Critical temperature and superfluid density suppression in disordered high-T_c cuprate superconductors

M. Franz
Department of Physics and Astronomy, Johns Hopkins University, Baltimore, Maryland 21218

C. Kallin,* A. J. Berlinsky,* and M. I. Salkola
Department of Physics, Stanford University, Stanford, California 94305

(Received 8 May 1997)

We argue that the standard Abrikosov-Gorkov (AG)-type theory of T_c in disordered d-wave superconductors breaks down in short coherence length high-T_c cuprates. Numerical calculations within the Bogoliubov–de Gennes formalism demonstrate that the correct description of such systems must allow for the spatial variation of the order parameter, which is strongly suppressed in the vicinity of impurities but mostly unaffected elsewhere. Suppression of T_c is found to be significantly weaker than that predicted by the AG theory, in good agreement with experiment. [S0163-1829(97)07334-7]

Sensitivity of the superfluid density and the critical temperature to moderate amounts of substitutional disorder served as an early indicator of the unconventional nature of the order parameter in high-T_c cuprate superconductors. The theory of dirty d-wave superconductors, pioneered in this context by Annett, Goldenfeld, and Renn[1] and by Hirschfeld and Goldenfeld,[2] successfully explained the observed T^2 dependence of the superfluid density $\rho_s(T)$ in $YBa_2Cu_3O_{7-\delta}$ (YBCO) films and single crystals, attributing it to the effect of the finite density of states at the Fermi level induced by impurities, treated as unitary scatterers. Crossover to T-linear behavior was predicted for lower impurity content, which was later observed in very clean YBCO single crystals by Hardy *et al.*[3] The theory was subsequently refined[4–6] to provide a good quantitative description of the low-temperature behavior of $\rho_s(T)$.

Similar models have been employed to predict the change of T_c from its clean value T_{c0} due to disorder,[5,7] essentially by extending the standard Abrikosov-Gorkov (AG) theory[8] to the case of d-wave superconductors with scalar impurities. It has been noted that the experimentally observed T_c is much more robust than one would expect from these simple models, when measured against the corresponding change in $\rho_s(0)$. Figure 1 illustrates this point by showing the experimental T_c versus $\rho_s(0)$ for YBCO samples disordered by different types of disorder as obtained by various groups[9–13] and the corresponding theoretical prediction.[5] While there exists a considerable spread in the experimental data, it is quite evident that the theory systematically overestimates the suppression of T_c, perhaps by as much as a factor of 2 in the cases of Refs. 9,11,13. In order to remedy this situation, more realistic models have been considered, taking into account strong coupling corrections within the Eliashberg formalism[7] together with realistic band structures,[14] proximity of the Fermi level to a van Hove point,[15] and the details of the pairing interaction within the spin-fluctuation model.[16] While some improvement over the simple model can be achieved for carefully selected parameters, the discrepancy between theory and experiment remains in place, suggesting that methods traditionally employed to determine the suppression of T_c by impurities are inadequate for the high-T_c cuprates. It has also been suggested that perhaps the effects of disorder on the transition from the rather peculiar and poorly understood normal state cannot be satisfactorily described within the framework of a simple BCS-like mean field theory.[17]

In the present article we show that one can, in fact, formulate an adequate description within mean-field theory, if the spatial variations of the order parameter caused by random disorder are accounted for in a fully self-consistent manner. We present a simple argument for the breakdown of the conventional AG-type theory (which enforces a uniform gap averaged over disorder) in superconductors with very short coherence lengths such as the high-T_c cuprates. This argument is then given substance by numerical calculations within the Bogoliubov–de Gennes (BdG) framework, carried

