Scaling of the specific heat and magnetization of $YBa_2Cu_3O_7$ in magnetic fields up to 7 T

O. Jeandupeux, A. Schilling, and H. R. Ott
Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule Hönggerberg, CH-8093 Zürich, Switzerland

A. van Otterlo
Theoretische Physik, Eidgenössische Technische Hochschule Hönggerberg, CH-8093 Zürich, Switzerland
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The specific heat $C_p(T,H)$ and the magnetization $M(T,H)$ of a large twinned single crystal of $YBa_2Cu_3O_7$ have been measured using a high-resolution differential-thermal-analysis calorimeter and superconducting quantum interference device magnetometry, in magnetic fields up to 7 T. An apparently reasonable scaling of the data may be obtained using either the three-dimensional (3D) XY model or the lowest-Landau-level approximation. The high relative accuracy of the data allows a direct evaluation of the critical temperature T_c and the critical exponent ν of the 3D XY model. This analysis indicates that the data do not obey 3D XY scaling in fields above $\approx$0.5 T. [S0163-1829(96)03618-1]

I. INTRODUCTION

The thermodynamic properties of the high-temperature superconductors (HTSC's) near the critical temperature T_c are quite different from those of conventional superconductors. This is mainly due to large anisotropies of physical properties and small superconducting coherence lengths at zero temperature, ξ_0, which allow for significant fluctuation effects in the range around the transition temperature. Consequently, a simple mean-field theory no longer applies. The temperature interval around T_c in which critical fluctuations are important is usually given by $T_c\mathrm{Gi}$,[1] with the Ginzburg number

$$\mathrm{Gi}=\frac{1}{2}\gamma^2\left(\frac{T_c}{H_{c0}^2\xi_0^2}\right)^2,$$

where H_{c0} is the thermodynamic critical field and γ is the anisotropy parameter. Although in conventional superconductors Gi is very small ($\approx 10^{-7}$), it may be of the order of 10^{-2} in HTSC's and therefore the critical regime is accessible to experiments.

On theoretical grounds and by analyzing experimental results it has been argued that the superconducting transition of cuprates in zero field may be assigned to the three-dimensional (3D) XY universality class.[2,3] In the standard picture of HTSC's the evolution of the superconducting state in a nonzero magnetic field may be regarded as a crossover of the normal metal to a vortex liquid at $H_{c2}(T)$ and, subsequently, a vortex liquid to vortex crystal (or glass) freezing transition at lower temperatures.[1] In low fields the crossover at H_{c2} has been claimed to show features of the zero-field 3D XY type of transition.[2,3] For higher fields it has been argued that the lowest-Landau-level (LLL) approximation of the Ginzburg-Landau theory is adequate.[4–6] The region of validity of these two approaches is controversial.[7] The scaling collapse of experimental specific-heat and magnetization data of $YBa_2Cu_3O_7$ in different magnetic fields up to 10 T onto single curves has been interpreted as evidence for the validity of the 3D LLL theory,[8–11] and the 3D XY model,[12,13] respectively.

The scaling hypothesis implies that a physical quantity, plotted versus some scaling variable, will be invariant if the critical behavior is the result of the divergence of the corresponding correlation length ξ. For example, applying the 3D XY model, Salamon and co-workers[12] note that the quantity $\mathscr{M}=M(T,H)/H^{1/2}$ should collapse onto a single curve if plotted versus the variable $x=(T/T_c-1)/H^{1/2\nu}$.

The field H_{LLL} above which the LLL approximation should be valid is usually estimated to be $H_{\mathrm{LLL}}\sim \mathrm{Gi}H_{c2}(0)\approx 1$ T. For fields higher than the field H_{LLL}, the interaction between Landau levels is small and only renormalizes parameters.[14] The field H_{XY} below which the 3D XY model can be justified to be valid may be estimated by realizing that the magnetic field breaks the XY symmetry if the correlation length $\xi_{XY}\approx\xi_0 t^{-\nu}$, with $t=|T/T_c-1|$ and $\nu=\nu_{XY}\approx\frac{2}{3}$, exceeds the magnetic scale $\sqrt{\Phi_0/(\pi H)}$. Thus the critical region is visible if $t^{2\nu}\geqslant H/[2H_{c2}(0)]$. With the zero-field Ginzburg criterion $t\leqslant\mathrm{Gi}$ this requires that $H\leqslant H_{XY}=2H_{c2}(0)\mathrm{Gi}^{2\nu}\approx 1$ T. Remarkably, it turns out that $H_{XY}\sim H_{\mathrm{LLL}}$. Away from the regions where LLL or XY scaling applies, the physics is dominated by Gaussian fluctuations which, e.g., give a contribution to the specific heat of the form $C\sim H[T/T_c(H)-1]^{-3/2}$.[15] The situation is summarized schematically in Fig. 1.

