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Anisotropy of the irreversibility lines for c-axis-aligned $HgBa_2Ca_2Cu_3O_{8+\delta}$ powders

Y. S. Song,* M. Hirabayashi, H. Ihara, and M. Tokumoto
Electrotechnical Laboratory, 1-1-4 Umezono, Tsukuba, Ibaraki 305, Japan

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Anisotropic irreversibility lines due to thermal fluctuation for quasi-two-dimensional high-T_c superconductors (HTS's) were observed in c-axis-aligned powders of the $HgBa_2Ca_2Cu_3O_{8+\delta}$ compound with $T_c=131\pm0.5$ K. The anisotropic ratio $H_{\rm irr}(\perp c)/H_{\rm irr}(\|c)$ below 2 was observed in all temperature regions. The simple three-dimensional-like power law $H_{\rm irr}=a(1-T/T_c)^n$ was observed only in the low-field region (≤ 2000 G) with $n=1.40\pm0.1$ for $H\perp c$ and $n=1.63\pm0.1$ for $H\|c$. In the higher field region up to 5 T, the temperature dependence of $H_{\rm irr}(T)$ lines changes into a two-dimensional-like exponential function $H_{\rm irr}=a\exp(-T/T_0)$ due to the breakdown of the interlayer coupling of the conduction channel, which consists of three Cu-O planes, with $T_0=16.0\pm1.3$ K for $H\perp c$ and $T_0=16.6\pm1.1$ K for $H\|c$. The exponent n is independent of the pinning strength or anisotropy related to interlayer coupling between CuO_2 planes. The characteristic crossover field giving deviation from the power function depends strongly on the anisotropic nature of materials; e.g., ≥ 5 T for Y HTS's, ~ 0.2 T for Hg HTS's, ~ 0.02 T for Bi HTS's, and ~ 0.01 T for Tl HTS's; this means that flux dynamics depend strongly on coupling strength between CuO_2 blocks in the HTS.

I. INTRODUCTION

Many experiments on superconducting layered copper oxides have revealed the existence of a distinct phase boundary in the magnetic phase diagram of the vortex state region between the lower critical field $H_{c1}(T)$ and the upper critical field $H_{c2}(T)$, which is due to the short coherence lengths and the weak interlayer coupling. This irreversibility line $H_{\rm irr}(T)$, which was first observed in $La_{2-x}Ba_xCuO_{4-y}$ by Müller, Takashige, and Bednorz,[1] separates two regions with distinctly different magnetic and resistive features. By crossing $H_{\rm irr}(T)$ from lower toward higher temperatures, the magnetization curve becomes reversible and the critical current densities vanish. Based on the observation that $H_{\rm irr}$ depends on T as $H_{\rm irr}(T)=a(1-T/T_c)^{3/2}$, they suggested the existence of a superconducting glass state.

After that, numerous results on the temperature dependence of the irreversibility field $H_{\rm irr}(T)$ have been fitted near T_c according to $H_{\rm irr}(T)=a(1-T/T_c)^n$ with the exponent n varying from 1.3 to 2.[2-8] But serious deviation from linear fitting in the ln $H_{\rm irr}$ versus ln $(1-T/T_c)$ plot starts to appear from $(1-T/T_c)\sim 0.2$ in all high-T_c materials except alkali-metal-doped C_{60} (Ref. 5) with an isotropic structure in which the deviation from the power function does not appear up to $(1-T/T_c)\sim 0.5$. The deviation from the power law indicates that other forms of temperature dependence in the high-field region are required. Proper functional forms in all temperature regions are still rather controversial; for example, the reported $H_{\rm irr}(T)$ varies in one case as $H\sim(1-T/T_c)^n$ for $H\leq 200$ G and $\exp(-T/T_0)$ for $H\geq 200$ G in the aligned powder sample[9] and randomly oriented sample[10] of $(Bi,Pb)_2Ca_2Sr_2Cu_3O_{10+\delta}$ and in another as $H\sim\exp(T_0/T)$ for $100\ {\rm G}<H<1000$ G in $(Bi,Pb)_xCa_2Sr_2Cu_3O_{10+\delta}$.[11]

