Unusual Thermodynamical and Transport Signatures of the BCS to Bose-Einstein Crossover Scenario below T_c

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In this paper we present predictions for thermodynamic and transport properties of a BCS to Bose-Einstein crossover theory, below T_c, which satisfies the reasonable constraints that it yields (i) the Leggett ground state and (ii) BCS theory at weak coupling and all temperatures T. The nature of the strong coupling limit is inferred, along with the behavior of the Knight shift, superfluid density, and specific heat. Comparisons with existing data on short coherence length superconductors, such as organic and high T_c systems, are presented, which provide some support for the present picture.

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Within the pseudogap regime of the cuprates, it has been widely argued that the excitations of the superconducting state are either fermionic [1] or bosonic [2] in character. In this paper we discuss a third scenario [associated with the BCS to Bose-Einstein condensation (BEC) crossover approach], in which the excitations contain a mix of bosonic and fermionic properties [3].

The BCS-BEC crossover scheme has been viewed as relevant to the cuprates and other "exotic" [4,5] superconductors where their short coherence lengths, ξ, naturally lead to a breakdown of strict BCS theory. The BCS-BEC scenario [3,6–9] owes its origin to Eagles [10] and Leggett [11] who proposed a ground state wave function of the BCS form, $\Psi_0 = \Pi_{\mathbf{k}}(u_{\mathbf{k}} + v_{\mathbf{k}} c^{\dagger}_{\mathbf{k}\uparrow} c^{\dagger}_{-\mathbf{k}\downarrow})|0\rangle$, which describes the continuous evolution between a BCS system, having weak coupling g and large ξ, and a BEC system with large g and small ξ. Here $u_{\mathbf{k}}, v_{\mathbf{k}}$ are the standard coherence factors of BCS theory, which are determined *in conjunction* with the number constraint.

The essence of this paper is a characterization of the excitations of Ψ_0 and their experimental signatures (for all $T \le T_c$). New thermodynamical effects stemming from bosonic degrees of freedom must necessarily enter, as one crosses out of the BCS regime, towards BEC. Here we show that the bosonic excitations (which appear in the gap equations and which we call "pairons") are different from the collective phase mode [12]: they generally have a quadratic dispersion similar to that of a *quasi-ideal* BEC system—a consequence of the mean field treatment of the pairs which, in turn, is dictated by the general mean field character of Ψ_0. These ideas are applied to the cuprates and other short ξ superconductors.

We consider fermions, with lattice dispersion $\epsilon_{\mathbf{k}}$ (measured with respect to the fermionic chemical potential μ), and with interaction $V_{\mathbf{k},\mathbf{k}'} = g\varphi_{\mathbf{k}}\varphi_{\mathbf{k}'}$, where $g < 0$; here $\varphi_{\mathbf{k}} = 1$ and $(\cos k_x - \cos k_y)$ for s- and d-wave pairing, respectively. We begin by reviewing BCS theory, in terms of a somewhat unfamiliar but very useful formalism [13] which is then generalized [3,14] to the crossover problem. For brevity, we use a four-momentum notation: $K = (\mathbf{k}, i\omega), \sum_K = T\sum_{\mathbf{k},\omega}$, etc. We also suppress $\varphi_{\mathbf{k}}$ until the final equations.

BCS theory involves the pair susceptibility $\chi(Q) = \sum_K G(K)G_0(Q-K)$, where the Green's function G satisfies $G^{-1} = G_0^{-1} + \Sigma$, with order parameter Δ_{sc} and $\Sigma(K) = -\Delta_{sc}^2 G_0(-K)$. In this notation, the gap equation is

$$1 + g\chi(0) = 0, \qquad T \le T_c. \tag{1}$$

At $Q = 0$, the summand in χ is the Gor'kov "F" function (up to a multiplicative factor Δ_{sc}) and *this serves to highlight the central role played in BCS theory by the more general quantity* $G(K)G_0(Q-K)$. Note that (for $Q \ne 0$) $\chi(Q)$ is *distinct from the pair susceptibility of the collective phase mode* which enters as $\sum_K\{G(K)[G(Q-K) + G(-Q-K)] + 2F(K)F(K-Q)\}$ [12,15]. Here, each Gor'kov F function introduces one GG_0, so that *the collective mode propagator depends on effectively higher order Green's functions than does the gap equation.*

