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Onset of phase correlations in $YBa_2Cu_3O_{7-x}$ as determined from reversible-magnetization measurements

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Isofield magnetization curves are obtained and analyzed for three single crystals of $YBa_2Cu_3O_{7-x}$, ranging from optimally doped to very underdoped, as well as the BCS superconductor Nb in the presence of magnetic fields applied both parallel and perpendicular to the ab planes. Near T_c, the magnetization exhibits a temperature dependence $\sqrt{M} \propto [T_a(H)-T]^m$. In accordance with recent theories, we associated $T_a(H)$ with the onset of coherent phase fluctuations of the superconducting order parameter. For Nb and optimally doped YbaCuO, $T_a(H)$ is essentially identical to the mean-field transition line $T_c(H)$. The fitting exponent $m \simeq 0.5$ takes its mean-field value for Nb and slightly varies from 0.5 for optimally doped YBaCuO. However, underdoped YBaCuO samples exhibit anomalous behavior, with $T_a(H) > T_c$ for $H \parallel c$, suggesting that the magnetization is probing a region of temperatures above T_c where phase correlations persist. In this region, the fitting exponent falls in the range $0.5 < m < 0.8$ for $H \parallel c$ compared to $m \simeq 0.5$ for $H \parallel ab$. The results are interpreted in terms of an anisotropic pairing symmetry of the order parameter: d wave along the ab planes and s wave along the $\hat{c}$ axis.

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I. INTRODUCTION

It is well known that underdoped high-T_c superconductors exhibit a rich variety of fluctuation phenomena in the pseudogap regime.[1,2] Many of these phenomena are thought to arise from amplitude fluctuations, which persist even without phase coherence.[3,4] However, the superconducting transition at zero magnetic field T_c represents the temperature at which phase coherence is lost.[5] For low-T_c superconductors, neither amplitude nor phase fluctuations play an important role, and the resulting transition is of the mean-field type.[5] On the other hand, high-T_c materials, as underdoped cuprates, have a low superfluid density,[5] leading to a small phase stiffness and an enhancement of phase fluctuations in the vicinity of T_c. In Ref. 6, a line of phase transitions $T_\phi(x) > T_c$ was obtained for $YBa_2Cu_3O_{7-x}$ as a function of doping x, marking the disappearance of phase coherence. However, Refs. 5 and 6 did not consider an applied magnetic field, although most superconducting phenomena above T_c in underdoped materials sensitively depend on the magnetic field[2–4,7] and fluctuating vortices. The dearth of experimental work in the literature, exploring the existence of the temperature T_ϕ, forms the motivation for the present experiments. As we shall explain below, our work was also motivated by the results of Ref. 8, where phase fluctuations are shown to have an effect on the superfluid density of states, reducing the gap in the vicinity of T_c.

In this paper, we present isofield measurements of reversible magnetization versus temperature, which is obtained from three single crystals of YBaCuO ranging from underdoped to optimally doped. We consider magnetic fields applied both parallel and perpendicular to the $\hat{c}$ axis. We also present a few isofield $\sqrt{M}$ versus T curves for Nb, which is a well known BCS superconductor,[9] for comparison. Our data are plotted as $\sqrt{M}$ versus T, where $\sqrt{M}$ is directly proportional to the amplitude of the order parameter $|\psi|$ near T_c.[10] In this way, we can directly probe fluctuations of the order parameter occurring below and above T_c in YBaCuO within the pseudogap region.[1,2,11]

