# Order of the fundamental vortex transformation in $YBa_2Cu_3O_{7-\delta}$

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In high-$T_c$ materials with low levels of static disorder, a small magnetization jump is thought to occur at the vortex solid to vortex liquid transformation. Using a null technique, the temperature dependence of the magnetization of an untwinned single crystal of $YBa_2Cu_3O_{7-\delta}$ has been investigated. No jump could be detected at the transformation, the data establishing an upper bound that is 1% of the value anticipated theoretically for a first-order melting transition. Residual disorder in the particular crystal used may have suppressed the jump to some extent. However, the result indicates that the first-order character of the fundamental transformation is likely to be weaker than anticipated.

## I. INTRODUCTION

The transformations exhibited by assemblies of vortices in high-$T_c$ materials have attracted much recent attention. There is strong evidence that an ordered vortex assembly (the flux "solid") is replaced by a less ordered configuration (the flux "liquid") above a well-defined temperature.[1] Monte Carlo simulations suggest that the transformation should be first-order in clean materials, with an associated entropy jump.[2] Sharp hysteretic resistive characteristics have been reported[3] in untwinned $YBa_2Cu_3O_{7-\delta}$ and identified with such a transformation. Subsequent experiments have been widely interpreted[4] as confirming the hypothesis that the transformation is first order. Although $YBa_3Cu_3O_{7-\delta}$ is the only material in which a sharp hysteretic resistive jump has been reported, a similar transformation has been invoked to explain the results of many recent experiments[5-8] on $Bi_2Sr_2CaCu_2O_8$.

The vortex solid to vortex liquid transformation in clean materials will be termed the fundamental transformation[1] in this paper. Despite the wide consensus that it is first order, transport experiments can only probe nonequilibrium aspects of the transformation. They cannot provide any direct information on changes in thermodynamic quantities. Indeed, it has been pointed out that the transport data can be qualitatively understood on the assumption that the transformation is continuous.[1] A recent quantitative study of the resistive characteristic also concluded that the presence of hysteresis does not establish that the transformation is first order.[9]

Aside of its intrinsic interest, the order of the fundamental transformation has implications for the widely accepted (vortex-glass) description[10] of the vortex solid that is realized when defects are introduced. There is strong evidence that the solid-liquid transformation in defected materials is second order.[11,12] A central tenet of the vortex-glass description is that this second-order character derives from the presence of static disorder. If the transformation in clean materials were to turn out to be second order, then the decisive role assumed for static disorder in vortex-glass theory would be open to doubt.

The issue of the transformation order can only be settled by probing an equilibrium property. An entropy jump implies that there should be an associated jump in the equilibrium magnetization. However, as discussed in detail in the next section, a difficulty encountered with any magnetization measurement is that the anticipated jump is small and superimposed on a much larger vortex contribution that itself varies strongly with temperature. Torque magnetometry[13] provides a probe of the magnetization that is sensitive to both its magnitude and direction. It turns out that this vector character can be exploited in a null experimental arrangement which effectively cancels out the temperature-dependent vortex contribution. Small superimposed magnetization changes can then be probed with significantly improved resolution.

## II. GENERAL EXPERIMENTAL CONSIDERATIONS

If the fundamental transformation is associated with a jump in the entropy, elementary thermodynamics implies that it should involve both a latent heat and a magnetization jump, $\Delta M$. The magnitude of the latter may be estimated from the magnetic analog to the Clausius-Clapyron relationship,[2] $\Delta M=[dT_m/dH]\Delta S$, where $\Delta S$ is the entropy jump per unit volume. (The field in studies of this sort is generally much greater than the lower critical field, so demagnetization effects are negligible.) The theoretical value suggested[2] for the entropy jump, $\Delta S_m=3\times10^{-1}k_B$, refers to the value of this quantity per vortex per CuO layer. The latent heat per unit volume $L$ and the magnetization jump $\Delta M_m$ anticipated for a sample exhibiting first-order melting are then given by

