Equilibrium magnetization of high-T_c superconductors below the irreversibility line

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(Received 21 November 2002; published 14 March 2003)

By scaling isothermal magnetization data measured at different temperatures in the mixed state of high-T_c superconductors, we show that in some cases the sample magnetization, measured in increasing magnetic field below the irreversibility line, is identical with the equilibrium magnetization even in magnetic fields well within the irreversible regime. This surprising behavior can hardly be explained in terms of traditional models of vortex pinning in the bulk of the sample.

One of the specific features of the field-induced magnetization in high-T_c superconductors (HTSC's) is that there is an extended range of external magnetic fields H below the upper critical field H_{c2}, where the sample magnetization M is reversible,[1] i.e., where the values of M measured in either increasing or decreasing magnetic fields coincide. The lower boundary of this range is the so-called irreversibility line (IRL) in the H-T phase diagram and the values of M measured above the IRL represent the equilibrium magnetization M_{eq}. There is no reliable way to evaluate M_{eq} from the experimental data below the IRL without some additional knowledge about the pinning mechanisms in the particular sample under investigation. Although different varieties of the critical-state model are often used for the analysis of experimental results, their applicability is very rarely justified and, therefore, the results of those analyses are not reliable. For instance, the simplest and most widely used critical-state model of Bean is based on the assumption that the critical current density j_c is independent of the magnetic induction.[2,3] Experiments show, however, that j_c in HTSC's strongly depends on the applied magnetic field; i.e., the Bean model is not really valid for describing the critical state of these materials. It has also been demonstrated that the equilibrium magnetization curves derived from magnetization data obtained below the IRL by employing the Bean model do not really represent M_{eq}.[4] In this work, we demonstrate how a scaling procedure, recently developed in Ref. 5 (see also Refs. 6 and 7), may successfully be used for the analysis of experimental magnetization $M(H)$ curves below the IRL and how, as a consequence, important information concerning the effective pinning of vortices may be obtained.

The scaling procedure is based on the single assumption that the Ginzburg-Landau (GL) parameter κ is temperature independent. In this case, the magnetic susceptibility of a superconductor in the mixed state $\chi(H,T)$ is a universal function of $H/H_{c2}(T)$ and the relation between the magnetizations at two different temperatures T and T_0 may be written as

$$M(H/h_{c2},T_0)=M(H,T)/h_{c2}, \qquad (1)$$

with $h_{c2}=H_{c2}(T)/H_{c2}(T_0)$ being the normalized upper critical field. Considering real HTSC's, we also have to take into account the temperature-dependent paramagnetic susceptibility χ_n of the normal vortex cores which, according to Ref. 5, leads to the relation

$$M_{eff}(H/h_{c2},T_0)=M(H,T)/h_{c2}-c_0(T)H, \qquad (2)$$

with $c_0(T)=\chi_n(T_0)-\chi_n(T)$. Equation (2) implies that the field dependence of the sample magnetization $M(H)$ at a chosen temperature T_0 may be obtained from $M(H)$ curves measured at different temperatures. The collapse of these individual $M(H)$ curves onto a single master curve may be achieved by a suitable choice of $h_{c2}(T)$ and $c_0(T)$, the adjustable parameters of the scaling procedure. The scaling procedure is only applicable to magnetization data collected above the IRL. In this case, $M_{eff}(H)=M_{eq}(H,T_0)$. At the same time, once $h_{c2}(T)$ and $c_0(T)$ have been established in the chosen range of temperatures, the transformation given by Eq. (2) may also be applied to magnetization data measured below the IRL. Because of the onset of irreversibility, $M_{eff}(H,T_0)$ generally no longer represents $M_{eq}(H,T_0)$. However, as will be shown below, a surprising asymmetry of the M_{eff} curves, calculated from $M(H)$ data taken in increasing and decreasing fields, with respect to the equilibrium magnetization curve offers to achieve important conclusions concerning the effective pinning mechanism.