On the other hand, there are several reports on the irreversibility lines for Hg 1201,[12,13] Hg 1201 before and after neutron irradiation,[14] Hg 1212 before and after oxygenation,[15] and Hg 1223.[16,17] In a recent report by Huang *et al.*,[15] they surmised that the irreversibility lines of Hg-based high-temperature superconductors (HTS's) lie between those of Y- and Bi/Tl-based HTS's and that n in the power function varies slightly within the 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 Bi/Tl HTS's.

Although the existence of an irreversibility line is well established, its underlying physics is being confused. A wide variety of models has been proposed including the Josephson-coupled glass model,[1] giant flux creep,[18] vortex lattice melting,[19] vortex glass melting,[20] and transitions within an entangled flux liquid state.[21] None of these models is yet developed well enough to provide clear quantitative predictions for the various experimental observations, and all have problems explaining some for the existing results.

In our previous study,[22] we presented the temperature dependence of the magnetic susceptibility for the c-axis-aligned Hg 1223 sample field cooled and zero-field cooled with a low field of 30 G parallel and perpendicular to the c axis. A superconducting transition temperature T_c of 131 ± 0.5 K was observed for this sample, and the Meissner and shielding factors of ~ 0.40 and ~ 0.47 for $\mathbf{H}\|c$ and ~ 0.13 and ~ 0.15 for $H\perp c$ have been obtained using the x-ray density $\rho=5.8$ g/cm^3 and powder mass $m=35.8$ mg. Also, we have presented the anisotropic ratio of about 3.0.

Now, to provide additional information regarding the motion of the flux lines, we will report on the observation and detailed examination of the temperature dependence of the anisotropic irreversibility properties for the c-axis-aligned powders of the $HgBa_2Ca_2Cu_3O_{8+\delta}$. This result is

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compared with the various functional forms for the $H_{\mathrm{irr}}(T)$ line reported in all high-T_c superconductors.

II. EXPERIMENTAL DETAILS

The source material for high-pressure synthesis was a mixture of precursor materials of $Ba_2Ca_2Cu_3O_7$ and yellow HgO. The precursor materials were prepared by calcining a well-ground mixture of $BaCO_3$, $CaCO_3$, and CuO powders with a nominal composition at 930 °C for 20 h in O_2. After regrinding and mixing with yellow powdered HgO, the pressed pellets were sealed in a gold capsule of 4 mm diameter and 6 mm length. The sample capsule was heated in an internal graphite tube heater at 850 °C for 1 h under a pressure of 5 GPa. The sample was subsequently quenched to room temperature before the pressure was released. Finally, the sample was annealed at 300 °C for 5 h in flowing oxygen in a similar way as in Ref. 23.

To obtain the *c*-axis-aligned powder sample, the method of Farrell *et al.*[24] was employed. The sintered single-phase superconducting pellets were ground to a powder with an average microcrystalline grain size of 2–3 μm, mixed with monomers of epoxy resin (Cemedine EP-007) in a Teflon tube of diameter 4 mm with a typical powder to epoxy ratio of 2:3 and then aligned for 14 h in a 7-T Bruker superconducting magnet at room temperature using anisotropic normal-state magnetic susceptibility. The degree[25] of *c*-axis alignment is checked from the intensities of the tetragonal (00*l*) line of the x-ray-diffraction (XRD) patterns obtained using a Rigaku RINT 1000 diffractometer with monochromated Cu $K\alpha$ radiation. We could confirm the anticipated *c*-axis orientation along the applied field at room temperature under ambient pressure.