In the following, we discuss the scaling of our data of the specific heat $C_p(T,H)$ and magnetization $M(T,H)$ of a large twinned single crystal of $YBa_2Cu_3O_7$ based on the 3D XY model. The scaling of the magnetization data is also investigated within the LLL approximation. We have chosen not to scale the specific-heat data in the LLL approximation because, in contrast to testing the 3D XY model, the LLL scaling relations require the uncertain evaluation of the dominant phonon contribution to the total specific heat. Note, however, that if the magnetization scales using one of these mentioned models, the specific heat must also scale, because both are

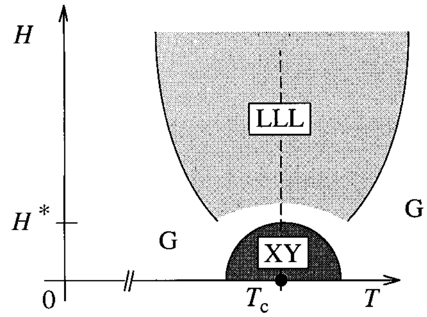

FIG. 1. A schematic sketch of the region of interest in the H-T phase diagram. The LLL approximation is valid above the field H^* and around the H_{c2} line. The isotropic 3D XY scaling applies for fields $H<H^*$ around T_c. The rest of the phase diagram is dominated by Gaussian fluctuations.

bulk thermodynamic quantities which are derived from the free energy. The problem with the uncertain lattice specific heat can be avoided by using the derivatives with respect to H and T of the scaling relations of the XY model. We show that with our procedure the critical temperature T_c and the 3D XY critical exponent ν may be evaluated directly. In the LLL approximation, the same procedure is not suitable for extracting directly the relevant parameters $T_c(H)$ and the dimensionality of the system.

II. EXPERIMENTS AND RESULTS

The dimensions of the single crystal of $YBa_2Cu_3O_7$ used for the experiments are approximately $4\times4\times0.5$ mm^3, with the c axis perpendicular to the largest surface and with a mass $m_s=16.8$ mg. It has previously been investigated in great detail.[16–18] The superconducting transition occurs at $T_c=91.4$ K, with a width $\Delta T_c\approx0.2$ K. For the experiments described here the crystal was oriented with the c axis parallel to the magnetic field.

A. Specific heat

The specific heat was measured using a high-resolution differential-thermal-analysis (DTA) calorimeter. This method has been described in detail by Schilling and Jeandupeux.[18] A reference sample, with a known heat capacity, and the sample are both thermally connected to a heat reservoir. Upon energy input, the temperature variations of the system are described by the equations

$$C_s\dot{T}_s=k_s(T_b-T_s), \tag{1a}$$

$$C_r\dot{T}_r=k_r(T_b-T_r), \tag{1b}$$

where the C's are the heat capacities, T's the temperatures, and k's the conductances of the heat links. The indices s, r, and b refer to the sample, the reference, and the heat reservoir, respectively. If we divide Eq. (1a) by Eq. (1b), we obtain

$$C_s=C_r\frac{k_s}{k_r}\frac{\dot{T}_r}{\dot{T}_s}\left(\frac{\Delta T}{\Delta T_r}+1\right), \tag{2}$$

