Pairing and Spin Gap in the Normal State of Short Coherence Length Superconductors

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(Received 27 May 1992)

We study the normal state of the 2D attractive Hubbard model using quantum Monte Carlo simulations. We show that singlet pairing correlations develop above T_c, and the normal state of a short coherence length superconductor deviates from a canonical Fermi liquid. In the intermediate U regime, the spin susceptibility χ_s is strongly temperature dependent, and the low-frequency spectral weight, as measured by the NMR relaxation rate $1/T_1T$, is shown to track χ_s. This provides a simple, qualitative explanation for the spin-gap behavior observed in several high-T_c systems.

PACS numbers: 74.65.+n, 67.50.Dg, 76.60.−k

The problem of the crossover [1] from a BCS superconductor to a condensate of pre-formed bosons has attracted renewed attention [2]. Given their very short coherence lengths, the high-T_c materials are likely to be in an intermediate regime between these two limits. Rather little is known [3] about how the normal (nonsuperconducting) state of a system of fermions with attractive interactions evolves from a Fermi liquidlike regime to a Bose regime as a function of increasing attraction. Independent of the microscopic mechanism, the question of how the resulting attractive interaction affects the *normal-state* properties is of considerable interest.

We address this question using quantum Monte Carlo simulations [4,5] for the attractive ("negative U") Hubbard model in 2D. We show that for intermediate coupling strengths the normal state clearly deviates from a canonical Fermi liquid. Our main results are the following: (1) The uniform, static spin susceptibility χ_s is strongly temperature dependent, with $d\chi_s/dT > 0$, for $T_c < T < T_p$, where T_c is the superconducting transition temperature, and T_p a "pairing" scale (defined later) below which strong singlet pairing correlations develop. (2) This is accompanied by a "spin gap," i.e., a reduction in the low-frequency spectral weight, leading to an NMR relaxation rate $1/T_1T$ which tracks $\chi_s(T)$. We emphasize that these results, which are perhaps easiest to rationalize in the nondegenerate, pre-formed boson limit at large U, persist well into the intermediate-U regime where one has a degenerate Fermi system.

These results are strikingly similar to the anomalous behavior of the NMR Knight shifts and relaxation rates observed [6,7] above T_c in many, but not all, high-T_c superconductors. Walstedt and co-workers [6] have particularly stressed the relationship $1/T_1T \sim \chi_s(T)$, which is very different from that expected in a Fermi liquid, namely, $1/T_1T \sim \chi_s^2$, with χ_s temperature independent. Theories of short-range antiferromagnetic fluctuations [8], which have been very successful in explaining many aspects of the normal state NMR, are, however, unable to account for these anomalies; see Ref. [9] for a clear discussion. Our work, which provides a natural explanation for these anomalies, is in a sense complementary to these earlier theories. We discuss below in more detail the implications of our results for experiments on the reduced oxygen $YBa_2Cu_3O_{6+x}$ (YBCO) systems.

The attractive Hubbard Hamiltonian is given by

$$H = -t \sum_{\langle i,j\rangle,\sigma} (c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.}) - U \sum_i n_{i\uparrow} n_{i\downarrow} + \mu \sum_{i,\sigma} n_{i\sigma} , \tag{1}$$

where $\langle i,j\rangle$ denotes a pair of nearest-neighbor sites on a square lattice, σ is a spin label, and $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$. From here on $t \equiv 1$, and all energies are measured in units of t. The chemical potential μ is tuned to fix [10] the average density $\langle n\rangle$. The superconducting (Kosterlitz-Thouless) transition [5] is expected to have a maximum T_c (as a function of U and $\langle n\rangle$) around 0.1. All of our numerical results will be in the normal state with $T > 0.1$ on systems of size up to 8×8. It is perhaps worth noting that the attractive Hubbard model does not suffer from the "sign problem" which often plagues fermion Monte Carlo calculations.

