Irreversibility line of the $HgBa_2CaCu_2O_{6+\delta}$ high-temperature superconductors

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(Received 19 July 1993; revised manuscript received 13 September 1993)

Reversible and irreversible magnetizations (M) have been measured on a polycrystalline sample of $HgBa_2CaCu_2O_{6+\delta}$ (Hg 1:2:1:2) before and after oxygenation. For the preoxygenated sample, the superconducting transition temperature (T_c) is $\sim$114 K with a transition width of $\sim$2 K. The irreversibility line (H_{irr}) is approximated by a power law $H_{irr}\sim(1-T_{irr}/T_c)^n$ with $n\sim\frac{5}{2}$ between 0.1 and 5.5 T. For the oxygenated sample, T_c is $\sim$120 K with a width $\sim$4 K and the n for H_{irr} is almost the same. The values of n, which are $\sim\frac{3}{2}$ for Y 1:2:3:, $\sim\frac{5}{2}$ for Hg 1:2:1:2, and $\sim\frac{11}{2}$ for the Bi and Tl families, seem to correlate well with the distance between adjacent CuO_2 blocks. A scaling law $M\propto[1-(T/T^*)^2]F(H)$, is observed for reversible M up to a few degrees below T_c with a $T^*\sim112$ K before oxygenation. This suggests that Hg 1:2:1:2 is a quasi-two-dimensional system, where the effects of thermal fluctuations have been previously demonstrated.

I. INTRODUCTION

The study of the magnetic properties of high-temperature superconductors (HTS's) has resulted in the observation of many interesting phenomena, including the irreversibility line (H_{irr}),[1] which separates the irreversible region from the reversible region in the H/T phase space. H_{irr} sets the upper H/T limit for most applications of HTS. In the highly anisotropic HTS's, i.e., the two BiO_2- or TlO_2-layer compounds, the rather low H_{irr} severely limits practical applications. Since the Hg family has a shorter distance between neighboring CuO_2 blocks (not layers), a higher H_{irr} at the same reduced temperature is expected. However, its precise position is yet to be determined.

Many models, including thermal depinning,[2] flux-line lattice (FLL) melting,[3] and vortex-glass transition,[4] have been proposed to explain the H_{irr} in HTS's. Experimental data seem[1] to suggest that the nature of H_{irr} depends on the anisotropy of the superconductors.[1] The functional dependence of $H_{irr}(T)$ and the I-V characteristics near H_{irr} are rather different between the two extremes of strongly coupled layered $YBa_2Cu_3O_{7-\delta}$ (Y 1:2:3) and loosely coupled layered $Bi_2Sr_2CaCu_3O_{8+\delta}$ (Bi 2:1:2:2). Hg 1:2:1:2, with moderate CuO_2-block separation, lies between these extremes and serves as an excellent subject for further study of the nature of H_{irr}.

In this work, we report the magnetic properties of a polycrystalline Hg 1:2:1:2 sample before and after oxygenation. It has a transition temperature (T_c) of $\sim$114 K and a transition width (ΔT) of $\sim$2 K prior to oxygenation, and a $T_c\sim120$ K and a $\Delta T\sim4$ K afterward. We found that its H_{irr} approximates a power law $H_{irr}\sim(1-T_{irr}/T_c)^n$ with $n\sim\frac{5}{2}$, somewhere between those of Y 1:2:3 and Bi 2:1:2:2, for the preoxygenated Hg 1:2:1:2 and a similar n after oxygenation. The H_{irr} is substantially higher than that of the two BiO_2- and TlO_2-layer HTS's, suggesting a superior property for large-current applications. A scaling law $M\propto[1-(T/T^*)^2]F(H)$ with $T^*\sim112$ K before and after oxygenation is observed for reversible M over a large-T range. This scaling behavior is rather similar to Bi 2:1:2:2 (Ref. 5) due to a quasi-two-dimensional nature.

