair–pair interaction \( \beta \) and the mini-gap \( \delta \) are maximal. Both \( \beta \) and \( \sim 7 \delta \) follow the dome-shaped curve: 1.8 \( k_B T_c \).
spectrum 1., a smaller gap width, with sharper QP peaks and in spectrum 5., a larger gap and very attenuated peaks. The spectra with strong SC features largely dominate on the optimally-doped sample, whereas the weak SC spectra are found in mesoscopic regions in the inhomogeneous underdoped sample. The series resembles the SC to PG transition at low-temperature as seen by Renner and Cren [25, 37] and in this context we consider the compatibility of our model.

In figure 4, left panel, we show the fits to the spectra, having removed the background slope and symmetrizing in each case. In the right panel we plot the values of the parameters in the DOS function equation (29) and their variation from spectrum to spectrum.

The first point is that our gap value $\Delta_p$ is 10–20% smaller than the nominal ones cited by McElroy et al. Indeed, the observed spectral gap is affected by the derivative of the gap function at $E_k \approx \Delta_p$, as illustrated in figure 5 lower panel. The measured peak is also affected by the parameter $\sigma$, which reaches $\sim$16 meV for spectrum 5. This damping effect is smaller in the case of sharp QP spectra.

Secondly, it is remarkable how several parameters scale with the spectral gap, in particular $\Delta_0$, $\sigma_0$, and the dip position $E_{\text{dip}}$. On the contrary, the pair interaction parameter $\beta$ moves in the opposite direction, concomitant with the lowering of spectral PHs.

This observed increase of gap width $\Delta_p$ with decreasing $\beta$ is a general consequence of the pair–pair interaction model, equation (16). Moreover, the discussion on the temperature dependance of $\beta$, where we have $\beta(T) \propto N_{\text{c}}(T)$, leads to the conclusion that the condensate occupation density $N_{\text{c}}$ is decreasing from spectrum 1 to 5. This is confirmed by the quantity $\varepsilon_c = \Delta_0 - \Delta_p$, which remains relatively robust in this sequence, and $\sigma_0$, which becomes large. The likely scenario is the effects of disorder in the underdoped regime [47, 50].

In summary, the simple QP spectrum, equation (29), matches remarkably well the measured local DOS of an inhomogeneous SC state, e.g. spectra 3–5. In a given region of the sample, the main effect is thus the change in magnitude of the condensate density $N_{\text{c}}(T)$ on the mesoscopic scale and, through the pair–pair interaction $\beta$, the damping effect on the QP features (PH and dip strength). Lastly, spectrum 5 shows very weak quasiparticle peaks, close to the PG state measured at low temperature by [25, 38, 39]. In our view, the bona fide PG state corresponds to the strict vanishing of both $\beta$ and $\delta$, the two parameters linked to the condensate.

5. Energy phase diagram

We have chosen a wide variety of tunneling DOS from the literature [18, 28, 29, 37, 39, 41, 55, 59] to study the energy scales, their doping dependence and the possible role of the pair–pair interaction. Five of these spectra labeled A–E in figure 5, have been selected for the apparent regularity of their parameters. In the upper panel, the QP DOS, using equation (29), corresponds to underdoped, with larger $\Delta_p \sim 50$ meV, to overdoped, with smallest $\Delta_p \sim 27$ meV.

In the lower panel, figure 5, we show the gap functions $\Delta_k(E_k)$ used to precisely fit each spectrum of the upper panel. The experimental data are only shown for spectra A and E for clarity and, as in figure 4, only the SC part of the spectrum is considered. The quasiparticle peaks in the DOS are fairly sharp, reaching $\sim$1.5–2 above the background level. These are determined by the negative slope of the gap function $\partial \Delta_k(E_k)/\partial E_k$ at $E_k = \Delta_p$. Higher QP peaks have sometimes been measured, in particular [59], in good agreement with our model for large $\beta$ and smaller $\sigma_0$.

Figure 5 reveals the regularity of the dip position, given by the positive derivative of the gap-function near $\sim 2 \Delta_p$, which increases from the overdoped to underdoped sides. As in [22, 41, 42], these characteristics are found without a dynamical collective mode. It remains to be investigated whether similar features seen in the pnictide LiFeAs [44] have the same origin.

The parameters of these five spectra are then plotted in the $T - p$ phase diagram, figure 6. The self-consistent gap $\Delta_p$ follows a linear trend decreasing from underdoped to overdoped sides and extrapolating to zero at $Q$. To serve as a guide, the dome-shaped plot 1.8 $k_B T_c$ is shown. To first approximation, for the doping near the top of the $T_c$-dome, the parameters $\Delta_0$, $\sigma_0$, and $E_{\text{dip}}$ are remarkably continuous and almost linear. This supports the conclusion that, excepting for highly underdoped samples, the parameters deduced from the spectra of figure 5 are reliable.