II. EXPERIMENTAL

The single crystals of YBaCuO were grown at Argonne National Laboratory[12] with fully developed transitions of width $\Delta T_c \simeq 1-2$ K. The optimally doped $YBa_2Cu_3O_{7-x}$ single crystal had a composition of $x \sim 0.05$ with $T_c = 91.7$ K, approximate dimensions of $1 \times 1 \times 0.3$ mm^3, and a mass of $\simeq 1.7$ mg. For the underdoped samples with $H \parallel \hat{c}$, we used magnetization data, which were previously obtained from two deoxygenated crystals[13] with compositions of $x = 0.6$ and 0.5 and corresponding transitions at $T_c = 41.5$ and 52 K, respectively (these two samples have approximate dimensions of $1 \times 1 \times 0.2$ mm^3 and mass $\simeq 1$ mg). For the Nb sample, we used the magnetization data previously presented in Ref. 14. All our magnetization data were obtained with a commercial magnetometer, which is based on a superconducting quantum interference device. The data were obtained after cooling the sample from temperatures above T_c in zero applied magnetic field to a desired temperature below T_c. Magnetic fields up to 50 kOe were then applied, reaching the desired value without overshoot. The data were obtained by heating the sample in fixed increments of temperature up to a temperature well above T_c. We also obtained field-cooled curves, which allowed to obtain the reversible (equilibrium) magnetization. Two field orientations were used, corresponding to $H \parallel \hat{c}$ and $H \parallel ab$ planes. For the latter geometry, we attached the samples with varnish to a tiny quartz-rod inserted along the axis of a straw. The magnetization data were obtained up to 150 K for the $T_c = 91.7$ K sample, up to 80 K for the $T_c = 52$ K sample, and up to 70 K for the $T_c = 41.5$ K. The background magnetization due to normal state electrons was determined (and removed) for each data set by

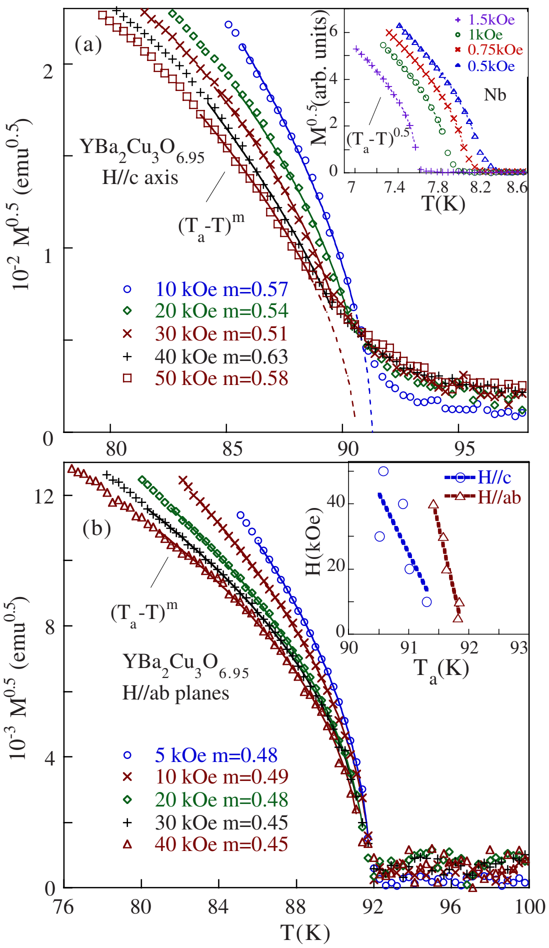

FIG. 1. (Color online) Isofield curves of $\sqrt{M}$ vs T where M is the reversible magnetization. (a) Optimally doped YBaCuO (T_c=91.7 K) with $H\|c$. The dashed lines show the extrapolation of the fittings to $T_a(H)$ for H=50 and 10 kOe. Inset: Nb data (note that Nb is isotropic). (b) Optimally doped YBaCuO with $H\|ab$. Inset: phase correlation temperature $T_a(H)$ vs H.

fitting to the form $M_b=c(H)/T-a(H)$ in a temperature window well above T_c.

III. RESULTS

The resulting reversible magnetization data are plotted as $\sqrt{M}$ versus T in Figs. 1–3. The Nb data are shown in the inset of Fig. 1(a). As a reference point, the $H\|\hat{c}$ data, as shown in Figs. 1(a), 2(a), and 3(a), all exhibit a crossing point, where the magnetization is independent of the field. This interesting feature occurs in two-dimensional (2D) systems[15–17] and in three-dimensional systems[17,18] and can be explained in terms of vortex fluctuations. The crossing point was previously investigated for the same deoxygenated samples studied here,[13] and will not be discussed further.

