Field-Induced Suppression of the Phase Transition in $Bi_2Sr_2CaCu_2O_8$

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Magnetization measurements in field on a $Bi_2Sr_2CaCu_2O_8$ single crystal do not reveal any sharp phase transition between the normal and superconducting states. The $M(H)$ curves for temperatures about T_c in fields up to 5 T all display diamagnetic behavior. Above the mean-field transition temperature T_c^{MF} , in helds up to 3.1 an uisplay diamagnetic behavior. Above the mean-field cransition temperature T_c , quasi-two-dimensional diamagnetic fluctuations are responsible. Below T_c^{MF} strong deviations from the conventional mean-field (Abrikosov) behavior lead to an apparent $T_c(B)$ which *increases* with field. This effect clearly demonstrates the inadequacy of the mean-field theory to describe the vortex state below the crossover line $H_{c2}(T)$.

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It has been a general belief that the in-field transition between the normal and mixed states in a type-II superconductor is given by a second-order phase transition defined as $H_{c2}(T)$. However, recent theoretical work [1] regarding the high- T_c cuprates rather characterized $H_{c2}(T)$ as a crossover line between the normal state and a vortex liquid. A similar prediction has been formulated based on the analogy with quasi-two-dimensional (2D) magnetic insulators [2]. In the high-temperature superconductors (HTS), the vortex-liquid regime in the (B, T) phase diagram is quite extended, especially when the material, because of its large anisotropy, is quasi-20. For example, for $Bi₂Sr₂CaCu₂O₈$ (Bi 2:2:1:2) with an anisotropic effective-mass ratio $\Gamma \geq 3000$ [3], above a field $B_{2D} \approx \Phi_0/\Gamma s^2$ in the c direction the pancake vortices in adjacent $CuO₂$ double layers with spacing s are essentially decoupled [4]. Consequently, the vortex-lattice melting temperature in the limit of weak pinning is expected to be the same as for a 2D superconductor of thickness s $(s = 1.5 \text{ nm})$; i.e., for $B > B_{2D} \approx 1 \text{ T}$ it melts at about 30 K. Below the melting line it is natural to consider the Abrikosov lattice as the ground state when investigating fluctuation effects [5]. For the liquid state it can no longer be expected that this is allowed [6]. The magnetic properties in the liquid state may therefore deviate from the well-known Abrikosov behavior. As we shall see below, considerable deviations are indeed observed due to fluctuations.

Note that the fluctuations suppress the melting line in a field by over 50 K from the mean-field transition $T_c \approx 90$ K. This can be contrasted with the fluctuation effects in zero field [7] where the Kosterlitz-Thouless (KT) transition at T_c is suppressed by only 2 K below the Ginzburg-Landau (GL) transition temperature T_c^0 [8].

In this paper we report on magnetization measurements on a high-quality Bi 2:2:1:2 single crystal in a commercial SQUID system with the field along the c direction. The data are all taken in the vortex-liquid and nornal regimes, i.e., in the fully reversible field-temperatur regime above the irreversibility or melting line. The crystal weight and dimensions are 11.4 mg and $4 \times 4 \times 0.11$ mm³ and its lattice parameters are $a = 0.5333$ nm, $b=0.5485$ nm, and $c=3.076$ nm. The composition as determined by microprobe analysis is $Bi_{2,2}Sr_{1,9}CaCu_{2}O_{x}$. Details of the preparation will be published in Ref. [9]. High-resolution electron microscopy confirmed the high purity of the crystal, for only minor traces of the 2:2:0:1 phase could be detected in a concentration of 2 out of 1000 CuO₂ double layers [10]. T_c has been determined from the ac susceptibility transition in a $2.8-\mu T$ ac field applied parallel to the $a-b$ planes in a shielded dc environment of less than 0.3 μ T. A linear extrapolation of the $\chi'(T)$ data to zero defined $T_c(0) = 88.1$ K, and that to $\chi' = -1$, the transition width of 1.5 K, demonstrating the uniformity of our sample.