The observations in italics were first made in Ref. [13] where it was noted that the BCS gap equation could be rederived by truncating the equations of motion so that only the one (G) and two particle ($\mathcal{T}$) propagators appeared. Here, G depends on Σ which in turn depends on $\mathcal{T}$. In general, $\mathcal{T}$ has two additive contributions [14], from the condensate (sc) and the noncondensed (pg) pairs. Similarly, the associated self-energy [13] $\Sigma(K) = \sum_Q \mathcal{T}(Q)G_0(Q-K)$ can be decomposed into $\Sigma_{pg}(K) + \Sigma_{sc}(K)$. The two contributions in Σ come, respectively, from $\mathcal{T}_{sc}(Q) = -\Delta_{sc}^2\delta(Q)/T$, and from the $Q \ne 0$ pairs, with $\mathcal{T}_{pg}(Q) = g/[1 + g\chi(Q)]$. In the leading order mean field theory $\Sigma = \Sigma_{sc} = -\Delta_{sc}^2 G_0(-K)$ which, from Eq. (1), yields the usual BCS gap equation.

More generally, at larger g, the above equations hold but we now include feedback into Eq. (1) from the finite momentum pairs, via $\Sigma_{pg}(K) = \sum_Q \mathcal{T}_{pg}(Q)G_0(Q-K) \approx G_0(-K)\sum_Q \mathcal{T}_{pg}(Q) \equiv -\Delta_{pg}^2 G_0(-K)$, which defines a pseudogap parameter, Δ_{pg}. This last approximation is valid only because [through Eq. (1)] $\mathcal{T}_{pg}$ diverges as $Q \to 0$. In this way, $\Sigma_{pg}(K)$ has a BCS-like form, as does the total self-energy $\Sigma(K) = -\Delta^2 G_0(-K)$, where $\Delta^2 = \Delta_{sc}^2 + \Delta_{pg}^2$. Thus, in the present approach, the energy gap for single electron excitations reflects the presence of both finite center-of-mass momentum pairs as well as the condensate. While the structure of the gap equation will be seen to be formally identical to that in BCS theory, the vanishing of the excitation gap, Δ, takes place at a higher temperature than that at which the order parameter, Δ_{sc}, vanishes. The latter defines T_c.

If we now expand $\mathcal{T}_{pg}^{-1}(\mathbf{q}, \Omega) \approx a_1 \Omega^2 + a_0 \Omega + \tau_0 - Bq^2 + i\Gamma'_{\mathbf{q}}$, we see that the chemical potential of the pairs μ_{pair} is proportional to τ_0, and, via Eq. (1), precisely zero at and below T_c. This provides an interpretation along the lines of ideal Bose gas condensation. (Here, also, at small $\mathbf{q}$, $\Gamma'_{\mathbf{q}} \to 0$.) As g increases, the term $a_0\Omega$ in $\mathcal{T}_{pg}^{-1}$ becomes progressively dominant with respect to $a_1\Omega^2$. For the physically relevant regime of moderate g, we have found, after detailed numerical calculations, that a_1 may be safely neglected. At weak coupling, there is no loss of generality in approximating $\mathcal{T}_{pg}$ in this more particle-hole asymmetric way, since its contribution is negligible. In this way, we can write

$$\mathcal{T}_{pg}^{-1}(\mathbf{q}, \Omega) = a_0(\Omega - \Omega_{\mathbf{q}} + \mu_{\text{pair}} + i\Gamma_{\mathbf{q}}). \quad (2)$$