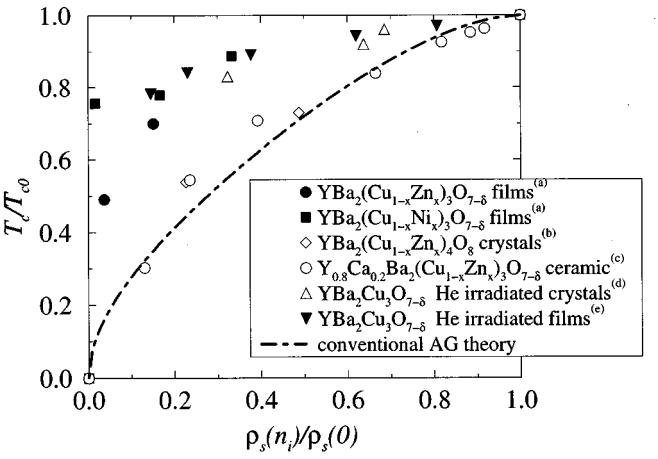

FIG. 1. Normalized critical temperature versus the normalized zero temperature superfluid density. Experimental data were obtained by (a) mutual inductance (Ref. 9), (b) (d) infrared reflectance (Refs. 10,11), (c) muon spin rotation (Ref. 12), and (e) field-current density analysis (Ref. 13). Theoretical curve is from Ref. 5.

out for a model d-wave superconductor with a random distribution of nonmagnetic point impurities. Such calculations convincingly demonstrate that the rapid drop in the predicted T_c is an artifact of AG theory resulting from spatial averaging of the order parameter. The true critical temperature obtained from a fully self-consistent solution of the BdG equations is always higher, in agreement with experiment.

In a d-wave superconductor nonmagnetic impurities are pair breaking and lead to suppression of T_c. A simple modification of the original AG approach[8] yields an equation for T_c of the familiar form[5,7,14]

$$\ln\left(\frac{T_{c0}}{T_c}\right)=\psi\left(\frac{1}{2}+\frac{\alpha T_{c0}}{2\pi T_c}\right)-\psi\left(\frac{1}{2}\right), \tag{1}$$

where ψ is a digamma function and $\alpha=1/2\tau T_{c0}$ is a pair breaking parameter. In the limit of unitary scatterers, which is relevant for Zn and other solutes in YBCO,[2] the scattering rate is given by $1/2\tau=n_i/\pi N(0)$, with n_i being the number density of impurities and $N(0)$ the normal-state density of states at the Fermi level. For α small we have

$$\frac{T_c^{\mathrm{AG}}}{T_{c0}}\simeq 1-\frac{\pi}{4}\alpha=1-\frac{n_i}{4N(0)T_{c0}}, \tag{2}$$

which is in fact an excellent approximation to the full solution of (1) for $\alpha\lesssim\alpha_c/3$, where $\alpha_c=0.88191$ is the critical pair breaking parameter $[T_c(\alpha_c)=0]$.

One of the crucial assumptions entering the derivation of Eq. (1) is that the position dependent order parameter $\Delta(\mathbf{r})$ can be replaced in the gap equation by its spatial average $\overline{\Delta}$. Such a procedure is valid when the gap varies over a length scale that is large compared to the average spacing between the impurities l_i. In the vicinity of T_c significant variations of the order parameter take place on the length scale set by the Ginzburg-Landau temperature dependent coherence length $\xi(T)$, which is related to the low-temperature BCS coherence length $\xi_0=v_f/\pi\Delta$ by $\xi(T)\simeq\nu\xi_0(1-T/T_c)^{-1/2}$, where $\nu=0.74$ in a conventional s-wave superconductor with a spherical Fermi surface.[18] For a d-wave superconductor ν will be a different number of order unity, depending on the precise k-space structure of the gap function.

Consider the problem of a single unitary impurity in a d-wave superconductor. The order parameter will be strongly suppressed at the impurity site and it will recover its bulk value over the distance $\sim\xi(T)$.[19,20] Since a single impurity cannot affect T_c of a macroscopic sample, this coherence length is given by

$$\xi(T)\simeq\nu\xi_0(1-T/T_{c0})^{-1/2}. \tag{3}$$

For a small finite density of impurities with average spacing $l_i\gg\xi_0$, except very close to T_c, the areas of depressed order parameter around individual impurities will not overlap, and most of the sample will remain completely unaffected. Thus it is still reasonable to use Eq. (3) with the unperturbed T_{c0} for $\xi(T)$. When these areas begin overlapping, i.e., when $\xi(T)\gtrsim l_i$, the entire sample is affected in the sense that the order parameter is suppressed everywhere. The temperature at which this happens provides a lower bound for the true T_c of the sample. Using $l_i=2a_0/\sqrt{\pi n_i}$ (where a_0 is the ionic lattice spacing) this condition becomes

$$\frac{T_c}{T_{c0}}\gtrsim 1-\frac{\pi}{4}\left(\frac{\nu\xi_0}{a_0}\right)^2 n_i. \tag{4}$$

If T_c^{AG} predicted from Eq. (2) falls below this lower bound, one may conclude that the AG theory is not self-consistent, since the true coherence length at $T=T_c^{\mathrm{AG}}$ is smaller than the average distance between the impurities and the averaging over $\Delta(\mathbf{r})$ is not allowed.