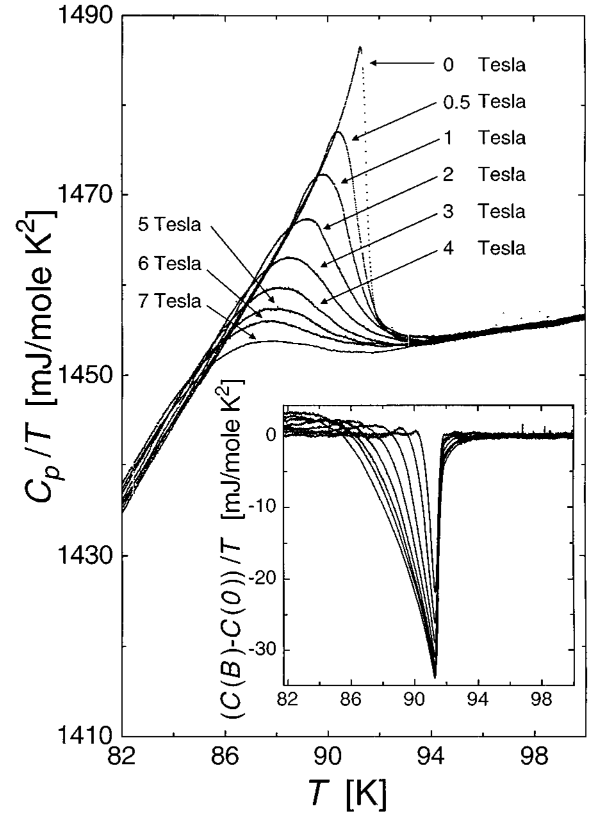

FIG. 2. Specific heat C_p/T vs T of the $YBa_2Cu_3O_7$ single crystal measured in different magnetic fields. The inset shows the difference between the data in nonzero fields and at $H=0$.

where $\Delta T=T_s-T_r$ and $\Delta T_r=T_r-T_b$. For the measurement of C_s, we vary the temperature T_b linearly with time t, regulating with a platinum thermometer, and measure ΔT and ΔT_r with copper-Constantan thermocouples. Except k_s/k_r, all quantities are measured. The heat links in the form of thin copper wires are selected so that nominally $k_s/k_r=1$. For the reference sample, we have chosen a polycrystalline copper specimen with a mass $m_r=15.6$ mg. Because of the monotonic T dependence of C_p of Cu, ΔT_r is a smooth function of time, which can conveniently be fitted. The essential quantity of the measurement is ΔT, which is accurately monitored with a picovoltmeter. Using polynomial fits of $\Delta T(t)$ and $T_r(t)=T_b(t)+\Delta T_r(t)$, we calculate the ratio $\dot{T}_r(t)/\dot{T}_s(t)=[\Delta\dot{T}(t)/\dot{T}_r(t)+1]^{-1}$. Due to parasitic thermal emf's, the absolute values of $\Delta T(t)$ suffer from small uncertainties between different runs. The data measured in different magnetic fields have thus been adjusted to the data taken in zero field using cubic polynomials, fitted above 100 K and from 50 to 70 K. The resulting data coincide above 100 K as expected, because the specific heats are identical if both the electronic and the lattice specific heats are unaffected by magnetic fields. We note, however, that below T_c the specific heat at constant temperature does vary with magnetic field, again as expected and verified in previous work.[19,20] In any case, the described procedure has a negligible effect on the results in the temperature range between 80 and 100 K. In Fig. 2, we show the specific-heat data C_p/T plotted versus T for magnetic fields up to 7 T after employing the procedure mentioned above, assuming

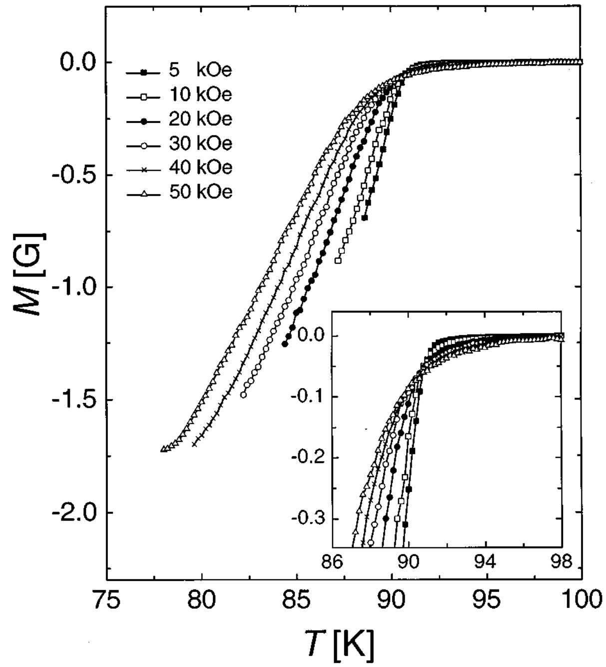

FIG. 3. Magnetization M vs T of the $YBa_2Cu_3O_7$ single crystal measured in magnetic fields of 5, 10, 20, 30, 40, and 50 kOe. The inset displays the data near the critical temperature on expanded scales.

