Critical fluctuations in the magnetization of $Bi_2Sr_2CaCu_2O_8$ near the $H_{c2}(T)$ line

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The magnetization measurements on $Bi_2Sr_2CaCu_2O_8$ single crystals with H parallel to the c axis are presented to show that the critical fluctuations in the magnetization near the $H_{c2}(T)$ line can be well studied within the two-dimensional nonperturbative scaling theory developed by Tešanović *et al.* By using this scaling theory and basic thermodynamic arguments, we obtained the Ginzburg-Landau parameter κ ($\simeq 115$) and dH_{c2}/dT ($\simeq -2.7$ T/K) near T_c for this material. In addition, we found that these derived parameters are in good agreement with those determined from the study of reversible magnetization in an intermediate field at low temperature.

Recently, it has been shown that the critical fluctuations in the thermodynamics of high-T_c superconductors near the upper critical-field $H_{c2}(T)$ line can be studied in terms of the Ginzburg-Landau (GL) field theory on a degenerate manifold spanned by the lowest Landau level (LLL) for Cooper pairs.[1-3] The GL-LLL theory implies that the free energy $F(H,T)$ near the $H_{c2}(T)$ line is of the form $F(T,H)=THf(At)$. $f(At)$ is a scaling function of the variable $t=[T-T_c(H)]/(TH)^n$, where A is a field- and temperature-independent coefficient, and n is $\frac{2}{3}$ for a three-dimensional (3D) system, and $\frac{1}{2}$ for a two-dimensional (2D) system. This scaling behavior for some thermodynamic quantities has been observed by several groups for many high-T_c superconductors. For instance, a 3D scaling behavior of the magnetization, Ettinghausen effect, and specific heat of the $YBa_2Cu_3O_7$ single crystal near the $H_{c2}(T)$ line was observed by Welp *et al.*,[4] while a 2D scaling behavior of the magnetization for a c-axis-oriented superconducting $Bi_2Sr_2Ca_2Cu_3O_{10}$ [Bi(2:2:2:3)] thin tape near the $H_{c2}(T)$ line was reported by Li *et al.*[5] However, there has been no report so far to our knowledge, that demonstrates a clear 2D scaling behavior for the magnetization of superconducting $Bi_2Sr_2CaCu_2O_8$ [Bi(2:2:1:2)] even though Bi(2:2:1:2) is thought to be a quasi-2D-type superconductor. It is the purpose of this paper to present our magnetization measurements on large Bi(2:2:1:2) single crystals with magnetic field parallel to the c axis, and to show that the critical fluctuations in the magnetization near the $H_{c2}(T)$ line do display a 2D scaling behavior. Furthermore, we shall demonstrate that the magnetization of Bi(2:2:1:2) near the $H_{c2}(T)$ line can be well studied within the two-dimensional nonperturbative scaling theory developed by Tešanović, *et al.*[2] By using this scaling theory and basic thermodynamic arguments, we obtained the value of the Ginzburg-Landau parameter κ ($\simeq 115$) for superconducting Bi(2:2:1:2), as well as dH_{c2}/dT ($\simeq -2.7$ T/K) near T_c. These values are found in excellent agreement with those derived by applying the modified Hao-Clem variational model (including the effect of vortex fluctuation in 2D superconductors) to our reversible magnetization data taken in intermediate field at low temperature.

Single-crystals Bi(2:2:1:2) were grown by the traveling solvent floating zone method.[6] The large crystal used in our study (cleaved from the single-crystal boule) weighs 76 mg with dimensions $4.1\times4.3\times0.64$ mm^3. The lattice parameters were measured by a four-circle diffractometer (Cu K_α radiation) and found to be $a=5.406$ Å, $b=5.411$ Å, $c=30.789$ Å. The composition of the crystals is $Bi_{2.14}Sr_{1.59}Ca_{1.13}Cu_{2.00}O_{7.83}$, in which all metal elements were determined by employing an inductively coupled plasma (ICP) technique, while oxygen content was measured by extraction of oxygen by carbon at a temperature above 2000 °C in an inert gas. A detailed optical microscopy study showed that the crystal that we studied consists of a few domains, but the c axes of each Bi(2:2:1:2) domain aligned perpendicular to the broad surface of our crystal with very small angle boundary of less than 5°. For the purpose of magnetic measurement along the c axis, we did not break this crystal into a single domain, since the magnetization measurement on this kind of crystal at fields parallel to the c axis is well known to have negligible difference with that of a *perfect* single crystal. The shield [zero-field-cooled (ZFC)] and Meiss-

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ner [field-cooled (FC)] data taken at a field of 2 Oe for this Bi(2:2:1:2) crystal are shown in Fig. 1. The shielding value of $4\pi M$ at 20K is -11.3 G which leads to the fraction of ideal shielding volume being a little over 100% after taking account the correction for the demagnetization factor[7] along the c direction. A linear extrapolation of $4\pi M(T)$ data to the zero magnetization line defines $T_c=84.2$ K, and that to the horizontal $4\pi M=-11.3$ G line gives a transition width of 1.5 K. The sharp transition demonstrates the uniformity of our crystal in quite a large scale (mass = 76 mg).