II. EXPERIMENTS

The technique for preparation of the polycrystalline Hg 1:2:1:2 used in this study is similar to that used in Ref. 6, which follows the two-step solid-state reaction technique. A precursor material with a nominal composition of $Ba_2CaCu_2O_5$ (not a compound) was prepared by calcinating and thoroughly mixing appropriate amounts of $BaCuO_2$ and CaO in an oxygen atmosphere for 16 h. The calcinated powder was then mixed with HgO powder and compacted into pellets which were sealed in evacuated quartz tubes. The tubes were, in turn, sealed in stainless-steel tubes. They were subsequently placed inside a furnace and slowly heated at a rate of $\sim$160 °C/h to 800 °C and maintained at this temperature for 9 h before being cooled to room temperature. Structure characterization was carried out by x-ray powder diffraction using a Rigaku D-MAX III powder diffractometer. Bar samples with dimension $3\times2\times6$ mm^3 were cut from these HBCCO pellets for electric and magnetic measurements. Resistance was determined using the standard four-lead technique with a LR-400 ac bridge operating at 16 Hz and current from 0.1 μA to 1 mA. A Quantum Design superconducting quantum interference device magnetometer was used to measure the magnetization.

III. RESULTS AND DISCUSSION

Figure 1 shows typical dc susceptibility as a function of T at a low field ($\sim$1 Oe) prior to and after oxygenation at

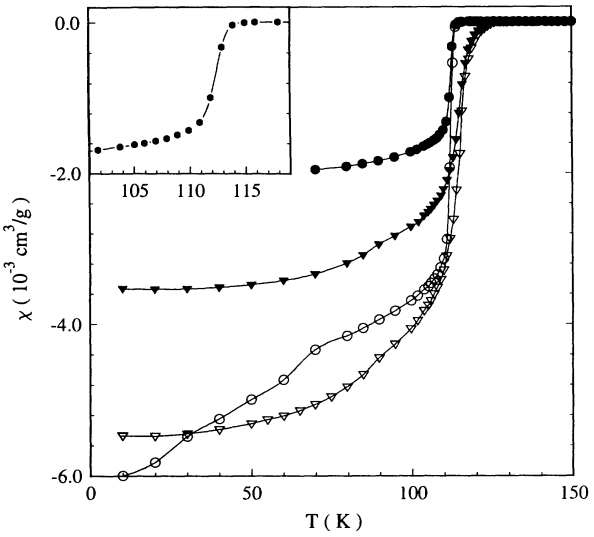

FIG. 1. dc magnetization as a function of temperature Hg 1:2:1:2 at low field (about 1 Oe): ● FC before oxygenation; ▼ FC after oxygenation; ○ ZFC before oxygenation; ▽ ZFC after oxygenation. Inset: An expanded view of the transition indicating the sharpness of the transition.

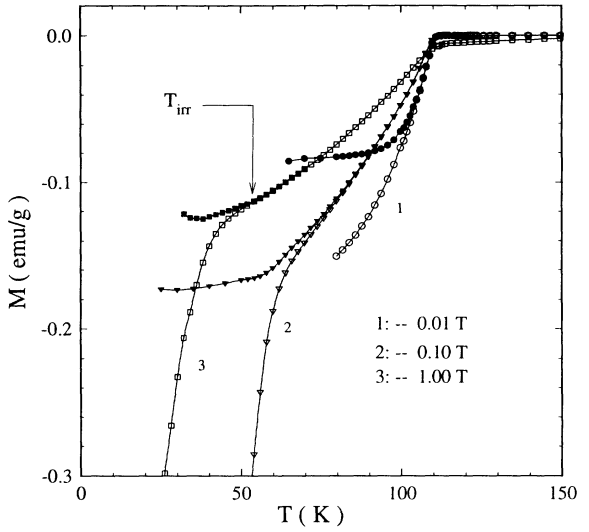

FIG. 2. ZFC and FC data of Hg 1:2:1:2 at 0.01, 0.1, and 1 T.