Magnetic measurements were performed on a Quantum Design superconducting quantum interference device (SQUID) magnetometer with conventional procedures to be described in more detail elsewhere.[5] Standard zero-field-cooling (ZFC) and field-cooling (FC) measurements as a function of temperature were performed for both directions under different magnetic fields up to 5 T.

III. RESULTS AND DISCUSSION

The irreversibility temperature T_{irr} for the aligned powder sample $HgBa_2Ca_2Cu_3O_{8+\delta}$ is obtained from the merging point for the FC and ZFC curves of the temperature dependence of the magnetization, $M(T)$. But as shown in Fig. 1, the determination of the irreversibility temperature has considerable ambiguity because the ZFC and FC curves approach each other asymptotically as a function of temperature. To remove such ambiguity, the trace deduced from the magnetic susceptibility ratio $\chi_{\mathrm{ZFC}}(T)/\chi_{\mathrm{FC}}(T)$ of the ZFC and FC curves at consistent temperature using an interpolation was drawn with a solid line from the low-temperature side and the intersection between the line and $\chi_{\mathrm{ZFC}}(T)/\chi_{\mathrm{FC}}(T)=1$ was defined as $T_{\mathrm{irr}}(H)$. The temperature dependences of the magnetic susceptibility ratio, $\chi_{\mathrm{ZFC}}(T)/\chi_{\mathrm{FC}}(T)$ in various applied fields parallel to the aligned *c* axis are shown collectively in Fig. 2.

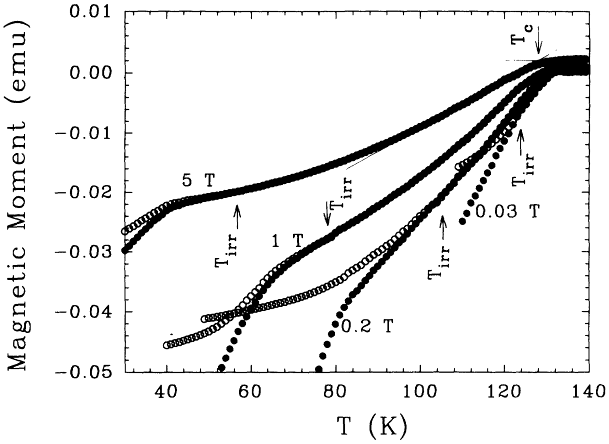

FIG. 1. Temperature dependence of the magnetic moments *m*(ZFC) and *m*(FC) for an aligned powder of Hg 1223 ($m = 35.8$ mg) in various applied fields parallel to the *c* axis.

The irreversibility lines $H_{\mathrm{irr}}(T)$'s for $HgBa_2Ca_2Cu_3O_{8+\delta}$ with applied field parallel and perpendicular to the *c* axis are shown in Fig. 3. The large reversible region in the *H*-*T* plane indicates very low flux pinning for this *c*-axis-aligned powder sample, which is similar to the published data of Hg-based materials.[12–17] Two lines for the lower critical field H_{c1} (dashed line) (Ref. 26) and upper critical field H_{c2} (dashed and dotted lines) (Ref. 27) are anisotropic in nature and are reference guides. The anisotropic ratio $H_{\mathrm{irr}}(\mathbf{H}\perp c)/H_{\mathrm{irr}}(\mathbf{H}\parallel c)$ below 2 was observed in all temperature regions we measured. These values are smaller than those of the more anisotropic systems such as Tl- and Bi-based materials.[9]

The irreversibility lines of $H_{\mathrm{irr}}(\mathbf{H}\perp c)$ and $H_{\mathrm{irr}}(\mathbf{H}\parallel c)$ for *c*-axis-aligned powder samples of $HgBa_2Ca_2Cu_3O_{8+\delta}$ are shown in the logarithmic plot of the H_{irr} versus $(1-T/T_c)$ (Fig. 4). Linear behavior in the low-field re-