$k_s/k_r=1$. The absolute values of our experimental data agree within less than 5% with previously published values, as discussed in Ref. 18. In the inset, we plot the difference between the data measured in various external fields $C(B)$ and those obtained in zero field $C(0)$. The shape of these curves is reminiscent of mean-field-type anomalies typical for continuous phase transitions. Far below T_c, $C(B)-C(0)$ increases with field, in agreement with calculations of Šášik and Stroud.[21]

B. Magnetization

The magnetization $M(T,H)$ was measured above the irreversibility line and for magnetic fields up to 5 T using a commercial superconducting quantum interference device (SQUID) magnetometer (Quantum Design, model MPMS). The magnetization and specific-heat measurements were performed on the *same* sample of $YBa_2Cu_3O_7$. From the raw data, we subtract a normal-state background, consisting of a constant offset plus a small Curie term, fitted above 100 K. The resulting $M(T)$ data are shown in Fig. 3 for different magnetic fields. The inset shows the magnetization data near T_c on expanded scales. We note a crossing of our magnetization curves as a function of temperature, but not in a single point, as has been observed in $Bi_2Sr_2CaCu_2O_8$.[22] The same feature has also been observed by Welp *et al.*[8] on single crystals of $YBa_2Cu_3O_7$, but not by Salamon *et al.*[12] on the same material. Recent numerical predictions for the magnetization of $YBa_2Cu_3O_7$, based on the Ginzburg-Landau theory, show a similar crossing in the same region of the phase diagram.[21]

III. DISCUSSION

A test of model predictions for the specific-heat characteristics of a superconducting phase transition in principle requires one to isolate the features of $C_p(T)$ of the electronic subsystem at the transition. In our case we intend to study whether the 3D XY model or the LLL approximation of the Ginzburg-Landau theory describes the anomalous behavior of the specific heat C_p and the magnetization M in the vicinity of T_c. In order to avoid the subtraction of an essentially unknown lattice contribution to the total specific heat, we prefer an approach which uses the specific-heat difference between measurements performed at different fields. Implicitly we assume that both the background electronic specific heat in the normal state and the lattice specific heat are not affected by external magnetic fields between 0 and 7 T. This *differential* approach reduces the number of fit parameters, thereby enhancing the level of confidence in the resulting fit parameters.

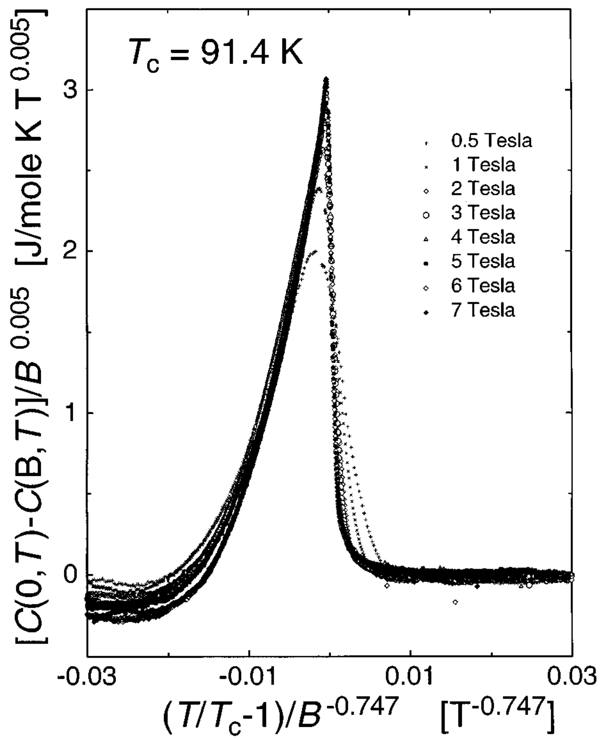

FIG. 4. 3D XY scaling of the specific heat according to Eq. (3) (see text).

