# Theory of the pseudogap in high-$T_c$ superconductors

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Several different experimental techniques have confirmed the existence of a ''pseudogap'' in normal high-$T_c$ superconductors with low hole densities. Previous quasispin Monte Carlo simulations by the authors have shown that a planar one-band model of the oxygen lattice, with a local pairing interaction, produces uncorrelated bound hole pairs above the superconducting transition temperature when the hole densities are low. Results of normal-state simulations are reported in this work. They provide evidence for the identification of the binding energy of intraplanar uncorrelated hole pairs with the observed pseudogap.
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Tallon *et al.*[1] and Williams *et al.*[2] have compared the results of several experimental determinations of the spin gap, the pseudogap, and the normal-state gap in the 123 and 124 high-$T_c$ superconductors. They argue that, as all these types of gaps appear to have the same dependence on hole density, they should be regarded as different aspects of the same phenomenon. The experimental evidence is consistent with the formation of an energy gap at the Fermi surface in normal-state superconducting cuprates at low hole densities. In the following we refer to this experimentally determined gap as the *pseudogap*. A simple theoretical explanation of the pseudogap is proposed in this work, based on results obtained from temperature-dependent simulations of a *planar* one-band Hamiltonian with a localized pairing interaction.

The present work is based on the ''hole-modulated hopping'' model[3] which, when referred to the oxygen lattice, has been shown to provide a good representation of many of the superconducting properties of copper oxide planes.[3–6] It has been suggested that, because this model produces only extended $s$-wave superconducting pairs, it does not provide as adequate a model of high-$T_c$ superconductivity in some cuprates as does, for example, the $t$-$J$ model. This is to overlook the fact that the $t$-$J$ model is only appropriate for the description of hole pairs on the copper lattice, with near half-filling. There is no contradiction in having local pair solutions which are extended $s$ wave when one of the holes is located on an oxygen ion and $d$ wave when one of the holes is located on a copper ion.

Previous simulation results[4–6] have shown that the superconducting transition takes quite different forms for materials with low and high hole densities. At high hole densities the superconducting transition is BCS-like, with the extended $s$-wave superconducting energy gap $\Delta_s$ going to zero and the coherence length becoming infinite as the system temperature rises above the superconducting transition temperature $T_c$. This is interpreted as a pair-breaking transition. At low hole densities another energy gap (denoted $\Delta_p$) opens up for temperatures above $T_c$. The hole pairs remain small and bound, giving a Bose-like transition, in which superconducting order is lost because there is insufficient correlation energy at low hole densities to maintain it.

We employ the **k**-space quasispin representation developed in Refs. 4–6. This incorporates electronic charges, but supposes all electrons to form singlet pairs, suppressing the spin degrees of freedom. A quasispin eigenstate with $s^z_{\mathbf{k}}=-1/2$ corresponds to a $(\mathbf{k}\uparrow,-\mathbf{k}\downarrow)$ pair; $s^z_{\mathbf{k}}=+1/2$ corresponds to this pair state being empty. Hence the $z$ component of the quasispin changes sign at the Fermi surface. Superconducting order at $T=0$ is represented by a gradual turning over of the quasispin directions near the Fermi surface in such a way that their components in the $x$-$y$ plane are all parallel.[7] When the dispersion is small, most of the quasispins in the Brillouin zone are found to contribute to the stabilization of the $x$-$y$ plane alignment. At finite temperatures, thermal fluctuations of the quasispin directions are superimposed on this simple picture.

The Hamiltonian employed in this work is expressed in terms of quasispin operators[4] as follows:

$$H=-2\sum_k\left(\epsilon_k-\mu+\frac{1}{2}\sum_{k'}V_{kk'}\right)s^z_k+\frac{1}{N}\sum_{k,k'}V_{kk'}s_k\cdot s_{k'}, \tag{1}$$

where $\mu$ is the chemical potential and $N$ is the number of lattice sites. The pairing potential for the hole-modulated hopping model is given by

$$V_{k,k'}=U-K(\cos k_x+\cos k_y+\cos k'_x+\cos k'_y), \tag{2}$$

where $U$ is the on-site (Hubbard) repulsion energy and $K$ is the pairing energy associated with the stimulated hopping of holes between nearest-neighbor sites. The dispersion

$$\epsilon_k=-2t(\cos k_x+\cos k_y)+t'(\cos k_x\times\cos k_y) \tag{3}$$

includes both nearest-neighbor hopping, with matrix element $t$, and next-nearest-neighbor hopping, with matrix element $t'$.