In order to get a rough idea of how restrictive this argument is, we note that the prefactor of n_i in Eq. (2) can be converted into a length scales ratio $1/N(0)T_{c0}\sim(\pi^2/1.13)(\xi_0/a_0)$.[21] Then, combining this with Eq. (4) one can derive a rough estimate for the range of validity of the AG theory of the form

$$\xi_0/a_0\gtrsim\pi/1.13\nu^2\simeq 5. \tag{5}$$

Clearly, for conventional low-T_c superconductors this condition is easily satisfied as ξ_0 is typically large compared to a_0. For high-T_c cuprates, however, the situation is quite different since typically ξ_0 is several lattice spacings ($\xi_0\approx 4a_0$ in YBCO). A more careful estimate using realistic values of T_{c0} and $N(0)$ for YBCO,[14] confirms that the result of our rough estimate (5) holds. This indicates that the usage of the AG theory for this material is probably not justified.[22]

While the argument presented above is admittedly crude, we believe that together with the experimental evidence discussed above it points to the necessity of studying the effects of spatial variations of the order parameter in disordered short-coherence-length superconductors. We now present results of a numerical calculation that strongly support the picture outlined above.

We employ an extended Hubbard model on a square lattice with nearest-neighbor attraction and on-site repulsion:

$$H=-t\sum_{\langle ij\rangle\sigma}c_{i\sigma}^{\dagger}c_{j\sigma}-\mu\sum_{i\sigma}n_{i\sigma}+\sum_{i\sigma}V_i^{\mathrm{imp}}n_{i\sigma}+V_0\sum_i n_{i\uparrow}n_{i\downarrow}+\frac{V_1}{2}\sum_{\langle ij\rangle\sigma\sigma'}n_{i\sigma}n_{j\sigma'}, \tag{6}$$

where $\langle ij\rangle$ stands for nearest-neighbor pairs, and the notation is otherwise standard. Such a model, treated within a self-consistent BdG theory, has been used previously to study vortices[23,24] and impurities[19,20,25] in d-wave superconductors. The impurities are modeled by $V_i^{\mathrm{imp}}=V^{\mathrm{imp}}\gg|t|$ at randomly chosen sites with density n_i and $V_i^{\mathrm{imp}}=0$ elsewhere. We solve this Hamiltonian within the standard mean-field theory as described in Ref. 20. All physical quantities of interest can be derived from the quasiparticle amplitudes $[u_n(\mathbf{r}),v_n(\mathbf{r})]$, which satisfy a system of BdG equations[26]

$$\begin{pmatrix}\hat{\xi} & \hat{\Delta}\\ \hat{\Delta}^* & -\hat{\xi}^*\end{pmatrix}\begin{pmatrix}u_n\\ v_n\end{pmatrix}=E_n\begin{pmatrix}u_n\\ v_n\end{pmatrix}, \tag{7}$$

with

$$\hat{\xi}u_n(\mathbf{r}_i)=-t\sum_{\delta}u_n(\mathbf{r}_i+\delta)+(V_i^{\mathrm{imp}}-\mu)u_n(\mathbf{r}_i),$$