According to the XY model, the scaling relation for the specific-heat data $C(T,H)$ is

$$[C(T,H)-C(T,0)]H^{\alpha/2\nu}=\mathcal{C}((T/T_c-1)H^{-1/2\nu}), \quad (3)$$

where $\mathcal{C}(x)$ is the unknown scaling function.[12] The parameters $\alpha\approx-0.007$ and $\nu\approx0.669$ are the 3D XY critical exponents of the specific heat and the coherence length, respectively. In Fig. 4 we show a compilation of our C_p data close to T_c plotted as required by Eq. (3). It may be seen that the scaling is not obeyed for low fields, exactly where the 3D XY model is expected to be valid. The deviations are not due to finite-size effects, as claimed by Salamon and co-workers,[12] because the superconducting coherence length ξ_{XY} is comparable to the size of the crystal only for $t\approx10^{-8}$.

For the magnetization, as mentioned in the Introduction, the XY model scaling relation is given by

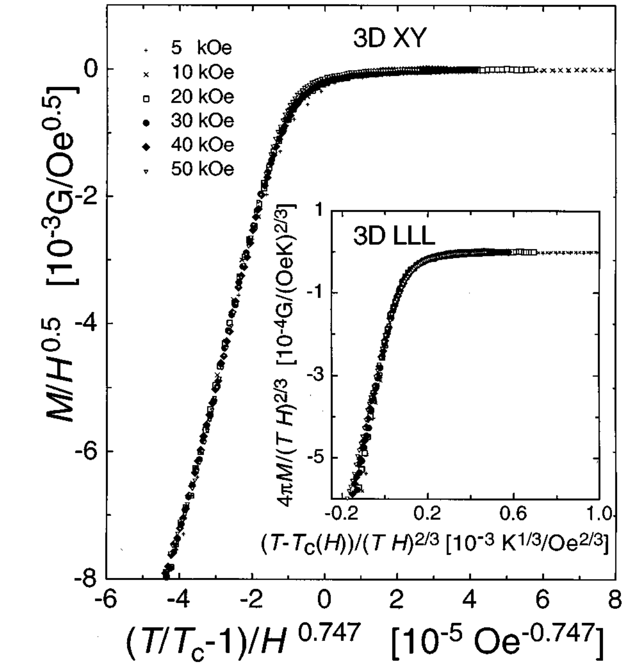

FIG. 5. 3D XY scaling of the magnetization according to Eq. (4) (see text). The inset shows the scaling of the same data based on the 3D LLL approximation.

$$\frac{M(T,H)}{H^{1/2}}=\mathscr{M}((T/T_c-1)H^{-1/2\nu}). \tag{4}$$

The 3D XY scaling of the magnetization $M(H,T)$ according to Eq. (4) is shown in Fig. 5. The parameter T_c has been adjusted for each curve, but is approximately field independent, as is assumed in the model. The inset of the figure shows a 3D LLL scaling of the same data. The relevant equation is

$$\frac{4\pi M(H,T)}{(HT)^{2/3}}=\mathscr{L}\left(\frac{T-T_c(H)}{(HT)^{2/3}}\right). \tag{5}$$

Here the forced scaling lets $T_c(H)$ vary quadratically with field, with a negative curvature. For both models the scaling seems to hold equally well but we note that the data scale best for $T>T_c(H)$ in the case of the 3D LLL approximation. We now turn to test the scaling relations via the differential approach.

Taking the derivative with respect to T of both the left- and right-hand sides of Eq. (3), we obtain

$$H^{\alpha/2\nu}\frac{\partial[C(T,H)-C(T,0)]}{\partial T}=\frac{\partial\mathscr{C}(x)}{\partial x}\frac{1}{T_cH^{1/2\nu}}.$$

The derivative of Eq. (3) with respect to H reads

$$H^{\alpha/2\nu}\frac{\partial C(T,H)}{\partial H}=\frac{\partial\mathscr{C}(x)}{\partial x}\frac{T_c-T}{2\nu T_cH^{1+1/2\nu}},$$

where the additive small term $\alpha/2\nu[C(T,0)-C(T,H)]H^{(\alpha/2\nu)-1}$ has been omitted because it is found to be smaller than the remaining term by two orders of magnitude. By eliminating the unknown function $\partial\mathscr{C}(x)/\partial(x)$ in the above equations, we obtain the relation