We begin by determining the regime in parameter space where one has a degenerate Fermi system, since it is important to establish that the anomalous normal-state properties discussed later persist in this regime. The U and T dependence of the chemical potential μ required to maintain a fixed density, $\langle n\rangle = 0.5$, is shown in Fig. 1. We find $d\mu/dT < 0$, as one usually expects, for all T and U, in contrast to the result of summing ladder diagrams [11] in 2D. At low T we find good agreement with a $T=0$ mean-field treatment [12] which self-consistently solves for the superconducting gap function and the renormalization of μ. To determine the degenerate regime (see Fig. 1) we use the following criterion. If the chemical potential, measured from the bottom of the band (at $-4t$), taking into account the Hartree shift, is larger than the temperature, namely,

$$\mu(T,U) + 4 + \langle n\rangle U/2 > T , \tag{2}$$

then the system is degenerate (strictly speaking, the in-

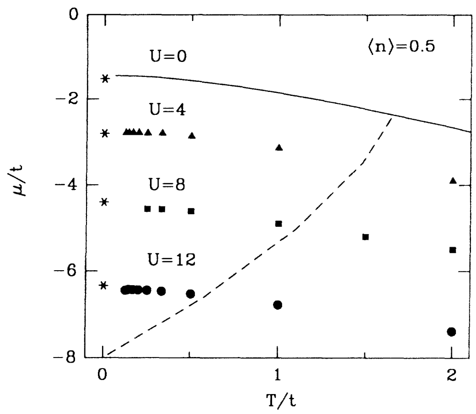

FIG. 1. Chemical potential as a function of temperature required to maintain a fixed density, $\langle n \rangle = 0.5$. The Monte Carlo data at $U=4$ and 8 are on 8×8 lattices, and the $U=12$ data are from a 4×4 system. The points (stars) at $T=0$ are the mean-field results from Ref. [12]. The degenerate region is above the dashed curve [see Eq. (2)].

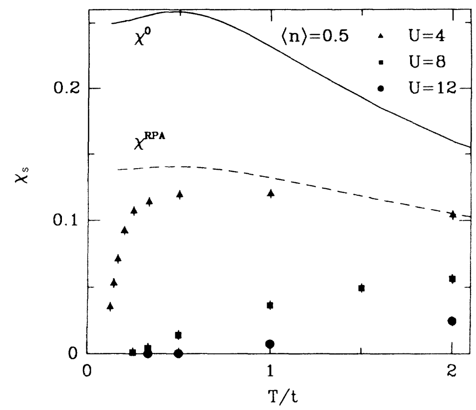

FIG. 2. The uniform, static spin susceptibility as a function of temperature, for various U values. The RPA form is fitted to the $U=4$ MC data (not shown) in the high-temperature regime $2<T<4$ by using a "renormalized" value $\tilde{U}=3.25$. System sizes are the same as in Fig. 1. Error bars not explicitly shown are of the size of the symbols.

equality should be replaced by $\gg$). We note that at sufficiently low temperatures all [13] of our Monte Carlo data are in the degenerate regime. (We estimate [14] that for $\langle n \rangle = 0.5$ we require $U>16$ to be nondegenerate at very low T.)

To see the formation of pairs as U increases, we look at the on-site correlation function $\langle n_{i\uparrow} n_{i\downarrow} \rangle$. This increases monotonically with U at any fixed T, and at low T goes from the totally uncorrelated value of $\langle n \rangle^2/4$ at $U=0$ to nearly 0.96 of $\langle n \rangle /2$, the value characteristic of on-site pairing, at $U=12$. From this point of view the $U=12$ case, even though degenerate at low T, appears to be close to the infinite-U limit.