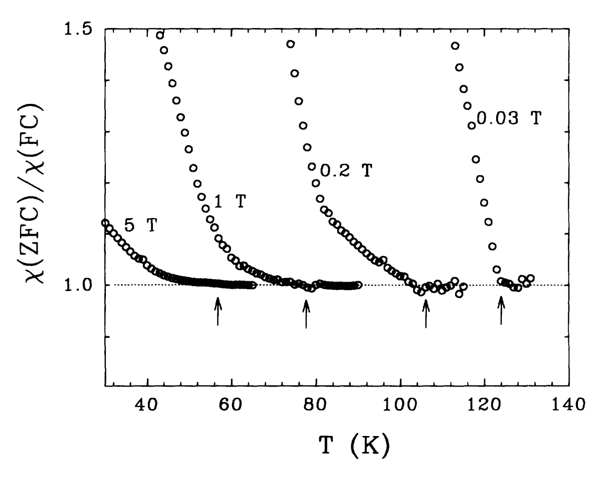

FIG. 2. Temperature dependence of the magnetic susceptibility ratio χ(ZFC)/χ(FC) for an aligned powder of Hg 1223 in various applied fields parallel to the *c* axis.

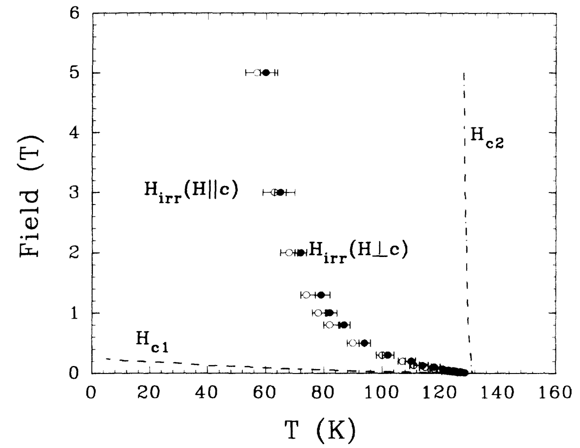

FIG. 3. Anisotropic irreversibility lines $H_{irr}(T)$ in the H-T plane for Hg 1223. The lower critical field H_{c1} (dashed line) (Ref. 26) and upper critical field H_{c2} (dashed and dotted line) (Ref. 27) are reference guides.

gion below 0.2 T indicates that both lines can be accurately fitted by the simple power law

$$H_{irr} = a(1 - T/T_c)^n ,$$

with $n = 1.40 \pm 0.1$, $a = 1.64 \pm 0.36$ T for $\mathbf{H} \perp c$ and $n = 1.63 \pm 0.1$, $a = 2.75 \pm 0.5$ T for $\mathbf{H} \| c$. The power value $n \sim 1.5$ for the $HgBa_2Ca_2Cu_3O_{8+\delta}$ compound is comparable to the value observed for all high-T_c superconductors, which is the same as predicted by the giant flux creep model.[18]

In a report by Civale *et al.*,[28] a systematic shift in the constant a in the power law for $H_{irr}(T)$ was observed due to creation of a pinning center by proton irradiation. [Note that these authors claimed no variation in $H_{irr}(T)$ after proton irradiation for single-crystal Y-Ba-Cu-O in spite of large increases in the critical current density.] This shift in $H_{irr}(T)$ for neutron-irradiated $HgBa_2CuO_{4+\delta}$,[14] for a set of specimens $YBa_2(Cu_{0.8}M_{0.2})_3O_7$ (Ref. 29) (where M = Al, Fe, Ni, and Zn) and for Ca-doped Y-Ba-Cu-O (Ref. 7) were also reported. In particular, a systematic shift of this line was observed in the change of sample size[30] or film thickness[31] as well.