As a consequence, we have

$$\Delta_{pg}^2 = -\sum_Q \mathcal{T}_{pg}(Q) = \frac{1}{a_0} \sum_{\mathbf{q}\neq 0} b(\Omega_{\mathbf{q}}). \quad (3)$$

We now rewrite Eq. (1), along with the fermion number constraint, as

$$1 + g\sum_{\mathbf{k}} \frac{1 - 2f(E_{\mathbf{k}})}{2E_{\mathbf{k}}} \varphi_{\mathbf{k}}^2 = 0, \quad (4)$$

$$\sum_{\mathbf{k}} \left[1 - \frac{\epsilon_{\mathbf{k}}}{E_{\mathbf{k}}} + \frac{2\epsilon_{\mathbf{k}}}{E_{\mathbf{k}}} f(E_{\mathbf{k}})\right] = n. \quad (5)$$

Here, $f(x)$ and $b(x)$ are the Fermi and Bose functions and $E_{\mathbf{k}} = \sqrt{\epsilon_{\mathbf{k}}^2 + \Delta^2 \varphi_{\mathbf{k}}^2}$ is the quasiparticle dispersion. Equations (3)–(5) are consistent with BCS theory at small g, and with the ground state Ψ_0 at all g; in both cases the right-hand side of Eq. (3) is zero. The simplest physical interpretation of the present decoupling scheme is that it goes beyond the standard BCS mean field treatment of the single particles (which also acquire a self-energy from the finite $\mathbf{q}$ pairs) but it *treats the pairs at a self consistent, mean field level.*

The dispersion $\Omega_{\mathbf{q}} = q^2 B/a_0$, as well as the coefficient a_0, are determined by a Taylor expansion of $\mathcal{T}_{pg}^{-1}$ [16]:

$$\mathcal{T}_{pg}^{-1}(\mathbf{q}, \Omega) = g^{-1} + \sum_{\mathbf{k}} \left[\frac{1 - f(E_{\mathbf{k}}) - f(\epsilon_{\mathbf{k}-\mathbf{q}})}{E_{\mathbf{k}} + \epsilon_{\mathbf{k}-\mathbf{q}} - \Omega} u_{\mathbf{k}}^2 - \frac{f(E_{\mathbf{k}}) - f(\epsilon_{\mathbf{k}-\mathbf{q}})}{E_{\mathbf{k}} - \epsilon_{\mathbf{k}-\mathbf{q}} + \Omega} v_{\mathbf{k}}^2\right] \varphi_{\mathbf{k}-\mathbf{q}/2}^2. \quad (6)$$

For small g and in three dimensions (3D), the poles of $\mathcal{T}_{pg}$ (at $T = 0$) occur at $\Omega = \sqrt{3}\, cq$, where c is the usual phase mode velocity. At moderate g, where the pairons become increasingly more relevant, and for quasi-2D dispersion $\epsilon_{\mathbf{k}}$, $\Omega_{\mathbf{q}} \approx q_{\parallel}^2/2M_{\parallel}^* + q_{\perp}^2/2M_{\perp}^*$. Here we find that the ratio $M_{\parallel}^*/M_{\perp}^* \propto (t_{\perp}/t_{\parallel})^2$, where $t_{\parallel}$ and $t_{\perp}$ are the in- and out-of-plane hopping integrals, respectively. Numerical calculations show that the masses, as well as the residue a_0, are roughly T independent constants at low T [17]. In the BEC regime at low density and with s-wave pairing in a 3D continuous model, M^* is $2m_e$ for all $T \le T_c$ [16], as found previously [7]. The examples in this paper, which apply to the fermionic regime, correspond to somewhat smaller $M_{\parallel}^*$.

It is important to note that in strictly 2D the logarithmic divergence on the right-hand side of the pseudogap equation (3) (which is essentially a boson number equation) implies $T_c = 0$, as in an ideal Bose gas. For large anisotropy, or small $t_{\perp}$, $T_c \propto -1/\ln(t_{\perp}/t_{\parallel})$, which vanishes logarithmically [16]. Finally, since both a_0 and the effective pair mass (tensor) M^* are constants at low T, Eq. (3) implies $\Delta_{pg}^2(T) = \Delta^2(T) - \Delta_{sc}^2(T) \propto T^{3/2}$. Moreover, because Δ depends on T only exponentially, $\Delta_{sc}^2(T) = \Delta^2(0) - AT^{3/2}$ at low T, where A is T independent.

