Measurement of the irreversibility boundary of superconducting $YBa_2Cu_3O_7$ single crystals

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Results from dc-magnetization $M(T)$ measurements on $YBa_2Cu_3O_7$ single crystals (with the magnetic field **H** parallel to the c axis) near the irreversibility boundary in the H-T plane suggest a temperature dependence of the irreversibility boundary $H^*(T) \approx H_0^*(1-T/T_c)^{3/2}$. It is shown that the measurement of $M(T,H)$ at temperatures T below the irreversibility temperature $T^*(H)$ is seriously affected by a spatial inhomogeneity of the applied field H, if the sample has to be moved through a detection-coil system. An alternative procedure for the $H^*(T)$ detection by dc magnetometry without moving the sample is suggested.

Many recent experiments on superconducting layered copper oxides have revealed the existence of a distinct phase boundary in the magnetic phase diagram of the vortex state (Refs. 1–3 and many others). The so-called irreversibility line $H^*(T)$ separates two regions with distinctly different magnetic and resistive features. By crossing $H^*(T)$ from lower toward higher temperatures, magnetization curves become reversible,[1] and the critical current densities vanish.

In previous work, the temperature dependence of the irreversibility field $H^*(T)$ has been fitted near T_c according to

$$H^*(T)=H_0^*(1-T/T_c)^n \ , \quad T \to T_c \ , \qquad (1)$$

where H_0^* is a model-dependent fitting parameter.

Theoretical predictions for the exponent n depend on the model chosen for describing the behavior of the vortex ensemble. An exponent $n=\frac{3}{2}$ is predicted from a description in terms of "giant-flux creep",[2,4] while $n=\frac{4}{3}$ is expected from a theory based on a vortex-glass formation[5,6] as well as from Tinkham's giant-flux-creep model,[4] if, using the latter, a characteristic length $d \sim H^{-1/2}$ related with the vortex shifts upon depinning is postulated.[7,8] An exponent $n=2$ is found by assuming a flux-lattice melting according to a nonlocal elastic theory using a Lindemann-type melting criterion.[9]

In this work, we report on dc magnetometry results obtained on $YBa_2Cu_3O_7$ single crystals for magnetic fields **H** parallel to the c axes of the crystals. An alternative method for determining the irreversibility temperature $T^*(H)$ without moving the sample is suggested. The method is based on using the pickup-coil system in a second-derivative configuration of a commercial superconducting quantum interference device (SQUID) magnetometer. We compare these results with those of magnetization $M(T)$ measurements obtained with the standard measuring technique, where the sample is moved through an array of detection loops.

First, we performed conventional magnetization $M(T)$ measurements in constant external magnetic fields H in the region of interest within the H-T plane of the considered superconductor. For a fixed applied field H, all $M(T)$ curves should merge in the reversible temperature region, irrespective of the magnetic and thermal history of the sample. The use of large single crystals in such experiments makes it possible to verify irreversible contributions ΔM to the total magnetization M as a function of temperature with good resolution. This is not the case when small crystallites of only the size usually met in polycrystals are used. Nevertheless, even with improved resolution, establishing the irreversibility line from $M(T)$ curves obtained in the usual way may not be straightforward, as we shall demonstrate below.

The investigations were performed on $YBa_2Cu_3O_7$ single crystals which were grown from a CuO-BaO flux in an SnO_2 crucible.[10] The crystals exhibited inductively measured T_c values of 91.4 K and transition widths $\Delta T_c \approx 0.2$ K, defined by the temperature interval in which 90% of the total negative low-temperature susceptibility is reached (see inset of Fig. 1). This latter value was determined to be 30% of $-1/4\pi$ when the crystals were cooled in $H=30$ Oe. The size of the crystals used in the experiments ranges from approximately $2\times2\times0.3$ to $4\times4\times0.5$ mm^3. Figure 1 shows susceptibility $\chi(T)$ data near the transition to superconductivity, measured in $H=50$ kOe parallel to the c axis of the crystal. A linear-approximation procedure yields an estimate of the nucleation temperature T_{c2} (50 kOe) $=87.2\pm0.5$ K. The

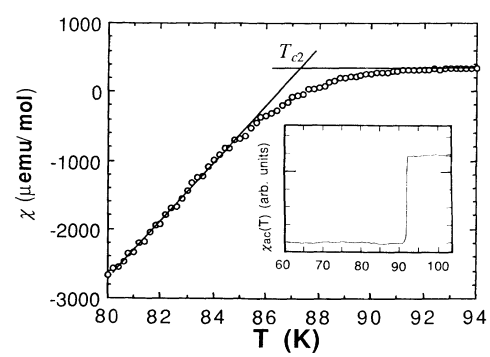

FIG. 1. Temperature dependence of the magnetic susceptibility $\chi(T)$, measured in an external field $H=50$ kOe parallel to the c axis of the crystal. The solid lines represent the linear approximation procedure to estimate the nucleation temperature T_{c2}. The normal-state susceptibility is $\chi_n \approx 330$ μemu/mol. The inductively measured transition to superconductivity in a field of a few Oersted is shown in the inset.

