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Spin-fluctuation-induced superconductivity in $YBa_2Cu_3O_{6.9}$

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We show that, independent of the choice of values for the carrier concentration n and specific heat in the normal state, existing experiments on the superconducting properties of polycrystalline $YBa_2Cu_3O_{6.9}$ are consistent with calculations carried out in the clean limit. We find, for a wide choice of n, a substantial exchange enhancement of the Pauli susceptibility. For $n<5\times10^{21}$ cm^{-3}, long-wavelength spin fluctuations, with a characteristic temperature, $T_{SF}\sim600$ K, can give rise to a strong-coupling superconducting transition to a triplet Balian-Werthamer phase. On the other hand, if $n>10^{22}$ cm^{-3} antiferromagnetic spin fluctuations must be invoked if the pairing is of magnetic origin.

In the search for the physical origin of superconductivity in the high-temperature superconductors,[1,2] an attractive interaction between electrons resulting from the virtual exchange of antiferromagnetic spin fluctuations has emerged as one of the leading possibilities.[3] Heavy-electron systems are, at present, the only known systems where this mechanism has been shown to be responsible for superconductivity.[4] The possibility that the high-temperature superconductors are part of the heavy-electron family has been discussed by Pethick and Pines.[4] They argue that, because the carrier concentration is low in the high-temperature superconductors, the characteristic spin-fluctuation temperatures will be much larger than those found in the heavy-electron systems, with a concomitant increase in the superconducting transition temperature.

In this Communication we consider the evidence for spin-fluctuation-induced superconductivity which is provided by experiments[5-8] on polycrystalline samples of $YBa_2Cu_3O_{6.9}$, if one makes the assumption that the behavior of these samples can be interpreted as resulting from quasiparticles which form, in first approximation, a three-dimensional (3D) one-component Fermi liquid in both the normal and superconducting states.

The relevant quasiparticle properties are then the density of states at the Fermi surface $N(0)$, the Fermi velocity v_F, and the Landau parameters F_0^a and F_1^s. Rather than extracting the density of states, $N(0)=(3/\pi^2)\gamma(T_c)$, from the specific-heat jump at the superconducting transition, ΔC_v, by assuming that the coupling parameter

$$\alpha=\frac{\Delta C_v}{1.43\gamma(T_c)T_c} \qquad (1)$$

takes its weak-coupling Bardeen-Cooper-Schrieffer (BCS) value of unity, we use the measurement of the temperature dependence of the London penetration depth, $\lambda(T)$ (Ref. 5), to obtain the product $N(0)v_F^2$. For a given choice of carrier concentration, one can then determine $N(0)$, v_F, and α.

In Leggett's treatment of a superconducting Fermi liquid,[9] the London penetration depth is given by

$$\lambda^2(0)/\lambda_L^2(T)=[\rho_s^0(T)/\rho]/1+\frac{F_1^s}{3}[\rho_n^0(T)/\rho] \ , \qquad (2)$$

where $\rho_s^0(T)$, the noninteracting superfluid density, is to a good degree of approximation for the strong-coupling superconductors[10] given by

$$\rho_s^0(T)/\rho\equiv[1-\rho_n^0(T)/\rho]\simeq[1-(T/T_c)^4] \qquad (3)$$

for a clean superconductor with an isotropic gap, where $\rho_n^0(T)$ is the noninteracting normal fluid density. The parameter F_1^s is the dimensionless measure of the strength of the backflow field which describes the quasiparticle current-current coupling; for a crystal with a spherical Fermi surface, it defines the dynamic mass, m_c, found in the absence of backflow, through the relation $m^*/m_c=(1+F_1^s/3)$. The dynamic mass m_c is measured directly by $\lambda(0)$, the zero-temperature penetration depth, which is given by

$$\lambda(0)=\frac{m_c c^2}{4\pi ne^2}=\frac{3c^2}{4\pi e^2}\frac{1}{N(0)v_F^2(1+F_1^s/3)} \ . \qquad (4)$$

Here n is the quasiparticle density and we introduce the explicit dependence of the penetration depth on the product, $N(0)v_F^2$, in the second form for $\lambda(0)$. In the clean limit $\lambda_L(0)=\lambda(0)$. In Fig. 1 we present two possible fits to the penetration depth data of Harshman *et al.*;[5] in Table I we list the resulting values of $N(0)v_F^2$.