 

$$L=\frac{H}{s\phi_0}T_{\rm Tr}\Delta S_m ,$$
$$\Delta M_m=\frac{H}{s\phi_0}\frac{dT_{\rm Tr}}{dH}\Delta S_m , \quad (1)$$

where $s$ is the spacing of the CuO planes, $T_{\rm Tr}$ is the transformation temperature, $\phi_0$ is the flux quantum and $H$ is the applied field in Oe. Both $L$ and $\Delta M_m$ turn out to be small quantities, so it is convenient to work in cgs units. As discussed in the next section, for $YBa_2Cu_3O_{7-\delta}$ in a field of 20 000 Oe along the $c$ axis, $T_{\rm Tr}=87.2$ K and $HdT_{\rm Tr}/dH=2.5$ K. With these values, and $s=1.2\times10^{-7}$ cm, Eq. (1) gives $L=3\times10^3$ ergs/cm$^3$ and $\Delta M_m=4\times10^{-3}$ erg/G cm$^3$. (In what follows, the magnetization units will be denoted in the usual way as emu/cm$^3$, an emu having the units erg per G).

In considering the experimental implications of these numbers, it is important to bear in mind that a first-order transformation is only anticipated in crystals having a low level of static disorder. It is still difficult to produce large crystals with a low defect density. To our knowledge, the one studied in this work (volume $\sim10^{-4}$ cm$^3$) is the largest to show a sharp hysteretic resistive transformation. The latent heat for a crystal of volume $10^{-4}$ cm$^3$ is $<1$ erg. Furthermore, judging by the transport data,[3] the fundamental transformation occurs over a temperature interval of the order of 0.1 K. In the vicinity of the transformation, the specific heat of $YBa_2Cu_3O_{7-\delta}$ is known[14] to be $\sim10^7$ erg/K/cm$^3$. About $10^2$ ergs must therefore be supplied to drive the vortex assembly through the transformation. This is more than two orders of magnitude larger than the latent heat. Experimentally, then, a latent heat would appear as a tiny anomaly in the specific heat.

Turning to the magnetization jump, for $YBa_2Cu_3O_{7-\delta}$ at $T=87$ K in a field of 20 000 Oe, it is reported[15] that $dM/dT\sim0.3$ emu/K cm$^3$. The change in the vortex magnetization associated with a temperature increase of 0.1 K is therefore $\sim8\times10^{-2}$ emu/cm$^3$. This is $\sim8$ times larger than the anticipated magnetization step, which is a smaller (i.e., more favorable) ratio than that for a latent-heat experiment. However, reproducible temperature sensors contain magnetic material. The sample in a high-resolution magnetization measurement must therefore be coupled to the thermometry by exchange gas. As a consequence, temperature measurement and control present difficulties. A method which somehow nulled out the large temperature variation of the vortex magnetization would therefore be useful. An effective cancellation of this temperature variation has been achieved with the scheme described below.

## III. PRINCIPLE OF THE METHOD

The overall temperature dependence of the magnetization in $YBa_2Cu_3O_{7-\delta}$ is well established and exhibits features that are important for understanding the method discussed here. Figure 1 shows schematically the behavior observed in conventional superconducting quantum interference device (SQUID) measurements when a field of 20 000 Oe is applied along the $c$ axis.[15] There is a nonlinear region extending to a few degrees below $T_c$ which is thought to be due to fluctuations. At lower temperatures, a linear region extending over several degrees is observed. The linear magnetization extrapolates to zero at a temperature $T_c^*$, whose angular dependence can be understood quantitatively within the framework of the anisotropic Ginsberg-Landau equations.[16] The fundamental transformation temperature $T_{\rm Tr}$, located as discussed below is also indicated in Fig. 1. In the vicinity of the transformation the magnetization is linear, to the ($\sim1\%$) resolution of a conventional SQUID measurement.