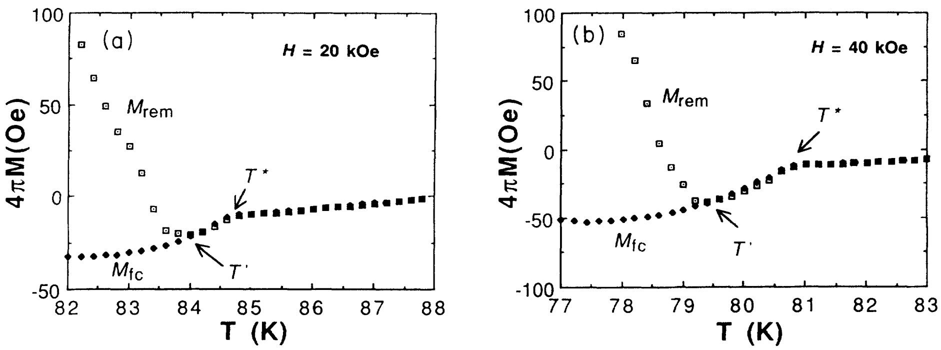

FIG. 2. (a) and (b) Representative $M_{rem}(T)$ and $M_{fc}(T)$ curves collected using a $YBa_2Cu_3O_7$ single crystal for external fields H parallel to the c axis. The magnetization values M have been calculated by the software of the magnetometer and do *not* necessarily represent the true values of M, namely at temperatures below T^* (see text). The temperatures T' and T^* (see text) are indicated by arrows.

rounding of analogous data near T_{c2} was previously observed by various authors[11] and can be ascribed to fluctuations of the order parameter near T_c. The resulting slope $\partial H_{c2}/\partial T = 12.2 \pm 1.5$ kOe/K is in quantitative agreement with the data in Ref. 12.

For all $M(T)$ measurements using the standard technique, a scan length of the sample of 2 cm through the magnet of a commercial SQUID magnetometer (Quantum Design, model MPMS) was used. In this range, the inhomogeneity $\Delta H/H$ is claimed to be smaller than $5\times 10^{-3}\%$. The magnetization M_{rem}, a superposition of a large remanent and a smaller reversible contribution, was obtained by cooling the sample from 100 to 20 K in $H=55$ kOe applied parallel to the c axis of the crystal. Then, a chosen measuring field H was set and, in order to avoid effects of possible temperature overshooting, the sample was warmed up to a temperature 2 K below the temperature from which the corresponding $M(T)$ scan was subsequently obtained by slowly increasing the sample temperature T. Each temperature scan was then performed using a step width $\Delta T=0.2$ K. A temperature overshoot of the order $\Delta T \approx 0.1$ K during temperature stabilization could not be avoided. For the subsequent field-cooled measurement to obtain $M_{fc}(T)$, the crystal was heated to temperatures above T_c and then cooled in the corresponding fields H from the normal state ($T=100$ K) to a temperature 2 K below the starting temperature of each $M(T)$ scan. For a *preliminary* analysis, the magnetization values M, calculated by the software of the magnetometer using the response curve of the pickup-coil system, were assumed to represent the true magnetization values $M_{rem}(T)$ and $M_{fc}(T)$. Typically obtained $M_{rem}(T)$ and $M_{fc}(T)$ curves for one crystal are shown in Figs. 2(a) and 2(b).

For each external field value H, $M_{rem}(T)$ and $M_{fc}(T)$ seem to merge at a well-defined temperature T'. A second, very well discernible feature in these data is a break in the slope of the magnetization $M(T)$ for all applied fields H at a temperature T^* exceeding T'. The coordinates $T^*(H^*=H)$ of this apparently abrupt change in magnetization $M(T)$ data define a "phase boundary" which is displayed in Fig. 3 (open squares).