In order to make simulations of superconducting phase transitions practicable it has been found necessary to employ a classical approximation for the quasispins,[4–7] just as has been done in real-space simulations of superconductivity,[8] and in simulations of ferromagnetism (e.g., Ref. 9). This approach has been validated[10] through finite temperature simulations of the original BCS Hamiltonian, for which the mean-field solution is exact. First of all, *classical* mean-field results were obtained by using statistics based on the Brillouin func-

 

tion $B_\infty(E_\mathbf{k}/k_BT)$, rather than $B_{1/2}(E_\mathbf{k}/k_BT)$, where $E_\mathbf{k}$ is the quasiparticle energy at $\mathbf{k}$. This showed that the main effect of using classical spins is to reduce the transition temperature by approximately a factor of 2. Both systems had the same ground state and showed similar behavior near to the transition temperature. Good agreement was obtained between the classical simulation for quite small lattices and the classical mean-field calculation.

The Metropolis algorithm is employed to simulate an ensemble of classical quasispins in $\mathbf{k}$ space at finite temperatures, corresponding to a grand canonical ensemble of hole pairs. Details of this procedure have been given elsewhere.[4] The work reported herein has all been carried out using a $100\times100$ square lattice in $\mathbf{k}$ space. This is the reciprocal of a real-space lattice with periodic boundary conditions, corresponding to the oxygen atoms in the $CuO_2$ planes. Parameter values (in eV) are $t=0.055$, $t'=-0.01$, $V=0.946$, and $U=5.0$. In each simulation, the temperature $T$ and the chemical potential $\mu$ are specified, and the hole density $n$ is determined.[4]

A detailed account of the large number of measurables determined in our simulations, including their interpretation, can be found elsewhere.[4,6] Apart from the hole density $n$, the following measurables are specially relevant to this work:

(i) The *active hole density* ($n_{\mathrm{ACT}}$), specifying the hole density associated with quasispin vector components in the $x$-$y$ plane, which are subject to superconducting order. This satisfies the inequality $n_{\mathrm{ACT}}\leqslant n$, and is very nearly equal to $n$ for low hole densities.

(ii) The *coherence length* ($\xi$), expressed in terms of oxygen lattice spacings. This is closely related to the inverse of the energy gap $\Delta$, whatever its origin.

(iii) The *extended s-wave ordering parameter* ($C_{\mathrm{exs}}$), which is finite in the superconducting phase.

Several methods have been employed to determine the hole density in high-$T_c$ materials. Of particular interest is the relationship obtained by Obertelli, Cooper, and Tallon[11] between the low-temperature hole density in the superconducting planes and the room-temperature thermopower. The main consequence of such work is the now generally accepted empirical relationship between the experimental value of $T_c$, and the hole density $n$ per oxygen atom, for all the high-$T_c$ superconductors. This is sometimes expressed in the form of an inverted parabola: $T_c=T_c(\max)[1-82.6\times(2n-0.16)^2]$. Using this expression, the maximum value of $T_c$, i.e., $T_c(\max)$, is seen to occur at an *optimal* hole density $n_{\mathrm{opt}}=0.08$.

Theoretical determinations of the superconducting transition temperatures, as a function of $n$, have been reported previously (see Ref. 4, and references therein). These reproduce the inverted parabolic form, at least for temperatures near to $T_c(\max)$. In the present simulations it is found that $n_{\mathrm{opt}}=0.07$. The small difference between this and the experimental value (0.08) could easily be bridged by adjusting the relative values of the parameters in the Hamiltonian. The calculated value of $T_c(\max)$ (i.e., 29 K) is not regarded as significant, for it scales with the energy parameters used in the Hamiltonian.

The form of the parabola can be characterized in terms of its width, $\Delta n=2|n'-n_{\mathrm{opt}}|$, where $n'$ is either of the hole densities corresponding to $T_c/T_c(\max)=3/4$. Using the above, empirically derived, formula one obtains $\Delta n=0.055$, while the theoretical value obtained from Fig. 4 of Ref. 4 is $\Delta n=0.07$. Again, this discrepancy could easily be bridged by adjusting the parameters of the Hamiltonian. New simulation results have been obtained in order to investigate the dependence of the energy gap on hole density at temperatures well above the superconducting transition temperature. Our main result is the phase diagram shown in Fig. 1. It defines three regions, denoted I (superconducting, with correlated pairs), II (uncorrelated pairs), and III (unpaired holes). The criteria employed to distinguish results which fall into each of these regions are