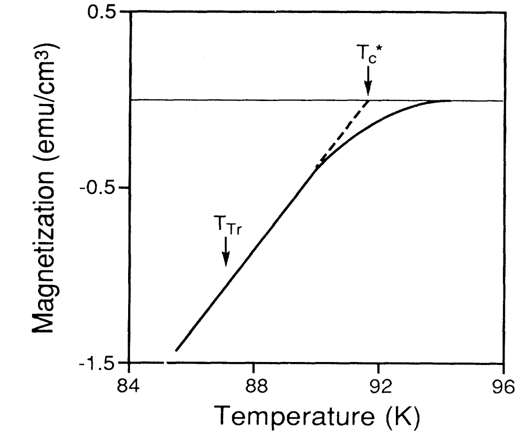


FIG. 1. Schematic plot of the main features of SQUID magnetization data for single crystal $YBa_2Cu_3O_{7-\delta}$ in a magnetic field of 20 000 Oe applied along the $c$ axis (Ref. 19). The vortex transformation in the same field occurs at the temperature $T_{\rm Tr}$. $T_c^*$ is the temperature at which the magnetization in the linear region extrapolates to zero.

Our approach to resolving small changes of magnetization at $T_{\rm Tr}$ involves a modification of conventional torque magnetometry, the principles[17] and practice[13] of which are well established. In conventional torque measurements on high-$T_c$ superconductors, a crystal of the material is placed in a uniform magnetic field **H** applied at an angle $\theta$ to the $c$ axis. If the field induces a magnetization **M**, the torque $\tau$ is given by $\tau=V\mathbf{M}\times\mathbf{H}$, where $V$ is the volume of the crystal. For the small values of $\theta(<10°)$ of interest here, the direction of the magnetization lies less than $\sim0.1°$ away form the $c$ axis.[18] A change in the magnitude of the magnetization $\Delta M$, therefore produces a change in the magnitude of the torque $\Delta\tau$ that is well approximated by $\Delta MVH\sin\theta$. In other words, changes in the torque provide a measure of changes in the $c$-axis magnetization.

In previous work along these lines,[19] using a standard tungsten wire magnetometer,[13] no changes could be detected at the fundamental transformation in $YBa_2Cu_3O_{7-\delta}$. However, for the reason mentioned previously, exchange gas was used to couple the sample to the thermometry. The resulting thermometry difficulty, together with the noise level associated with the standard instrument ($\sim 10^{-4}$ dyn cm), limited the effective resolution of the measurement to $\sim \Delta M_m$. The null method, realized with the high-resolution magnetometer described below, permits changes of the order of $10^{-3}\Delta M_m$ to be resolved.

The vortex torque discussed above has the symmetry property $\tau(\theta) = -\tau(-\theta)$. Hence, if two identical crystals are mounted so that the magnetic field lies in a direction bisecting the angle between their $c$ axes, the total torque on the pair, $\tau = \tau_1 + \tau_2$, would be zero at all temperatures. Such a null arrangement would therefore completely cancel the temperature-dependent vortex magnetization, achieving the goal outlined above. Unfortunately, equal and opposite jumps in $\tau_1$ and $\tau_2$ would occur at the fundamental transformation temperature, and so would be cancelled out. Our null method utilizes a less symmetric experimental arrangement which shifts the transformations in the two crystals to different temperatures. At the same time, although $\tau$ is no longer equal to zero for $T < T_c$, it is *independent of temperature* in the vicinity of the transformation. Thus the most important feature of the completely symmetrical arrangement is retained.

The method exploits the anisotropy of the fundamental transformation temperature. Anisotropy is anticipated for all high-$T_c$ vortex transformations governed by equilibrium thermodynamics.[20] In the case of the fundamental transformation in $YBa_2Cu_3O_{7-\delta}$, the transformation temperature increases if the angle $\theta$ is increased. The detailed angular dependence is well understood, both theoretically[20,21,22] and experimentally.[22,23] In our scheme, the volumes of the crystals are chosen to be different, the field making different angles, $\theta_1$ and $\theta_2$, with the $c$ axes of crystals with volumes $V_1$ and $V_2$, respectively. As discussed below, if the angles are properly adjusted, a small torque exists in the vicinity of the vortex transformations. However, since it is independent of temperature, it does not limit the experimental resolution in any way.