300 °C. Before oxygenation, the zero-field-cooled (ZFC) data at low T's corresponds to ~50% complete shielding and the field-cooled (FC) data corresponds to ~15%. Since, the ZFC data may overestimate the superconducting volume fraction due to the shielding effect and the FC data may underestimate the superconducting volume fraction due to the possible presence of a pinning effect, the real superconducting volume fraction in our sample is expected to be between 15 and 50 %. This is consistent with the x-ray data, which indicate that some minor quantities of impurity phases are present, including $CaHgO_2$, Hg 1:2:0:1, $BaCu_3O_{5.9}$, and $BaCuO_2$, in order of decreasing volume fraction. The narrow transition in our FC data (from 113.5 to 111.2 K, $\Delta T = \sim 2$ K) demonstrates how homogeneous the sample is. After oxygenation, the onset T_c shifts to ~120 K and ΔT is slightly broader. These are in agreement with our resistivity measurements.

At high fields, the paramagnetic contribution of the impurity phases is significant. In this experiment, the magnetization between 175 and 250 K at 5 T was fitted to the Curie law $\chi = \chi_0 + C/T$. The fitting parameters were used to subtract the background. As a verification, the magnetization at 0.2 T was compared with the calculated background, and no discrepancy was found within the experimental resolution. A set of typical ZFC and FC data at higher fields is displayed in Fig. 2 for the sample before oxygenation. The irreversibility temperature (T_{irr}) is defined as the deviation point between the ZFC and FC magnetization curves at a fixed field (Figs. 2 and 3). A T^2 dependence of the reversible M was also observed as shown in Fig. 4, indicative of the existence of fluctuations in Hg 1:2:1:2.

Since the discovery of H_{irr}, extensive work has been

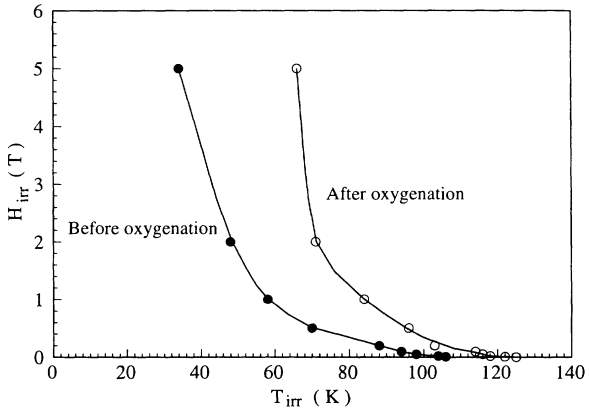

FIG. 3. The irreversibility line $H_{irr}(T)$ for Hg 1:2:1:2: ● before oxygenation, ○ after oxygenation.

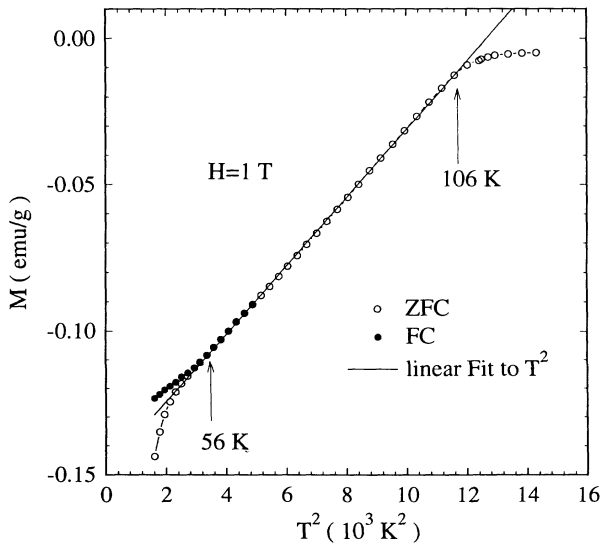

FIG. 4. The magnetization M at 1 T as a function of T^2.