In the conventional theory of the upper critical field H_{c2},[19] the magnetic induction B obtained from the Ginzburg–Landau equation can be expressed as[10]

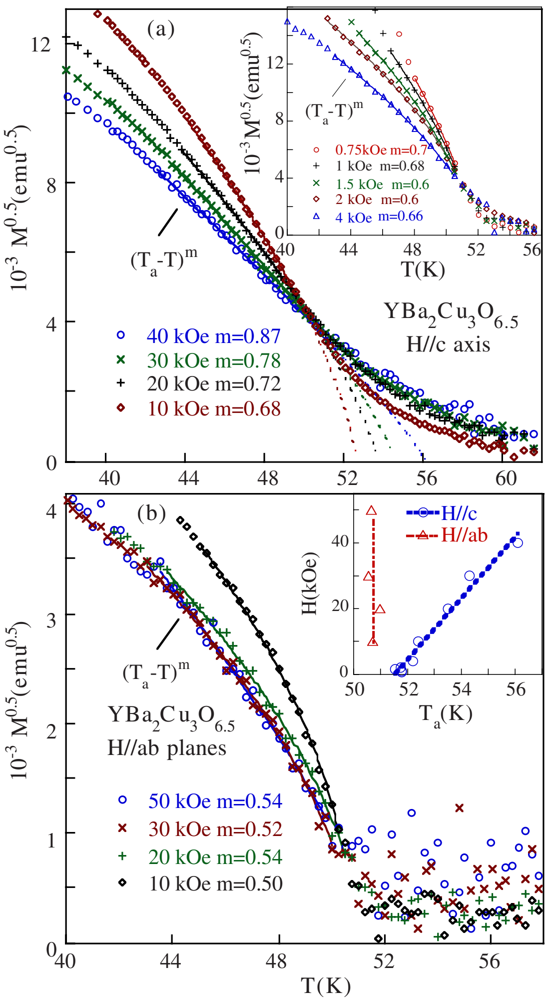

FIG. 2. (Color online) Isofield curves of $\sqrt{M}$ vs T for underdoped YBaCuO (T_c=52 K), where M is the reversible magnetization. (a) $H\|\hat{c}$, higher fields. The dashed lines show the extrapolation of the fittings to $T_a(H)$. Inset: lower fields. (b) The same sample, $H\|ab$. Inset: phase correlation temperature $T_a(H)$ vs H.

$$B = H - \frac{4\pi e\hbar}{mc}|\psi|^2, \tag{1}$$

where ψ is the superconducting order parameter. The magnetization $M=(B-H)/4\pi$ is then given by

$$M = -\frac{e\hbar}{mc}|\psi|^2. \tag{2}$$

Within the Abrikosov approximation,[19] it therefore follows that $\sqrt{M}$ is directly proportional to the average amplitude of the order parameter. Near the superconducting transition, the temperature dependence of the magnetization can then be expressed as $\sqrt{M}\propto[T_c(H)-T]^m$ in terms of the mean-field transition temperature $T_c(H)$. The mean-field exponent is given by $m=1/2$ both for s-wave BCS superconductors[10] and for d-wave superconductors within a Ginzburg–Landau theory.[20]