The origin of this temperature-independent offset can be understood on the basis of previous magnetization studies. As discussed above, with measurements of conventional resolution, the magnetization is observed to vary linearly with $(T_c^* - T)$ in the vicinity of the fundamental transformation.[15] The temperature $T_c^*$ depends on both the field and the angle, $\theta$, of the field to the $c$ axis.[18] If the field makes angles $\theta_1$ and $\theta_2$ with the $c$ axes of two different crystals, the temperature dependence of the total torque on the pair $\tau$ will be given by

$$\tau = |\tau_1 + \tau_2| = \alpha(T_{c1}^* - T) - \beta(T_{c2}^* - T) \ . \quad (2)$$

None of the four parameters in this expression depend on temperature. All of them depend on the magnitude of the applied field. In addition, $\alpha$ and $\beta$ are functions of $V_1, \theta_1$ and $V_2, \theta_2$, respectively, while $T_{c1}^*$ and $T_{c2}^*$ are functions of $\theta_1$ and $\theta_2$, respectively. In the arrangement adopted in this work, the crystals were mounted with their $c$ axes a fixed angle $(=\theta_1 + \theta_2)$ apart. The field orientation was then iterated until a temperature-independent torque was observed in the vicinity of the transformations, i.e., until $\alpha = \beta$. Equation (2) indicates that there must then be a constant offset torque, $\alpha(T_{c1}^* - T_{c2}^*)$, in the same temperature region.

Our method is designed to search for the torque jump calculated from Eq. (1). The temperature separation of the jumps in the two crystals is determined by the field and the ratio of their volumes. For the parameters of the present experiment, the transformation temperatures for the large and small crystals are 87.2 and 87.8 K, respectively, and $T_{c1}^* = 91.75$ K, $T_{c2}^* = 92$ K. The line in Fig. 2 shows the form anticipated for the temperature dependence of the total torque $\tau$ on the basis of Eqs. (1) and (2). (In the temperature interval from 91.75 to 92 K the contribution from the large crystal is zero and the sharp increase of torque over this interval arises entirely from the small crystal).

Previous SQUID magnetization studies[15] of $YBa_3Cu_3O_{7-\delta}$ were done with a resolution of $\sim 1\%$. At this level, the linear temperature dependence assumed by Eq. (2) held up to $\sim 89$ K (see Fig. 1). The present work probes the magnetization with a resolution of $\sim 10^{-3}\%$. The dashed line in Fig. 2 indicates the *sense* of the rounding that must occur at higher temperatures due to the fluctuation broadening in both crystals. However, the temperature at which deviations from Eq. (2) will occur in our experiment cannot be predicted from the previous studies, an uncertainty acknowledged by the question mark in Fig. 2.

The fundamental transformations could also be moved to different temperatures by using crystals with different transition temperatures. If the crystals have equal volumes a null regime would then be achieved with $\theta_1 = \theta_2$. If the choice of crystals is limited, more complicated arrangements are possible in which the transition temperatures, volumes and angles are all different. However, whatever arrangement is chosen, both Eq. (1) and an equation similar to Eq. (2) will hold, so a temperature dependence similar to that shown in Fig. 2 is anticipated.

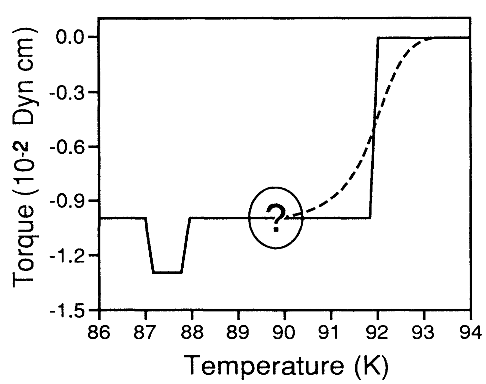


FIG. 2. The full line represents the temperature dependence of the total torque anticipated on the basis of Eqs. (1) and (2) for the balanced pair of crystals studied in this work. The drop at 87.2 K is associated with the large crystal, and the rise at 87.8 K with the small one. The dashed line indicates the sense of the anticipated rounding due to fluctuation broadening at higher temperatures in both crystals (see Fig. 1). As indicated by the question mark, the onset of this broadening can not be predicted on the basis of previous work.