carried out on various HTS's and low-temperature superconductors (LTS's). There exist some discrepancies in the H_{irr} positions. For example, the T_{irr} in Ref. 7 is about 10 K higher than that in Ref. 8 for the Bi 2:2:1:2 compound with similar T_c's. However, the functional dependence of H_{irr} on $1-T_{irr}/T_c$ is rather similar for the same compound and insensitive to its crystalline state and pinning strength. In fact, the measured magnetization for a random grain orientation only comes from the $H\|c$ components, as demonstrated in Ref. 9. Therefore, it is expected that the measured H_{irr} of a ceramic sample will be nearly the same as that of a grain-aligned samples with $H\|c$. This functional dependence has been used to identify vortex melting in Nb_3Sn,[10] Nb-Ti,[10] and Nb.[11] An approximate power law, $H_{irr}\sim(1-T/_{irr}/T_c)^n$ with $n\sim\frac{3}{2}$ is obtained for Y 1:2:3.[2] In the more anisotropic Tl- and Bi-based materials, the H_{irr}'s are far more complicated. At low fields (<0.1 T), an exponential law $H\sim\exp(1-T/T_c)$ was observed in Bi-2:2:1:2.[12] At high fields (>0.1 T), the H_{irr} can approximately be described as a power law within $n\sim5.5$. For the newly discovered Hg 1:2:1:2 prior to oxygenation, the H_{irr} obeys the power law with $n\sim2.5$, or $\frac{5}{2}$ between 0.1 and 5.5 T. Upon oxygenation, the value of n is almost the same. A similar n exists for our mixed-phase Hg-Ba-Ca-Cu-O sample displaying superconductivity up to ~140 K (Ref. 13) and single-phase Hg 1:2:0:1 with $T_c\sim94$ K (Ref. 14) (Fig. 5). It seems that n varies slightly within each family of Y-, Hg-, and Bi/Tl-based HTS's, but is very different from one family to another, e.g., $\frac{3}{2}$ for Y-HTS's, $\frac{5}{2}$ for Hg-HTS's, and $\frac{11}{2}$ for Tl/Bi-HTS's.

Flux dynamics depend strongly on anisotropy which, in turn, depends on coupling between CuO_2 blocks in the HTS (e.g., the block of CuO_2-Y-CuO_2 in Y 1:2:3 where the CuO chain is regarded as a pure charge reservoir).

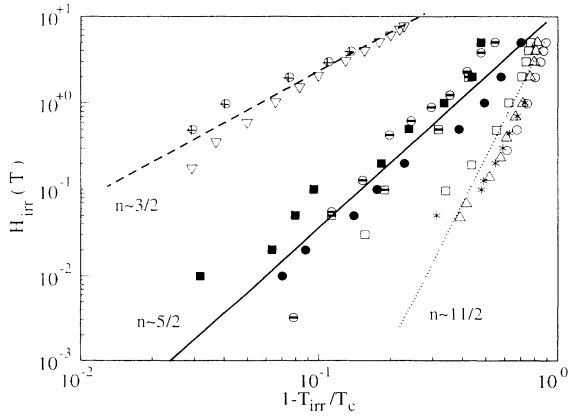

FIG. 5. The irreversibility line $H_{irr}(1-T/T_c)$ in log-log plot for several HTS's: ● Hg 1:2:1:2 before oxygenation; ■ Hg 1:2:1:2 after oxygenation; ⊟ Hg 1:2:2:3 (Ref. 13); ⊖ Hg 1:2:0:1 (Ref. 14); ▽ Y 1:2:3 single crystal (Ref. 21); ⊕ melt-textured Y 1:2:3 (Ref. 22); ⋆ Tl 2:2:2:3 (Ref. 23); □ Tl 2:2:1:2 (Ref. 24); ○ (BiPb) 2:2:2:3 grain-aligned polycrystal (Ref. 8); △ Bi 2:2:1:2 single crystal (Ref. 8).

The distance between two CuO_2 blocks can be used to characterize the coupling strength. It is 9.51 Å for Hg 1:2:1:2 which lies between those of Y 1:2:3 (8.245 Å) and Bi 2:2:1:2 (11.8 Å). This indicates that the coupling strength of Hg 1:2:1:2 is between those of Y 1:2:3 and Bi 2:2:1:2. Our observation suggests that there is a correlation between n and anisotropy and/or coupling. It should be noted that there are other factors, e.g., the carrier concentration in the oxygen deficient Y 1:2:3 (Ref. 15) and the defects in Sn-irradiated B 2:2:1:2 (Ref. 16), which also affects the H_{irr}/anisotropy.

