Magnetization jumps and irreversibility in $Bi_2Sr_2CaCu_2O_8$

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Using torque magnetometry, the bulk magnetization of single-crystal $Bi_2Sr_2CaCu_2O_8$ has been investigated for temperatures close to the superconducting transition temperature. The results establish that the irreversibility field coincides with the field at which the vortex solid melts. It is also reported that the magnitude of the magnetization jump that can appear at the melting transformation is strongly correlated with the irreversibility displayed by the vortex solid. The accepted interpretation of the magnetization jumps observed in both local and bulk magnetic experiments on high-T_c superconductors is that they provide evidence for an entropy jump at the melting transformation. The results reported here suggest that the magnetization jumps may have an artifactual origin.

I. INTRODUCTION

The vortex transformations occurring in high-T_c materials have recently attracted widespread attention.[1,2] In the most studied transformation, an ordered flux lattice is thought to ''melt'' into a disordered assembly of flux lines.[2] The pioneering theoretical study[3] concluded that this transformation should be first order. Subsequent theoretical work,[4–9] although differing in detail, has supported that conclusion. Since the transformations at the upper and lower critical fields in conventional type-II superconductors are continuous,[10] a first-order melting transformation would represent an important addition to the vortex phase diagram in superconductors.

Hysteretic resistive characteristics have been reported for $YBa_2Cu_3O_{7-\delta}$,[11,12] and widely accepted[13] as evidence for a first-order transformation. Unfortunately, since resistance is a nonequilibrium property it cannot provide any quantitative thermodynamic information.[2,14] A study of an equilibrium property in $YBa_2Cu_3O_{7-\delta}$ (the magnetization) reported[15] an upper bound for any entropy jump of $0.003k_B$ per vortex per layer, an order of magnitude weaker than the lowest available theoretical estimate.[9]

A similar melting transformation has also been invoked to explain the results of many experiments on $Bi_2Sr_2CaCu_2O_8$.[16–23] Early experimental work established an upper bound of $0.02k_B$ per vortex per layer for any entropy jump at low temperatures.[16] In a recent study, the field and temperature dependencies of the magnetic induction B have been investigated over a wide temperature range using a miniature Hall probe.[22] A sharp jump in the induction was reported, a result that has been widely interpreted[24] as unequivocal evidence that the melting transformation just below T_c is strongly first order. In this work, the magnetization signature has been investigated in bulk single-crystal $Bi_2Sr_2CaCu_2O_8$. Our results suggest that the jump observed using either a local or bulk probe may have an artifactual origin.

No consensus exists that the transition of interest in $Bi_2Sr_2CaCu_2O_8$ corresponds to a simple melting transformation. In fact, the so-called ''decoupling'' scenario seems to describe the available phase boundary data equally well.[22] The less specific term ''transformation'' will therefore be used in the rest of this paper.

II. THEORY

A. The ellipsoidal geometry

The specimens used in both local and bulk magnetic experiments on $Bi_2Sr_2CaCu_2O_8$ are usually thin flat plates. Nonetheless, the physics of such experiments can be clarified by first considering the case of a sample in the form of an ellipsoid with the field applied along one of the principal axes. If it is further supposed that the material exhibits no flux pinning of any sort, then the field H inside the sample is uniform, and a reversible function of the applied field H_{app}:

$$H = H_{app} - 4\pi n M, \tag{1}$$

where n is the demagnetization factor for the chosen field direction, and M is the magnetization, also uniform through-

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out the sample. Hence the (uniform) induction B is given by

$$B=H_{\mathrm{app}}+4\pi(1-n)M. \tag{2}$$

Consider first an ellipsoid with $n=0$, so that $H=H_{\mathrm{app}}$. This geometry is approximated by the textbook ''long thin needle,'' with the field applied along the axis of the needle. It is assumed that a first-order transformation occurs at some field $H_m>H_{c1}$. If the field is increased, a magnetization jump ΔM will occur at $H_{\mathrm{app}}=H_m$. The value of ΔM is related to the entropy jump per vortex per layer, ΔS, by the magnetic analog to the Clausius-Clapeyron relationship,[4]

$$\Delta M=-(B/s\phi_0)(dT/dH_m)\Delta S, \tag{3}$$

where s is the spacing of the CuO double layers in the material and the derivative (dT/dH_m) represents the slope of the phase boundary. It should be noted that this relation is true for *all* geometries, provided M represents the average magnetization of the specimen. Since measurements indicate that the sign of dT/dH_m is negative,[22] ΔM is a positive quantity.

Associated with the change of M given by Eq. (3) is an increase, ΔB, in the induction throughout the specimen. In this special limiting geometry, the applied field interval over which the transformation takes place, ΔH_{app}, is equal to zero. In the local-probe literature,[22,23] $B-H_{\mathrm{app}}$ is reported as a function of H_{app}, and the change in the quantity $B-H_{\mathrm{app}}$ is used to evaluate the entropy jump. When $n=0$, Eqs. (2) and (3) indicate that $\Delta B=-4\pi(B/s\phi_0)(dT/dH_m)\Delta S$. Hence, in this special case, the experimental quantity of interest for an induction measurement is given by

$$\Delta(B-H_{\mathrm{app}})=-4\pi(B/s\phi_0)(dT/dH_m)\Delta S. \tag{4}$$

In contrast to Eq. (3), this relation is only valid for the limiting case of a long thin needle.

In principle, the response of an ellipsoid with $n>0$ is physically different from the case $n=0$. A similar distinction arises for the first-order normal-to-superconducting transformation that takes place in a type-I superconductor. In the type-I case, for $n>0$, the specimen spontaneously splits up into a domain structure consisting of normal and superconducting regions. In the present case, assuming that the transformation is also first order, a structure consisting of an intimate mixture of solid and liquid domains *must* occur, as discussed further in the Appendix. It is shown there that, in contrast to the conventional domain structure in type-I superconductors, the applied field interval over which this structure is anticipated to exist is small, even assuming a strong first-order transformation of the sort suggested in the literature.[22] The bulk transformation will therefore still take place at a fairly sharply defined field, and the bulk magnetization jump will still be related to the entropy jump by Eq. (3).