In Fig. 1 our $M(T)$ data are shown for various fields (H||c) between 0.1 and 5 T. The gradual decrease above 90 K indicates the effect of diamagnetic fluctuations [1 ll. Since this effect is negligibly small above 120 K, we corrected for a background signal by subtracting M at 120 K. The data sets between 70 and 87 K display an essentially linear behavior. Assuming linear $M(T, H)$ behavior to be valid near $H_{c2}(T)$, as is the case in Abrikosov's solution, extrapolation to $M = 0$ would determine $T_c(B)$. These intercepts are shown in the inset of Fig. 1. It is seen that the resulting " $T_c(B)$ " increases with increasing B , opposite to the usual behavior. We stress here that we do not believe these data represent the mean-field $H_{c2}(T)$, but rather that they demonstrate that the simple linear construction fails, for reasons to be discussed below.

It has been recently pointed out [12] that the linear

FIG. 1. Compilation of M-vs-T data of a Bi 2:2:1:2 single crystal in fields of 0.1, 0.3, 0.5, 1, 2, 3, 4, and 5 T (bottom to top) measured in both increasing and decreasing T runs. Inset: " $T_c(B)$ " as determined by linearly extrapolating $M(T)$ back to $M = 0$.

Abrikosov regime near H_{c2} is very narrow in T because of the large dH_{c2}/dT values of the HTS. Our measurements might therefore display the subsequent logarithmic regime in which $-M \propto (\Phi_0/\lambda^2) \ln(\beta H_{c2}/H)$, where β is a constant of order unity, $\lambda(t) = 0.7\lambda_L(0)(1-t)^{-1/2}$ is the Ginzburg-Landau penetration depth, and $\lambda_L(0)$ is the London penetration depth at $T=0$. From the slope of dM/dT vs ln(H) the value of $\lambda_L(0)$ can be determined; see the inset of Fig. 2, where $YBa₂Cu₃O₇$ (YBCO) data [13] are also shown. The YBCO data give λ_1 (0) $=140\pm9$ nm, in good agreement with the literature [14]. Our data, however, show a kink at about ¹ T which would lead to two values, namely, $\lambda_L(0) = 170$ and 260 nm. In fact, a detailed comparison of our Bi $2:2:1:2$ results with the computations of Ref. [12], taking $S = -\mu_0 dH_c \frac{2}{dT}\Big|_{T_c} = 1$ or 2 T/K, shows that neither the upturn of " $T_c(B)$ " nor the field dependence of dM/dT can be satisfactorily explained.

Additional evidence for deviations from the mean-field theory is seen in Fig. 2 where the $M(H)$ data above 0.1 T are plotted for a series of temperatures about T_c . Results below 0.1 T and below 86 K (e.g., at 80 K) show an initial sharp decrease to $M = -7200$ A/m followed by a steep rise to -2700 A/m at 20 mT (without correcting for the demagnetization effect). This result seems in accord with the conventional magnetization about H_{c1} . The high-field behavior at 80 K also agrees with the Abrikosov prediction. Evidently, the results increasingly deviate from the canonical behavior when T is increased. A clearly detectable diamagnetic signal remains above $T_c(0)$. It shows a fast increase which levels off at about 1 T and remains almost constant up to 5 T.

The data depicted in Figs. ¹ and 2 are the major results of this paper. They suggest that strong fluctuation effects

FIG. 2. Compilation of M -vs- H data above 0.1 T at 80 K $(0, 82 \text{ K } (\blacksquare), 84 \text{ K } (\square), 86 \text{ K } (\lozenge), 88 \text{ K } (\lozenge), 90 \text{ K } (\spadesuit), \text{ and}$ 92 K (\triangle) . Inset: A semilogarithmic plot of the slopes of the linear parts of the $M(T)$ data vs field (circles). Note the kink at about $B_{2D} \approx 1.0$ T. For comparison the data of YBCO [13] are shown as well (triangles).

