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Enhanced Irreversibility by Crystal Defects in the Bi-Sr-Ca-Cu-O System

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Abstract

We measured magnetic irreversibility, $H^*(T)$, in liquid-quenched Bi-Sr-Ca-Cu-O samples with various microstructures at high applied field (up to 7 T). We found that $H^*(T)$ is considerably increased in the $\text{Bi}_2\text{Sr}_2\text{Ca}_3\text{Cu}_4\text{O}_x$ (2234) sample with a large amount of crystal defects, including severe lattice distortion and calcium and copper rich precipitates. We argue that the lattice distortion may affect the degree of anisotropy and the Josephson coupling between the Cu-O layers, which in turn enhance the irreversibility in the Bi-Sr-Ca-Cu-O system. A critical current density, J_c , of $8 \times 10^4 \text{ A/cm}^2$ at 8 kG and 40 K was obtained in the 2234 sample with many defects, while the J_c dropped significantly at the same temperature and field in the near stoichiometry $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ (2223) sample with a uniform microstructure.

Introduction

Irreversibility lines have been identified in both conventional and high- T_c superconductors (1-6). However, the reversible regime is more pronounced in the latter. Using zero-field-cooled (zfc) and field-cooled (fc) magnetization measurements, Yeshurun and Malozemoff identified irreversibility lines in both Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O single crystals, and they interpreted the irreversibility based on the classical Anderson-Kim thermally activated flux creep theory (2,7,8). In transport resistivity experiments, Palstra et al. (5,9,10) found a similar flux motion in both Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O single crystals. They pointed out that the broadening of the resistive transition tails was attributed to high thermal activation, which could be described by an Arrhenius law. These studies emphasized high thermal energy in the high- T_c system and concluded that the flux motion was dominated by thermally activated flux creep. With a different approach, Feigel'man et al. (11) developed a so-called collective-creep theory. In contrast to the Anderson-Kim flux creep model in which a linear logarithmic time decay of current should occur, the collective-creep model predicts that the time decay of current density, $J(t)$, is not logarithmic but is proportional to $[T/U \ln(t/t_0)]^{-1/\mu}$. Although these two models describe flux motion and current decay in different ways, their common physical ground is the thermally activated flux dynamics.

Several other models have also been proposed to interpret the irreversibility line in high- T_c superconductors. For example, M. P. A. Fisher (12) developed a vortex-glass model in which the vortex line freezes to a new vortex-glass phase at low temperatures. The model predicts a second-order phase transition at the glass transition boundary. Other models include the Josephson-coupling model (13) and flux entanglement (14).

Most recently, Ling , Shi, and Budnick (15) proposed a fundamentally different interpretation for the vortex behavior in type II superconductors, based on the concept of the self-organized criticality (SOC) (16). They pointed out that the flux motion in the vortex state should be spatially and temporally correlated, while only uncorrelated individual flux hoppings occur in the classical thermally activated flux-creep theory. SOC gives an entirely different physical picture of flux motion for type II superconductors, which has been shown to be related to the dynamics in the sandpile process (16).

In high- T_c superconducting systems, two major superconductors (Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O) exhibit quite different flux-creep effects and irreversibility lines in the vortex state (3,5). The irreversibility line in the Y-Ba-Cu-O lies in a much higher temperature and field regime close to H_{c2} boundary compared with the Bi-Sr-Ca-Cu-O system in the H vs. T phase diagram. It is not clear whether the irreversibility line is only intrinsically connected to the superconducting anisotropy and flux pinning in the system or whether it is related to the microstructure.

In this paper we present magnetic irreversibility data from a liquid-quenched Bi-Sr-Ca-Cu-O system. We show that flux pinning is strongly affected by microstructural changes, which in turn enhance the irreversible region in the system. The critical current densities were estimated for the samples with different microstructures. We discuss the possible correlation between the irreversibility lines and the microstructures.

Experimental Details

Starting materials of Bi_2O_3 , SrCO_3 , CaCO_3 , and CuO were weighed in the atomic proportions of 2:2:2:3 and 2:2:3:4. The weighed powders were thoroughly mixed by a ball mill in an agate container for 2-3 h. After ball milling, the mixed powders were dried and calcined in air at 800°C for 20 h.

The calcined powders were melted in a platinum crucible and splat quenched between two copper blocks preheated at 200°C. Extremely dense glass sample plates with dimensions of 50mm × 40mm × 0.5mm were formed for subsequent crystallization annealing. The glass samples with the 2:2:2:3 (2223) and 2:2:3:4 (2234) nominal compositions were annealed in air at 870°C for 24 h and slowly cooled to room temperature. Detailed information about the sample preparation can be found elsewhere (17,18).