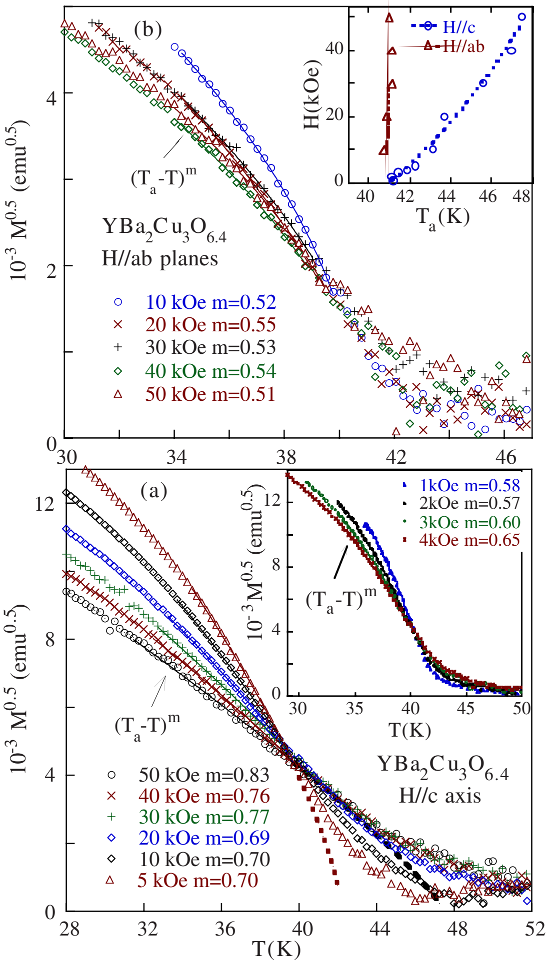

FIG. 3. (Color online) Isofield curves of $\sqrt{M}$ vs T for underdoped YBaCuO (T_c=41.5 K), where M is the reversible magnetization. (a) $H\|\hat{c}$, higher fields. The dashed lines show the extrapolation of the fittings to $T_a(H)$ for H=50 and 5 kOe. Inset: lower fields. (b) The same sample, $H\|ab$. Inset: phase correlation temperature $T_a(H)$ vs H.

Here, we perform a generalized scaling analysis of the magnetization data. We infer the presence of a phase-mediated transition from the fitting relation $\sqrt{M}\propto(T_a(H)-T)^m$, where $T_a(H)$ denotes the apparent transition temperature and m is the fitting exponent. For underdoped samples, in particular, the phase fluctuations may be quite large, and the exponent m may vary significantly from its mean-field value.

To focus on phase-mediated behavior, we must distinguish it from the complementary fluctuations of the amplitude of the order parameter, which appear as anomalous enhancements of the magnetization above T_c, particularly for the field orientation $H\|\hat{c}$. Like the phase fluctuations,[6] these amplitude fluctuations predominantly occur in the pseudogap phase of underdoped materials due to their extreme anisotropy or two dimensionality.[21] Indeed, deoxygenated crystals with the same transition temperatures studied here have previously been shown to exhibit strong 2D critical fluctuations.[22] The phase-mediated behavior of interest here occurs at temperatures below the amplitude fluctuation regime in the vicinity of the crossing point. For our analysis, we determine a temperature fitting range for each data set. For the curves obtained with $H\|\hat{c}$, we take the inflection point of the magnetization as our high temperature cutoff since it marks a change in the temperature dependence (curvature) of the magnetization. Typically, the inflection point remains within the superconducting scaling regime since it lies within a degree or two below the zero-field transition. For most of the curves, the inflection point occurs at the crossing point. For the curves with $H\|ab$, the crossing point is not well defined (neither a change in curvature), and the high temperature cutoff was visually taken from the smoothness of the asymptotic behavior of each curve near T_c. For the low temperature cutoff, we choose a point where the reversible magnetization approximately becomes linear. The resulting fitting curves are shown as solid lines in Figs. 1–3. The dashed lines in Figs. 1–3 (only for $H\|\hat{c}$) represent the extrapolation of the fittings to $T_a(H)$ and also helps us to visualize the change in the curvature in each curve. We note that the fitting ranges have a typical width of 5–8 K, which tends to increase for higher fields with $H\|\hat{c}$ and to decrease for more underdoped samples. For the narrower fitting ranges, we included some magnetization data lying in the linear $M(T)$ regime, corresponding to Abrikosov's solution of the Ginzburg–Landau equations.[19] Such linear data have previously been used to estimate mean-field transition temperatures $T_c(H)$ from isofield $M(T)$ data.[23] Figure 4(a) illustrates the linear region of the reversible-magnetization curves for selected fields and the respective $T_c(H)$, which are obtained from the linear extrapolation. Figures 4(b) and 4(c) show plots of $\sqrt{M}$ vs T (and fittings) obtained from two M vs T curves of Fig. 4(a), evidencing the differences between $T_c(H)$ and $T_a(H)$ for these samples. We mention that values of $T_c(H)$ obtained from this linear method produced consistent values of $dH_{c2}/dT<0$ for all samples in reasonable agreement with the values listed in Ref. 13. Results for the fitted values of $T_a(H)$ and m are shown for different samples and field orientations in Figs. 1–3. We note that most of the resulting fitting curves can be extended for several degrees below their fitting range (up to 5 K for high field curves), while still providing a good description of the experimental data. For the Nb sample in the inset of Fig. 1(a), the resulting