The condition that $\chi(H,T)$ depends only on the ratio $H/H_{c2}(T)$, which is the essential background of the scaling procedure, remains valid for any configuration of the mixed state. The vortices may form a vortex lattice, a vortex liquid, or, as has recently been proposed, a system of superconducting filaments embedded in the matrix of the normal metal.[7] This circumstance provides the possibility to use the scaling procedure even if there is a step in the $M(H)$ curves, marking the so-called first-order phase transition in the mixed state of HTSC's, which usually is attributed to the melting of the vortex lattice.[8–12] Although the vortex lattice melting represents a rather plausible hypothesis, to the best of our knowledge, there is no direct experimental evidence for this claim. For our discussion, however, the real nature of the phase transition does not need to be known. It is only important that in the H-T phase diagram there is a boundary $H_{PT}(T)$ between two possible configurations of the mixed state. In this case, at a fixed temperature and with increasing magnetic field, a phase transition leads from one configura-
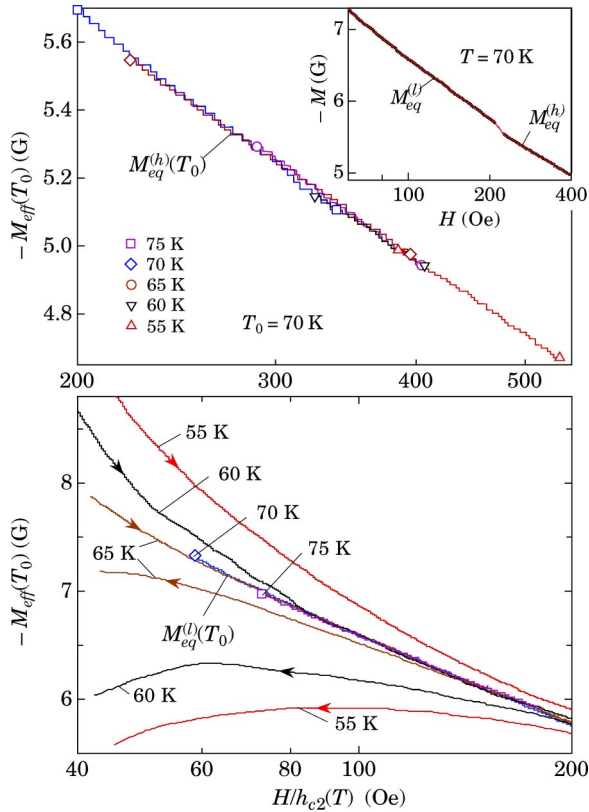
FIG. 1. $M_{eff}(H,T_0)$ for sample Bi-1 [original $M(H)$ data taken from Ref. 14], (a) above and (b) below the first-order transition. The open symbols mark the end points of the covered field ranges at the indicated temperatures. The inset illustrates the definitions of $M_{eq}^{(h)}$ and $M_{eq}^{(l)}$, taking the $M(H)$ curve at $T=70$ K as an example. The $M_{eff}(H,T_0)$ curves were calculated using Eq. (2) with $T_0 = 70$ K (see text).

FIG. 2. (a) $M_{eff}(H,T=70$ K) for sample Bi-2 (original data taken from Ref. 15), above the phase transition. (b) $M_{eff}(H,T=70$ K) below the phase transition. The $M_{eff}(H,T_0)$ curves were calculated using Eq. (2) (see text).

tion (low-field phase) to the other (high-field phase). By $M_{eq}^{(l)}(H)$ and $M_{eq}^{(h)}(H)$ we denote the equilibrium field-induced variations of M in the low-field and high-field phases, respectively. An example is shown in the inset of Fig. 1(a).[13] Of course, $M_{eq}^{(l)}(H)$ and $M_{eq}^{(h)}(H)$ do not coincide, but they both should scale with the same values of $h_{c2}(T)$ and $c_0(T)$. In this work we concentrate on the features of the magnetization curves distinctly above and below the phase transition. A detailed analysis of the magnetization very close to the phase transition will be published elsewhere.

Below we consider results of the magnetization measurements for three $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-1, Bi-2, and Bi-3) single crystals that were reported in Refs. 14, 15, and 16, respectively. In all three cases only the magnetization data from above the IRL were used to establish the parameters $h_{c2}(T)$ and $c_0(T)$.