The annealed 2223 and 2234 samples chosen for the magnetization measurements had the same dimension of 1.7mm × 1.1mm × 0.5mm. Magnetic hysteresis data were taken with a Princeton Applied Research 4500 vibrating sample magnetometer. The sample was cooled to a temperature below T_c in a zero magnetic field. The field was then continuously increased for the magnetic hysteresis measurements.

Results

Our resistivity and magnetization measurements and x-ray diffraction data show that the 2223 and 2234 samples have mostly the 85 K superconducting phase with different amounts of calcium- and copper-rich impurities. Our transmission electron microscopy (TEM) and x-ray diffraction experiments showed that the 2223 sample had a nearly single 85 K phase, with an extremely small amount of calcium- and copper-rich precipitates (17,18). In contrast, we observed a large amount of finely dispersed calcium- and copper-rich precipitates in the 2234 sample matrix (19). With high-resolution TEM, we also identified high density of lattice defects in the 2234 sample, while a nearly perfect lattice structure was observed in the 2223 sample. Our magnetic hysteresis data indicated that these microstructural changes strongly affected the flux pinning strength of the crystallized samples. As shown in Fig. 1, the magnetization curves of the 2223

and 2234 samples taken at 40 K exhibit a large difference in the magnetization.

Figure 2 shows the hysteresis difference, ΔM , vs. field, H at 25 K (Fig. 2a), 32 K (Fig. 2b), and 40 K (Fig. 2c). As can be seen in this figure, the hysteresis width decreases with increasing field differently in 2223 and 2234 samples. For example, at 40 K, ΔM drops to a extremely small value below our VSM's sensitivity near 8 kG, while about 1.22 emu/cm^3 of ΔM remains at the same field for the 2234 sample. By applying the Bean critical state model (19,20), we estimate the critical current density (J_c) at 40 K for these samples. As shown in Fig. 3, at 40 K, J_c reaches a value of $8 \times 10^4 \text{ A/cm}^2$ at 8 kG in the 2234 sample, but is significantly reduced at the same field for the 2223 sample.

In Fig. 4, we plot the irreversibility line, $H^*(T)$, for both 2223 and 2234 in the H vs. T phase diagram. The irreversibility line is determined by the hysteresis curves at various temperatures. The boundary values are obtained by a criterion of $\Delta M \sim 1 \times 10^{-5} \text{ emu}$. As shown in Fig. 4a, the irreversibility line of the 2234 sample is considerably shifted up to the high temperature and field region. For comparison, we also show the irreversibility line for a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ (SC 2212) single crystal with the field parallel to the c axis. It can be seen that the irreversibility lines of SC 2212 and 2223 coincide at low temperature and high field regions ($T < 20 \text{ K}$, $H > 40 \text{ kG}$). However, at relatively high temperature region ($T > 20 \text{ K}$), the irreversibility line of 2223 is considerably higher than that of SC 2212. If we consider the irreversibility line of the 2212 crystal to be determined by the "pure" intrinsic structure, the enhanced irreversibility in the 2234 sample must be associated with the microstructure and crystal orientation, which are extrinsic to the materials. In Fig. 4b, we plot $H^*(T)$ in the logarithmic scale against $1 - t$, where $t = T/T_c$. In this figure, the shift of the irreversibility line can also be seen clearly as a

result of differences in microstructure and crystal orientation. For instance, the difference in $H^*(T)$ between 2223 and SC 2212 is mainly associated with the crystal orientation, whereas precipitate pinning causes the further shift of the irreversibility line in the 2234 sample.

Discussion

It has been reported that the degree of resistive broadening effect in high- T_c oxides is proportional to anisotropy of the materials. The considerably low values of $H^*(T)$ in the Bi-Sr-Ca-Cu-O and Tl-Ba-Ca-Cu-O systems are associated with high superconducting anisotropy (13,21). In the $YBa_2Cu_3O_x$ (123) system, the Cu-O layers are strongly coupled as a result of closer interlayer spacing and the vortices exhibit three-dimensional behavior. Therefore, the transport resistive broadening under a magnetic field is not severe in 123, and consequently the irreversibility line can extend to a region near the H_{c2} boundary. However, the Cu-O layers are weakly coupled in the Bi-Sr-Ca-Cu-O and Tl-Ba-Ca-Cu-O systems. Because of a slight misalignment and crystal defects along the a-b plane, the superconducting currents are established only through tunneling between the Cu-O layers. Thermal fluctuation and applied field can both strongly affect the tunneling process, thus causing strong resistive broadening and reversible magnetization. This fact is also evidenced in the transport measurements, where the Tl- and Bi-based thin films exhibit a huge anisotropy of H_{c2} as a result of an extremely small coherence length along the c-axis (22).

