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Anisotropic Irreversibility Lines for *C*-Axis Aligned (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ Powders

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Anisotropic irreversibility lines due to thermal fluctuation for quasi-two-dimensional high-T_c superconductors were observed in *c*-axis-aligned powders of the (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ Bi(2223) compound with $T_c=108$ K. The anisotropic ratio $H_r(\perp c)/H_r(/\!/ c)$ decreases sharply from 13.6 at 100 K to low values of 3.2 at 80 K and 3.1 at 70 K. The simple 3D-like power law $H_r=a\cdot(1-T/T_c)^n$ was observed only in the low field region (≤ 200 G) with $n=2.19$ for $H\perp c$ and 2.99 for $H/\!/ c$. In the higher field region up to 4 kG, the temperature dependence of $H_r(T)$ lines changes into a 2D-like exponential function $H_r=b\cdot\exp(-T/T_0)$ due to the breakdown of the interlayer and/or intralayer coupling of the conduction channel which consists of three Cu-O planes, with $T_0=14.2$ K for $H\perp c$ and 13.7 K for $H/\!/ c$. The magnetic susceptibility ratio χ_c/χ_{ab} for this highly anisotropic superconductor in a low applied field of 8 G increases sharply from 9.8 at 5 K to 17.9 near T_c.

KEYWORDS: *c*-axis-aligned (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ powder, irreversibility line

One of the most intriguing properties of quasi-two-dimensional high-T_c superconductors is the occurrence of the irreversibility line $H_r(T)$ due to thermal fluctuations in the vortex state region between the lower critical field $H_{c1}(T)$ and the upper critical field $H_{c2}(T)$. This irreversibility or "quasi-de-Almeida-Thouless" line was first observed in the La$_{2-x}$Ba$_x$CuO$_{4-y}$ superconductor where $H_r(T)$ can be fitted by a simple power law of $H_r(T)=a\cdot(1-T/T_c)^n$ with $n\cong 1.5$.[1)] The irreversibility line $H_r(T)$ was later observed in all high-T_c superconductors with the power n varying from 1.3 to 2.[2-6)] However, in most cases the power law can be applied only in the low-field ($<10^2\sim 10^3$ G) region; serious deviation from linearity in the logarithmic plot indicates that other forms of temperature dependence in the higher field region are required. Recently, in the (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ Bi(2223) bulk sample, an exponential law of the form $H_r(T)=b\cdot\exp(-T/T_0)$ for $T<80$ K was reported.[7)]

The effects of increasing disorder or other imperfections on the position of $H_r(T)$ in the H-T plane were also explored. After 3-MeV proton irradiation, which creates random local point defects, the irreversibility line of the YBa$_2$Cu$_3$O$_{7-x}$ single crystal remained unchanged.[8)] However, a large shift was reported for the Bi$_2$CaSr$_2$Cu$_2$O$_{8+\delta}$ Bi(2122) single crystal after neutron irradiation.* A shift in $H_r(T)$ due to the thickness of YBa$_2$Cu$_3$O$_{7-x}$ thin film was also observed.[9)]

The theoretical interpretations of the origin of the irreversibility line $H_r(T)$ due to strong thermal fluctuations for these quasi-two-dimensional high-T_c superconductors are confusing. Various models ranging from the giant flux creep model to the vortex lattice (low random pinning) or vortex glass (strong random pinning) melting models were proposed.[10-13)]

Regardless of the origin of $H_r(T)$, anisotropic irreversibility is expected for these anisotropic quasi-two-dimensional high-T_c superconductors. Here, we report on the observation and detailed examination of temperature dependence of the anisotropic irreversibility properties for the *c*-axis-aligned powders of the Bi(2223) (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ compound.