All magnetization measurements were carried out in a field applied parallel to the c axis by using a superconducting quantum interference device (SQUID) magnetometer (by Quantum Design) with a 2-cm scan length, where the field inhomogeneity is estimated to be no greater than 0.005%. The reversible magnetization data were taken by measuring the magnetic moment versus temperature from irrreversible temperature up to 200 K. A 10-min delay was introduced after each temperature change to stabilize the system so that the system temperature was always within ± 0.02 K of the target temperature prior to measurement. An accuracy of better than 2×10^{-6} emu (equivalent to 3×10^{-3} G for the value of $4\pi M$) for magnetic moments was obtained. Due to the strong fluctuation effect observed in Bi-based superconductors,[5,8] we corrected for both background and normal-state contributions by subtracting the measured magnetization at 200 K. It was also noted that background substraction by using the measured magnetization at 150 K instead of that at 200 K made negligible difference in the absolute value of the magnetization near the $H_{c2}(T)$ line used in this study.

Figure 2(a) shows the magnetization of superconducting Bi(2:2:1:2) near T_c in various fields parallel to the c axis. The strong fluctuation effect is clearly demonstrated by the crossover of various $4\pi M(H,T)$ versus T curves. The crossing point for each $4\pi M(H,T)$ versus T curve at fields between 500 and 55 000 Oe was found at the same location within the experimental error, where the crossing-point temperature $T^*=83.1\pm0.2$ K, and crossing-point magnetization $4\pi M^*=-3.7\pm0.3$ G. This well-defined crossing point for magnetization recently has been ascribed to one of the characteristics of fluctuation effects of a quasi-2D superconductor near mean-field transition temperature $T_c(H)$. In low fields near

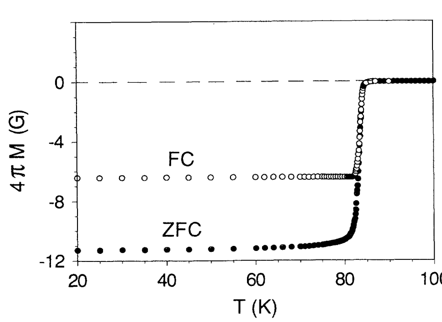

FIG. 1. The magnetic-shielding (ZFC) and Meissner effect (FC) measurements of the Bi(2:2:1:2) crystal for a field of 2 Oe applied parallel to the c axis.

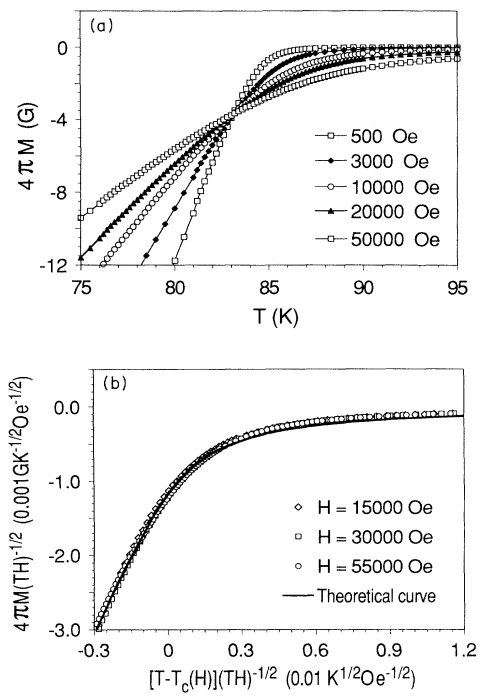

FIG. 2. (a) The T dependence of the magnetization of superconducting Bi(2:2:1:2) measured in various fields (parallel to the c axis). (b) 2D scaling of the magnetization data of Bi(2:2:1:2), where the theoretical curve is obtained from scaling function $-f'(x)$ given by Eq. (2).

$T_c(H)$, this crossover behavior is due to the phase fluctuation of order parameters[9] (or the positional fluctuation of vortices), while in high fields near the $H_{c2}(T)$ line it is largely caused by the amplitude fluctuation of order parameters.[2] In this work, we concentrated on the critical fluctuations at high field in the vicinity of $H_{c2}(T)$.