We now calculate physical quantities such as the magnetic penetration depth (λ) and related superfluid density (n_s), the Knight shift (K_s), and the NMR relaxation rate (R_s) using techniques similar to those used to study fluctuation effects in normal metal superconductors [18]. Here the usual (lowest order) Maki-Thompson and Aslamazov-Larkin diagrams are extended to be compatible with Σ and $\mathcal{T}$ by applying the generalized Ward identity to incorporate the pairon vertex correction [12,18]. Finally, the specific heat C_v can be computed following a similar analysis as for the paramagnon problem [19].

While in the BCS limit the expressions for λ (or n_s), K_s, and R_s contain only the total gap Δ, here, they, in principle, depend on both the quasiparticle (via Δ^2) and the pairon (via Δ_{pg}^2) contributions [12,14]. This decomposition leads to a form of "three fluid" model (including the condensate, fermionic quasiparticles, and bosonic pairons). In the same way, C_v can also be decomposed into a sum of two contributions corresponding to an ideal Bose gas of pairons, with dispersion $\Omega_{\mathbf{q}}$, and an ideal Fermi gas of quasiparticles, with dispersion $E_{\mathbf{k}}$. The pairon contributions to n_s enter as follows [3,14]: the general expression is identical to its BCS counterpart, but with the overall multiplicative factor of Δ^2 replaced by $\Delta_{sc}^2 = \Delta^2 - \Delta_{pg}^2$. By contrast, for spin-singlet pairing, there is no explicit pairon contribution to K_s and R_s, and the corresponding expressions reflect the generalized excitation gap Δ, as might have been expected physically. The single most important conclusion of this analysis is that *the presence of low lying pair excitations will introduce new low temperature power law dependences with ideal Bose gas character into physical* *quantities.* Below, we explore these power laws in the context of highly anisotropic 3D, i.e., quasi-2D systems.

Figures 1(a) and 1(b) present a comparison between an s-wave short ξ pseudogap (PG) superconductor and an s- and d-wave BCS system. It should be noted that the short ξ superconductors are still far from the BEC limit. For the parameters illustrated by the figures, μ deviates from E_F by roughly 3%. Here, and throughout this paper, we take $t_\perp/t_\parallel = 0.01$. The main body of Fig. 1(a) indicates that the Knight shift (and NMR relaxation rate, not shown) at T_c are substantially reduced relative to their high T asymptotes, i.e., K_n, as is illustrated by the solid line (for the PG s wave). Because pairon effects are not explicit, the low T behavior is exponentially activated as for the BCS s-wave case, but here the ratio $\Delta(0)/T_c$ is significantly enhanced over the BCS value. Overall, the behavior of R_s will yield rather similar plots; however, the s-wave BCS limit exhibits the well-known Hebel-Slichter peak, which is absent below T_c for the other two cases.

In the inset of Fig. 1(a), we plot the behavior of the low T specific heat "coefficient," $\gamma(T) \equiv C_v/T$, for the same parameters as above. In the short ξ, quasi-2D case, slightly above $T = 0$, $\gamma(T)$ will appear to be a constant $\gamma(T) = \gamma^*$, although it vanishes strictly at $T = 0$ as $T^{1/2}$. This intrinsic γ^* effect, which may have been seen in both organic and cuprate (layered) superconductors [20], has, in the past, been related to extrinsic effects. Figure 1(b) plots the normalized superfluid density n_s, or λ^{-2}, vs T/T_c, which for the PG (s-wave) case exhibits a $T^{3/2}$ dependence. Here, λ (unlike C_v) is not particularly sensitive to the mass anisotropy ratio, and the boson power law dependence is more 3D.