## IV. EXPERIMENTAL DETAILS

### A. Torque magnetometer

The torque magnetometer described by Condon and Marcus[24] forms the basis of the instrument used here. An updated tungsten wire version of their design, achieving a noise level (defined below) of $\sim 10^{-4}$ dyn cm, has been described in detail elsewhere.[13] The special requirements of the present work, including a further substantial reduction in this noise level, necessitated the additional modifications described in this section.

Figure 3 shows schematically the elements of the instrument. The sample holder consists of two rectangular pieces of glass ($2\times1\times1/10$ mm) glued together with epoxy so that their large flat surfaces make an angle of 45° with each other. Using a thin layer of silicone grease, the flat surface of one of the crystals is glued to one of these plates; the second crystal is similarly attached to the other plate, fixing the $c$ axes at an angle of 45 degrees to each other. Helium exchange gas ($P\sim1$ Torr) couples the crystals thermally to the immediately adjacent thermometry. The sample holder is glued with epoxy to the lower end of a quartz rod that is 1 mm diameter and 70 cm long. The upper end of the rod is at room temperature and supported by a tungsten wire 25 $\mu$m in diameter. The holder and thermometry are enclosed in a copper can (not shown) with a small access hole in the top of the can for the quartz rod.

A commercial Ga-As temperature sensing diode and temperature controller[25] are used to measure and control the temperature of the experimental chamber. The transition temperature of a high-$T_c$ crystal may be measured by noting the temperature at which a torque first develops in a weak field. Transition temperatures of a variety of crystals in the 80–95 K range have been measured in this way. A comparison of the results with those obtained on the same crystals by the SQUID technique in other laboratories suggests that our measurements are reliable to $\pm0.1$ K. The temperature difference between the two crystals of the null pair is estimated to be completely negligible ($<10^{-4}$ K).

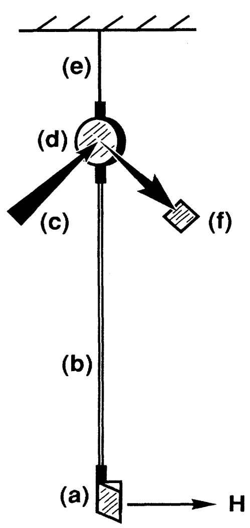


FIG. 3. Schematic (not to scale) of the null torque method described here. (a) Sample holder, (b) quartz rod, (c) Laser beam, (d) mirror, (e) Tungsten support wire, (f) Optoelectronic sensor.

As the temperature of the experimental chamber is varied, any change in torque experienced by the crystals is cancelled out by an equal and opposite torque applied at the upper end of the quartz rod. The nulling torque is produced by passing a current through a coil (not shown) that is fixed to the upper end of the rod and situated in the field of a permanent magnet. The current through the coil therefore provides a measure of the torque on the sample. An accurate null is achieved by using the optoelectronic feedback system indicated schematically in Fig. 3 and described in detail in Ref. 9. This maintains the angle of the sample fixed with respect to the direction of the sensing beam, with an uncertainty of $\sim10^{-4}$ degrees.

A magnetic field is produced by a conventional iron-core electromagnet with a current supply stabilized to 10 ppm.[26] The magnet can be rotated by hand, but changes smaller than $\sim0.03°$ are not practical. To allow the null orientation discussed in the previous section to be established more precisely, the optoelectronic detection system was mounted on a computer-controlled linear translation stage.[27] With the feedback system in operation, a translation of the stage by $10^{-4}$ cm rotates the quartz rod by $5\times10^{-4}$ degrees. Thus, the initial adjustment of the angle of the magnetic field to the balanced pair is done by mechanically rotating the electromagnet, with the translation stage being used for the final adjustment.

An important modification that was made for the present investigation involved the tungsten suspension. It was found that changes in apparent torque are produced by changes in the temperature of the wire. The temperature coefficient for the particular suspension used was measured to be $3\times10^{-4}$ dyn cm/K. Laboratory temperature fluctuations of $\pm1$ K therefore introduce significant measurement noise. As with the unidirectional drift phenomenon discussed below, no useful theory exists for this property of tungsten wire. In the present work, the phenomenon was eliminated as a source of noise by enclosing the suspension in an isothermal chamber. A thermistor was used in conjunction with commercial temperature control instrumentation[25] to hold the temperature at 305 K, with fluctuations limited to $\pm0.01$ K.