We next turn to the effect of pairing in the normal state on various observable quantities; we shall focus mainly on magnetic correlations in this paper. We first look at the uniform, static spin susceptibility $\chi_s \equiv \chi(q \to 0, \omega=0)$ as a function of U and T, shown in Fig. 2. For $U=0$, χ^0 has the expected Pauli behavior for $T<1$ crossing over to a Curie-like form at higher temperatures. (The small bump at $T \simeq 0.5$ is due to the logarithmic singularity in the density of states at the band center.) With increasing U the overall magnitude of χ_s decreases, which may be understood (at high T) within RPA with $\chi^{\mathrm{RPA}} = \chi^0/(1+U\chi^0)$.

For $U=4$, χ_s resembles the RPA result at high temperatures, flattens out at intermediate T, and then for $T<0.5$ shows a remarkable temperature dependence. Note that $T_c \simeq 0.05$ for this case [5]. This low-temperature drop in χ_s is a result of the tendency towards pair formation in the normal state (and is, of course, completely absent in the RPA). For $U=8$ and 12, we find that χ_s is an increasing function of T for all $T<4$. For large U one obtains tightly bound singlet pairs which contribute to χ_s only when they are ionized.

The temperature at which the Monte Carlo data deviate from the RPA can be used to define a pairing scale T_p which is clearly larger (for the values of U studied) than the transition temperature T_c at which coherence is established. The anomalous behavior in the normal state discussed here is in the regime $T_c < T < T_p$. For small U, T_p is expected to be the same as the mean-field transition temperature T_c^0, and one can describe the regime $T_c < T < T_c^0$ in terms of superconducting fluctuations [15]. For large U the two scales are widely separated [16] with $T_c^0 \sim t^2/U$ while $T_p \sim U$, the pair binding energy. The anomalous temperature dependences in the normal state then clearly arise from the formation of singlet pairs without any coherence, and *not* from superconducting fluctuations.

A static susceptibility with $d\chi_s/dT>0$, though suggestive, is by itself not [17] sufficient to establish a spin gap [9,18], namely, a reduction in the low-frequency spectral weight $\chi''(\mathbf{q},\omega)$, which is probed by the NMR relaxation rate $1/T_1$. In general, it is very difficult to obtain $\chi''(\mathbf{q},\omega)$ from Monte Carlo calculations, since it requires analytic continuation from Matsubara to real frequencies. However, for our present purpose one can use the following strategy.

We can write [8] $\chi''(\mathbf{q},\omega) = \chi(\mathbf{q},\omega=0)\omega f(\mathbf{q},\omega)$ where f is even in ω and, using Kramers-Kronig relation, $\int f(\mathbf{q},\omega) d\omega/\pi = 1$ for each $\mathbf{q}$. Defining $1/\Gamma_q = f(\mathbf{q},0)$, $1/T_1$ can be written as

$$\frac{1}{T_1 T} = \lim_{\omega\to 0} \sum_{\mathbf{q}} \frac{\chi''(\mathbf{q},\omega)}{\omega} = \sum_{\mathbf{q}} \frac{\chi(\mathbf{q},0)}{\Gamma_q} . \tag{3}$$

The imaginary time correlation function $S(\mathbf{q},\tau) = \langle s_{\mathbf{q}}(\tau) \times s_{-\mathbf{q}}(0) \rangle$, where $s_{\mathbf{q}}$ is the Fourier transform of $s_i^z = n_{i\uparrow} - n_{i\downarrow}$, is related to the spectral weight via

$$S(\mathbf{q},\tau) = \int_{-\infty}^{\infty} d\omega \frac{\exp(-\omega\tau)}{1-\exp(-\omega/T)} \chi''(\mathbf{q},\omega) . \tag{4}$$

For $\tau = 1/2T$, the thermal factor in (4) is $1/\sinh(\omega/2T)$ and the integral gets cut off on the scale of T. Using $\chi''(\mathbf{q},\omega) = \chi(\mathbf{q},0)\omega/\Gamma_q$ for $\omega < 2T$, an assumption which is reasonable for T less than the scale for spin fluctuations, we find

$$\sum_{\mathbf{q}} S(\mathbf{q},\tau = 1/2T) = \pi^2 T^2 \sum_{\mathbf{q}} \frac{\chi(\mathbf{q},0)}{\Gamma_q} . \tag{5}$$

We are thus able to extract $1/T_1T$ from an imaginary-time spin correlation function measured directly in the Monte Carlo simulations.