In order to address d-wave effects in short ξ superconductors, we turn to the cuprates. Note, the dimensionless coupling is $g/t_\parallel$. To be consistent with the observed metal-insulator transition at half filling ($x = 0$), we introduce a hole concentration x dependent renormalization of the in-plane hopping integral $t_\parallel(x) = t_0 x$ deriving from Coulomb correlations, and presume, in the absence of any more detailed information about the pairing mechanism, that g is x independent. Our quasi-2D band structure is taken from the literature [21]; the one free parameter $-g/4t_0$ is chosen ($= 0.045$) to optimize agreement with the energy scales in the cuprate phase diagram. This calculated phase diagram [3], deduced from Eqs. (3)–(5), can be shown to yield reasonable agreement with experimental data. Two important points should be stressed: (i) The chemical potential μ/E_F differs from unity by at most a few percent over the entire range of x. (ii) While the band mass increases with underdoping due to Coulomb effects, the thermodynamically measured mass, obtained from, for instance, the Knight shift $K_s(T_c)$ (or specific heat C_v/T at T_c^-), *decreases* with underdoping, as a consequence of the opening of the pseudogap [see Fig. 2(c)].

Figures 2(a) and 2(b) illustrate the predicted behavior of the Knight shift for the cuprates as a function of x and T. Because it depends only on (d-wave nodal) quasiparticle excitations, K_s exhibits a scaling with $T/\Delta(0)$ which is illustrated in Fig. 2(b) via plots of K_s (normalized to its high T asymptote K_n) for the entire range of x, and for temperatures below each respective $T_c(x)$. An alternate scaling form is shown in Fig. 2(a) where we plot K_s normalized at T_c as a function of T/T_c for various x (with x increasing from top to bottom). The near-collapse of the different x dependent curves is similar to that found in the experimental data [22] shown in the inset. The normalization factor $K_s(T_c)$ for this figure [which varies as the band mass multiplied by $T_c/\Delta(0)$] is plotted as a function of x in Fig. 2(c). Also plotted here are our specific heat (C_v/T) predictions for the pairon contribution to γ^* as compared with the usual d-wave quasiparticle term [20] αT_c as a function of x. The pairon term becomes increasingly more important with underdoping.

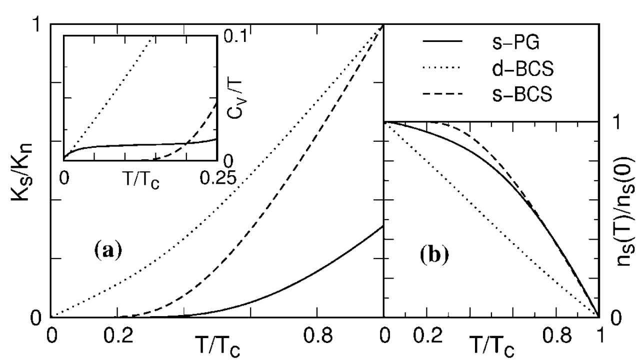

FIG. 1. Temperature dependence of (a) Knight shift, K_s, specific heat C_v/T (inset), and (b) superfluid density n_s in conventional s- and d-wave (BCS) and short ξ (PG) s-wave cases, calculated at $(n, -g/4t_\parallel)$ as follows: s-BCS: (0.5, 0.5); s-PG: (0.5, 0.7); d-BCS: (0.8, 0.225); d-PG: (0.92, 0.56).