In order to test the reliability of the magnetization $M(T)$ values calculated by the commercial software of the magnetometer, we performed additional investigations to be described below for $H=2.5$–50 kOe. The idea is to eliminate the possible influence of the magnetic-field inhomogeneity $\Delta H<2.5$ Oe on the crystal during a measuring cycle within the used scan length. For this purpose we fixed the crystal position at the center of the detection-loop system. The use of a so-called second-derivative loop configuration with three pickup coils makes it possible to detect a signal $V(x)$, which is related to the magnetic-flux difference

$$\Delta\Phi(x)=2\Phi(x)-\Phi(x-x_0)-\Phi(x+x_0)\ , \qquad (2)$$

where x_0 denotes the distance between the upper and the lower single turns and the counterwound two-turn center coil, Φ is the magnetic flux through each turn, and x is the distance between the sample and the symmetry plane of the detection-loop system. The value $\Delta\Phi$ is nonzero even if the sample *rests* at the center of the coil system (i.e., $x=0$). The corresponding response voltage V_0 from the electronics of the SQUID setup is therefore a measure of the *relative change* ΔM of the magnetization M of the *immobile* sample, although V_0 itself is not necessarily proportional to M. The detection of V_0 thus allows to qualitatively investigate the variation of the magnetization M of the sample without moving it through the slightly inhomogeneous field of the solenoid. Although

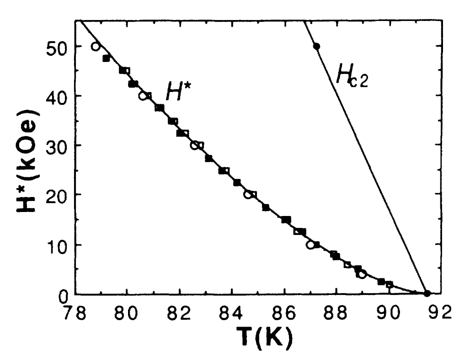

FIG. 3. Phase diagram of the superconducting state of a $YBa_2Cu_3O_7$ crystal for applied fields H parallel to the c axis. The open-square data points represent the H^* values determined from the apparent breaks in the slope of the $M(T)$ data (see text). A second corresponding data set of $H^*(T)$, taken on a different crystal of $YBa_2Cu_3O_7$, is represented by open circles. The solid squares represent the irreversibility temperatures T^* determined from an evaluation without moving the crystal (see text). The solid line corresponds to a least-squares fit according to Eq. (1). The $H_{c2}(T)$ line is a linear interpolation between the $T_{c2}(H)$ value determined from Fig. 1 (solid circle), and $T_c(H=0)$. Its slope is in quantitative agreement with the data in Ref. 12.

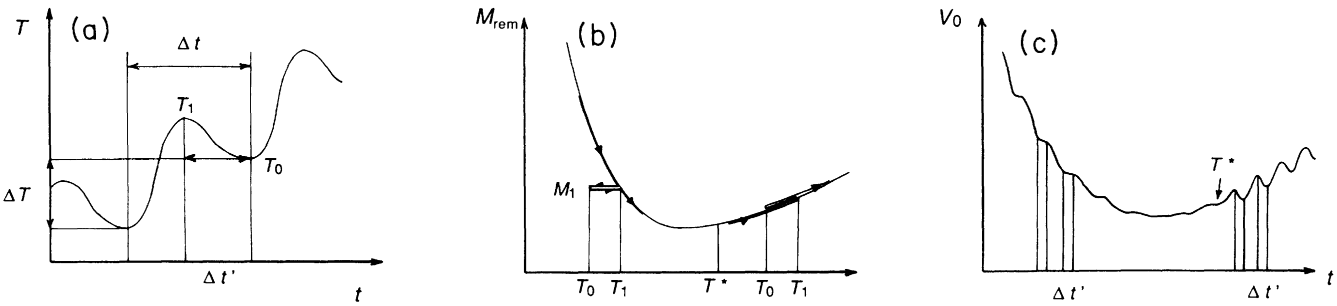

FIG. 4. (a) Schematical temperature vs time $T(t)$ characteristics used to obtain the voltage $V_0(t)$ curves shown in Fig. 5. The temperature increases steplike to an overshoot temperature T_1, and decreases again within the time $\Delta t'$ to approach the equilibrium temperature T_0. (b) Schematic sketch of a typical decay of the remanent magnetization M_{rem}, as a function of temperature T. Below T^*, the magnetization remains at a value M_1 defined by the overshoot temperature T_1, although the temperature T slightly decreases as a function of time to the equilibrium temperature T_0 [see Fig. 4(a)]. Above T^*, the magnetization M is a definite function of T and does not show any hysteretic behavior. (c) Typical signal vs time $V_0(t)$ characteristics in an experiment, according to Figs. 4(a) and (b). The voltage V_0 is a measure of the magnetization variation ΔM of the sample resting in the center of the detection-coil system (see text). Below T^*, the magnetization remains constant within $\Delta t'$ during the temperature equilibration after each temperature step ΔT. Characteristic small-amplitude oscillatory features in $V_0(t)$ appear above the transition to reversibility at T^*.

such measurements of ΔM can, in principle, be performed in magnetometers with simpler pickup-coil design (see, e.g., Ref. 13), the procedure described here offers the possibility to use the same experimental platform with identical magnet, thermometers, and sample holder as one uses for conventional magnetization M measurements.