On the other hand, the fact that it requires a finite increase in the applied field to pass through the transformation does modify the character of the induction signature. Using Eqs. (A2) and (3), the quantity of experimental interest for an induction measurement will be given by

$$\Delta(B-H_{\mathrm{app}})=-4\pi(1-n)(B/s\phi_0)(dT/dH_m)\Delta S. \tag{5}$$

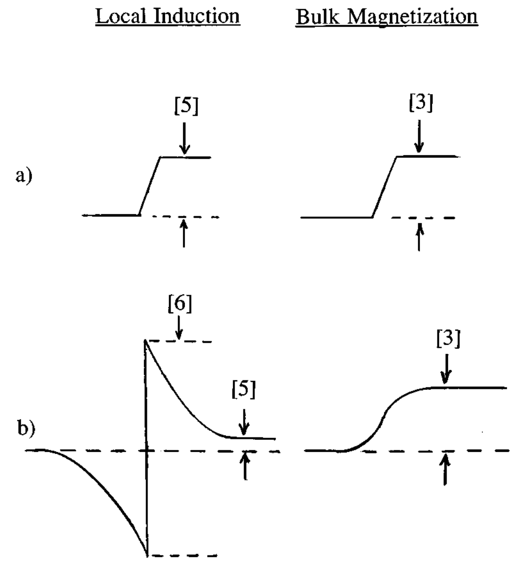

FIG. 1. Sketches of the dependencies of the local signature $(B-H_{\mathrm{app}})$ and the bulk magnetization M on the applied field, for two different types of sample geometry: (a) an ellipsoid; (b) a flat plate. The vortex transformation is assumed to be associated with an entropy jump ΔS. In each case, the numbers in brackets refer to the equations in the text that allow ΔS to be calculated from the jumps in the observed quantities. The background dependencies arising from the vortex solid and liquid have been ignored in making these sketches.

According to this expression, the induction signature depends sensitively on the shape of the ellipsoid, in sharp contrast to the magnetization signature [Eq. (3)] which is shape independent. The magnetization and induction signatures anticipated for an ellipsoid are sketched in Fig. 1(a).

The standard approach to calculating the magnetic properties of nonellipsoidal samples such as plates is to model the actual geometry by an inscribed ellipsoid. However, an example indicates that this approach fails for the phenomenon of interest here. Consider the case of a plate sample of $Bi_2Sr_2CaCu_2O_8$ at a temperature of 80 K, with $T_c=90$ K. Suppose that the inscribed ellipsoid approximation indicates an effective demagnetization factor $\sim$0.8, a value that is typical for the local induction experiments. A typical observed value[22] of $\Delta(B-H_{\mathrm{app}})$ at 80 K is 0.4 Oe. Inserting this into Eq. (5), noting that the melting field is reported[22] to increase linearly with (T_c-T), and making the approximation $B\sim H_{\mathrm{app}}$, gives $\Delta S/k_B\sim 3$ at this temperature. The literature estimate for $\Delta S/k_B$ is quite different, namely, $\sim$0.6.[22] The origin of this discrepancy is that Eq. (5) was *not* employed in Ref. 22 to evaluate the entropy change, but rather the quite different relationship

$$\Delta(B-H_{\mathrm{app}})=-4\pi(B/s\phi_0)(dT/dH_m)\Delta S. \tag{6}$$

For a given experimental value of $\Delta(B-H_{\mathrm{app}})$, the value of $\Delta S/k_B$ deduced from Eq. (5) is a factor $(1-n)^{-1}$ larger than that given by Eq. (6), accounting for the discrepancy just noted.

From the discussion given above, it should be clear that Eq. (6) is *not* a consequence of equilibrium thermodynamics.

As explained below, it represents an *approximation* to the complex physical situation that arises in the presence of flux inhomogeneity.

B. The consequences of inhomogeneity

For $Bi_2Sr_2CaCu_2O_8$ at temperatures close to T_c, it has been established that the interaction of vortices with bulk static disorder plays a negligible role in vortex pinning.[25] However, in a flat-plate geometry, flux pinning still occurs via an interaction with a ''geometrical'' barrier.[26] (A qualitatively similar effect occurs in type-I materials.[27]) This barrier consists of (persistent) currents that appear at the edges and surface of the material as a consequence of the nonellipsoidal geometry. The same currents cause the equilibrium flux density to reach a maximum in the center of the plate.[26] For a bulk magnetization measurement, the vortex transformation will take place at different applied fields for different locations inside the sample. This means that the bulk signature will be broadened, as sketched in Fig. 1(b). Still, if a magnetization jump ΔM is observed, the entropy change can be estimated directly by using Eq. (3).

The consequences of inhomogeneity for a local induction measurement are much more subtle.[28] As the applied field is increased, the transformation field will first be reached at the center of the sample. Hence the vortex liquid phase will first form as a ''droplet'' at the sample center. For further increases of field, the droplet will increase in size and eventually fill the whole sample. During its growth, the liquid-solid phase boundary will therefore pass underneath any noncentral surface probe. If the phase transition is first order, then, as discussed below, the passage of this phase boundary will generate a large change in the induction that is most naturally described as a ''wiggle'' of amplitude ΔB_w.

A number of assumptions are required to estimate ΔB_w. In our view, the most important of these is that the inhomogeneity discussed above has no effect on the local properties of either the flux solid or the liquid. In the case of the solid, it seems possible that the stress associated with the existence of a density gradient might lead to the presence of nonthermal defects (vacancies and dislocations). The liquid should not be affected in this way. Hence, even if there were no equilibrium jump in the entropy, the density of the solid could be reduced from its equilibrium value. The sense of this distinction between the the solid and liquid densities is the same as that observed with a local probe and interpreted as being associated with an entropy jump. (Note that the same assumption is implicit in our discussion of the bulk measurement. If the assumption fails, for the reason just suggested, then one will observe a jump in the bulk magnetization even in the absence of an entropy jump.)

However, let us proceed for the moment on the assumption that flux gradients do not introduce any artifacts of this nature. Elementary thermodynamics indicates that the local field H must be continuous across the interface between the solid and liquid. Since the local values of B, H, and M are related by $B=H+4\pi M$, the difference in B on crossing the interface, ΔB_w, is equal to $4\pi\Delta M$. Assuming that a negligible change in the applied field is required to drive the interface past the probe, we have $\Delta(B-H_{\rm app})=\Delta B_w=4\pi\Delta M$. Using Eq. (3), this leads directly to Eq. (6). In practice, the interface may be curved and of finite thickness, and the induction is monitored outside the specimen. $\Delta(B-H_{\rm app})$ will therefore depend on both the sample and probe geometries. Still, if the probe is close to the sample surface, and has a lateral dimension much smaller than the sample thickness, Eq. (6) should provide a first approximation for $\Delta(B-H_{\rm app})$.