The relaxation rate $1/T_1T$, obtained from (3) and (5), plotted in Fig. 3(a), is found to decrease as T is lowered. In Fig. 3(b) we show that $1/T_1T$ indeed tracks the static susceptibility χ_s, thus establishing spin-gap behavior in the normal state. It is worth noting that, for the two values of U studied, the data for $1/T_1T$ vs χ_s appear to lie on the same curve. Further, we emphasize that the spin-gap arises from a temperature-dependent Γ_q. To see this we have checked that $\chi(\mathbf{q},0)$, for typical values of $q \neq 0$, is weakly T dependent, as one might expect [9] from the moment sum rule combined with Kramers-Kronig relation. The only way that $1/T_1T$ can then have strong temperature dependence is through Γ_q.

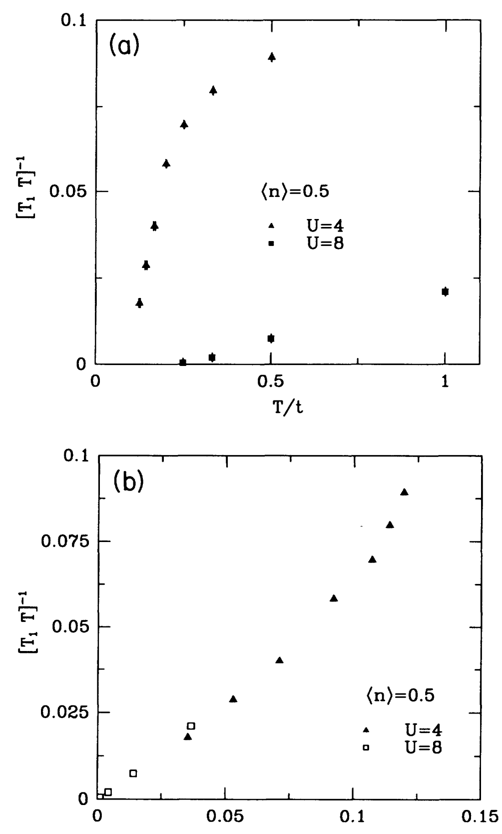

FIG. 3. (a) The relaxation rate $1/T_1T$ (in arbitrary units) as a function of temperature for $U = 4$ and $U = 8$ obtained from 8×8 lattices. (b) Parametric plot of $1/T_1T$ (in arbitrary units) vs spin susceptibility χ_s for $U = 4$ and $U = 8$ to show that the two quantities track each other. For a Fermi liquid all the data, for a given U, should collapse to a single point.

It is interesting to ask if the opening up of a spin gap is accompanied by a charge gap, or a pseudo gap. In this model the answer appears to be yes, although more detailed work is necessary. The single-particle density of states obtained via analytic continuation in Ref. [19] gives some evidence for a gaplike structure developing above T_c. We have also studied the momentum distribution function $\langle n_k\rangle$; these results will be published separately. While $\langle n_k\rangle$ is clearly broader than what would be expected for a Fermi gas ($Z = 1$) with thermal smearing, it is hard to conclude (from fitting the Monte Carlo data to various functional forms) if the observed behavior necessarily implies a gap.

Finally, we discuss the applicability of our results to normal state NMR experiments [6,7] on the YBCO systems; while these are the best studied systems, spin-gap behavior is not [6(c)] restricted to them. (At least) two issues need to be discussed: (1) the role of antiferromagnetism (AFM), and (2) why spin gaps are seen in some materials, and not others.