Critical fluctuations in the thermodynamics of quasi-2D type-II superconductors can be studied in terms of nonperturbative scaling theory developed by Tešanović *et al.*,[2] as long as the GL-LLL description for Cooper pairs is valid. The main result given by this theory is that the scaling function for free-energy density $F(B,T)$ of a 2D type-II superconductor in the critical region around the $H_{c2}(T)$ line can be expressed in an *explicit closed* form [Eq. (1)]:

$$F_s(B,T)=\frac{B^2}{8\pi}+F(B,T)\ , \quad \frac{F(B,T)}{TB}s\phi_0=f(x)\ , \quad x=At, \tag{1}$$

$$f(x)=-\tfrac{1}{2}x^2+\tfrac{1}{2}x\sqrt{x^2+2}+\sinh^{-1}\left[\frac{x}{\sqrt{2}}\right],$$

where $t=[T-T_c(B)]/(TB)^{1/2}$, A is a field- and temperature-independent constant given in Ref. 2, s is the effective spacing of superconducting layers, and ϕ_0 is the flux quantum $hc/2e$. Since the magnetization of an extreme type II superconductor near $H_{c2}(T)$ is very small as compared to applied field H, we can put $B=H$ in the argument of function $F(B,T)$ in Eq. (1). Then, the scaling function for magnetization takes the following form:

$$\frac{M(H,T)}{\sqrt{HT}}\frac{s\phi_0 H'_{c2}}{A}=-f'(x)=x-\sqrt{x^2+2}\ , \qquad (2)$$

where the leading derivative is kept, $H'_{c2}=|dH_{c2}/dT|$ at $T=T_{c0}$, and T_{c0} is the mean-field transition temperature at zero field. The scaling-function $-f'(x)$ for magnetization implies that all $M(T)$ curves for different H cross at temperature T^*, at which the value of magnetization $M^*(T^*)$ is given by $M^*(T^*)=k_B T^*/(s\phi_0)$ and $(T_{c0}-T^*)/T^*=H'_{c2}/(2A^2)$, where k_B is the Boltzmann constant (1.38×10^{-16} erg K^{-1}). In an earlier study of superconducting Bi(2:2:2:3), we found excellent agreement between our measured magnetization of Bi(2:2:2:3) near the $H_{c2}(T)$ line and this scaling theory.[2,10] Reference 10 gives a detailed description on the application of this scaling theory to study free energy, magnetization, and specific heat of a quasi-2D superconductor in the critical fluctuation region.

The critical region near the $H_{c2}(T)$ line, where the scaling theory applies, is taken approximately as $H\geq\frac{1}{3}H_{c2}(T)$ at fixed temperature, or in the equivalent temperature region of $T\geq T_{c0}-3H/H'_{c2}$ at fixed field. We fitted Eq. (2) to our entire magnetization data taken at fields from 15 000 to 55 000 Oe, and temperatures in the critical region of $T\geq T_{c0}-3H/H'_{c2}$, by using the fitting sequence given in Ref. 10. The only adjustable parameters involved are T_{c0} and H'_{c2}. The result of fitting is shown in Fig. 2(b), where a 2D scaling of these magnetization data and the corresponding fitting curve are displayed. An excellent agreement between the theory and our data is clearly illustrated in Fig. 2(b). The best fit yields $T_{c0}=87.32$ K and $H'_{c2}=2.73$ T/K. It was also observed that the 2D scaling behavior of our data was much more sensitive to the choice of T_{c0} than the choice of H'_{c2}. For instance, we found that a 1% change in T_{c0} could significantly distort the scaling plot, while a 7% change in H'_{c2} only slightly altered the appearance of the scaling plot. In order to verify that the 2D scaling of the magnetization of Bi(2:2:1:2) is unique, we also tried to scale our magnetization data in terms of variables $4\pi M/(TH)^{2/3}$ and $[T-T_c(H)]/(TH)^{2/3}$, as suggested for a 3D superconductor.[4,5] A rather poor scaling behavior was observed, with any choice of T_{c0} and H'_{c2}.

The *real superconducting* volume fractions presented in a particular quasi-2D superconducting specimen can be estimated based on the measured values $M^*(T^*)$ and T^*. $M^*(T^*)/T^*$ depends only on s, the effective interlayer spacing, which characterizes the type of 2D superconducting system. For Bi-based superconductors,[2,10] s can be taken to be half the unit cell length along the c direction. Then, in our Bi(2:2:1:2) crystal, we should have $s=15.395$ Å, which leads the ideal value of $4\pi M^*(T^*=83.1$ K) to be -4.53 G. However, our measured value for $4\pi M^*(T^*=83.1$ K) is -3.70 G so that the superconducting volume fraction in our crystal is about 82%, even though the low-field T_c measurements shows a 100% shielding. Actually, it is not surprising to find that some nonsuperconducting phases are enclosed in our crystal, since the Bi(2:2:1:2) single crystals used in almost all experiments are grown with an off-stoichiometry composition. Hence, these crystals may not be 100% superconducting in the mixed state. But, the fact that only 82% of our crystal contributes to the total measured diamagnetic moment does not affect the scaling behavior and the derived values of T_{c0} and H'_{c2} because the ratio of $M(T,H)/M^*$ was used in the fitting (for details, see Ref. 10).