Both Fisher's theory[20] on the glass-liquid transition temperature and the explanation for $H_{irr}(T)$, which is based on the thermally activated flux-line depinning, predicted that the values of $T_{irr}(H)$ or the constant a are influenced by variations in the pinning strength measured by the hysteresis width $\Delta M(T,H)$. Thus it appears that the observed relationship between the irreversibility temperature and the strength of flux pinning is a general phenomenon in all superconductors. But a systematic change of the exponent n in the power law for $H_{irr}(T)$ was not observed in any system; this means that n is independent of the strength of flux pinning or anisotropy, which depend on the coupling between CuO_2 blocks in HTS's.

In the higher-field region above 0.2 T, a serious deviation from the power law starts to appear. However, using the semilogarithmic plot of H_{irr} versus T as shown in Fig. 5, the irreversibility lines can be fitted by the exponential function

$$H_{irr} = b \exp(-T/T_0) ,$$

with $T_0 = 16.0 \pm 1.3$ K, $b = 182 \pm 60$ T for $\mathbf{H} \perp c$ and $T_0 = 16.6 \pm 1.1$ K, $b = 118 \pm 32$ T for $H \| c$. The presence of this irreversibility line at low temperatures has been in-

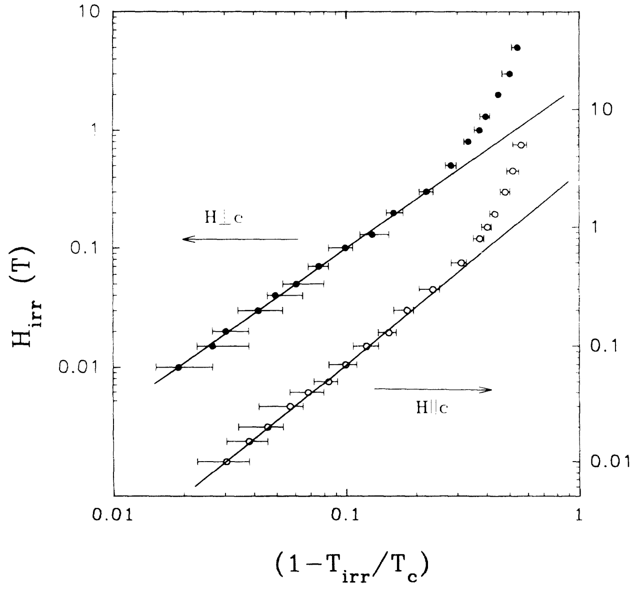

FIG. 4. Magnetic field H_{irr} vs reduced irreversibility temperature $(1 - T/T_c)$ of the c-axis-aligned power sample for both principle directions. Linear behavior was observed in the low-field range up to 0.2 T.

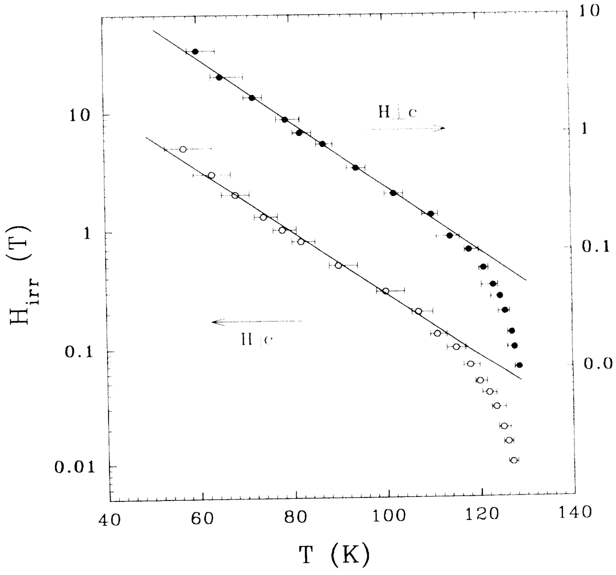

FIG. 5. H_{irr} vs T of the c-axis-aligned power sample. Exponential dependence of the irreversibility lines for $H > 0.2$ T was observed.

terpreted in terms of a change in dimensionality, in which the motion of the external flux results from a thermally activated crossover from three-dimensional vortex lines to two-dimensional vortices that can move independently in the Cu-O multilayers.[8,32]

A remarkable feature is the fact that in the temperature range below $(1-T/T_c)\sim 0.2$, irreversibility lines are well fitted with a power function, and then serious deviation from the fitting function starts to appear at the temperature of $(1-T/T_c)\sim 0.2$ for all high-T_c materials, where the exponent n is almost consistent with ~ 1.5.