Figure 1 shows $M_{eff}(H,T_0)$ data for the sample Bi-1. At $T \geq 60$ K the IRL line for this sample is substantially below $H_{PT}(T)$.[14] The scaled magnetization curves above the phase transition are depicted in Fig. 1(a). Because these data were collected above the IRL, the resulting curve in Fig. 1(a) represents the equilibrium $M_{eq}^{(h)}(H)$ curve for $T_0 = 70$ K. The magnetization data collected below the phase transition are shown in Fig. 1(b). The magnetization of this sample mea-
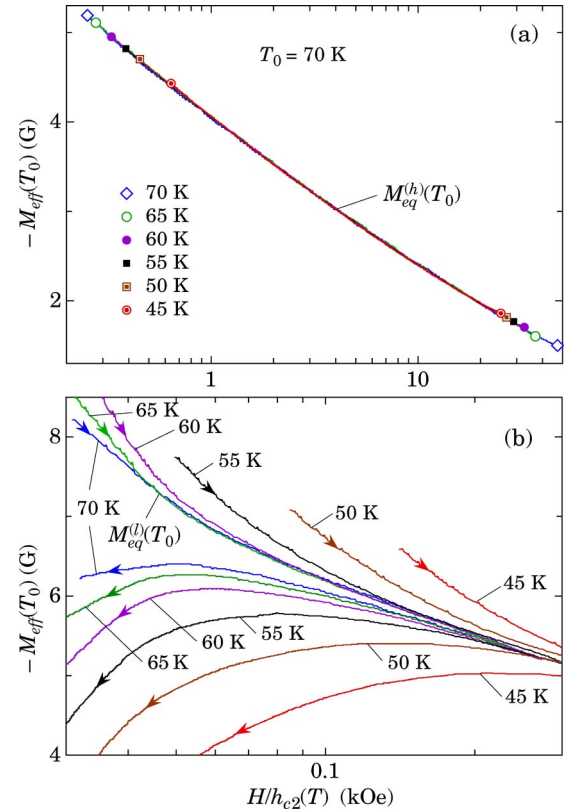
sured at temperatures of 70 K and 75 K is reversible in the entire covered range of fields and, therefore, the corresponding curves in Fig. 1(b) represent the $M_{eq}^{(l)}(H,T_0)$ curve. The merging of the individual $M(H)$ curves to $M_{eq}^{(h)}(H,T_0)$ and $M_{eq}^{(l)}(H,T_0)$, as displayed in Figs. 1(a) and 1(b), was achieved with the same values of $h_{c2}(T)$ and $c_0(T)$ on both sides of the transition, thus confirming our claim above. Although the magnetization at $T \leq 65$ K is irreversible in low magnetic fields, the $M_{eff}(H)$ curves calculated from the magnetization data measured in increasing magnetic field at 60 and 65 K merge into the equilibrium magnetization curve in magnetic fields considerably below the corresponding values of the irreversibility field H_{irr}. This is obviously not the case for $M_{eff}(H,T_0)$ calculated from $M(H)$ data taken in decreasing field, revealing an asymmetry of the magnetization process.

Analogous results for the sample Bi-2 are shown in Fig. 2. In contrast to the previous case, $H_{irr}(T)$ for sample Bi-2 is practically identical with $H_{PT}(T)$, marking the phase transition, at all temperatures.[15] Because the relative magnetic field range covered in Ref. 15 is extremely wide, accurate and reliable values of $h_{c2}(T)$ and $c_0(T)$ were obtained. As is demonstrated in Fig. 2(a), the scaling procedure results in a perfect overlap of the $M(H)$ curves above the IRL and deviations between the data measured at different temperatures are of the order of the width of the line. Figure 2(b) emphasizes the features of $M_{eff}(H,T_0)$ below the transition. Simi-
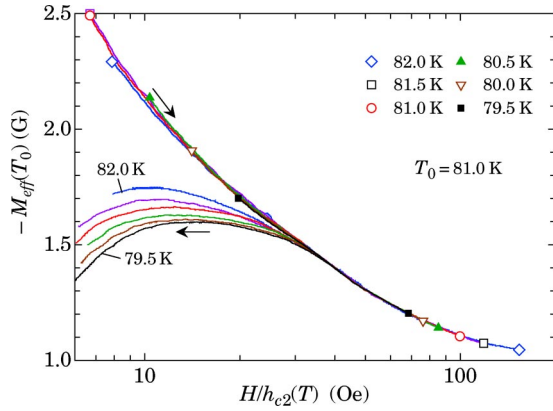
FIG. 3. $M_{eff}(H, T_0)$ curves calculated using Eq. (2) with T_0 = 81 K (see text) for sample Bi-3. Original $M(H)$ data are taken from Ref. 16. The symbols mark the end points of the covered field ranges at the indicated temperatures.

lar to the previous case, the $M_{eff}(H, T_0)$ curves calculated from the measurements in increasing field at $T \geqslant 55$ K coincide already in a magnetic field range which extends to substantially below H_{irr}. This is only possible if each of the coinciding parts of the curves is calculated from the equilibrium magnetizations at the respective temperatures. Again, due to irreversibility, the $M_{eff}(H)$ curves deviate from the equilibrium magnetization curve at lower temperatures, but these deviations are again noticeably smaller for the measurements made in increasing field than for those made with decreasing field.

A third set of data is shown in Fig. 3. For this plot we have chosen only the $M(H)$ data measured at several temperatures rather close to the critical temperature T_c. Although the first order phase transition clearly manifests itself on the magnetization curves at lower temperatures,[16] it is practically invisible in this high-temperature range. As may be seen in Fig. 3, the $M_{eff}(H)$ curves, calculated from the measurements in increasing field, all merge in the entire covered ranges of fields, thus clearly indicating that this curve represents the equilibrium magnetization curve for $T = T_0$.