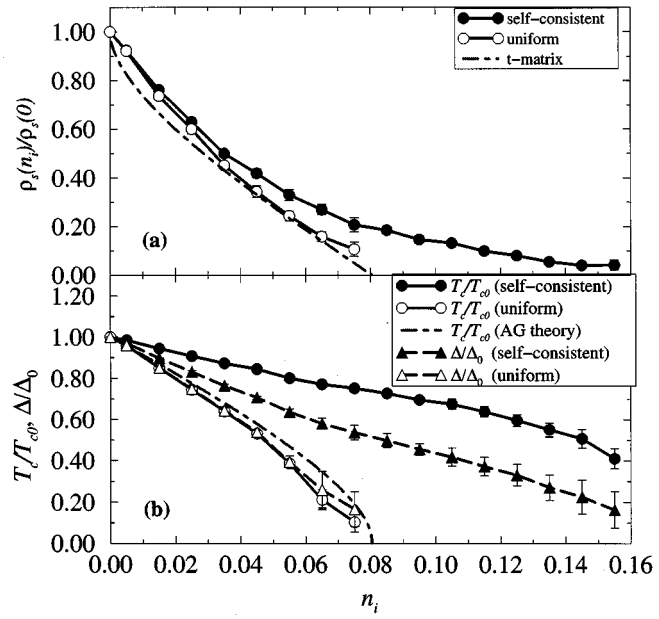

FIG. 2. (a) Normalized zero-temperature superfluid density as a function of impurity concentration: fully self-consistent solution of BdG equations (solid symbols), solution with uniform order parameter (open symbols), and analytic t-matrix solution with unitary scatterers from Ref. 5 (dash-dotted line). (b) Normalized critical temperature and average gap. Dash-dotted line is numerical solution of the AG equation (1).

$$\hat{\Delta} v_n(\mathbf{r}_i) = \Delta_0(\mathbf{r}_i) v_n(\mathbf{r}_i) + \sum_{\delta} \Delta_{\delta}(\mathbf{r}_i) v_n(\mathbf{r}_i + \delta), \qquad (8)$$

subject to the constraints of self-consistency

$$\Delta_0(\mathbf{r}) = V_0 \sum_n u_n(\mathbf{r}) v_n^*(\mathbf{r}) \tanh(E_n/2k_BT),$$

$$\Delta_{\boldsymbol{\delta}}(\mathbf{r}) = \frac{V_1}{2} \sum_n [u_n(\mathbf{r}+\boldsymbol{\delta}) v_n^*(\mathbf{r}) + u_n(\mathbf{r}) v_n^*(\mathbf{r}+\boldsymbol{\delta})] \tanh(E_n/2k_BT). \qquad (9)$$

Here Δ_0 and Δ_δ are the on-site and nearest-neighbor pairing amplitudes respectively with $\delta = \pm\hat{x}, \pm\hat{y}$ and the summation is over positive eigenvalues E_n only. For a finite $L \times L$ system we solve the BdG equations (7) by exact numerical diagonalization using a suitable guess for the initial gap functions. We then compute new gap functions from Eq. (9) and iterate this process until self-consistency is established[20,23].

The superfluid density $\rho_s(T)$ is evaluated from the standard linear response formula appropriate for lattice models[19,27]

$$\rho_s(T)/4 = \langle -K_x \rangle - \Lambda_{xx}(q_x = 0, q_y \to 0, \omega = 0), \qquad (10)$$

where $\langle -K_x \rangle$ is the average kinetic energy along the $\hat{x}$ direction, and $\Lambda_{xx}(\mathbf{q},\omega)$ is a diagonal element of the current-current correlation function. Both $\langle -K_x \rangle$ and $\Lambda_{xx}(\mathbf{q},\omega)$ can be written in terms of (u_n, v_n) and E_n, resulting in lengthy expressions which we omit here for brevity.

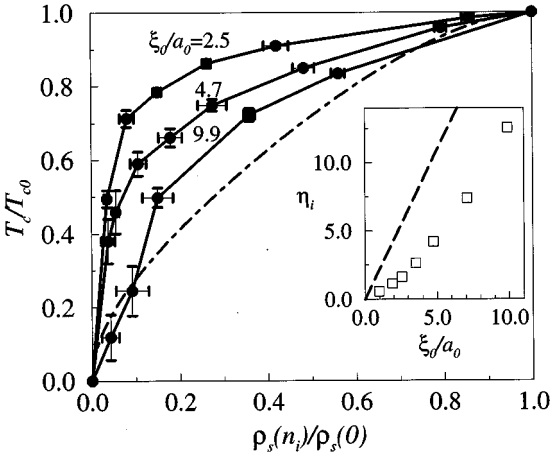

FIG. 3. Normalized critical temperature versus normalized zero-temperature superfluid density as computed from a fully self-consistent solution of BdG equations for systems with different coherence lengths. The error bars reflect the statistical scatter of data for six different impurity configurations. Parameters used are $L=22$, $V^{\rm imp}=-100t$, $\mu=-0.36t$, and $V_0=-V_1=(0.80,1.05,1.40)t$ for $\xi_0=(9.9,4.7,2.5)a_0$, respectively. Dash-dotted line is a conventional AG solution from Ref. 5. Inset: the rate of change η_i versus ξ_0/a_0 (open squares), and the expected behavior from AG theory $\eta_i = 2.18(\xi_0/a_0)$ (dashed line).