A closer look reveals the convex shape of $\Delta_0$, with the consequence on $\varepsilon_c = \Delta_0 - \Delta_p$ as well as $E_{\text{dip}}$, also being convex. The dip position [18, 22] evolves from below $2 \Delta_p$ to above $2 \Delta_p$ for increasing $p$, as resolved by many tunneling spectra. On the right-hand side, all curves seem to converge to a single point $Q$.

Of major significance to the present model, the pair–pair interaction follows $T_c$, along with the attractive mini-gap $\delta$, as plotted in the lower part of figure 6. We find $\beta \approx 1.8$ $k_B T_c$ and $\delta \approx 0.26$ $k_B T_c$ with $\beta / \delta \approx 7$. Admittedly, $\beta$ reveals some scatter, sometimes reaching $\sim 2.2$ $k_B T_c$. We note the loss of precision in the fits on the far under-doped side, where the coherence peaks are attenuated. In this regime where $\beta$ decreases sharply, the number of excited pairs, related to $\sigma$, increases markedly, so that $\sigma$ practically joins $\sigma_0$. The available data indicates that, at this extreme left-hand side of the $p$-diagram, both $\beta$ and $\delta$ are vanishing.

In our view, the overall scenario is as follows. A pair–pair interaction, governed by the coupling $\beta_{ij}$, allows for incoherent pairs, with distribution $P_0(\Delta'_k)$ to begin aligning their energies, ultimately to the amplitude $\Delta_0$. Since the interaction is mediated by the quasiparticles, it provides the mechanism for establishing long-range order. The final pair–pair interaction in the SC state, being proportional to the condensate density, $\beta(T) = \beta_0 N_{\text{c}}(T)$, supports this hypothesis.

At low-doping, the coherent state cannot be formed if the distance between pre-formed pairs is too large to be efficiently coupled, since the spatial inhomogeneity that breaks the coherence will result in a cut-off in the interaction. With increased doping, the interaction favors long-range order, the
system becomes more homogeneous, and $T_c$ increases. At the same time, the pair–pair interaction weakens the pair self-energy ($\Delta_p$, in the anti-nodal direction) which decreases uniformly from under-doped to over-doped sides (see figure 6). Thus the SC state becomes weakened again.

In the context of pre-formed pairs, the question of a non-BCS condensation mechanism arises. Indeed, in a 2D system, a conventional Bose condensation is not possible without an inter-particle interaction. We proposed that a mini-gap $\delta$ in the pair excitation spectrum allows for the condensation. The condensate density $N_{cc}(T)$ using this model follows very nicely the experimental data obtained both by single particle tunneling and by Josephson effect (figure 3). The temperature dependence thus has the properties of an order parameter; at the critical temperature, $N_{cc}(T_c) = 0$ and we get back the incoherent pairs of the PG state. The mini-gap $\delta$ is the direct measure of the stability of the coherent state and, just like $\beta$, it follows the $T_c$ versus $p$ curve.

Since $\delta$ is the energy to excite a single pair with respect to the condensate, it can also be viewed as the energy needed to remove a pair from the condensate. No thermal QP excitations, condensation at all and no coherent pair associated with the decrease of $\Delta_p$, is involved. With this mechanism, the properties of the condensate occupation number $N_{cc}(T)$ are in good agreement with available experiments. Moreover, the Fermi-level spectral gap $\Delta_p$ remains constant in this temperature range.

The problem is rendered complex by two additional properties of the pair–pair interaction: $\sim 2$ $\beta(T)P_0(\Delta_p)$. First, after examining a wide range of experimental spectra, we proposed that the excited states are coupled to quasiparticles $\Delta_{\mathbf{k}} \leftrightarrow E_{\mathbf{k}}$, which modifies the final gap equation, equation (26). Once in the superconducting state, these QP excitations become well defined, revealing the characteristic peak-dip structures in the spectral function. The non-retarded energy-gap function described herein is thus the direct consequence of this QP-mediated interaction.

Secondly, the pair–pair interaction also depends directly on the condensate: $\beta(T) = \beta_0 N_{cc}(T)$, which we identify as the order parameter. Not only does it vanish at $T_c$, but it is directly linked to long-range order. This interaction varies spatially in the inhomogeneous superconductor, but also as a function of concentration $p$. In the latter case, the interaction is larger with increasing $p$ which leads to a decrease in the self-consistent gap $\Delta_p$ and consequently to the lowering of $T_c$ as well. Paradoxically, the very potential that is responsible for superconducting order eventually, in the high-density limit, provokes its demise.

6. Conclusion

In this work we have shown that the phase transition to superconductivity from a pseudogap state of preformed pairs can be understood in relatively simple terms and few parameters. It consists of a Bose-like condensation with a small mini-gap $\delta$ above $\Delta_p$ which represents the minimum energy to remove a pair from the condensate.

The low-level pair excitations above $\Delta_p$ are thus critical for the condensation mechanism. No thermal QP excitations, à la BCS, are needed; only the pair excitation distribution $P_0(\Delta_p)$ is involved. With this mechanism, the properties of the condensate occupation number $N_{cc}(T)$ are in good agreement with available experiments. Moreover, the Fermi-level spectral gap $\Delta_p$ remains constant in this temperature range.

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