play an important part in the experimental $T_c(B)$. The fluctuation diamagnetization has been reviewed in Ref. [1]. The scaling of M and χ' follows from the singular part of the free energy per unit volume, or per unit area per coupled layer when it is used in 2D:

$$
F_s \propto k_B T \xi + D F(B/\overline{B}) \,, \tag{1}
$$

where $\bar{B} \equiv \Phi_0/\xi_+^2$ and D is the dimensionality. The scaling function F is different for each of the four regimes: 2D or 3D, weak or strong fluctuations (the latter being the critical regime). The coherence length ξ + diverges at T_c in the critical regime between T_c and T_c^0 ; for $D = 2$
this divergence is exponential, while for $D = 3$, $\xi_+ \propto (T$ $(T_t - T_c)^{-\nu}$, with $\nu \approx \frac{2}{3}$. The coherence length behaves classically in the weak-fluctuation regime (WFR) above T_c^0 , namely, as $\xi_+ \approx \xi_{GL}(0)\tau_0^{-1/2}$, with $\tau_0 = (T - T_c^0)/T_c$ T_c^0 . In 2D a convenient interpolation formula has been proposed [8]: $\xi_+ \approx b^{-1/2} \xi_c \sinh[(b\tau_c/\tau)^{1/2}]$, with b a dimensionless constant of order unity, $\tau = (T - T_c)/T_c$, $r_c \equiv (T_c^0 - T_c)/T_c$, and $\xi_c \approx \xi_{GL}(0) \tau_c^{-1/2}$. The susceptibility in zero field in the 2D WFR is [8]

$$
-\chi_s^{2D} = (\pi \mu_0 k_B T / 3 \Phi_0^2) \xi_+^2 \,. \tag{2}
$$

The scaling of M is given by

$$
-M_s = \partial F_s / \partial B \propto (k_B T \xi_+^{2-D} / \Phi_0) F'(B/\overline{B})
$$
 (3)

The scaling function F' saturates at large arguments for $D=2$ and grows proportional to $(B/\overline{B})^{1/2}$ in 3D. For small B , F' is linear in the argument. Low-field contours of constant M would thus reveal the prevailing dimensionality, because they are equivalent to contours of constant $B\xi_+^{4-D}$. For $D=3$ this leads to $B \propto (T-T_c^0)^{1/2}$ in the WFR and $B \propto (T - T_c)^v$ in the critical regime. Both would then have downward curvature of such contours for 3D. For $D=2$, on the other hand, the contours are straight in the WFR and curve upwards in the critical regime.

The inset of Fig. 3 shows contours of $-M=10$, 30, and 100 A/m at T above 86 K. The curvature shows that D = 2. The dashed lines indicate that T_c^0 = 88.4 \pm 0.4 K in agreement with our experimental definition of $T_c(0)$. For 2D, $M_s(B)$ should saturate to about $-\overline{M}=k_B T_c^0/$ Φ_0 s, so $M = -390$ A/m for Bi 2:2:1:2: This agrees reasonably well with the value $M = -310$ A/m at 88 K and 5 T (see Fig. 2). Note also that down to 0.3 T, M indeed is constant. In Fig. I this corresponds to the point at which all the lines coincide. The combination of $M(B)$ being a nonzero constant at T_c^0 and the essentially linear behavior of M vs T with increasing slope for smaller fields causes " $T_c(B)$ " as defined above to increase with B.

In the WFR the coupling strength between the superconducting layers was explicitly taken into account by Gerhardts [15] and Klemm, Beasley, and Luther [16]. The dimensionality is represented by a parameter $r = (8/\pi^2)\Gamma^{-1}[\xi_{GL}(0)/s]^2$ which, with $\Gamma = 3000$ and $\xi_{\text{GL}}(0) \approx 2$ nm, is estimated to be $r = 4.6 \times 10^{-4}$. We now compare our data with the theoretical $M(B)$ curve at T_c depicted in Fig. 5 of Ref. [15]. In the regime between $B = 1$ and 5 T the magnetization is given by $-M$ $=0.3k_BT_c/s\Phi_0$, which yields $M = -120$ A/m. This value corresponds to our experimental data slightly above $T=92$ K suggesting that this might be the mean-field (BCS) transition temperature T_c^{MF} . According to a re-