The free energy, thermodynamic critical field $H_c(T)$, and specific heat of Bi(2:2:1:2) in the critical region can be estimated by means of Eq. (1) and the derived parameters (T_{c0}, H'_{c2}, T^* and $4\pi M^*$).[10] Figure 3 shows the temperature dependence of the calculated free-energy density $f_s(H,T)-f_n(H,T)$ for the Bi(2:2:1:2) crystal near T_c, where solid squares and circles are for $H=15\,000$ and 55 000 Oe, respectively. Also shown in Fig. 3 are the changes in Gibbs free-energy density $[g_s(H,T)-g_s(0,T)]$ (open squares and circles are for $H=15\,000$ and 55 000 Oe, respectively) for the superconducting state of the Bi(2:2:1:2) crystal in magnetic fields, obtained by integrating the magnetization data (including low field).[10] By using these results, we can calculate the superconducting condensation energy $f_s(T)-f_n(T)$ for the Bi(2:2:1:2) crystal, which in turn could be utilized to compute $H_c(T)$, via $f_s(T)-f_n(T)=H_c^2(T)/8\pi$. The inset of Fig. 3 shows such a derived $H_c(T)$ as a function of temperature. Here it should be noted that the data shown in Fig. 3 are calculated results without the correction for the actual superconducting volume fraction (82%) present in our sample. If we account for this correction, the corresponding values for the free energy of an ideal Bi(2:2:1:2) should increase by 22%, while $H_c(T)$ increases by 11%. Using the derived value of $-dH_c(T)/dT$ near T_c (170±20 Oe/K) and the relation $H_{c2}(T)=\kappa\sqrt{2}H_c(T)$, we found the Ginzburg-Landau parameter $\kappa\simeq115$ for Bi(2:2:1:2). It is surprising to note that these derived values of $H'_{c2}(\simeq2.73$ T/k) and $\kappa(\simeq115)$ based on this scaling analysis are in good agreement with the values $H'_{c2}(\simeq2.8$ T/K) and $\kappa(\simeq130$ at $T=30$ K), determined independently[11] by studying the magnetization of the same crystal in the intermediate fields at lower temperatures, where the reversible magnetization data were fitted to the Hao-Clem model[12] modified by taking account of

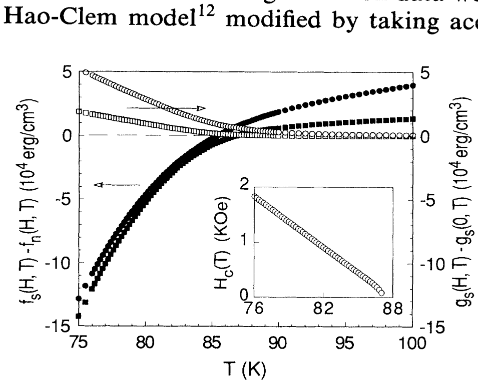

FIG. 3. The free-energy density $f_s(H,T)-f_n(H,T)$ (solid symbols) and $g_s(H,T)-g_s(0,T)$ (open symbols) for Bi(2:2:1:2) plotted as a function of temperature near T_c for $H=15\,000$ Oe (squares) and 55 000 Oe (circles). The inset shows the derived T dependence of $H_c(T)$.

the vortex fluctuation in a quasi-2D superconductor.[13] The detailed results of the low-temperature magnetization study will be reported elsewhere.[11]

The maximum specific-heat jump $\Delta C_p/T_c$ at T_c of Bi(2:2:1:2) at zero field can be estimated via the expression $\Delta C_p/T_c = H'_{c2}M^*/(T_{c0}-T^*)$. The value of $\Delta C_p/T_c$ for our Bi(2:2:1:2) crystal is ~ 31 mJ/mol/K^2. If we take into account the 82% superconducting volume fraction in our sample, the maximum specific-heat jump for an ideal Bi(2:2:1:2) would be around 38 mJ/mol/K^2. This value is substantially smaller than the corresponding value of $\Delta C_p/T_c$ (~ 63 mJ/mol/K^2), estimated for an ideal superconducting Bi(2:2:2:3) (Ref. 10).

In conclusion, we have shown a clear 2D scaling behavior for the high-field magnetization of a Bi(2:2:1:2) crystal near the $H_{c2}(T)$ line. This scaling relation was found to be in an excellent agreement with the nonperturbative 2D GL-LLL scaling theory, developed by Tešanović *et al.* Based on the scaling analysis, we also presented our study on the temperature and field dependences of some of the thermodynamic quantities for this material in the critical region. Superconducting parameters derived in this study are found very close to their values determined in a different approach based on the magnetization in intermediate fields at low temperature.

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