Because of the relatively isolated Cu-O layers in the Bi-Sr-Ca-Cu-O and Tl-Ba-Ca-Cu-O systems, vortices are predicted to be pancake-like, and the flux motion in adjacent layers is independent of each other (23). The two-dimensional vortices may cause significant flux creep, as the field is parallel to the c axis and any defect pinning is also greatly reduced. According to the

anisotropy and Josephson-coupling arguments, it seems that $H^*(T)$ would be intrinsically determined by the superconducting parameters of the materials.

However, it has been clearly observed in this experiment that the irreversibility line in the Bi-Sr-Ca-Cu-O system is dependent on the microstructure. This result agrees well with previously reported data that the irreversibility line is also strongly affected in neutron-irradiated 2212 single crystals (24). As we pointed out earlier, the 2234 sample has mostly the 85 K phase with a high density of defects (calcium and copper-rich precipitates) in the matrix. More important, we also observed a great amount of lattice defects in the 2234 samples (19). A typical one is a highly distorted lattice structure. We found that the lattice varied about 8% between two local regions separated by approximately 50 nm. Interestingly, this severe lattice distortion did not result in any suppression in T_c , but contributed greatly to strong flux pinning. In contrast, very uniform lattice structure was observed in the 2223 sample, and practically no calcium and copper-rich precipitates were found. We believe the lattice distortion and the finely distributed precipitates are both responsible for the enhanced $H^*(T)$ and increased critical current density. If the Josephson coupling argument is valid, the high lattice distortion may affect the anisotropy and coupling between the Cu-O layers. Clearly, the coupling becomes stronger when the distance (lattice constant) between the Cu-O layers is shortened as a result of local lattice deformation. The calcium and copper rich precipitates have poor crystal coherency with the 85 K phase structure and, therefore can deform the lattice. The mechanism of forming these lattice defects has been previously reported (19). The calcium and copper-rich precipitates have extremely small dimensions ($< 0.01 \mu\text{m}$) and are finely dispersed in the sample matrices. Even when the field is parallel to the c axis, these precipitates can act as strong flux-pinning centers,

especially at high magnetic fields. It should be noted that $H^*(T)$ can be further enhanced as the microstructure is optimized by adding appropriate amounts of calcium and copper into the materials and adjusting the annealing procedures.

We attempted to fit the experimental $H^*(T)$ values of SC-2212 with the flux-lattice melting (FLM) model proposed by Houghton, Pelcovits, and Sudbo (25). However, the fitting gave poor agreement between the experimental data and the flux-lattice melting model (Eq. 3.9 of ref.25). For example, with $c = 0.4$ in the Lindemann melting criterion (as obtained for $\text{YBa}_2\text{Cu}_3\text{O}_x$ and $\text{Bi}_{2.2}\text{Sr}_2\text{Ca}_{0.8}\text{Cu}_2\text{O}_8$ single crystals in ref. 25), we found the Ginzburg-Landau parameter, κ , varied from 100 to 160 as the temperature changed from 20 K to 50 K. Further, the FLM predicts a melting behavior associated only with the intrinsic superconducting parameters, while the $H^*(T)$ observed in this study clearly depends upon the microstructure.

Conclusion

We conclude that the irreversibility line can be affected by the microstructure in the Bi-Sr-Ca-Cu-O system. The enhanced $H^*(T)$ and critical current density in the 2234 sample indicate that a high density of crystal defects (lattice distortion and precipitate) greatly increases the pinning strength. The enhancement of $H^*(T)$ may also be related to the changes in anisotropy of the material and strengthened coupling between Cu-O layers.

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Figure Captions

- Fig. 1 Magnetization curves obtained at 40 K for the liquid quenched 2223 and 2234 samples
- Fig. 2 Magnetic hysteresis difference, ΔM vs. applied field, H , for the 2223 and the 2234 samples, at (a) $T = 25$ K, (b) $T = 32$ K, and (c) $T = 40$ K.
- Fig. 3 Critical current density vs. field at 40 K for (a) the 2223 sample and (b) the 2245 sample.
- Fig. 4 (a) $H^*(T)$ versus T , and (b) $H^*(T)$ versus $1-t$, where $t = T/T_c$ (The solid lines are the a guide for the eye), determined by the magnetic hysteresis loops for the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ single crystal, the 2223 and 2234 samples.

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