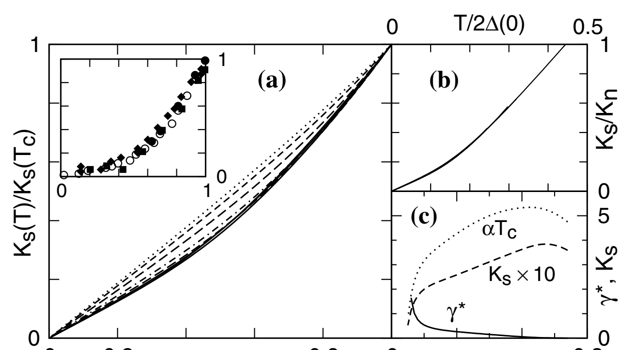

FIG. 2. (a)–(b) Scaling behavior of the T dependence of Knight shift, K_s, for the cuprates with respect to doping x, and (c) doping dependence of $\gamma(T) = \gamma^* + \alpha T$ and K_s at T_c^-. In (a) [and (b)], x varies from 0.05 to 0.2 from top to bottom. For comparison, shown in the inset are experimental data from Ref. [22] on underdoped (■), optimally doped (○, ●), and overdoped (◆) $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals. As per the experimental convention for $\varphi_{\mathbf{k}}$, we use $2\Delta(0)$ in (b).

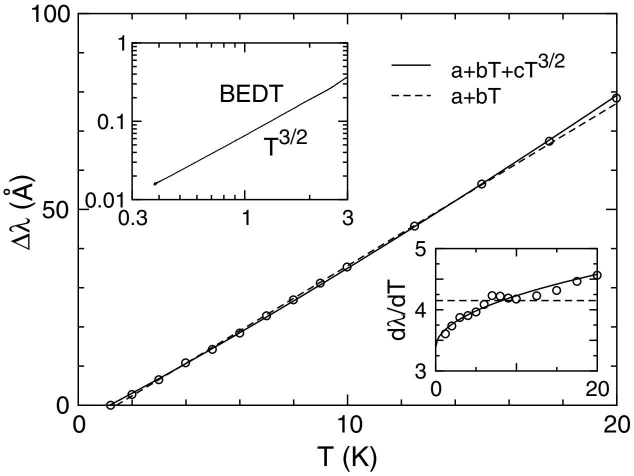

FIG. 3. Comparison of penetration depth data [23], $\Delta\lambda$, along the a axis, in nominally pure $YBCO_{6.95}$ single crystal, with different theoretical fits corresponding to BCS d-wave (dashed curve) and to BCS-BEC (solid curve) predictions. The corresponding derivatives are plotted in the lower inset. In the upper inset are experimental data ($\Delta\lambda$ vs T) for the organic superconductor BEDT from Ref. [24].

In Fig. 3 we present a-axis penetration depth data, $\Delta\lambda(T)$, in a nominally clean optimally doped $YBa_2Cu_3O_{7-\delta}$ (YBCO) single crystal, from Ref. [23], along with d-wave fits to our BCS-BEC theory and to a straight line associated with a BCS superconductor. Because these two fitted curves are essentially indistinguishable, in the lower inset we plot the slopes $d\lambda/dT$ where the difference between the two sets of curves is more apparent. Here it is shown that the low temperature downturn of the derivative, seen to a greater or lesser extent in all $\Delta\lambda(T)$ measurements, fits our predicted $T^{1/2}$ + const dependence rather well. This downturn has been frequently associated with impurity effects, which yield a linear in T slope for $\Delta\lambda$ at very low T, and, in this case, provide a poorer fit. While these cuprate experiments were performed on a nearly optimal sample, the same analysis of an underdoped material yielded similarly good agreement, but with a $T^{3/2}$ coefficient about a factor of 2 larger. Future more precise and systematic low T experiments on additional underdoped samples are needed. Plotted in the upper inset are data [24] on the organic superconductor κ-$(ET)_2Cu[N(CN_2]Br$ (BEDT, $T_c \approx 11$ K) which fit a pure $T^{3/2}$ power law over a wide temperature regime; in contrast to the cuprates, there is no leading order linear term. At present, there seems to be no other explanation (besides the pairon mechanism presented here) for this unusual power law at the lowest temperatures.

In summary, within a BCS-BEC crossover theory (based on the Leggett ground state), we find that new low T power laws associated with a quasi-ideal gas of bosonic pair excitations appear in the thermodynamic and transport properties, which may be generally relevant to short ξ superconductors.

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