FIG. 3. Comparison of $-M\Phi_0/k_BT(\Phi_0/4\pi\Gamma B)^{1/2}$ $-T_c^{\text{MF}}/\mu_0H$ plots for experimental data at 0.3 T (\bullet), 2 T (\Box), and 5 T (\blacksquare) with the WFR theory [15] for layered compounds with $r = 10^{-4}$ and $B/B_s = 0.3$ and 5 (solid lines) and anisotropic 3D behavior (dashed line). The experimental data fit is obtained with $S=1.1$ T/K, $T_c^{\text{MF}}=92.2$ K, and $B_s=1$ T. Inset: Contours of constant M above 86 K for $-M=10$, 30, and 100 A/m. The dashed lines display the expected 2D WFR behavior and define T_c^0 .

mark in Ref. [8], T_c^{MF} is shifted with respect to T_c^0 by an amount of order $\tau_c T_c^0 \ln[\xi_{\text{GL}}(0)/\xi_c] = 0.5 \tau_c T_c^0 \ln \tau_c$.
With $\tau_c = 2.5 \times 10^{-2}$ [7] this amounts to $T_c^{\text{MF}} - T_c^0 = 4.0$ With $\tau_c = 2.5 \times 10^{-2}$ [7] this amounts to $T_c^{\text{MF}} - T_c^0 = 4.0$ K, so that $T_c^{\text{MF}}=92$ K is in accord with the above analysis.

In addition, we compare with the temperature dependence of M at $B = 0.3B_s$ and $B = 5B_s$, shown in Fig. 7(b) of Ref. [15]. For $r < 10^{-2}$, the scaling field B_s is given by $B_s = 3.7\Phi_0/\Gamma s^2 = 3.7B_{2D}$, yielding for Bi 2:2:1:2, $B_s = 1.07$ T. In Fig. 3 the theory for $r = 10^{-4}$ is represented by the solid lines. The symbols depict our data at 0.3, 2, and 5 T. For comparison a theoretical anisotropic 3D result is depicted by the dashed line. As was concluded in Refs. [15] and [16] the shape of the curves is not unique, but the scaling with B_s is. This horizontal scaling is very sensitive to the choice of T_c^{MF} , and not so much to the value of S. Given the value of $B_s = 1.07$ T, the best fit to the 0.3- and 5-T data was obtained with $S = 1.1$ T/K and T_c^{MF} =92.2 K (solid lines in Fig. 3). The deviation from the 0.3-T data occurs within a I-K interval above T_c^{MF} . For 5 T this interval is 15 times smaller. The 2-T data show behavior similar to the 5-T results in accord with the 2D nature of the fluctuations.

Both comparisons with the weak-fluctuation theory point to $T_c^{\text{MF}} = 92.2 \pm 0.5$ K. Combining this with the trend in the $T_c(B)$ plot in Fig. 1 we estimate $S_{MF}=3$ to 7 T/K. Such a large value seems to disagree with the best fit in Fig. 3. However, it would be in line with Palstra's result $S=7$ T/K for YBCO [17]. The corresponding coherence length would point to local pairing, since $\xi(0)$ \leq 1 nm.

The temperature dependence of the Ettinghausen effect in YBCO [17] about T_c could be well described by Ullah and Dorsey [18]. Starting from the Lawrence-Doniach model [19] and incorporating the interaction between fluctuations, good agreement with the data was achieved for $S=7$ T/K. However, a breakdown of scaling below T_c could not be explained by the mean-field approach. We think this breakdown is similar to the deviating behavior of our in-field $M(T)$ data in Fig. 1. Recently, Ikeda and Tsuneto [20] computed the effect of fluctuations on $M(T)$ and found reasonable agreement with the YBCO results [13] using $S = 3.7$ T/K. However, a similar computation for Bi 2:2:1:2 by only changing Γ yields $M(T)$ curves which clearly disagree with our data, especially because " $T_c(B)$ " decreases with B. On the other hand, Ikeda, Ohmi, and Tsuneto [21] were able to fit their theory nicely to the in-field $R(T)$ data on Bi 2:2:1:2 single crystals [22]. Because the interpretation of $R(T)$ curves may also involve thermally activated flux motion, this agreement may be fortuitous. We therefore argue that the magnetization data provide a stricter test for theoretical models [23].