Proposed theoretical interpretations for the nature of H_{irr} include thermal depinning,[2] FLL melting,[3] and vortex-glass transition.[4] For some LTS's, such as Nb film,[11] Nb_3Sn,[10] and Nb-Ti,[10] the H_{irr} is identified as the melting line by their flatter T dependence of H_{irr}. In Y1:2:3, experiental data on I-V scaling near H_{irr} suggests that the H_{irr} is a 3D vortex-glass transition line. For the highly anisotropic HTS's, such as Bi/Tl 2:2:1:2 and 2:2:2:3, the exponential H dependence of T_{irr} at low fields is interpreted as thermal depinning.[12] At higher fields, the H_{irr} is explained in terms of a thermally activated 3D-2D crossover[18] and glass transition.[19] In the 3D-2D crossover picture, the H_{irr} is expected to depend on the distance between CuO_2 blocks. Since that distance in Hg 1:2:1:2 is between those of Y 1:2:3 and Bi 2:2:1:2, it is expected that the H_{irr} of Hg 1:2:1:2 lies between those of Y 1:2:3 and Bi 2:2:1:2, as observed (Fig. 5).

We observed that the reversible M can be fitted by the products of two functions of T and H separately, such that $M(T,H)\sim M_0[1-(T/T^*)^2]F(H)$ with $T^*\sim112$ K for all fields studied. This separation of H and T dependence was previously observed in Bi 2:2:1:2 single crystals as well as Bi 2:2:2:3,[5] but not in Y 1:2:3. In the case of Bi 2:2:1:2/Bi 2:2:2:3, this factorization of the T and H dependence leads to an unphysical divergence of the Ginzburg-Landau parameter κ and H_{c2} near T^* and was suggested as a manifestation of the quasi-2D nature of Bi 2:1:2:2.[5] In this sense, Hg 1:2:1:2 is similar to the Bi-based HTS's and may also be quasi-2D. The reversible $M(T,H)$ was also fitted to the Hao-Clem model.[20] The distribution of the grain orientation and the uncertainty in the superconducting volume fraction makes complete data analysis difficult at this time. However, it was found that κ is mainly determined by the H dependence of M and is rather insensitive to the absolute value of M in the proposed variation model. Therefore, the value of $\kappa\approx100$ so extracted might still be valid. It should be pointed out that this extracted value of κ is, again, between those of Y 1:2:3 and Bi 2:2:1:2, just as might be expected based on their CuO_2-block distances.

In conclusion, magnetization has been measured in the reversible and irreversible regions of a high-quality Hg 1:2:1:2 polycrystalline sample before and after oxygenation. The H_{irr}'s of the sample in both the pre- and post-oxygenation states are much higher than those of Bi 2:2:1:2/Bi 2:2:2:3. An approximate power law $H_{irr}\sim(1-T_{irr}/T_c)^n$ with $n\sim\frac{5}{2}$ has been observed for the H_{irr} of Hg 1:2:1:2. The value of the index n for Hg 1:2:1:2 seems to be between those of Y 1:2:3 ($\frac{3}{2}$) and Bi/Tl

based HTS's (~5.5), as might be expected based on the CuO_2-layer coupling strength. Oxygenation shifts the H_{irr} significantly, but the functional dependence of H_{irr} on T remains the same. A T^2 dependence of reversible M was also observed. This can be attributed to a quasi-2D nature in Hg 1:2:1:2, as in the case of Bi 2:2:1:2 superconductor.

ACKNOWLEDGMENTS

This work is supported in part by NSF Grant No. DMR 91-22043, USAFOSR Grant No. F49620-93-1-0310, the State of Texas, the Texas Center of Superconductivity at the University of Houston, and the T. L. L. Temple Foundation.

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