We again induced a magnetization M_{rem} in the crystal which was previously fixed in the center of the coil system, in a way similar to that described above. Then, from a chosen starting value, the temperature was raised in steps of $\Delta T = 0.2$ K. A schematic sketch of the corresponding time evolution $T(t)$, showing also the unavoidable temperature overshoot after each step ΔT, is shown in Fig. 4(a). The temperature steps were chosen such as to take $\Delta t = 430$ sec, including the time $\Delta t'$ for temperature equilibration. The output voltage V_0 of the SQUID electronics was then recorded as a function of time. Some typically recorded $V_0(t)$ curves are shown in Fig. 5.

Our interpretation of the $V_0(t)$ characteristics is as follows: In the irreversible region of the H-T plane, the magnetization $M(T)$ decays with increasing temperature [see Fig. 4(b)]. However, when the sample temperature after a temperature step ΔT slightly decreases from the overshoot temperature T_1, to finally approach the equilibrium temperature T_0, the magnetization remains constant at a value M_1 defined by the amount of trapped flux at T_1 [see Fig. 4(b)]. This is reflected in the $V_0(t)$ characteristics by the occurrence of time intervals $\Delta t'$ with almost constant or only slightly varying response voltage $V_0(t)$ [see Fig. 4(c)]. At temperatures beyond the irreversibility boundary, however, the magnetization is reversible and thus well defined as a function of temperature. Sequences of temperature steps ΔT, overshoots to T_1, and subsequent equilibrations to T_0 will then appear in $V_0(t)$ [and $\Delta M(t)$, respectively], as an oscillating feature of small amplitude [see Fig. 4(c)]. The onsets of these small-amplitude oscillations in the experimental data of $V_0(t)$ (see Fig. 5) must therefore be interpreted as the respective transitions to reversible behavior. These temperatures, which coincide with the temperatures $T^*(H)$ deduced from the $M(T)$ data shown in Figs. 2(a) and 2(b), are also displayed in Fig. 3 (solid squares).

The obvious breaks in the slope at T^* in the preliminary $M_{rem}(T)$ curves, however, are not observed in the corresponding $V_0(t)$ data. The appearance of such a feature in the $M(T)$ data shown in Figs. 2(a) and 2(b) must therefore be interpreted as being due to an experimental artifact, which can be qualitatively explained as follows. Moving the sample in a slightly inhomogeneous magnetic field corresponds to the application of a super-

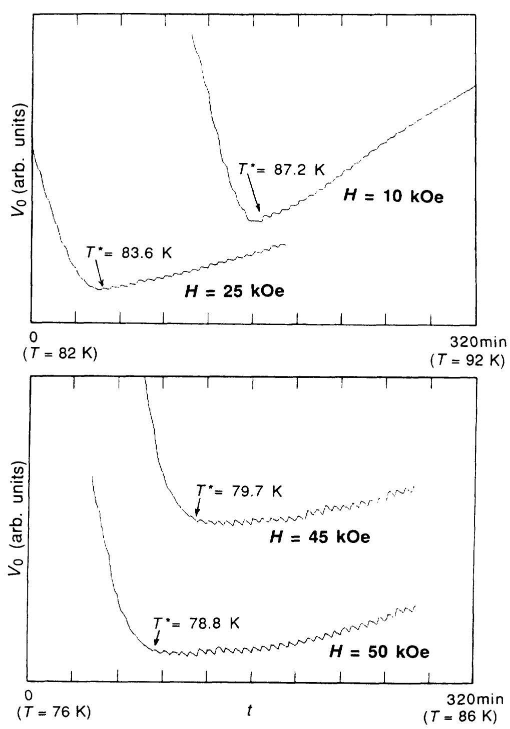

FIG. 5. Typically observed $V_0(t)$ characteristics. The signal V_0 is a measure of the relative change ΔM of the magnetization M of the sample, although V_0 is *not* proportional to M (see text). The $V_0(t)$ curves include a significant voltage-vs-time drift of the SQUID setup, which does not originate from the sample.

position of a large constant field and a small disturbing low-frequency field due to the field inhomogeneity ΔH. This latter field is expected to substantially change the magnetization M of the sample in an irreversible way *during the scan,* which will result in a distorted response curve of the pickup coils. From such a response curve, which does *not* correspond to an ideal response curve that one would obtain from a corresponding scan of a pointlike constant magnetic dipole moment, the software most likely calculates unreliable values for the magnetization M of the sample. The magnetization $M(T)$ data for temperatures T exceeding T^*, however, are expected to represent the true values of the reversible magnetization M of the single crystal, which is virtually constant during a measuring scan.