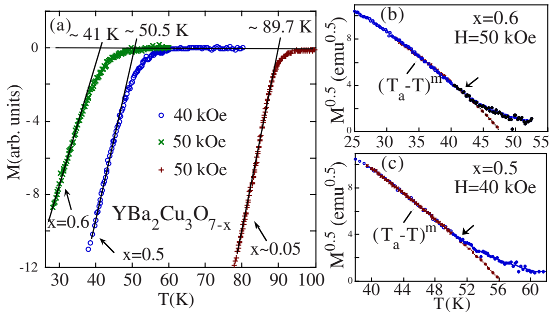

FIG. 4. (Color online) (a) Linear extrapolation of the reversible magnetization for YBaCuO isofield curves with $H\|\hat{c}$. [(b) and (c)] $\sqrt{M}$ vs T for two M vs T curves of (T_c=41.5 and 52 K). The dashed lines represents the extrapolation of the fittings to $T_a(H)$. The arrow in each curve indicates the position of the inflection point.

values of $T_a(H)$ are essentially identical to the mean-field transition $T_c(H)$ obtained in Ref. 14, with $[dT_a(H)/dH]<0$. For the optimally doped YBaCuO sample in Fig. 1(b), we also obtain transition temperatures consistent with the linear $M(T)$ extrapolation method used in Ref. 23 for both field orientations. One may expect this conventional behavior for both of these samples, which exhibit no pseudogap phase. We note that the observed fitting exponents for optimally doped YBaCuO were slightly enhanced for $H\parallel\hat{c}$ and slightly suppressed for $H\parallel ab$ relative to their mean-field values. However, these deviations are relatively weak compared to the underdoped samples.

The underdoped samples in Figs. 2 and 3 show behavior that is not characteristically mean field and reflect enhanced fluctuation effects. For $H\perp\hat{c}$, $T_a(H)$ is essentially constant, with no resolvable slope. However, for $H\parallel c$, we observe a distinct, anomalous slope with $dT_a(H)/dH>0$. In fact, for high field curves, the inflection points occur above the respectives crossing points, but below T_c, as shown in the plots of Figs. 4(a) and 4(b), which partially explains why $T_a(H)$ increases with field. For these curves, the fitting exponents m also appear to anomalously increase with the field.