Based on the above picture, the anticipated field dependencies for the local and bulk signatures in the presence of inhomogeneity are sketched in Fig. 1(b). The sharp rise in the local signature corresponds to the passage of the interface current sheet directly beneath the probe. The small difference between the initial and final values of $B-H_{\rm app}$ corresponds to the small change that would be estimated from Eq. (5) using an effective demagnetization factor approach. Figure 1(b) is drawn for the case of $n=0.9$. One of the published local signatures[22] appears to approximate the form shown in Fig. 1(b). [We also note that if the distance from the probe to the center of the specimen were small compared with the specimen thickness, there would be no initial decrease in $B-H_{\rm app}$ as the interface approached the probe. However, Eq. (6) should still apply for the jump $\Delta(B-H_{\rm app})$, and the rest of the signature should still be obtained. This possibility appears to describe the rather different results reported in another local investigation.[23]]

In summary, the signature reported in the local induction experiments could arise in (at least) two physically distinct ways. It could correspond to a true jump in the equilibrium entropy or to an artifactual distinction between the solid and liquid associated in some way with the presence of flux inhomogeneity. The first objective of our work was to obtain high-resolution bulk magnetization data close to T_c, where $\Delta S/k_B$ has been reported to rise to values in excess of unity.[22] These data are reported in the next section and establish that the apparent bulk entropy jump agrees quite well with the one obtained from the local probe. Our main goal was to study the bulk jump as a function of flux inhomogeneity, and these experiments are reported in Sec. IV.

III. MEASUREMENT OF MAGNETIZATION AND IRREVERSIBILITY

The magnetization close to T_c is roughly proportional to (T_c-T). Because of this strong background temperature dependence, it is difficult to resolve structure in $M(T)$. This experimental problem can be addressed using a differential technique, as discussed elsewhere.[15] Fortunately, the background field dependence of M is much weaker, making it possible to resolve small structure without the additional complication of a differential technique. The present investigation was therefore confined to a study of the field dependence of M.

The magnetization was obtained from $M=m/V$ where m is the magnetic moment and V the sample volume. It is technically impossible to make superconducting quantum interference device (SQUID) measurements of m as a function of the applied field without moving the sample. As discussed below, sample motion is known to be associated with a serious artifact. In this investigation, m was therefore measured with torque magnetometry, the sample remaining fixed throughout the measurement. The field $H_{\rm app}$ was applied normal to the plate (along the c axis of the crystal). An additional field $H_\parallel$ was applied parallel to its flat surface. The magnetic moment is equal to $\tau/H_\parallel$, where τ is the measured torque. In all the work discussed in the next section the value of $H_{\parallel}$ was 50 Oe. The effect of varying $H_{\parallel}$ is discussed in Sec. IV. Sample volumes were obtained by measuring the mass and using the formula density (=6.8 g/cm^3).

In general, if the magnetization is measured in increasing and decreasing fields, the two data sets coincide above a field that is termed the irreversibility field H_{irr}. Unfortunately, as explained in detail elsewhere,[2] the measurement of H_{irr} presents serious experimental problems for *all* type-II superconductors. As an example of just one difficulty, early work[29,30] on conventional (low-pinning) materials established that H_{irr} is always depressed by the application of an ac field. The standard SQUID technique involves the displacement of the sample in an inevitably nonuniform field, so an effective ac field is always present. For a given ac field, the depression of H_{irr} becomes larger as the flux pinning is reduced.[2] High-T_c materials exhibit extremely low pinning so measurements on them are particularly susceptible to this artifact.

Despite this difficulty, and others, it has generally been considered[2] that H_{irr} is identical to the transformation field H_m. However, it was recently claimed that H_{irr} and H_m can differ substantially in $Bi_2Sr_2CaCu_2O_8$.[23] On this basis, the same report argued that the irreversibility line and the melting transition have a completely different physical origin. In serious conflict with that report, it is shown here that the field at which irreversibility is destroyed always coincides with the appearance of the flux liquid.

The magnetization was measured at $T=77.5$ K on a sample of $Bi_2Sr_2CaCu_2O_8$ in the form of a disk with diameter 3 mm and thickness 0.04 mm, and with a T_c of 84.5 K. Stray 60 Hz fields in the vicinity of the sample were $\sim 10^{-1}$ Oe, a not uncommon level for a busy urban laboratory. The data are shown in Fig. 2(a). Although it can barely be seen with the resolution of this figure, a small structure, discussed below, occurs in vicinity of the field marked H_m.

Examining the data shown in Fig. 2(a), the irreversibility field appears to be about 30 Oe. Figure 2(b) shows data obtained under identical experimental conditions, except that screening was employed to reduce the stray 60 Hz fields in the vicinity of the sample to $\sim 10^{-3}$ Oe. The overall irreversibility is dramatically increased. In particular, the apparent H_{irr} is increased, although, at this resolution, it still appears to lie somewhat below H_m.

As mentioned previously, the ac artifact for conventional materials is well documented in the literature. The only unusual aspect of the data presented in Fig. 2 is a quantitative one—the sort of ambient ac field present in a typical laboratory causes a very large depression of H_{irr}. It was found that only the component along the *c* axis is effective—the transverse component producing no measurable depression. Almost completely reversible magnetization curves have been obtained with ac fields of 0.2 Oe amplitude applied along the *c* axis. Significant depressions of H_{irr} have been observed for fields as low as 10^{-2} Oe. This finding indicates that the power supply used to generate H_{app} must be free of ripple to about a part in 10^4 if the irreversibility is to be reliably measured. (In the experiments reported here, the ripple was less than one part in 10^5.)

Figure 3 presents magnetization data in the vicinity of H_m with a fivefold increase in resolution. The data are reversible above a field H_{irr}~80 Oe. The dotted line in this figure represents an extrapolation of the mean magnetization below 80 Oe. This field is identified as H_m because a reversible jump in magnetization, ΔM, occurs over a narrow (~4 Oe) interval of field just above 80 Oe. The relationship $H_{irr} \sim H_m$ has been verified in this way for a number of different sample geometries in the temperature range investigated, namely, $0.82 < T/T_c < 0.98$.

Using ΔM defined in Fig. 3, and the phase boundary data

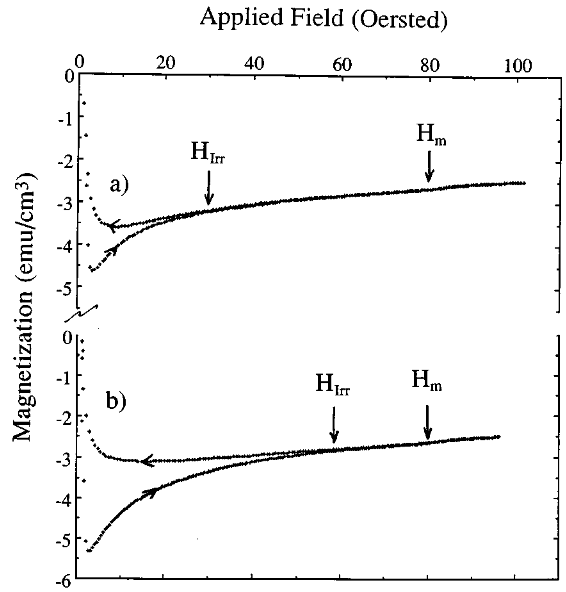

FIG. 2. Data for the field dependence of the magnetization of single-crystal disk of $Bi_2Sr_2CaCu_2O_8$ at a temperature of 77.5 K in two different environments. In each case, H_{irr} marks the apparent irreversibility field, and H_m the transformation field. (a) No precautions taken to shield the sample from stray ac fields. (b) Stray (~0.1 Oe) ac fields were screened from the vicinity of the sample. Note the substantial increase in the apparent value of H_{irr}.