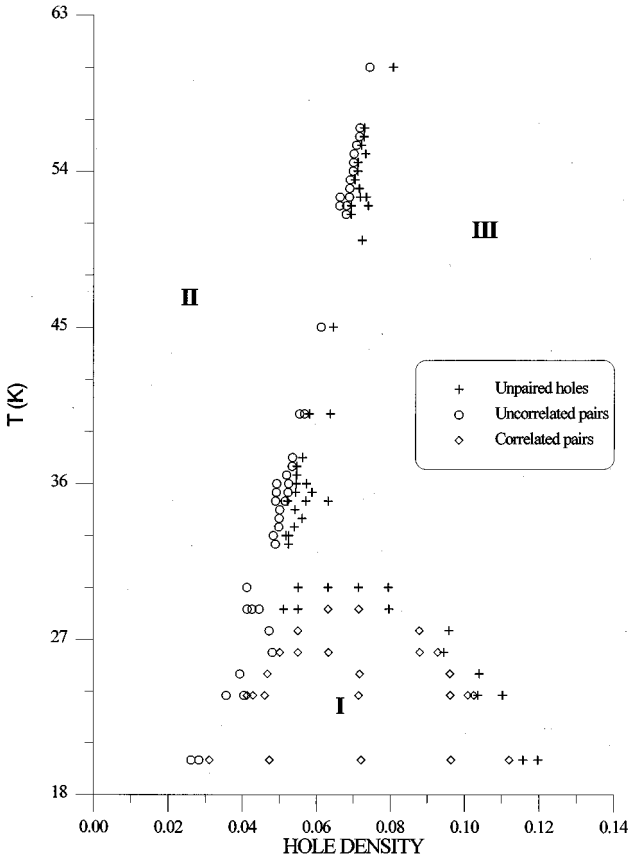


FIG. 1. Phase diagram derived from the simulations, showing the boundaries between phases. I. superconducting, with correlated hole pairs, II: uncorrelated hole pairs, and III: unpaired holes.

I: $C_{\mathrm{exs}}>10^{-4}$, shown as diamonds,
II: $C_{\mathrm{exs}}<10^{-4}$, $\xi<25$ lattice spacings, shown as circles,
III: $C_{\mathrm{exs}}<10^{-4}$, $\xi\geqslant25$ lattice spacings, shown as plus signs.

The 25 lattice spacing criterion is used because, with the finite lattice used in our simulations, the coherence length cannot exceed $30\pm5$ lattice spacings. Most of the results shown in the diagram have been selected so as to define precise boundaries between the three regions. Within the limits imposed by the simulation statistics, the boundary between phases II and III is a straight line above $T_c(\max)$. This phase boundary has been found to continue as a straight line up to even higher temperatures than those shown in Fig. 1. The transition from phase I to phase III is BCS-like, and the transition from phase I to phase II is Bose-like.

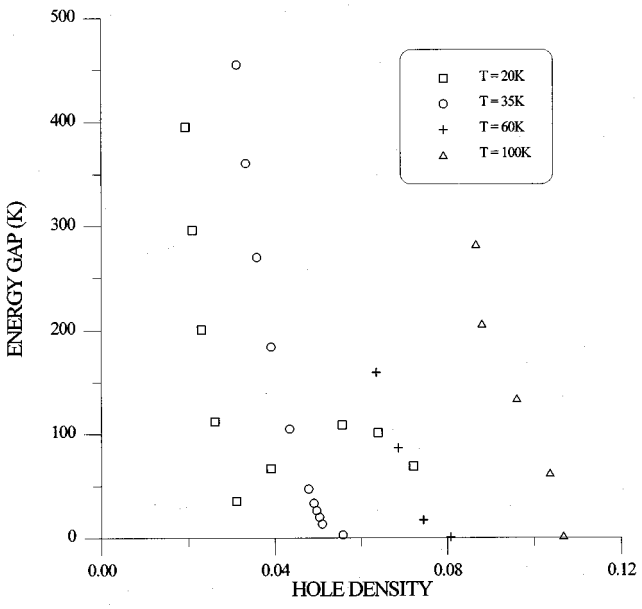


FIG. 2. Dependence of the energy gap $\Delta_p$ on hole density for several temperatures.

The pairs in region II of the phase diagram are small, with a binding energy $\Delta_p$. They exist at all temperatures for sufficiently low hole densities. As shown in Fig. 2, $\Delta_p$ increases rapidly with decreasing hole density for results in region II. This is qualitatively similar to the behavior of the observed pseudogap as a function of the hole density.[1,2] Given the small quantitative differences between theory and experiment, it is reasonable to identify $\Delta_p$ with the pseudogap. Just as in the case of the dependence of $T_c$ on hole density, it is expected that the quantitative discrepancies could be reduced by the use of a more realistic Hamiltonian. No explicit determinations of the temperature dependence of the pseudogap have been found in the literature, so it is not possible to make direct comparisons between experiment and the temperature dependence of $\Delta_p$ shown in Fig. 2.