Bulk samples were synthesized using the solid-state reaction method. High-purity powders of Bi$_2$O$_3$Pb$_3$O$_4$, CaCO$_3$, SrCO$_3$ and CuO were used with the ratio (Bi+Pb):(Ca+Sr):Cu=(1.85+0.15):(2.2+1.8):3 with excess PbO and CuO in order to preserve the entropy-stabilized metastable Bi(2223) phase with the nominal composition (Bi$_{1.85}$Pb$_{0.15}$)Ca$_{2.2}$Sr$_{1.8}$Cu$_3$O$_{10+\delta}$. Well-mixed powders were calcined at 800°C in air for 1 day with several intermediate regrindings. These powders were then pressed into pellets, sintered at 859°C in air up to 3 days and then furnace-cooled.

For the *c*-axis-aligned powder sample, Farrell's method was employed.[14)] Pellets were ground into powders with an average microcrystalline grain size of 1-10 μm, mixed with SPAR 5-minute epoxy/hardener in an 8-mm quartz holder with typical powder:epoxy ratio of 1:7, then aligned in a 9.4 T magnetic field at room temperature using the anisotropic normal state magnetic susceptibility. The degree of *c*-axis alignment is higher than 90% as can be checked from the intensities of the orthorhombic (001) lines from X-ray diffraction patterns.[14,15)]

Superconducting data were obtained using a Quantum Design MPMS SQUID magnetometer from 2 to 300 K. For zero-field-cooled (ZFC) measurements, the "magnetic reset" option was used to quench the superconducting magnet and reduce the residual or rem-

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*W. Kritscha, F. M. Sauerzopf, H. W. Weber, G. W. Crabtree, Y. C. Chang and P. Z. Jiang: unpublished.

nant field to less than 1 G.

The temperature dependence of the zero-field-cooled (ZFC) anisotropic magnetic susceptibility ratio $\chi_c(T)/\chi_{ab}(T)$ for a *c*-axis-aligned powder sample of (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ in low applied fields of 8, 30 and 80 G are shown in Fig. 1. A high anisotropic ratio χ_c/χ_{ab} of 9.8 was observed at low temperature. The effective anisotropic lower critical field H^*_{c1} (lower bound from the deviation of linearity in the initial $M(H)$ magnetization measurements) data indicate that H^*_{c1} (//*c*, 0 K) ≈ 60 G and H^*_{c1} (⊥*c*, 0 K) ≈ 30 G. These values are lower than actual H_{c1} values due to powder size, shape and flux pinning.* The anisotropic ratio χ_c/χ_{ab} in a low applied field of 8 G increases sharply when the temperature approaches the superconducting transition temperature T_c of 108 K, reaches a maximum value of $\chi_c/\chi_{ab} \approx 17.9$ near T_c. In a higher applied field of 80 G, the anisotropic ratio decreases steadily due to field penetration when 80 G is larger than the lower critical field $H_{c1}(T)$ above certain temperature T.

The irreversibility temperature T_r for the aligned powder sample (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ was obtained from the merging point of the field-cooled (FC) and zero-field-cooled (ZFC) curves of the temperature dependence of mass magnetic susceptibility χ_g. The temperature dependences of mass magnetic susceptibility ratios χ_g(ZFC)/χ_g(FC) in various applied fields parallel to the aligned *c*-axis are shown collectively in Fig. 2. The irreversibility temperatures T_r's can be easily pinpointed using the ratio χ_g(ZFC)/χ_g(FC), which decreases steadily to 1 when the field-cooled and zero-field-cooled curves merge together for $T \geq T_r$.

The irreversibility lines $H_r(T)$'s for (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ with applied fields up to 4 kG parallel and perpendicular to the *c*-axis are shown in Fig. 3. Dashed lines for the lower critical field H_{c1} and upper critical field H_{c2} are anisotropic in nature and are reference guides. The anisotropic ratio $H_r(\perp c)/H_r(/\!/ c)$ decreases rapidly from 13.6 at 100 K ($T_c = 108$ K) to 5.9 at 90 K, 3.2 at 80 K, and 3.1 at 70 K. The anisotropic ratio $H_r(\perp c)/H_r(/\!/ c) > 1$ was also observed for the anisotropic upper critical field H_{c2} of all high-T_c superconductors due to the anisotropic coherence length ξ with $\xi_c < \xi_{ab}$. Although $H_r(T)$'s are closely related to pinning/depinning mechanisms and are sample dependent, in-depth studies of the relationships between anisotropic properties of the irreversibility lines and anisotropic superconducting intrinsic parameters are necessary and are in progress.