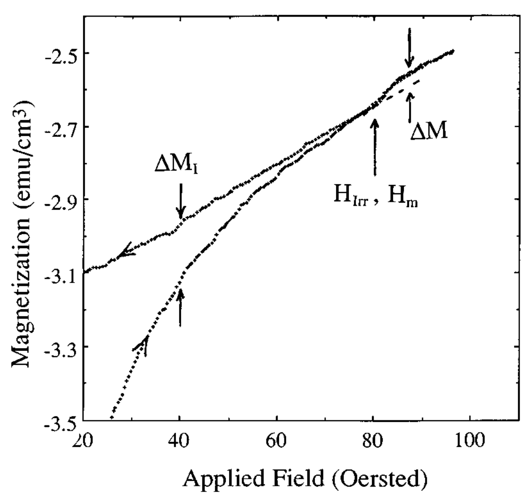

FIG. 3. Details of the data in Fig. 2(b) shown with higher resolution. In the absence of an ac field, a finite irreversibility ΔM_I can be resolved all the way up to H_m so that H_{irr} and H_m are one and the same field. A sharp reversible magnetization jump ΔM occurs just above the change of slope at H_m.

presented in Sec. V, the apparent entropy jump deduced from Eq. (3) is $\Delta S/k_B = 0.7$, in good agreement with the local-probe value for the same (reduced) temperature. The other quantity of interest for our subsequent discussion is the irreversibility ΔM_I, defined as the difference in magnetization between the two branches of the magnetization curve (see Fig. 3). It is too small to measure directly just below H_m, but a useful measure is the (percentage) irreversibility δ, defined by

$$\delta = 100\Delta M_I/M, \tag{7}$$

where ΔM_I is evaluated at a reference field $H_m/2$, and M is the mean of the two magnetization branches at the same field. This parameter provides a convenient relative measure of the overall irreversibility.

IV. VARYING THE FLUX INHOMOGENEITY

In experiments on conventional type-II materials with bulk pinning, the application of an ac field is thought to produce a homogeneous flux distribution approximating the thermodynamic equilibrium state that would be observed in the absence of pinning.[29,30] In the previous section, it was shown that the application of a weak ac field to $Bi_2Sr_2CaCu_2O_8$ produces a highly reversible state, so it is tempting to conclude that the flux inhomogeneity is eliminated by such a field. However, this ignores the physical origin of pinning and inhomogeneity in the case of $Bi_2Sr_2CaCu_2O_8$ and in the temperature range of interest.

As mentioned previously, both pinning and flux inhomogeneity derive from a ''geometrical barrier'' which is a consequence of the nonellipsoidal geometry of the sample.[26] However, the way in which they depend on the barrier is completely different from the way in which pinning and inhomogeneity are produced by bulk static disorder in conventional type-II materials. In the present context, the most important distinction is that the characteristic ''dome'' flux inhomogeneity persists above H_{irr}, i.e., it exists in the *absence* of any pinning;[31] inhomogeneity is a direct equilibrium consequence of the geometrical barrier, for either the solid or the liquid. By contrast, the results discussed above indicate that pinning only appears if the vortex assembly is in the solid state. From these facts, we infer that the ac field does not remove the flux inhomogeneity present in the nonellipsoidal geometry.

The geometrical barrier itself is a consequence of the particular configuration of Meissner screening currents that flow in a nonellipsoidal sample. An ellipsoidal geometry is difficult to achieve, but we have found that the irreversibility can be drastically reduced by increasing the transverse field $H_\parallel$. Note that, as $H_\parallel$ is increased, the angle that the total magnetic field makes with the flat surface of the sample becomes smaller. Our approach is therefore reminiscent of the slanting-field technique introduced by Sharvin[32] for studying the intermediate-state flux domains in flat plates of type-I superconductors. In the type-I case, an inclined field appears to reduce the pinning experienced by the domains, allowing the intermediate-state structure to approach more closely that predicted by equilibrium thermodynamics.[32–34] The mechanism involved in the present case will be discussed in Sec. VI.

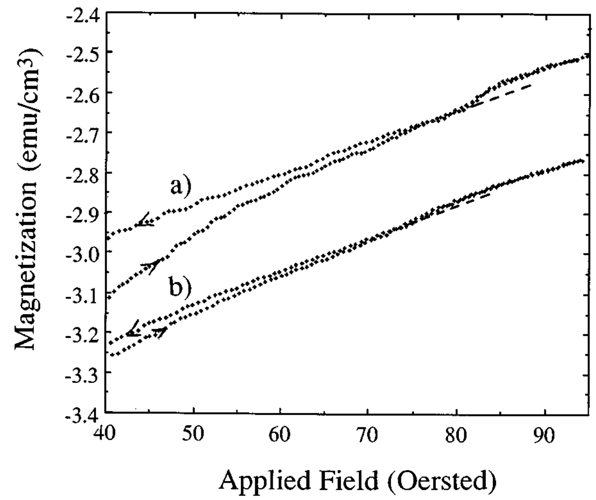

FIG. 4. Data for the field dependence of the magnetization at 77.5 K for two different values of the transverse field $H_\parallel$. (a) $H_\parallel = 50$ Oe; (b) $H_\parallel = 300$ Oe. (For clarity, 0.3 emu/cm^3 has been subtracted from these data.) The larger transverse field reduces *both* the irreversibility and the magnetization jump.

Figure 4 shows magnetization curves in the field region of interest at $T = 77.5$ K for the disk whose results are reported in the previous section. Data are shown for two different values of $H_\parallel$, 50 and 300 Oe. These data have a number of interesting features. First, at $H_\parallel = 300$ Oe, H_m is shifted downward in field, by ~4 Oe. According to the conventional scaling of the transformation field with angle,[35] one would expect a shift of order $\sim H_m(H_\parallel/\gamma H_{app})^2$, where γ is the anisotropy parameter. Although this parameter is not very well known, it is thought[36] to be of order 10^2; hence the predicted shift is of order 0.1 Oe, significantly smaller than is observed. The sense of the observed shift is consistent with a field-induced increase of γ. However, no (currently accepted) theory predicts such an effect so we are unable to offer an explanation for this particular feature of our data.