To explore the consistency of these results with measurements of the other superconducting parameters, we make the following simple ansatz,

$$\Delta(T)=\alpha^{1/2}\Delta_{BCS}(T) \ , \qquad (5)$$

where $\Delta_{BCS}(T)$ is the weak-coupling BCS value for the gap, with $\Delta_{BCS}(0)=1.76kT_c$. This, in fact, is just the model successfully applied by Padamsee, Neighbor, and

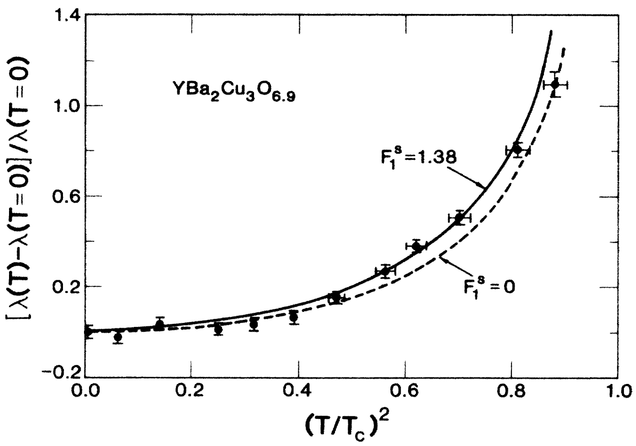

FIG. 1. The temperature dependence of the penetration depth with $F_1^s = 0$, the dashed line, and $F_1^s = 1.38$, the solid line. The experimental points are from Ref. 5.

Shiffman[11] to study the properties of Pd−x at.% In and other strong-coupling superconductors. Near T_c Eq. (5) is exact; however, at lower temperatures, to the extent α is temperature dependent,[12] it is an approximation. On combining Eqs. (1) and (5) we can calculate the coherence length, $\xi(0)$ in the clean limit,

$$\xi(0) = \frac{0.91 v_F}{\pi\Delta(0)} = 0.36 \left[\frac{v_F^2 N(0)}{T_c \Delta C_v} \right]^{1/2} , \qquad (6)$$

and from this we can obtain a number of properties of the superconductor. These include the upper critical field, $H_{c2}(0) = \Phi_0/2\pi\xi^2(0)$, and its slope near T_c, $H'_{c2} = -H_{c2}(0)/0.69T_c$, with $\Phi_0 = hc/2e$. $\xi(0)$ and the measured value of $\lambda(0)$ yield the Ginzburg-Landau parameter $K_{GL} = \lambda(0)/\xi(0)$; this in turn yields the thermodynamic critical field $H_c(0) = H_{c2}(0)/\sqrt{2}K_{GL}$ and the lower critical field $H_{c1}(0) = H_{c2}(0)\ln K_{GL}/2K_{GL}^2$. These superconducting properties are likewise given in Table I.

A physical quantity of interest in connection with spin-fluctuation-induced superconductivity is the Pauli susceptibility, χ_P, which in Fermi-liquid theory is given by

$$\chi_P = \mu_B^2 N(0)/(1+F_0^a) , \qquad (7)$$

where μ_B is the Bohr magneton. To extract χ_P from the susceptibility, $\chi_{expt} = 3.9\times10^{-4}$ emu/mol, measured by Cheong *et al.*,[6] it is necessary to consider corrections coming from core electron and quasiparticle diamagnetism. The latter, the Landau-Peierls term, turns out to be negligible, since when account is taken of Fermi-liquid effects, $\chi_{LP} = -\frac{1}{3}(1+F_0^a)(m/m^*)^2\chi_P$, and for all choices of carrier density (see Table II) gives rise to corrections $\sim 1\%$. The core correction used by Cheong *et al.*[6] is -1.7×10^{-4} emu/mol, so that $\chi_P = 5.6\times10^{-4}$ emu/mol.

To determine the exchange enhancement parameter, F_0^a and $N(0)$, we consider three possible values for n: 3.5×10^{21} cm^{-3}, 4.5×10^{21} cm^{-3}, and 10^{22} cm^{-3}. These densities span a range of values obtained from simple counting arguments and Hall measurements assuming a one-band equivalent isotropic model. The resulting values of the thermal effective mass, m^*, γ, F_0^a, α, and $\Delta(0)$ are given in Table II. There one sees a considerable range of quasiparticle masses and exchange enhancements consistent with the measured values of the superconducting parameters displayed in Table I. If the quasiparticle density of states is ~ 8 mJ/mol K^2, then F_0^a is sufficiently negative that the coupling of electrons to ferromagnetic spin fluctuations can be shown to give rise to a superconducting transition temperature close to that observed. This result follows from a calculation of the transition temperature using the Patton-Zaringhalam method for a charged system,[13]