As discussed elsewhere,[13] suspensions utilizing tungsten wire always exhibit a curious unidirectional angular drift. With the feedback loop described above in operation, this produces an apparent time-dependent torque. Fortunately, the drift rate becomes small and constant a few days after mounting the suspension.[19] In our case, this terminal drift rate was $\sim10^{-5}$ dyn cm/h. With no sample present, the noise over any measurement period is defined as the standard deviation of the torque during the period, after this uniform drift has been subtracted out.

After eliminating the noise source discussed in this paper and others discussed previously,[13] the largest remaining source is associated with mechanical disturbances of the building housing the magnetometer. This source has roughly a $1/f$ character, so the noise as defined above depends on the period of measurement. Over a period of one second, it was below our measurement limit ($<10^{-7}$ dyn cm). However, to ensure thermal equilibrium, 1 h is required to sweep through the 1-K temperature interval of interest. All the data reported here were acquired by computer in the time period from midnight to 4 a.m., when building disturbances fall to a minimum. In this early morning period, the magnetometer noise over a 1-h period was typically $\sim 3\times 10^{-6}$ dyn cm. For comparison, the torque jump anticipated at the fundamental transformation is $3\times 10^{-3}$ dyn cm. Detailed torque magnetometer noise characterizations are not available in the literature. However, it appears that our magnetometer noise is nearly two orders of magnitude lower than lowest previously employed[9] ($10^{-4}$ dyn cm) for torque work in condensed-matter physics.

### B. Crystal characterization

The crystals for the balanced pair were chosen to have the same transition temperatures. As discussed above, equal and opposite jumps in the torque should occur at their transformation temperatures. For reasons that will be clarified later, it turns out that our method has significantly greater resolution for probing the transformation associated with the larger of the two crystals. Accordingly, a resistive characterization was only done for the larger crystal, which had a volume of $2.2\times 10^{-4}$ cm$^3$ was in the form of a flat plate of thickness $\sim 30$ $\mu$m.

The temperature dependence of the resistance of this crystal was measured as described previously,[28] using the standard four-lead method and a current density of $\sim 1$ amp/cm$^2$. Previous work[3,28] on the resistance of untwinned YBCO in a magnetic field has shown that the final $\sim 10\%$ of the resistance measured at $T_c$ is lost on cooling over a narrow ($\sim 0.1$ K) temperature interval. As shown in Fig. 4, in a field of 20 000 Oe along the $c$ axis the sharp drop occurs at 87.2 K, identified as the transformation temperature $T_{\rm Tr}$ in this field. From the observed field dependence of the resistive characteristic at $H=20\,000$ Oe, the value $HdT_{\rm Tr}/dH=2.5$ K, was obtained, in good agreement with estimates that can be derived from other literature data.[1]

A small untwinned crystal of volume $4.8\times 10^{-5}$ cm$^3$ was used to balance the torque on the larger one discussed above. The transition temperatures of both crystals were measured separately by the torque onset in a weak 80 Oe field and found to be identical ($=93.0$ K) within the uncertainties of measurement ($\pm 0.1$ K). In the null regime, the field made angles of 8° and 37° with the $c$ axis of the large and small crystal, respectively. The transformation temperature for the large crystal is raised by only 0.03 K from the $c$-axis result, while that for the small crystal lies 0.6 K higher.[22]

## V. RESULTS AND DISCUSSION

The experimental data for the torque in a field of 20 000 Oe are presented in Fig. 5. These were obtained by ramping the temperature down at the rate of 1 K/h. Data obtained with the temperature ramped up reproduced these to within the size of the data points, establishing the equilibrium character of the measurements. Two features are of interest.