Of more interest for the present paper is the strong effect of $H_\parallel$ on both δ and the magnetization jump that is evident in Fig. 4. Compared with their values at the lower field, both δ and the jump are significantly reduced at 300 Oe. Figure 5

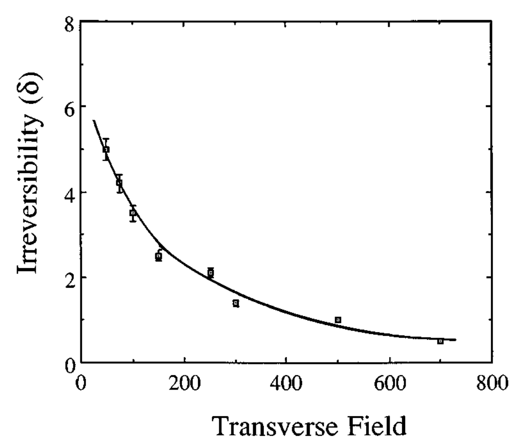

FIG. 5. Data for the suppression of the irreversibility δ (defined in the text) by the transverse field $H_\parallel$ for the disk sample at $T = 77.5$ K. The curve through the points is to guide the eye.

shows data for the dependence of δ on $H_{\parallel}$ at $T=77.5$ K. As $H_{\parallel}$ is increased, δ initially falls rapidly, with a more gradual fall apparent at higher fields. Data for the apparent entropy jump as a function of δ at $T=77.5$ K are shown in Fig. 6. These results are very striking, particularly the fact that the apparent entropy jump extrapolates to *zero* at zero irreversibility. This strongly suggests that the jump is indeed associated with flux inhomogeneity.

Additional support for this viewpoint is provided by the variation of both $\Delta S/k_B$ and δ with temperature. Figure 7 shows data for the dependence of δ on $H_{\parallel}$, at both 77.5 and 82.0 K. For $H_{\parallel}$ greater than $\sim$100 Oe, there is little difference between the two data sets. However, at lower fields, the irreversibility is significantly larger at 82.0 K. Data for the entropy jump as a function of δ are plotted in Fig. 8, together with those reported in the previous section at 77.5 K.

The data in this figure form the central result of our work. They clearly establish that the apparent bulk entropy jump is controlled by irreversibility, making it difficult to see how it can represent a true equilibrium thermodynamic property. The apparent jump (for small values of $H_{\parallel}$) is numerically close to the value reported for the local probe, obtained with $H_{\parallel}=0$. This implies that the bulk and local jumps almost certainly have the same physical origin.

A further experimental finding is of interest for our later discussion. The mean magnetization was found to be essentially independent of field sweep rate, but the value obtained for ΔM_I was found to depend on the time taken to complete the hysteresis loop. Smaller values of ΔM_I were obtained if the sweep rate was reduced. The empirical criterion used to fix the sweep rate was as follows. ΔM_I was measured as a function of sweep rate at each temperature and $H_{\parallel}$ value. The value found for ΔM_I was accepted as our best experimental estimate if a halving of the sweep rate increased ΔM_I by less than 5%. Using this criterion, the data shown in Fig. 4(a), with $H_{\parallel}=50$ Oe, were obtained with a sweep rate of 1 Oe/min. Those in Fig. 4(b), obtained with $H_{\parallel}=300$ Oe, required a rate of 0.25 Oe/min. Note that increasing $H_{\parallel}$ by a factor of 6 produces a substantial (factor of 4) increase in the relaxation time associated with changes of H_{app}. This curious experimental finding will be discussed further in Sec. VI.

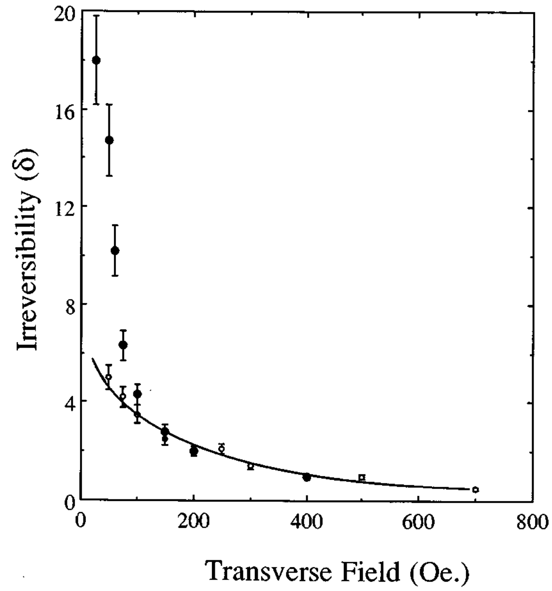

FIG. 7. Data for the suppression of the irreversibility δ by the transverse field $H_{\parallel}$ for the disk sample. Filled circles: $T=82.0$ K. Open circles: $T=77.5$ K. The line is drawn through the 77.5 K data to guide the eye. At high fields the two data sets fall close together, but the 82 K data move to markedly higher irreversibilities at lower fields.

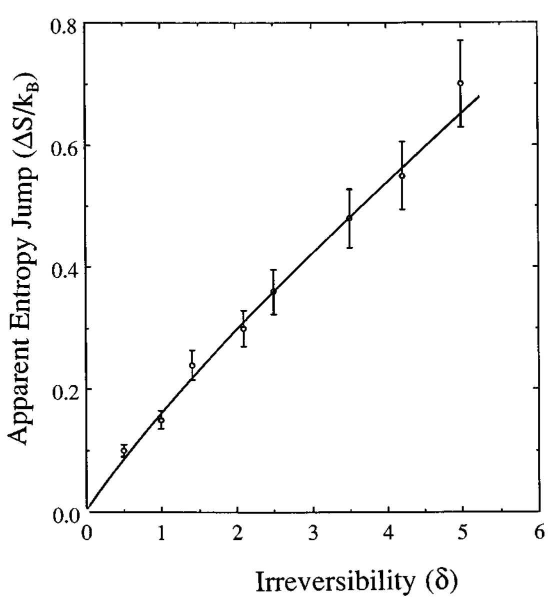

FIG. 6. Data for the dependence of the apparent entropy jump $\Delta S/k_B$ on the irreversibility δ for the disc sample at $T=77.5$ K. The curve through the points is to guide the eye. Note that in the limit of zero irreversibility, the apparent entropy jump tends to zero.