IV. DISCUSSION

It is shown in Ref. 8 that phase fluctuations of the order parameter with d-wave symmetry can have a net effect on the superfluid density of states reducing the gap in the vicinity of T_c. An inspection of Fig. 2 of Ref. 8 shows that the change in the shape of the gap near T_c is accompanied by a change in the value of the exponent m. We observed that we can apply the later result to explain the large values of the fitting exponent m found for YBaCuO with $H\parallel c$. By following this conjecture, the temperature dependence of the order parameter in the curves of Figs. 1–3 is defined by phase fluctuations, and the values of $T_a(H)$, which are obtained from the fittings, represent temperature values at which phase coherence is lost. Then, the temperature dependence of the magnetization observed here in several different samples is generally consistent with the proposed theories of Curty and Beck[6] and Emery and Kivelson.[5] In this picture, the inferred transition temperature $T_a(H)$ represents the onset of phase coherence and may be associated with the zero-field temperature T_ϕ. Although $T_a(H)$ does not correspond to a true phase transition, it appears to control the scaling properties of the magnetization $M(T)$ in the asymptotic regime, which is just below the regime of strong amplitude fluctuations. For conventional superconductors such as Nb, phase and amplitude fluctuations are both weak, and the magnetization exhibits mean-field behavior. For optimally doped YBaCuO, phase fluctuations are expected to be important in the vicinity of T_c,[5] but the absence of a pseudogap phase produced a mean-fieldlike behavior for $T_a(H)$ as for Nb. However, underdoped high-T_c superconductors exhibit a very different behavior, with significant phase fluctuations, and a line of phase-mediated transitions $T_a(H)$ quite distinct from the mean-field transition $T_c(H)$. The observation of phase coherence above T_c correlates with the presence of a pseudogap in the underdoped materials.[6] The persistence of phase coherence above $T_c(H)$ [up to $T_a(H)$] may explain why fluctuation effects observed above T_c in underdoped materials resemble superconducting properties normally observed below T_c, as claimed in Refs. 3, 4, and 7. The distinction between $T_a(H)$ and $T_c(H)$ may also explain why the observed fitting exponent deviates from $m=1/2$. Since doping and the magnetic field both effect phase fluctuations,[24] we can anticipate that the value of m will depend on the field.

A possible explanation for the anomalous behavior of dT_a/dH and m in the underdoped samples arises from the d-wave pairing symmetry of the order parameter. As mentioned above, it is shown in Ref. 8 that phase fluctuations of a d-wave order parameter can have a net effect on the superfluid density of states, producing a change in the value of the fitting exponent m. In this picture, the anisotropic dependence of m on H can also be explained in terms of the order parameter symmetry. Differences of the effect of phase fluctuations depending on the symmetry, i.e., s wave or d wave, of the order parameter are expected since the d-wave symmetry presents node and antinode where the effects of phase and amplitude fluctuations are distinct.[8] This fact suggests that differences observed in the values of the exponent m, depending on the direction of the magnetic field with respect to the c axis, are consequences of the pairing symmetry of the order parameter in YBaCuO, which is likely to be d wave in the ab planes and s wave along the c axis (as in Nb). We note that this pairing symetry of the order parameter was suggested by Mannhart *et al.*[25] for high-T_c superconductors and a recent result in a LaSrCuO crystal also suggests s-wave pairing along the $\hat{c}$ axis.[26]

An alternative explanation for the anistropic fitting exponent m may arise from the anisotropy of the vortex critical current in the quasi-2D underdoped cuprates. For these highly anisotropic superconductors, the high-field vortex state is formed of pancake vortices when $H\parallel\hat{c}$ and Josephson vortices when $H\perp\hat{c}$, with dissipation that is field independent for the case of $H\parallel ab$.[27] The zero-field-cooled (reverslble) magnetization may therefore provide a direct measurement of the anisotropy of the critical current, which sustains the vortex state, and its temperature dependence. We point out, however, that the observed behavior is strongly influenced by fluctuations, leading to complications for interpretations based on mean-field arguments.

In conclusion, we have determined the apparent transition temperature $T_a(H)$, which is associated with phase coherence above the mean-field transition, by fitting the temperature dependence of the magnetization $\sqrt{M}\propto[T_a(H)-T]^m$ for several high-T_c and conventional superconductors. For deoxygenated YBaCuO samples with $H\parallel\hat{c}$, we observe that $T_a(H)>T_c$ with $dT_a/dH>0$. Such anomalous behavior suggests that the magnetization is probing a regime where phase coherence persists above T_c, even in the absence of true long-range order. These same samples also exhibit significant amplitude fluctuations without phase coherence extending to well above $T_a(H)$. For all our samples, the values of the fitting exponent $m\simeq 0.5$ are consistent with mean-field theory when $H \| ab$ but deviate from this conventional behavior when $H \| c$ axis, particularly for the underdoped cuprates. We have suggested several interpretations for this anomalous behavior, including an anisotropy of the symmetry of the order parameter: s wave along the $\hat{c}$ axis vs d wave in the ab planes.