Determination of T_c is more complicated since the number of iterations needed to self-consistently solve Eqs. (7),(9) to a required accuracy increases dramatically near the transition (and presumably diverges at $T=T_c$). We use two different methods to overcome this difficulty: (i) we improve our initial guess for $\Delta(\mathbf{r})$ at each new temperature by extrapolating from previous temperature points, and (ii) we locate T_c iteratively by following the development of the gap initialized to infinitesimal value.

Figure 2 summarizes the results of our numerical calculation, for parameters resulting in $\xi_0 \approx 4a_0$ (we use $V_0=-V_1=1.13t$, $\mu=-0.36t$, and $V^{\rm imp}=100t$ for a system size $L=22$). Panel (a) shows the normalized zero-temperature superfluid density as a function of n_i obtained from a fully self-consistent solution of Eqs. (7), (9) and from a solution with an enforced uniform order parameter. For comparison a conventional analytic solution within the t-matrix formalism is also shown [we use Eq. (15) of Ref. 5)]. As one would expect the uniform solution is very close to the analytical one for all values of n_i. The self-consistent solution agrees well with the two, except for large n_i. Our results for T_c [panel(b)] are much more interesting. While T_c computed for the uniform solution tracks the analytic solution of the AG equation (1), the true critical temperature obtained using a self-consistent solution is much higher. In fact at $n_i \approx 0.08$ where $T_c^{\rm AG}=0$, the true critical temperature is still more than 70% of T_{c0}. Also note that the average zero-temperature gap does not scale with T_c, as one would expect from BCS theory.

We have carried out calculations for several different sets of parameters, all showing similar behavior (see Fig. 3). As the coherence length grows, the discrepancy between the true critical temperature and $T_c^{\rm AG}$ diminishes, as expected from the analysis presented above. For $\xi_0/a_0 \approx 4$ as in YBCO, the

discrepancy is substantial, and can easily account for the experimentally observed robustness of T_c. The critical impurity density for this case is $n_i^c \approx 0.17$. Since Zn substitutes primarily for the planar copper sites in YBCO (Ref. 28) and only 2/3 of all Cu atoms reside in the planes, our result implies a bulk critical density of $n_{\rm bulk}^c \approx 0.10$. This is in reasonable agreement with the experimentally observed $n_{\rm bulk}^c = 0.08 - 0.10$ for Zn doped YBCO.[29] By contrast conventional models tend to underestimate $n_{\rm bulk}^c$ by a factor of 2 or more.[15] The inset to Fig. 3 shows the dependence of the initial rate of change of T_c with n_i, $\eta_i = T_{c0}^{-1}(dT_c/dn_i)_{n_i=0}$, on ξ_0. This quantity also deviates significantly from the linear $\eta_i \sim (\xi_0/a_0)$ behavior expected from the AG prediction (2), showing instead a quadratic behavior consistent with Eq. (4).

In closing we comment on the appropriateness of using BdG theory to calculate T_c in short coherence length materials. As with all conventional theories of superconductivity, BdG theory is a mean field theory and hence is incapable of modeling the important effects of phase fluctuations on the ordering temperature in short coherence length, highly anisotropic materials such as the high-T_c oxides. On the other hand, unlike many more specialized mean field theories, including AG theory, BdG theory is well-suited to calculating the effects of static spatial disorder on the order parameter amplitude and on the mean field T_c. Since the static random potential does not couple to the phase, the effects of spatial randomness and thermal phase fluctuations are largely decoupled. To a good approximation, at least for the dilute case, the effects of impurities can be calculated within mean field theory, while the effects of phase fluctuations, in reducing T_c from its mean field value, require a proper theory of critical behavior. A complete theory for the combined effects is not presently known. However, it is apparent, even at the mean field level, that such a theory for the effect of inhomogeneous pair breaking on T_c must take proper account of the resulting inhomogeneity of the gap function. It is our hope that the present calculations will provide the physical motivation for such a theory.