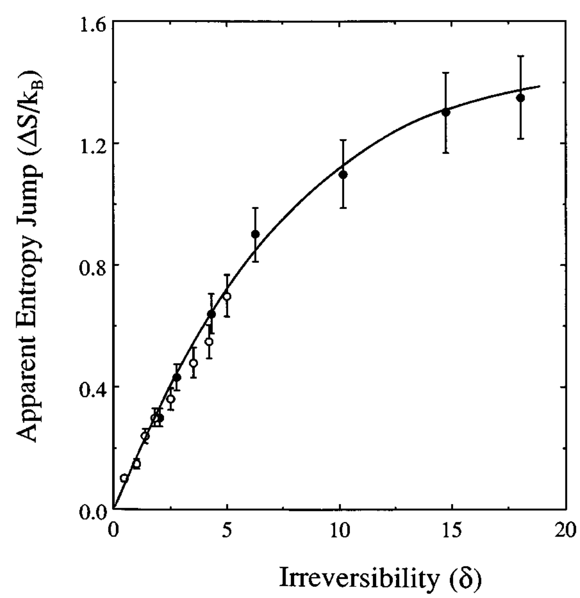

FIG. 8. Data for the dependence of the apparent entropy jump $\Delta S/k_B$ on the irreversibility δ for the disk sample at two different temperatures. Open circles: $T=77.5$ K; closed circles: $T=82.0$ K. The curve through the points is to guide the eye. This figure demonstrates that the apparent entropy jump is determined solely by the magnetic irreversibility.

V. ADDITIONAL EXPERIMENTS

A. Influence of the demagnetization factor

The experiments discussed above were done on geometries with effective demagnetization factors in excess of 0.9. The possibility that ΔM might depend on the demagnetization factor has been checked using a second crystal with $T_c = 84.6$ K. This was cut into nine squares, each with dimensions $\sim 1 \times 1 \times 0.1$ mm^3. Using a small amount of grease to glue them to a glass substrate, the squares were assembled into the plate configuration with dimensions $3 \times 3 \times 0.1$ mm^3. After magnetization measurements (with $H_\parallel = 50$ Oe) on this geometry were completed, the grease was dissolved with acetone and the squares reassembled into a pile with approximate dimensions $1 \times 1 \times 0.9$ mm^3. The effective demagnetization factors for both the plate and the pile were obtained from identifying dM/dH_{app} with $(4\pi[1-n])^{-1}$ in the Meissner region, giving $n_{\mathrm{plate}} = 0.94$ and $n_{\mathrm{pile}} = 0.43$. Despite this large range, the irreversibility and apparent entropy jumps were found to be roughly ($\pm 30\%$) equal, both to each other and to the jumps reported above for the disk.

Equation (6) predicts that the local induction jump should be similarly independent of geometry. However, as pointed out in Sec. II, a number of approximations are required to arrive at that equation. A series of local induction experiments have therefore also been performed to test the effect of varying the sample shape.[37] It was found that the local signature was strongly shape dependent. In particular, the amplitude of the signature was significantly reduced as the demagnetization factor was increased.

B. Measurements close to T_c

A rectangular plate crystal was available for this work whose volume was about ten times that of the disk discussed in the previous section. Although far removed from an ellipsoid geometry, the substantial increase in torque signal allowed the temperature dependence of the apparent entropy jump to be measured at temperatures significantly closer to T_c than was feasible for the disk.

The disk sample discussed in Sec. III was actually cut from this rectangular plate prior to the discovery that the irreversibility could be controlled by varying $H_\parallel$. Hence no such experiments were possible for the plate. However, on the basis of a set of experiments (not discussed here) on square plates, we believe that the same general behavior would have been obtained, but that significantly higher values of $H_\parallel$ would have been required to reduce the irreversibility parameter to the same extent as for the disk.

The plate had dimensions $6 \times 3 \times 0.22$ mm^3 and a T_c of 84.5 K. Figure 9 displays the magnetization data at $T = 77.5$ K, using $H_\parallel = 50$ Oe, both with and without an additional 0.2 Oe ac field. These data are qualitatively similar to those shown in Fig. 2, although the irreversibility is significantly larger than that for the disk, and the ac field less effective in reducing it. Figure 10 shows the same data at higher resolution, showing the magnetization jump near H_m. Note that a small irreversibility can be discerned right up to H_m, *even in the ac field data*, despite an apparent irreversibility field of ~ 50 Oe for the (lower-resolution) data in Fig. 9.

Results for the temperature dependence of the apparent entropy jump for the plate are shown in Fig. 11, and compared with those obtained using a local probe.[22] The agreement is good over the whole temperature range examined.

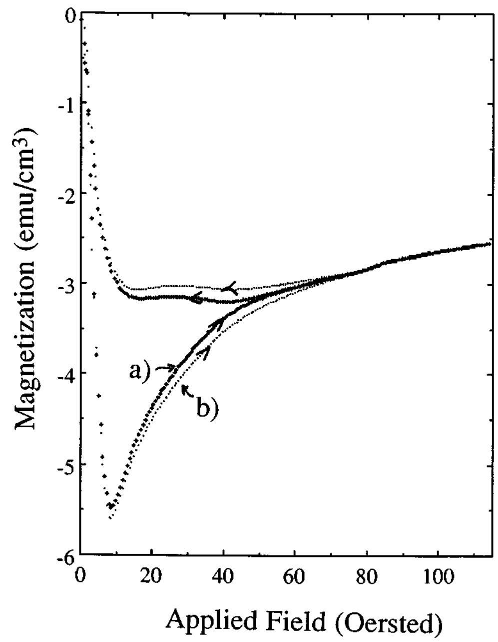

FIG. 9. Data for the field dependence of the magnetization of the rectangular plate sample of $Bi_2Sr_2CaCu_2O_8$ described in the text, at $T = 77.5$ K. (a) An ac field (5 Hz, 0.2 Oe) applied along the c axis. (b) ac fields screened to below 10^{-3} Oe.