Firstly, torque jumps of the magnitude shown in Fig. 2 are conspicuous by their absence. The temperature dependence of the torque in the vicinity of the transformation of the small crystal ($T=87.8$ K) complicates any attempt to resolve a small jump that may be due to it. We have therefore concentrated on the region close to

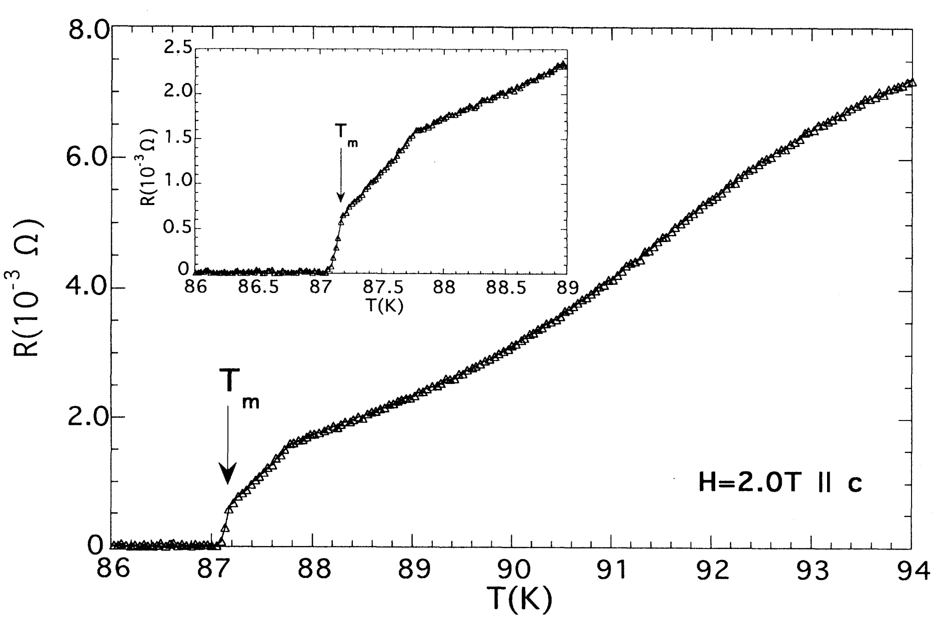


FIG. 4. The temperature dependence of the resistance of the large untwinned single crystal of $YBa_2Cu_3O_{7-\delta}$ studied in this work. A field of 20 000 Oe was applied along the $c$ axis, and the measuring current density was 1 amp/cm$^2$. The arrow indicates the temperature ($T=87.2$ K) at which a sharp resistive jump occurs, locating the fundamental transformation in this field. The inset displays data in the vicinity of the transformation.

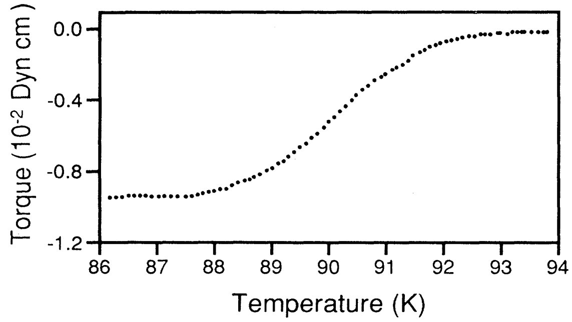


FIG. 5. The observed torque for a field $H = 20\,000$ Oe. The anticipated jumps at the vortex transformations (see Fig. 2) are not observed.

the transformation of the larger one. Figure 6(a) shows high-resolution data in a temperature interval centered on 87.2 K. These data represent the average of eight temperature sweeps made at a rate of 1 K/h, four with the temperature increasing and four with it decreasing. No systematic hysteresis was observed between the two directions.[29] The marker in this figure represents the torque jump anticipated for an entropy change of $10^{-2}\Delta S_m$. There is a slight decrease corresponding to a change of $\sim 3\times10^{-3}\Delta S_m$ in the temperature interval from 87.1 to 87.3 K. This feature is reproduced from run to run, and has the correct sense to represent a vestigial entropy change. However, it is close to the noise level of the present measurement and further work with larger crystals is required before any firm physical interpretation would be justified.