This interpretation strongly suggests that the measured $H^*(T)$, defined by the break in the slope of the $M(T)$ data [see Figs. 2(a) and 2(b)], i.e., the onset of "miscalculated" $M(T)$ data as described above, coincides with the true irreversibility boundary of the considered $YBa_2Cu_3O_7$ single crystal. The apparent merging of the magnetization $M(T)$ curves at temperatures T' below T^*, however, must be interpreted as an accidental coincidence due to the unreliable measuring technique of moving the sample in an inhomogeneous external magnetic field.

It seems natural to test our experimental data for $H^*(T)$ with respect to a fit using Eq. (1). We performed a least-squares fit to the data for $H^*(T)$ in a logarithmic-field scale. This procedure minimizes the *relative* error of H^*, while a conventional least-squares fit would give too much weight to the data points with high magnetic-field values and, therefore, temperatures T^* farthest apart from T_c. Although we realize that the applicability of Eq. (1) is only justified to experimental data near the critical temperature T_c (e.g., $0.9T_c < T \leq T_c$), it turned out that the resulting fit parameters T_c^*, n, and H_0^* essentially do not vary upon extending the fitting procedure to the full data set ($T > 0.85T_c$).

Taking into account all data for $H^*(T)$ of one crystal from Fig. 3, we obtain $H_0^* = 935 \pm 94$ kOe, $T_c^* = 91.31 \pm 0.13$ K, and $n = 1.46 \pm 0.05$ for the irreversibility field $H^*(T)$. The specified errors are 90% probability limits given by the fitting program. The open-circle data points in Fig. 3 represent $H^*(T)$ values that we obtained on a second $YBa_2Cu_3O_7$ crystal, but were not taken into account in the above calculation. The value of T_c^* coincides with the onset temperature of diamagnetism, $T_c \approx 91.4$ K, while the exponent $n \approx \frac{3}{2}$ has often been obtained in experiments probing the irreversibility boundary for different types of materials (e.g., Ref. 1). It is in agreement with the predictions of Tinkham's giant-flux-creep model,[4,6] but not with a vortex-glass hypothesis ($n = \frac{4}{3}$),[5,6] or the lattice-melting description using a Lindemann-type melting criterion ($n = 2$).[9]

In the strictly reversible region between $H^*(T)$ and $H_{c2}(T)$, experimental evidence for a further structure in the H-T phase diagram was reported by Chien, *et al.*[12] from resistivity and Hall-resistivity measurements. The authors claimed the existence of a field $H_k(T)$ in the so-called vortex-liquid state beyond the irreversibility line, which separates the in-plane resistivity $\rho_{ab}(T)$ characteristics of a $YBa_2Cu_3O_7$ single crystal in a strongly activated region ($H^* = H_g < H < H_k$) from a more diffusive one ($H_k < H < H_{c2}$). Furthermore, Crabtree and coworkers[14] reported the appearance of additional structures in resistivity $\rho(T)$ curves due to the presence of twin boundaries, depending on strength and orientation of the external magnetic field. This was interpreted as an occurrence of an "irreversibility line" specifically for twin-boundary pinning. Our dc magnetometry data, however, give no evidence for additional boundaries between $H^*(T)$ and $H_{c2}(T)$ within the H-T plane.

From our investigations on the irreversibility boundary $H^*(T)$ in $YBa_2Cu_3O_7$, we can conclude that the results from magnetometry in the static limit, namely the detection of magnetization $M(T)$ curves, yield reasonable results for the irreversibility boundary $H^*(T)$. Below the irreversibility temperature $T^*(H)$, however, magnetization M data, obtained by moving the considered sample in the magnetic field **H** of a solenoid, have to be carefully examined. The evaluation of magnetization values M can seriously be affected at temperatures below T^* by even small inhomogeneities of the applied field H, and may depend crucially on the algorithm used to extract the M values from the pickup-signal characteristics.

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