C. The phase boundary

The temperature dependence of the transformation field displayed no particularly striking features, so the results will just be briefly summarized. The transformation fields for the plate were identical to those for the disk, which is not sur-

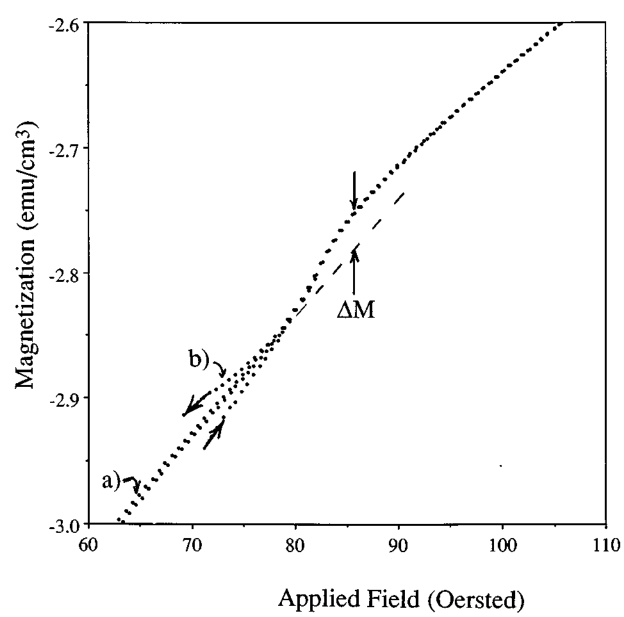

FIG. 10. Details of the data in Fig. 9, shown with higher resolution in the vicinity of the transformation field. A magnetization jump ΔM occurs just above the sharp change of slope at H_m.

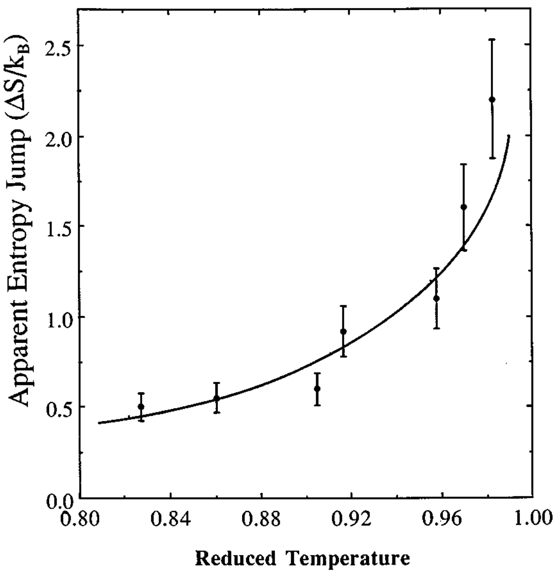

FIG. 11. The points are data for the dependence of the apparent entropy jump on reduced temperature ($=T/T_c$) measured for the rectangular plate sample. The line represents the data reported for the entropy jump using a local induction probe (Ref. 22).

prising, since, as mentioned, the disk was cut out from the plate. With an uncertainty of about ± 2 Oe, all the data were described by

$$H_m(T)=A(T_c-T), \tag{8}$$

where $A=11.4$ Oe/K. The phase boundary reported by Zeldov *et al.*[22] for a crystal with $T_c=90$ K is also linear close to T_c, but the constant A has roughly half this value. However, it is known that crystals with higher T_c's tend to have higher anisotropies.[38] If the underlying transformation corresponds to either melting or decoupling, an increase in the anisotropy will reduce $H_m(T)$, consistent with the sense of the difference just noted. Supporting this interpretation, the constant A was measured in the present work for a number of additional crystals. The data support the suggested correlation between A and T_c.

VI. DISCUSSION AND CONCLUSIONS

Despite the fact that the order of the transformation from vortex solid to vortex liquid is a central issue in the vortex field, to our knowledge only one thermal measurement of the entropy jump at the transformation has been reported.[39] This was for $YBa_2Cu_3O_{7-\delta}$ and gave an *upper* bound for $\Delta S/k_B$ of 0.03. However, a number of magnetic investigations have recently been reported that give entropy jump estimates close to T_c for the same material. To place the present work in a broader perspective, it is helpful to summarize the results of these experiments.

The magnetic estimates[15,40–42] for $\Delta S/k_B$ scatter over about four orders of magnitude, from an upper bound[15] of 0.003 to a value[42] of 25. Note that these experiments not only all studied the same physical quantity (the bulk magnetization), but also employed the same analysis [Eq. (3)]. This means that the magnetization jumps themselves varied by about four orders of magnitude. The scatter is therefore not a question of interpretation, but reflects a remarkable experimental uncertainty about the value of a well-defined physical quantity in a particular material.

By contrast, from a purely experimental viewpoint the situation for $Bi_2Sr_2CaCu_2O_8$ appears to be well settled. All the data on the bulk magnetization jump reported here (for small $H_\parallel$) agree fairly well with the induction jumps reported using a local probe, as to both magnitude and temperature dependence. It has been suggested here that these jumps do not reflect a real entropy change. Nonetheless, at least compared with the case of $YBa_2Cu_3O_{7-\delta}$, the *experimental* jump uncertainty for $Bi_2Sr_2CaCu_2O_8$ appears to be negligible.

Applying an additional field component $H_\parallel$ corresponds to tipping the total magnetic field away from the c axis of the crystal. As mentioned in Sec. III, the slanting-field technique was earlier used to study the domain structure in plates of type-I superconductors. In the type-I case it is known[32–34] that the flux structures that are obtained in this way better approximate those anticipated by equilibrium thermodynamics. Although this also appears to be the case for the vortex assembly realized in $Bi_2Sr_2CaCu_2O_8$, the mechanism involved must be quite different.

As discussed in Sec. IV, in addition to suppressing the magnetization jump, an increase of the transverse field component $H_\parallel$ produces two marked experimental effects. It increases both the reversibility *and* the relaxation time. Arguing from the large anisotropy of $Bi_2Sr_2CaCu_2O_8$ and torque data obtained at higher fields, it has previously been thought that the magnetic response of this material is entirely determined by the field component along the c axis.[36] It is therefore not at all obvious how the field $H_\parallel$ can play any role in the present experiments. The following reasoning (due to Kogan[43]) indicates physically how the three experimental effects reported here might arise.

In the presence of a finite transverse field $H_\parallel$, and in the low fields of interest, the Abrikosov vortices are thought to coexist with Josephson vortices, the latter having a density $H_\parallel/\phi_0$.[44] In the London limit, with core effects neglected, the Abrikosov and Josephson vortex systems do not interact. In reality, within the core of an Abrikosov vortex the Josephson coupling is suppressed by an amount on the order of $(\hbar j_0/e)\xi^2$, where j_0 is the Josephson critical current density $(=c\phi_0/8\pi^2 s\lambda_c^2)$ and ξ is the size of the (Abrikosov) vortex core. This constitutes a (weak) potential barrier that an Abrikosov vortex must overcome in order to pass through a Josephson vortex.