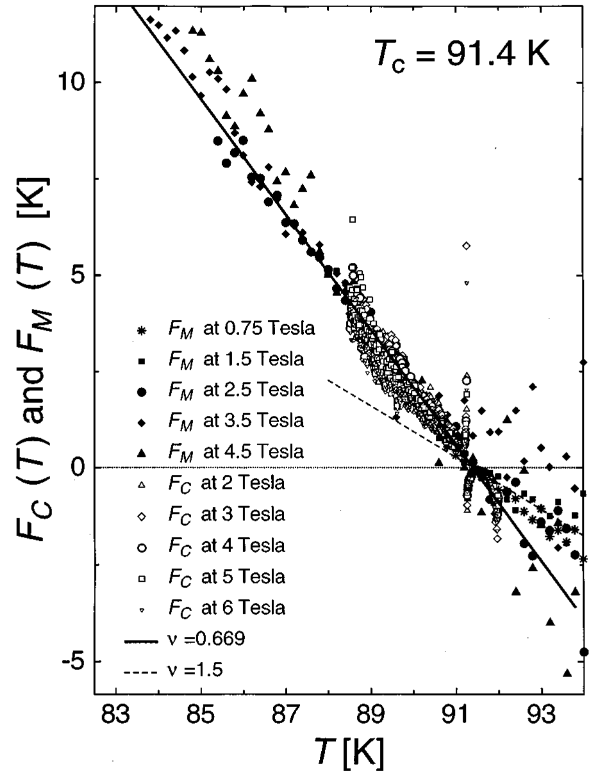

FIG. 6. Scaling of the specific heat and of the magnetization according to Eqs. (6) and (7), respectively (see text). The slope of the solid line is $1/\nu$, where $\nu=0.669$. The broken line is a fit to the data above T_c.

$$2H\frac{\partial C(T,H)/\partial H}{\partial[C(T,H)-C(T,0)]/\partial T}=\frac{T_c-T}{\nu}\equiv F_C. \tag{6}$$

Using the same procedure as for the specific heat, we obtain from Eq. (4)

$$2H\frac{\partial M(T,H)/\partial H-1/2M(T,H)/H}{\partial M(T,H)/\partial T}=\frac{T_c-T}{\nu}\equiv F_M. \tag{7}$$

Both F_C and F_M of Eqs. (6) and (7) depend solely on experimentally accessible quantities and vary linearly with temperature, provided the XY model gives an adequate description of the transition. In Fig. 6, we show the result of this analysis for *both* specific-heat *and* magnetization data. The derivatives of $M(T,H)$ and $C(T,H)$ with respect to T are evaluated using a temperature interval of ≈ 1 K. The derivative of $M(T,H)$ with respect to H at the fields $\langle H\rangle=0.75$, 1.5, 2.5, 3.5, and 4.5 T is approximated by $[M(T,H_{i+1})-M(T,H_i)]/(H_{i+1}-H_i)$ with the pairs of data measured at $\{0.5;1\}$, $\{1;2\}$, $\{2;3\}$, $\{3;4\}$, and $\{4;5\}$ T, respectively. For calculating $\partial C(T,H)/\partial H$ at $H=0.5$, 1, 2, 3, 4, 5, and 6 T, we use the corresponding C_p values at the pairs of fields $\{0;1\}$, $\{0;2\}$, $\{1;3\}$, $\{2;4\}$, $\{3;5\}$, $\{4;6\}$, and $\{5;7\}$ T, respectively. We show $F_C(T)$ only between 88.5 and 92.0 K, because both the numerator and the denominator of the left-hand side (LHS) of Eq. (6) are close to zero beyond these values (see Fig. 2). We also omit the F_C values at $H=0.5$ and 1 T where already the scaling according to Eq. (3) is not well obeyed. The solid line in Fig. 6 emphasizes the linear variation of F_C and F_M with T and it is