In summary, the apparent increase of " $T_c(B)$ " with B actually reflects the 2D nature of the fluctuations. As the system is cooled in a field, a gradual transition occurs from the normal state to a kind of mixed-state behavior (the vortex liquid) that clearly deviates from the meanfield behavior. These deviations are greatly enhanced over what is seen in YBCO because of the extreme anisotropy of Bi 2:2:1:2. We leave it for further theoretical research to clarify these intriguing features.

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- [1] D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B 43, 130 (1991).
- [2] L. J. de Jongh, Solid State Commun. 70, 955 (1989).
- [3] D. E. Farrell *et al.*, Phys. Rev. Lett. 63, 782 (1989).
- [4] V. M. Vinokur, P. H. Kes, and A. E. Koshelev, Physica (Amsterdam) 168C, 29 (1990); M. V. Feigelman, V. B. Geshkenbein, and A. I. Larkin, Physica (Amsterdam) 167C, 177 (1990).
- [5] G. Eilenberger, Phys. Rev. 164, 628 (1967).
- [6] E. Brezin, A. Fujita, and S. Hikami, Phys. Rev. Lett. 65, 1949 (1990).
- [7] S. Martin, A. T. Fiory, R. M. Fleming, G. P. Espinoza, and A. S. Cooper, Phys. Rev. Lett. 62, 677 (1989).
- [8] B. I. Halperin and D. R. Nelson, J. Low. Temp. Phys. 36, 599 (1979).
- [9] M. J. V. Menken, A. J. M. Winkelman, and A. A.

Menovsky (to be published).

- [10] H. W. Zandbergen (private communication).
- [11]D. C. Johnston and J. H. Cho, Phys. Rev. B 42, 8710 (1990).
- [12] Z. Hao, J. R. Clem, M. W. McElfresh, L. Civale, A. P. Malozemoff, and F. Holtzberg, Phys. Rev. B 43, 2844 (1991);Z. Hao and J. R. Clem (unpublished).
- [13] U. Welp, W. K. Kwok, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu, Phys. Rev. Lett. 62, 1908 (1989).
- [14] S. Sridhar, D-H. Wu, and W. Kennedy, Phys. Rev. Lett. 63, 1873 (1989).
- [15] R. R. Gerhardts, Phys. Rev. B 9, 2945 (1974).
- [16] R. A. Klemm, M. R. Beasley, and A. Luther, Phys. Rev. B 8, 5072 (1973).
- [17]T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. 64, 3090 (1990).
- [18] S. Ullah and A. T. Dorsey, Phys. Rev. Lett. 65, 2066 (1990).
- [19]W. Lawrence and S. Doniach, in Proceedings of the Twelfth International Conference of Low Temperature Physics, edited by E. Kanda (Academic Press of Japan, Kyoto, 1971), p. 361.
- [20] R. Ikeda and T. Tsuneto, J. Phys. Soc. Jpn. 60, 1337 (1991).
- [21] R. Ikeda, T. Ohmi, and T. Tsuneto, J. Phys. Soc. Jpn. 60, 1051 (1991).
- [22] B. Batlogg et al., Physica (Amsterdam) 153-155C, 1062 (1988).
- [23] In a private communication R. Ikeda and T. Tsuneto pointed out to us that a technical difficulty in their calculations causes the magnetization results to be less reliable for $T < T_c$. This problem does not occur in their computation of the fluctuation conductivity; the results for the resistance below T_c are therefore believed to be much more accurate.