$$T_c = 1.13\varepsilon T_F e^{6/(1+A_0^a)} , \qquad (8)$$

where $A_0^a = F_0^a/(1+F_0^a)$. In Table II we give the T_c values computed using the cutoff, $\varepsilon T_F = (1+F_0^a)T_F = T_{SF}$. In most applications of this method[13] for calculating T_c, $\varepsilon T_F < T_{SF}$, and as such the T_c values we find should be viewed as upper bounds. What we would conclude from this analysis is that triplet pairing resulting from the exchange of ferromagnetic spin fluctuations is a potential candidate for the high-T_c superconductor $YBa_2Cu_3O_{6.9}$. Since the measurements of $\lambda_L(T)$ provide no evidence for nodes in the energy gap, the resulting phase would be the Balian-Werthamer[14] (BW) triplet phase. Given the size of the strong-coupling corrections one might have expected the Anderson-Brinkman-Morel (ABM) state to be stabilized as in liquid ^{3}He (Ref. 15). However, as in the heavy-fermion superconductors, naive arguments, that ignore band structure, crystal symmetry, etc., can give the wrong phase. We further note that for this choice of $N(0)$, the strong-coupling parameter, α is large, so large indeed as to rule out a phonon mechanism, if one adopts the limiting values of $\Delta C_v/\gamma T_c$ computed by Blezius and Carbotte[16] using the Eliashberg equations.

On the other hand, for $\gamma(T_c)$ values in excess of 20 mJ/mol K^2, corresponding to $n \sim 10^{22}$ cm^{-3}, the exchange enhancement, while substantial, is not sufficient

TABLE I. Self-consistent parameters of the superconducting state, with $\lambda(0) = 1365$ Å.

$F_1^s/3$	$\xi(0)$	K_{GL}	$H_{c2}(0)$ (T)	$H'_{c2}(T_c)$ (T K)	$H_c(0)$ (T)	$H_{c1}(0)$ (G)	$N(0)v_F^2$ (10^{49} cm^{3}g^{-1})
0	14.4	94.7	156	−2.5	1.16	396	0.50
0.46	12	114	224	−3.6	1.39	408	0.34
Expt.	· · ·	· · ·	· · ·	1.3–3.0[a]	· · ·	400[a]	· · ·

[a]References 2 and 7.

TABLE II. Dependence of the properties of the normal and superconducting state on the density of carriers.

n_h (10^{21} cm^{-3})	$F_1^s/3$	m^*/m	γ (mJ/mol K^2)	F_0^a	α	$\frac{\Delta_a(0)}{K_BT_c}$	T_c (K)	T_{SF} (K)
3.5	0	2.32	5.7	−0.86	5.1	3.98	205	590
3.5	0.46	3.38	8.3	−0.8	3.5	3.31	88	590
4.5	0	3.0	8.0	−0.8	3.6	3.34	122	770
4.5	0.46	4.38	11.7	−0.71	2.5	2.76	13.6	770
10	0	6.6	23	−0.43	1.3	1.97	...	1700
10	0.46	9.7	34	−0.18	0.86	1.63	...	1700

for ferromagnetic spin fluctuations, i.e., paramagnons, to produce high-temperature superconductivity. This regime of parameter space would be consistent with antiferromagnetic spin fluctuations, whose coupling to quasiparticles can both enhance the effective mass and give rise to superconductivity.[4] The presence of strong antiferromagnetic correlations in this system would also tend to suppress the p-wave pairing interaction, g_1. To see how this works qualitatively we write

$$g_1 \sim -\int_0^{2k_F} \frac{q\,dq}{k_F^2}\left(1-\frac{q^2}{2k_F^2}\right) v_q^2\chi(q) \ , \tag{9}$$

where $v_q^2\chi(q)$ is the spin-fluctuation exchange interaction and $(1-q^2/2k_F^2)$ is the $l=1$ Legendre polynomial. If χ_q exhibits antiferromagnetic tendencies it will be peaked at finite q. This would tend to suppress a p-wave state and favor, for example, d-wave pairing, in which case one would expect to find nodes in the energy gap. For the p-wave state to be strongly favored $\chi(q)$ should be peaked for small q. The suppression of antiferromagnetic correlations away from a half-filled band and the appearance of a ferromagnetic metal phase in the large-U Hubbard model was first proposed by Nagaoka.[17] This tendency toward a ferromagnetic metal phase is possible in 2D and even more likely in 3D when the system moves away from half filling. This behavior was also seen in the two-band model recently proposed by Prelovsek[18] where moving away from half filling occurs by doping with holes on the oxygen sites.