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[1] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).
[2] P. A. Lee, N. Nagaosa, and X.-G. Wen, Rev. Mod. Phys. **78**, 17 (2006).
[3] Y. Wang, L. Li, M. J. Naughton, G. D. Gu, S. Uchida, and N. P. Ong, Phys. Rev. Lett. **95**, 247002 (2005); L. Cabo, J. Mosqueira, and F. Vidal, Phys. Rev. Lett. **98**, 119701 (2007).
[4] Y. Wang, L. Li, and N. P. Ong, Phys. Rev. B **73**, 024510 (2006).
[5] V. J. Emery and S. A. Kivelson, Nature (London) **374**, 434 (1995).
[6] P. Curty and H. Beck, Phys. Rev. Lett. **91**, 257002 (2003).
[7] S. Salem-Sugui, Jr., M. M. Doria, A. D. Alvarenga, V. N. Vieira, P. F. Farinas, and J. P. Sinnecker, Phys. Rev. B **76**, 132502 (2007).
[8] H.-J. Kwon, Phys. Rev. B **59**, 13600 (1999).
[9] I. Giaever and K. Megerle, Phys. Rev. **122**, 1101 (1961).
[10] P. G. deGennes, *Superconductitivity of Metals and Alloys* (Addison-Wesley, New York, 1989).
[11] H. Ding, T. Yokoya, J. C. Campuzano, T. Takahashi, M. Randeria, M. R. Norman, T. Mochiku, K. Kadowaki, and J. Giapintzakis, Nature (London) **382**, 51 (1996).
[12] B. W. Veal, A. P. Paulikas, H. You, H. Shi, Y. Fang, and J. W. Downey, Phys. Rev. B **42**, 6305 (1990).
[13] S. Salem-Sugui, Jr., A. D. Alvarenga, B. Veal, and A. P. Paulikas, J. Low Temp. Phys. **143**, 131 (2006).
[14] S. Salem-Sugui, Jr., M. Friesen, A. D. Alvarenga, F. G. Gandra, M. M. Doria, and O. F. Schilling, Phys. Rev. B **66**, 134521 (2002).
[15] L. N. Bulaevskii, M. Ledvij, and V. G. Kogan, Phys. Rev. Lett. **68**, 3773 (1992).
[16] Z. Tešanović, L. Xing, L. Bulaevskii, Q. Li, and M. Suenaga, Phys. Rev. Lett. **69**, 3563 (1992).
[17] B. Rosenstein, B. Ya. Shapiro, R. Prozorov, A. Shaulov, and Y. Yeshurun, Phys. Rev. B **63**, 134501 (2001).
[18] S. Salem-Sugui, Jr. and E. Z. da Silva, Physica C **235**, 1919 (1994).
[19] A. Abrikosov, Zh. Eksp. Teor. Fiz. **32**, 1442 (1975) [Sov. Phys. JETP **5**, 1174 (1957)].
[20] J.-H. Xu, Y. Ren, and C. S. Ting, Phys. Rev. B **52**, 7663 (1995).
[21] M. R. Norman, *Handbook of Magnetism and Advanced Magnetic Materials* (Wiley, New York, in press), Vol. 5.
[22] S. Salem-Sugui, Jr., A. D. Alvarenga, V. N. Vieira, and O. F. Schilling, Phys. Rev. B **73**, 012509 (2006).
[23] U. Welp, W. K. Kwok, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu, Phys. Rev. Lett. **62**, 1908 (1989).
[24] U. Welp, S. Fleshler, W. K. Kwok, R. A. Klemm, V. M. Vinokur, J. Downey, B. Veal, and G. W. Crabtree, Phys. Rev. Lett. **67**, 3180 (1991).
[25] J. Mannhart, H. Hilgenkamp, G. Hammerl, and C. W. Schneider, Phys. Scr., T **T102**, 107 (2002).
[26] R. Khasanov, A. Shengelaya, A. Maisuradze, F. La Mattina, A. Bussmann-Holder, H. Keller, and K. A. Muller, Phys. Rev. Lett. **98**, 057007 (2007).
[27] L. I. Glazman and A. E. Koshelev, Phys. Rev. B **43**, 2835 (1991).