In our opinion, the variation of n is independent of the pinning strength or anisotropy related to the interlayer coupling between CuO_2 planes. The characteristic crossover field H_{irr}, giving deviation from power function, depends strongly on the interlayer coupling strength closely related to the thickness of the normal blocks, e.g., ≥ 5 T for Y HTS's, ~ 0.2 T for Hg HTS's, ~ 0.02 T for Bi HTS's, and ~ 0.01 T for Tl HTS's.

IV. CONCLUSION

Anisotropic irreversibility lines due to thermal fluctuation for quasi-two-dimensional high-T_c superconductors were observed in *c*-axis-aligned powders of the $HgBa_2Ca_2Cu_3O_{8+\delta}$ compound with $T_c=131\pm 0.5$ K. The anisotropic ratio $H_{\mathrm{irr}}(\perp c)/H_{\mathrm{irr}}(\parallel c)$ below 2 is observed in all temperature regions. The simple three-dimensional-like power law $H_{\mathrm{irr}}=a(1-T/T_c)^n$ is observed only in the low-field region (≤ 0.2 T) with $n=1.40\pm 0.1$ for $\mathbf{H}\perp c$ and $n=1.63\pm 0.1$ for $H\parallel c$. In the higher-field region up to 5 T, the temperature dependence of $H_{\mathrm{irr}}(T)$ lines changes into a two-dimensional-like exponential function $H_{\mathrm{irr}}=a\exp(-T/T_0)$ as a result of the breakdown of the interlayer coupling of the conduction channel which consists of three Cu-O planes, with $T_0=16.0\pm 1.3$ K for $\mathbf{H}\perp c$ and $T_0=16.6\pm 1.1$ K for $\mathbf{H}\parallel c$. We believe that at least the exponent n is independent of the pinning strength or interlayer coupling between CuO_2 planes related to the anisotropic superconducting intrinsic parameter and that the characteristic crossover field depends strongly on the anisotropic nature of the HTS.

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[*] Permanent address: Research Institute for Basic Sciences, Seoul National University, Seoul 151-742, Korea.

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[25] In previous study (Ref. 22), we confirmed that all peaks can be identified by the Hg 1223 tetragonal structure with $a=3.852$ Å and $c=15.792$ Å. The $(00l)$ peaks are predominant in the aligned sample, where the degree of *c*-axis alignment was checked by the equation $P(006)=1-\beta$, where $\beta=(I_{110}/I_{006})^{\mathrm{aligned}}/(I_{110}/I_{006})^{\mathrm{unaligned}}$, and I_{110} and I_{006} are the relative intensities of the (110) and (006) peaks. The value of $P(006)$ was obtained to be 0.994. Rocking curve analysis of the (006) reflection of the sample reveals a full width at half maximum (FWHM) of about 3.5°.

[26] In a report by Y. S. Song *et al.* [Proceedings of the Interna-

tional Conference on Science and Technology of Synthetic Metals (ICSM'94), Seoul 1994, Synth. Met. (to be published)] we presented the temperature dependence of a lower critical field for this sample in both directions. These values are larger than one reported on the other system, while the anisotropic ratio of a lower critical field $H_{c1}\|c)/H_{c1}(\perp c)$ is low, which are expected from a structure with the mercury atom connected by two oxygen atoms.

[27] The $T_c(H)$ was evaluated from the extrapolation of the maximum slope as depicted in Fig. 1.

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