The data shown in Figs. 1–3 demonstrate that the effect of pinning is strongly dependent on the direction of the flux motion. The pinning effects are obviously much weaker for the magnetic flux entering the sample. We are not aware of any model that explains this kind of pinning force asymmetry, if these forces are related to pinning centers in the bulk of the sample. A reasonable explanation for this type of behavior might be, however, that in these high quality samples, the intrinsic pinning is weak and the main obstacle for the magnetic-flux motion is a barrier near the sample edges, the so-called geometrical barrier. The existence of this type of barriers is actually known since the early studies of the intermediate state in type-I superconductors employing a magnetic powder technique.[17] These experiments have shown that the concentration of the normal phase in the intermediate state of type-I superconductors is considerably smaller near the sample edges.[18] It was immediately recognized that this happens because of the nonellipsoidal shape of the sample. Indeed, as is well known, if the magnetic susceptibility χ is

nonzero, the magnetic induction B is uniform only in ellipsoidal samples.[20] In superconducting samples this nonuniformity of B is magnified by a strong dependence of χ on the magnetic induction. The resulting distribution of shielding currents effectively pushes the normal domains in the intermediate state of type-I superconductors as well as vortices in the mixed state of type-II superconductors towards the center of the sample. It was also demonstrated that this edge barrier for the flux motion in type-I superconductors may substantially be reduced by proper shielding of the sample edges[21] or by altering the sample shape.[19] The importance of this edge barrier for correct interpretations of experimental data was also recognized for HTSC's.[22–24]

The geometrical barrier reaches its maximum height very close to the sample edges and the corresponding potential decreases only gradually towards the center of the sample.[22] This asymmetry of the potential profile implies the corresponding asymmetry of its effect on the vortex motion. The geometrical barrier naturally represents a stronger obstacle for the vortex motion out of the sample because it keeps the vortices at some considerable distance from the sample edges and, therefore, thermal activation is ineffective for the exit of vortices. Because of the proximity of the potential maximum to the sample edges, the thermally activated entrance of vortices is much more likely than their exit. This simple model explains why the data presented in Figs. 1–3 are consistent with the assumption that the pinning in the bulk of the sample is negligible and that the irreversibility of the magnetization is due to the mentioned geometrical barrier. Because the height of the geometrical barrier is strongly dependent on the shape of the sample edges, it may vary significantly from sample to sample.

In many experimental studies, including that of Ref. 15, the irreversibility line in the H-T phase diagram practically coincides with the line marking the first-order phase transition. The standard interpretation of this onset of irreversibility rests on the nonzero shear modulus of the vortex lattice, causing it to be much stronger pinned than the vortex liquid. This may well be true for the bulk pinning, but the shear modulus of the vortex lattice is irrelevant for the entry or exit of the vortices across the geometrical barrier. In other words, if the bulk pinning is weak compared to the pinning arising from the sample edges, which seems to be the case at least for our three examples and possibly many other HTSC's, the onset of the irreversibility at the mentioned phase transition does not necessarily follow from postulating the melting of the vortex lattice.

With all this in mind, we suggest an alternative cause for the occurrence of the first-order phase transition in the mixed state of HTSC's. As was argued in Ref. 7, it seems possible that in high enough magnetic fields, the mixed state is formed by a system of superconducting filaments embedded in the matrix of the normal metal instead of the formation of Abrikosov vortices. Upon reducing the magnetic field, the system of superconducting filaments loses its stability and must undergo a transition to the traditional mixed state consisting of Abrikosov vortices in a superconducting matrix. This transition requires a complete change of the topology of

the system, and although it is not exactly a first-order phase transition, its principal features will include the occurrence of a latent heat, a discontinuity in the magnetization, and hysteresis effects. It should be noted that the sample magnetization for a mixed state consisting of superconducting filaments is always reversible, independent of whether the filaments are pinned or not. In this case, of course, the geometrical barrier has no influence on the reversibility of the sample magnetization. The transition to the traditional mixed state with Abrikosov vortices changes this situation completely and, if the vortices are pinned, H_{irr} naturally coincides with the phase transition.[25] The same is true with respect to the sample resistivity. It is clear that, because there is no direct superconducting link between one electrode and the other for the system of superconducting filaments, the

sample resistivity should drop with the transition to Abrikosov vortices. The magnitude of this resistance jump depends on the strength of the vortex pinning and, for strong pinning, the sample resistance may vanish at the transition point.

As demonstrated above, in a number of cases $M(H)$ measured in increasing magnetic field coincides with the equilibrium magnetization curve even in magnetic fields well below the IRL, which is a strong evidence that the geometrical barrier arising near the sample edges is the main obstacle for the motion of magnetic flux. If this is indeed the case, the onset of irreversibility and the resistivity jump at the transition point do not necessarily follow from the hypothesis of the vortex lattice melting. In this sense we also promote an alternative scenario for explaining the first order phase transition in the mixed state of HTSC's.

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