While the nodal structure of the energy gap as determined by $\lambda(T)$ can distinguish between the BW phase and a d-state pairing phase, the electromagnetic response and sound attenuation in the BW phase will be those of a s-wave singlet state. However, the latter two phases differ in their magnetic properties. The long-wavelength static Pauli susceptibility, $\chi_{\rm BW}$, is, for $T \ll T_c$, given by[15]

$$\frac{\chi_{\rm BW}}{\chi_P} = \frac{\frac{2}{3}(1+F_0^a)}{1+\frac{2}{3}F_0^a} \simeq 0.22,\ 0.29,\ 0.37 \ , \tag{10}$$

where the three values of this ratio come from the F_0^a's given in the first four rows of Table II. In neutron scattering this limiting value should be observed for wave vectors, $q < 1/\xi(0) \simeq 0.08$ Å^{-1}. As q increases the wave-vector-dependent susceptibility $\chi_{\rm BW}(q)$ reaches a maximum around $q=0.4$ Å^{-1}, since $\chi_{\rm BW}(q)$ approaches $\chi_P(q)$, as q increases while $\chi_P(q)$ falls off with increasing q.

We have assumed that $YBa_2Cu_3O_{6.9}$ is an isotropic, one-component 3D system. Given the important role which dimensionality plays in band structure[19] and in various theories[3,20] it is desirable to have an experimental measure of it. In the theory of layered superconductors[21] the dimensionality parameter t can be obtained from the layer spacing d and the measured slopes of the upper critical field in the parallel, $H'_{c2}(T_c;\parallel)$, and perpendicular, $H'_{c2}(T_c;\perp)$, directions,[21]

$$t = \frac{4c\hbar}{\pi e d^2 T_c}\frac{H'_{c2}(T_c;\perp)}{[H'_{c2}(T_c;\parallel)]^2} \ . \tag{11}$$

If $t \ll 1$ the system is 2D and single-particle hopping between the layers is much smaller than the coherent Josephson tunneling; while for $t \sim 1$, the system resembles a 3D anisotropic system. From the measurements of Worthington, Gallagher, and Dinger[22] on single-crystal $YBa_2Cu_3O_{6.9}$, using $d=11.65$ Å, where d is the distance between identical Cu–O layers, we find that $t \simeq 0.6$; thus this system is far from being a layered superconductor. A consequence of this large value of t is that fluctuations above T_c will be 3D over a wide temperature range, as has been observed in the conductivity measurements of Freitas, Tsuei, and Plaskett.[23] A possible way to reconcile the 2D band structure with the 3D superconducting properties has been proposed by Tesanovic,[24] who suggest that both 2D single-particle propagation and a strong Josephson interlayer coupling are present.

Given the importance of the strong-coupling corrections, the anisotropy observed by Worthington *et al.*[22] may not originate solely in an anisotropy in the effective mas, or more generally, in the density of states, since any strong-coupling corrections will also be anisotropic. For example, if the strong-coupling corrections to $H'_{c2}(T_c;\perp)$ are smaller than to $H'_{c2}(T_c;\parallel)$ a large mass anisotropy would be needed to explain the data, while the BW phase would be favored over the ABM phase, since the gap in the perpendicular direction would then be closer to the weak-coupling value than that in the plane.

We now turn to the question of whether the use of the clean limit is justified for $YBa_2Cu_3O_{6.9}$. From the polycrystalline[6] data the resistivity extrapolated to $T=0$ is $\sim 10^2$ $\mu\Omega$ cm which gives a mean free path, l, that lies between 20 Å ($n = 10^{22}$ cm^{-3}) and 55 Å ($n = 3.5 \times 10^{21}$ cm^{-3}). Clearly the clean limit analysis is valid for low densities and only marginally valid for higher densities. An analysis of the data that does not assume a clean limit has been carried out by Quader and Salamon,[25] who reach similar conclusions about the importance of exchange enhancement and strong-coupling corrections, but who calculate smaller values of T_c.

Our use of an isotropic one-component 3D Fermi-liquid model for the normal state is more difficult to justify, in light of the substantial resistivity anisotropy found by Tozer *et al.*[26] However, until realistic band-structure structure calculations are combined with appropriate anisotropic Fermi-liquid and Ginzburg-Landau theories, and one has a complete range of single-crystal measurements, including the anisotropic spin susceptibility, an isotropic one-component model to treat polycrystalline samples provides a useful framework for a systematic analysis of experimental data.

To test the Fermi-liquid model we have proposed, it would be useful to have measurements of $\lambda(T)$, χ_P, $H_{c2}'(T)$, ΔC_v, etc., as functions of the oxygen content ranging for example from $YBa_2Cu_3O_{6.5}$–$YBa_2Cu_3O_7$. This would tell us if the spin-fluctuation pairing mechanism we are proposing correlates with the susceptibility enhancement. Measurements of these properties as functions of pressure would likewise be instructive. Finally, a similar analysis is being carried out on Sr-doped La_2CuO_4 and will be reported in a subsequent communication.

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