In the $T_c = 60$ K system ($x \simeq 0.65$) the Knight shift, which probes χ_s, increases by a factor of 4 as T increases from T_c to 300 K, which we would identify as T_p. For the O and Y sites (where the form factors [8] filter out the AFM contribution) $1/T_1T$ indeed tracks χ_s below 300 K. Our model, which has no AFM, is able to naturally explain these spin-gap features. For the Cu site, $1/T_1$ is larger in magnitude and shows spin-gap behavior only below 150 K. This would appear to require a combination of AFM fluctuations and a spin gap opening up due to enhanced pairing correlations.

In the $T_c = 90$ K ($x \simeq 1.0$) material, spin-gap behavior is not seen (or seen over a very narrow regime). To discuss the range $T_c < T < T_p$ over which these anomalies will be seen, one needs to know how T_c and T_p change as a function of doping (x). Since the attractive Hubbard model is *not* a *microscopic* model for YBCO (e.g., half filling in this model has nothing to do with the magnetic insulator at $x = 0$), we can only make qualitative remarks. Let us assume that once YBCO is metallic, increasing x is analogous to increasing the carrier concentration n in the model. For fixed U, T_c increases with n, until $n \simeq 0.85$, and then drops to zero at $n = 1.0$ in 2D, as shown in Ref. [5]. On the other hand, T_p is expected to be relatively independent of n for small n, to the extent that one has independent pair formation, and could even drop with increasing n due to the Pauli principle blocking the formation of independent pairs. This crude argument suggests why the system with the highest T_c, and highest carrier concentration, might have the smallest window between the pairing temperature and T_c.

In conclusion, we have studied the crossover in the normal state of a simple model 2D superconductor as it evolves from a Fermi-liquid-like regime to a Bose regime.

We find that for moderate values of the attraction (one half the bandwidth) the normal state, even though it is a degenerate Fermi system, already shows anomalous magnetic correlations. These effects are strikingly similar to the anomalies seen in normal state NMR experiments on several high-T_c systems. Our results provide a qualitative explanation of these experiments quite independent of the microscopic origin of the attractive interaction.

We would like to thank Jan Engelbrecht, Hartmut Monien, and Rajiv Singh for useful discussions, and to acknowledge an especially stimulating conversation with Sriram Shastry. M.R. and N.T. were supported by the U.S. Department of Energy, Office of Basic Energy Sciences under Contract No. W-31-109-ENG-38. M.R. and N.T. would like to thank the Institute for Theoretical Physics, Santa Barbara, for hospitality during the final stages of this work. Work at the ITP was supported by NSF PHY 89-04035. R.T.S. acknowledges the donors of The Petroleum Research Fund, administered by the American Chemical Society, for partial support of this research.

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[14] At $T=0$ the chemical potential goes below the bottom of the band for $U>8/(1-\langle n\rangle)$, which is obtained from (2) using $\mu \simeq -\frac{1}{2}U$ for $U \gg 1$. In the *superconducting case*, the dependence of the energy gap on the order parameter changes qualitatively at this point; see Refs. [1,2,12].

[15] In 2D where T_c is much lower than T_c^0, it is interesting to ask if deviations from Fermi-liquid behavior persist even as $U\to 0$. Finite-size errors, which are more pronounced at small U, make if difficult to address this question via Monte Carlo calculations.

[16] In the large-U limit the attractive Hubbard model can be mapped onto a hard-core Bose system with effective hopping t^2/U. See Ref. [10].

[17] The 2D $S=\frac{1}{2}$ Heisenberg antiferromagnet provides a counterexample. $d\chi/dT>0$ for $T<J$, but $1/T_1T$ increases with decreasing T. See S. Chakravarty and R. Orbach, Phys. Rev. Lett. **64**, 224 (1990); R. R. P. Singh and M. Gelfand, Phys. Rev. B **42**, 996 (1990).

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