Suppose that a field $H_{\rm app}$ in the range $H_p<H_{\rm app}<H_{c1}$ is applied normal to the plane of a disk with no bulk static disorder, where H_p is the field for the first entry of flux. Vortices penetrate at the sample edges and are propelled inward by the Meissner screening currents with no impediment to their motion, assembling into the ''dome'' structure discussed in Sec. II. In the presence of Josephson vortices, motion in a direction along $H_\parallel$ is still free. However, motion in a direction perpendicular to $H_\parallel$ will be slowed down by a dissipation (per unit length) that is of order $(\hbar j_0/e)\xi^2 vH_\parallel/\phi_0$ where v is the component of the vortex velocity perpendicular to $H_\parallel$. An immediate consequence is that flux relaxation processes will slow down as $H_\parallel$ is increased, in agreement with the relaxation time results reported in Sec. III.

In addition, it is evident that an interaction (weak and unidirectional) exists between the Abrikosov and Josephson vortex systems. The associated force on an Abrikosov vortex is proportional to $H_{\parallel}$ and lies in a direction perpendicular to $H_{\parallel}$. Detailed calculations would be required to estimate the flux profile in the presence of this interaction between the different vortex species. However, since the interaction of the Abrikosov vortices with static disorder produces a flux gradient in the opposite sense to that of the dome, it is a reasonable conjecture that the additional interaction discussed here might reduce the dome inhomogeneity. According to the discussion given in Sec. II, it would therefore also reduce both the irreversibility and the magnetization jump.

To summarize, this paper has explored the possibility that the magnetization jump observed in both bulk and local measurements in $Bi_2Sr_2CaCu_2O_8$ may be an artifact associated with the unavoidable flux gradients present in nonellipsoidal samples. We suggest that field gradients may induce defects in the vortex solid, but not in the liquid. In zero transverse field, these defects will create an artifactual distinction between the solid and the liquid, and produce a magnetization jump. It is suggested that the presence of a transverse field tends to reduce the major source of field inhomogeneity (the dome), so that the artifactual magnetization jump is also reduced.

Note that this same scenario might also help to explain the striking scatter in the magnetization jump data for $YBa_2Cu_3O_{7-\delta}$ that was noted earlier. The extremely low (0.003) upper bound for $\Delta S/k_B$ was obtained[15] with the magnetic field tilted from the c axis. While the tilt angle (8°) was smaller than that employed here, the applied fields for $YBa_2Cu_3O_{7-\delta}$ are $\sim 10^2$ larger than for $Bi_2Sr_2CaCu_2O_8$. It is therefore possible that an inhomogeneity artifact is also producing the large jumps observed in $YBa_2Cu_3O_{7-\delta}$.

An ordinary atomic lattice can only accommodate a certain density of defects. For the Abrikosov lattice, this would imply that an artifactual jump of the sort considered here should saturate as the inhomogeneity and irreversibility are increased. This feature is certainly displayed by the data in Fig. 8, and helps to explain why a variety of samples may exhibit roughly comparable apparent entropy jumps at small transverse fields. All that is required is that the defect density is close to the saturation region.

Setting all interpretations aside, the most important *experimental* finding reported here is that the apparent entropy jump is strongly correlated with the irreversibility in the vortex solid. It particular, it tends to zero as the irreversibility is reduced to zero. *Whatever* the correct interpretation of the magnetization jump turns out to be, it seems very difficult to avoid the conclusion that it cannot represent a genuine thermodynamic phenomenon. Of course, a small fraction of the jump might still be thermodynamic in origin. Judging by the closeness of the intercept in Fig. 6 to zero, any such contribution to $\Delta S/k_B$ must be significantly less than 0.1.

As mentioned in Sec. I, early theoretical study[3] of the solid-liquid vortex transformation concluded that it should be first order and subsequent theoretical work[4–9] has supported that conclusion. In view of the results reported here, it is of interest to note that a very recent fundamental reexamination of the order issue has concluded that the transformation should be continuous.[45]

In conclusion, we have reported experimental evidence for a strong correlation between the apparent entropy jump and irreversibility at the transformation from vortex solid to vortex liquid in $Bi_2Sr_2CaCu_2O_8$. This result indicates that the magnetic jumps that can be observed in both local and bulk experiments on high-T_c superconductors may have an artifactual origin. Our interpretation successfully accounts for a good deal of experimental data, but further systematic studies will be needed to place it on a firm experimental and theoretical footing.

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APPENDIX: FIRST-ORDER MAGNETIC TRANSITIONS IN ELLIPSOIDAL SPECIMENS

As mentioned in Sec. II, if the solid-to-liquid vortex transformation is first order, it will occur over a finite range of field, even in an ellipsoidal geometry. This behavior is analogous to that of the first-order normal-to-superconducting transformation exhibited by a type-I superconductor in the presence of a magnetic field. In that case, the role of the ''transformation field'' is played by the thermodynamic critical field H_c. For $n=0$, the normal-superconducting transformation occurs when the applied field equals H_c, and the jump in the magnetization at the transformation is just $H_c/4\pi$. For $n>0$, it is known[9] that the intermediate-state structure exists within an applied field range $(1-n)H_c<H_{\mathrm{app}}<H_c$, i.e., in a field interval of $\Delta H_{\mathrm{app}}=nH_c$. For this entire field interval, the local field H throughout the specimen remains equal to H_c. Since it is related to the applied field by Eq. (1), the total change in magnetization within this interval is given by $\Delta H_{\mathrm{app}}/4\pi n=H_c/4\pi$. Note that this is the same as the sharp jump in the magnetization for the case $n=0$.

The same thermodynamic reasoning demands that in the present case a liquid/solid domain structure should persist over an applied field range given by

$$\Delta H_{\mathrm{app}}=4\pi n\Delta M, \tag{A1}$$

where ΔM is the jump in the magnetization. At a temperature of 77.5 K where $H_m\sim 80$ Oe, we observed $\Delta M\sim 0.03$ emu/cm^3 (see Fig. 4). It is argued in the paper that this jump is unlikely to be thermodynamic in origin. However, let us suppose that this value of ΔM represents a *bona fide* thermodynamic jump. Inserting it into Eq. (A1) and setting $n=1$ (to obtain an upper bound), one finds that $\Delta H_{\mathrm{app}}=0.4$ Oe. The observed width of the bulk transformation is ~ 4 Oe (see Fig. 4). Hence the largest conceivable broadening due to the presence of the solid/liquid domain structure is an order of magnitude smaller than the observed transformation width and over two orders of magnitude smaller than the melting field. We conclude that the effect of an intermediate-state type of structure on the magnetization signature is negligible.

On the other hand, the fact that the transformation is not completed until H_{app} has changed by a finite amount has important consequences for an induction measurement. Using Eqs. (2) and (A1), the quantity of experimental interest, $\Delta(B-H_{app})$, is given by

$$\Delta(B-H_{app})=4\pi(1-n)\Delta M. \tag{A2}$$

As discussed in Sec. II, in contrast to the magnetization signature, which is independent of the shape of the ellipsoid, this signature is strongly geometry dependent.

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