One aspect of the results shown in Figs. 1 and 2 is clearly counterintuitive. We would normally expect the hole pairs to become *less* stable with increasing temperature, but our results show them to become *more* stable. Also, for values of $n$ above 0.05 holes per oxygen ion, uncorrelated bound pairs *form* from unpaired holes as the temperature *increases* across the boundary between region III and region II (see Fig. 1). Further increases of temperature lead to a decrease in the pair size and a corresponding increase in the energy gap $\Delta_p$ (and hence an increase in stability). This behavior can be understood by noting that $\Delta_p$ measures the gap between the Fermi energy and the energies of the filled states at top of the band. The hole density is determined by the value of this gap and the Boltzmann distribution. In order to maintain a fixed low hole density in the single band, $\Delta_p$ is forced to rise with temperature. At the same time, it is found that $n_{\mathrm{ACT}}$ becomes closer to the total hole density $n$ as the temperature rises, so that all the holes are provided by quasispins with $\langle s_{\mathbf{k}}^z \rangle = 0$. These half-filled hole pair states are nearly degenerate, with energies close to the Fermi energy.

Several theoretical predictions of phase diagrams of the type shown in Fig. 1 have already appeared in the literature, some of which were published well before the discovery of high-$T_c$ superconductors.[12,13] In particular, Robaszkiewicz, Micnas, and Chao[13] developed a bipolaron model which exhibits superconductivity for low hole densities. Their theory is expressed in terms of a quasispin formalism *in real space*. This has the effect of fixing the size of the hole pairs, but allows inhomogeneities in the hole distribution on the lattice. As a result, their phase diagrams [Fig. 4(c) in Ref. 9] do not exhibit a BCS-like transition, but are in agreement with the phase diagram obtained in this work at low hole densities (where pair sizes are small).

More recently, a considerable amount of theoretical work has been carried out on possible ''crossover'' effects between BCS and composite boson superconductivity. We note, in particular, the work of Casas *et al.*[14] which discusses crossover in terms of pair size, and the more general discussion of Gorbar, Loktev, and Sharapov.[15] Randeria *et al.*[16] explicitly link pairing above $T_c$, obtained in a quantum Monte Carlo study of the negative $U$ model, with the spin gap. All of this work is in agreement with our main result: simple one-band models predict uncorrelated hole pairs to occur above $T_c$ for low hole densities.

In simulations of underdoped systems, $n < n_{\mathrm{opt}}$, with a fixed chemical potential $\mu$, the hole density is found to increase with temperature. Constraints on $\mu$ in *real* high-$T_c$ systems with low hole densities, could therefore lead to an increase, with temperature, of the hole density in the superconducting band. The lowest lying unoccupied band in the copper oxide planes of the high-$T_c$ cuprates is formed predominantly from copper $3d$ states. This would limit the increase in the chemical potential because, at sufficiently high temperatures, electrons excited into the copper $3d$ band would increase the hole density in the oxygen $2p$ band. Homes *et al.*[17] determine the upper bound of the chemical potential to be approximately 300 K above the oxygen $2p$ band. Above temperatures of this order, therefore, we would expect real systems to have an effectively constant chemical potential. While at very low temperatures the usual assumption of constant hole density would be correct, this could no longer be expected to hold at temperatures above $T_c(\max)$. Further simulations are being carried out to investigate this matter quantitatively.

Inelastic neutron-scattering results of Rainford *et al.*[18] show a broadening in the crystal-field linewidths of erbium in 123 and 124 compounds with $n$ near to $n_{\mathrm{opt}}$ at two temperatures: a little below $T_c$, and 30 to 40 % above $T_c$. Such an observation suggests the occurrence of resonances between more than one energy gap characteristic of the system and the crystal-field splitting energy. This is in qualitative agreement with development of a pseudogap above $T_c$ as shown in Fig. 1. As the temperature is raised, the crystal field first comes into resonance with superconducting gap $\Delta_s$, just below $T_c$ where it passes from region I into region III. As the temperature increases above $T_c$ the system will come into resonance with $\Delta_p$ as it passes from region III to region II. This scenario depends, of course, on there being only a small increase of hole density with temperature over this range.

In summary, the normal-state pseudogap, observed in sev-

eral of the cuprate superconductors, has been associated with the Fermi surface moving away from the top of the oxygen $2p$ band responsible for superconductivity. Comparison of simulated and experimental results show that this gap can be identified with the binding energies of intraplanar uncorrelated hole pairs. This interpretation is consistent with there being a Bose-like superconducting transition at low hole densities.

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