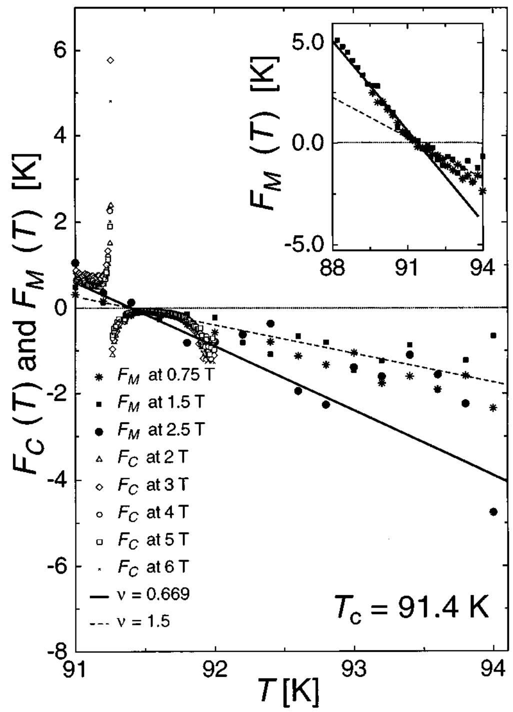

FIG. 7. $F_C(T)$ and $F_M(T)$ close to and above T_c on expanded scales. The data are better approximated by the broken line. Its slope corresponds to $\nu = 1.5$. The inset shows the $F_M(T)$ data for $H = 0.75$ and 1.5 T in the whole temperature range. The deviation from the linearity predicted by the 3D *XY* model is clearly observed above T_c.

drawn with a slope that corresponds to $\nu = 0.669$, the critical exponent of the 3D *XY* model. The crossing of the data with the horizontal axis provides a very well-defined experimental value for $T_c = 91.4$ K. The slope of the solid line and the plot of the experimental data are in excellent agreement, for temperatures below T_c, but the situation is less clear for temperatures above T_c. We show the temperature variation of F_C and F_M close to and above T_c on expanded scales in Fig. 7. Because of enhanced scattering, the $F_M(T)$ values in fields exceeding 3 T are not shown above T_c. Nevertheless, we may see that above T_c both $F_C(T)$ and $F_M(T)$ are inconsistent with a value $\nu = 0.669$ but are rather compatible with a value of $\nu > 1$. This is better documented in the inset of Fig. 7 where for low fields $H = 0.75$ and 1.5 T, the F_M values above T_c strongly deviate from the linearity predicted by the 3D *XY* model. These last observations invalidate the claim that the 3D *XY* scaling hypothesis is applicable for describing the superconducting phase transition of $YBa_2Cu_3O_7$ in external magnetic fields up to 7 T. Because in the LLL model T_c is assumed to vary with field, the same analysis as outlined above, involving the derivatives of the scaling relations, does not lead to similarly simple and transparent relations.

Before we state our conclusions some comments concerning the plots in Figs. 6 and 7 are in order. The strong upward and downward divergences of the F_C values just below T_c are due to a division by zero on the LHS of Eq. (6), when the derivatives of $C(T,H) - C(T,0)$ with respect to T pass from negative to positive values at the temperature where $C(T,H) - C(T,0)$ is minimum (see the inset of Fig. 2). We note that the temperature at which F_C diverges and T_c obtained from the fit procedure differ by 0.2 K. We ascribe this to numerical uncertainties in the evaluation of the derivatives F_C and F_M. Above T_c, at $T \approx 92$ K, the F_C data diverge downwards because, due to the numerical approximations, the numerator and the denominator of the LHS of Eq. (6) do not approach zero at the same rate. These diverging values are not relevant for the correct evaluation of the critical exponent ν, however, because the errors diverge as well.

IV. CONCLUSIONS

The analysis of the derivatives with respect to H and T of the usual 3D *XY* scaling relations for the specific heat and the magnetization allows a direct comparison of both these quantities with experiment and a direct evaluation of the critical temperature T_c and of the critical exponent ν of the coherence length. It is mainly the high resolution of the DTA method for measuring the specific heat of high-temperature superconductors which allows one to perform such an analysis. We have found for *both* specific-heat *and* magnetization data in magnetic fields exceeding ≈ 0.5 T that the critical exponent ν evaluated above T_c is not compatible with the prediction of the 3D *XY* model. A plausible explanation is that our analysis is done in high fields, in a region of the phase diagram where the 3D *XY* model does not apply and therefore its scaling relations are not valid. This interpretation is in agreement with our theoretical estimate of the limits of the validity of different approximations in the critical regime. Therefore our results should remove the controversies about the region of validity of the 3D *XY* model. From this extended analysis of the 3D *XY* relations we further conclude that an apparently good scaling of the magnetization is not sufficient for proving the validity of the model. This observation is also true in the case of the LLL approximation where the magnetization data scale well. Unfortunately, the complicated LLL scaling relation prohibits a similar analysis invoking T and H derivatives, and a direct comparison of the two models.

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