The authors are indebted to D. N. Basov, C. Bernhard, H. Kim, and S. H. Moffat for providing their data for Fig. 1, and are grateful to A. V. Balatsky, V. J. Emery, D. L. Feder, R. Fehrenbacher, K. Levin, J. S. Preston, and T. Timusk for inspiring discussions. This work has been partially supported by NSERC, OCMR, and by NSF Grant No. DMR-9415549 (M.F.).

*Permanent address: Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada L8S 4M1.

[1] J. F. Annett, N. D. Goldenfeld, and S. R. Renn, Phys. Rev. B **43**, 2778 (1991).
[2] P. J. Hirschfeld and N. D. Goldenfeld, Phys. Rev. B **48**, 4219 (1993).
[3] W. N. Hardy *et al.*, Phys. Rev. Lett. **70**, 3999 (1993).
[4] M. Prohammer and J. P. Carbotte, Phys. Rev. B **43**, 5370 (1991).
[5] H. Kim, G. Preosti, and P. Muzikar, Phys. Rev. B **49**, 3544 (1994).
[6] Y. Sun and K. Maki, Phys. Rev. B **51**, 6059 (1995).
[7] R. J. Radtke *et al.*, Phys. Rev. B **48**, 653 (1993).
[8] A. A. Abrikosov and L. P. Gorkov, Zh. Éksp. Teor. Fiz. **39**, 1781 (1960) [Sov. Phys. JETP **12**, 1243 (1961)].
[9] E. R. Ulm *et al.*, Phys. Rev. B **51**, 9193 (1995).
[10] D. N. Basov *et al.* (unpublished).
[11] D. N. Basov *et al.*, Phys. Rev. B **49**, 12 165 (1994).
[12] C. Bernhard *et al.*, Phys. Rev. Lett. **77**, 2304 (1996).
[13] S. H. Moffat, R. A. Hughes, and J. S. Preston, Phys. Rev. B **55**, 14 741 (1997).
[14] P. Arberg and J. P. Carbotte, Phys. Rev. B **50**, 3250 (1994).
[15] R. Fehrenbacher, Phys. Rev. Lett. **77**, 1849 (1996).
[16] P. Monthoux and D. Pines, Phys. Rev. B **49**, 4261 (1994).
[17] P. J. Hirschfeld, J. Phys. Chem. Solids **56**, 1605 (1995).
[18] M. Tinkham, *Introduction to Superconductivity* (Krieger, Malabar, 1975).
[19] T. Xiang and J. M. Wheatley, Phys. Rev. B **51**, 11 721 (1995).
[20] M. Franz, C. Kallin, and A. J. Berlinsky, Phys. Rev. B **54**, R6897 (1996).
[21] Here we use $1/N(0) \approx \pi v_f/2a_0$, valid for a tight-binding model away from half filling and from the bottom of the band, and a BCS relation $T_{c0}/\Delta \simeq 1.13/2$. For a d-wave superconductor this ratio will depend on the structure of the gap function, but will remain of order 1. For concreteness and simplicity we use the BCS result.
[22] In a different context the breakdown of the AG theory has been discussed by A. V. Balatsky, M. I. Salkola, and A. Rosengren [Phys. Rev. B **51**, 15 547 (1995)].
[23] P. I. Soininen, C. Kallin, and A. J. Berlinsky, Phys. Rev. B **50**, 13 883 (1994).
[24] Y. Wang and A. H. MacDonald, Phys. Rev. B **52**, R3876 (1995).
[25] Y. Onischi *et al.*, J. Phys. Soc. Jpn. **65**, 675 (1996).
[26] P. G. de Gennes, *Superconductivity of Metals and Alloys* (Addison-Wesley, Reading, MA, 1989).
[27] D. J. Scalapino, S. R. White, and S. C. Zhang, Phys. Rev. Lett. **68**, 2830 (1992).
[28] R. Villeneuve *et al.*, Physica C **235-240**, 1567 (1994).
[29] K. Ishida *et al.*, J. Phys. Soc. Jpn. **62**, 2803 (1993).