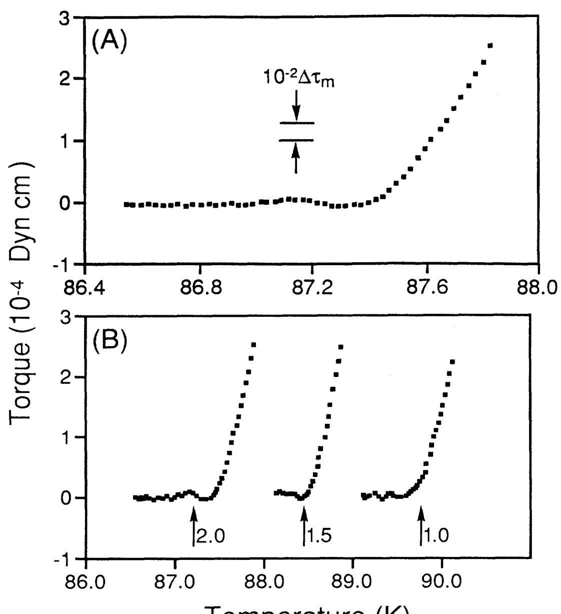


FIG. 6. (a) The data points represent the temperature dependence of the torque in a field of $H = 20\,000$ Oe and in a 1-K temperature interval centered on the transformation temperature of the larger crystal, indicated by the arrow. Note that the scale on the torque axis has a resolution that is two orders of magnitude higher than that in Fig. 5. The torque marker represents the change expected if a vestigial entropy jump equal to 1% of that anticipated theoretically were to occur. (b) Temperature dependence of the torque observed at three different fields. From left to right, the data sets were obtained in fields of 20 000, 15 000, and 10 000 Oe. The resistively determined transformation temperatures for the same fields are indicated by the labeled arrow.

The second feature of interest, clearly apparent in Fig. 6(a), is the rapid increase in the torque just above the transformation temperature of the large crystal. Further evidence suggesting that this feature is associated with the transformation is shown in Fig. 6(b). For three different fields, the onset of the increase tracks the transformation temperature obtained directly from transport measurements. In other words, the onset of fluctuation broadening, indicated by the question mark in Fig. 2, appears to occur close to the fundamental transformation.

The resistive transformation for the large crystal shown in Fig. 3 appears to be sharp and other measurements also indicate that it is hysteretic, with a field hysteresis of 75 Oe at 20 000 Oe. On the other hand, both the sharp drop and the hysteresis vanish for fields above $\sim 50\,000$ Oe. In other crystals, both these features have been observed to persist to higher fields.[30] In the same study it was shown that the resistive jump can be suppressed by the introduction of defects, so it is possible that the magnetization jump may be reduced by defects in our crystal. Further work on crystals in which the resistive jump persists to higher fields will be required to settle this point.

Recent SQUID magnetization data[8] on $BiBa_2CaCu_2O_y$ have been interpreted as establishing the existence of a relatively large ($\sim 0.2\Delta S_m$) entropy jump at the fundamental transformation in that material. However, in view of some uncertainties[31] in interpreting the SQUID data, it would be premature to draw any firm conclusions from this distinction.

The null technique approach discussed in this paper might also be applied in a differential thermal experiment. As discussed in Sec. II, a direct measurement of the latent heat appears to be impractical. However, in zero magnetic field, differential calorimetry has already proved useful for extracting the electronic component of the specific heat of high-$T_c$ superconductors.[32] A differential approach exploiting the anisotropy of the fundamental transformation might eventually make it possible to put a useful experimental bound on any latent heat.

To conclude, we have discussed the principles and practice of a new method for investigating small changes in the magnetization of high-$T_c$ superconductors. The technique employs a null variant of torque magnetometry that allows one to resolve small (1 part in $10^5$) magnetization changes superimposed on a strong temperature-dependent background. In this first application of the method, no magnetization jump could be detected at the fundamental transformation in untwinned $YBa_2Cu_3O_{7-\delta}$, at the level of 1% of that anticipated theoretically. The jump in the crystal employed may have been suppressed to some extent by the presence of defects. However, our work raises the possibility that the entropy jump accompanying the fundamental transformation may be significantly smaller than that anticipated theoretically.

Finally, a clear-cut result of this study is the finding that the onset of fluctuation broadening of the magnetization occurs at the fundamental transformation. This represents an equilibrium signature of the fundamental transformation in $YBa_3Cu_3O_{7-\delta}$.

## ACKNOWLEDGMENTS

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