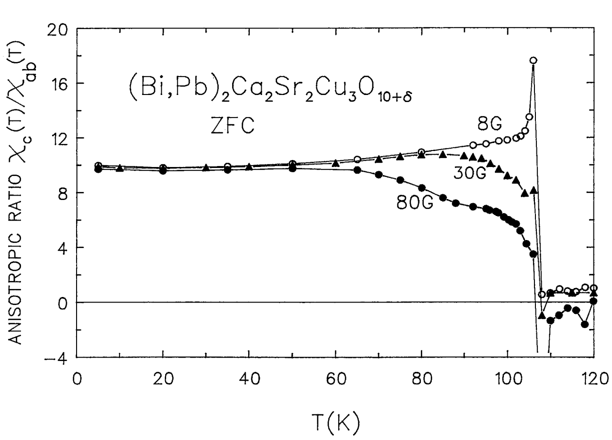

Fig. 1. Temperature dependence of zero-field-cooled (ZFC) anisotropic magnetic susceptibility ratio $\chi_c(T)/\chi_{ab}(T)$ for aligned powders of (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ in three low applied fields of 8, 30 and 80 G.

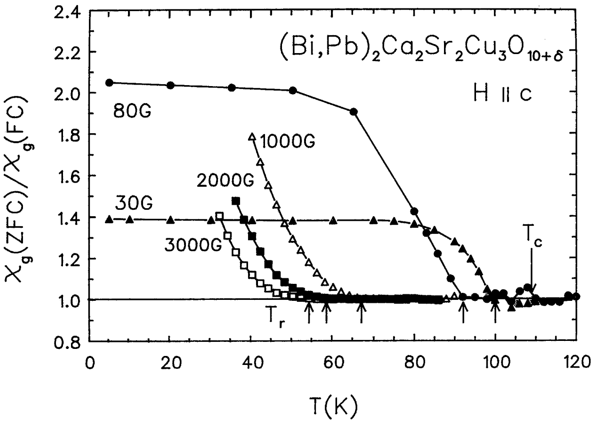

Fig. 2. Temperature dependence of mass magnetic susceptibility ratio χ_g(ZFC)/χ_g(FC) for aligned powders of (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ in various applied fields parallel to the *c*-axis.

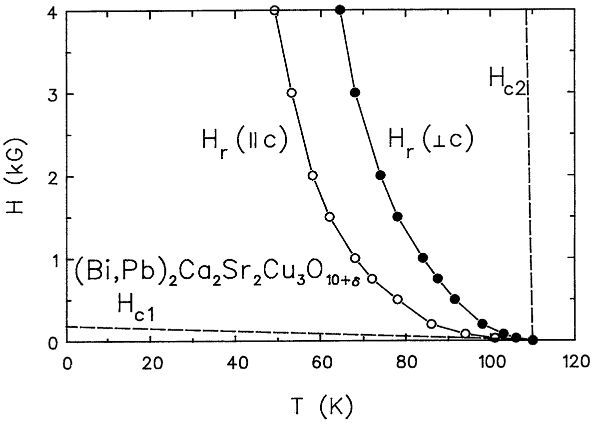

Fig. 3. Anisotropic irreversibility lines $H_r(T)$ in the *H-T* plane for (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$. Solid line, lower critical field H_{c1} and upper critical field H_{c2} dashed lines are reference guides showing no anisotropy.

The temperature dependence of the irreversibility lines $H_r(\perp c)$ and $H_r(/\!/ c)$ can be determined using the logarithmic plot. The irreversibility lines of *c*-axis-aligned powder sample (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ using the ln H_r versus ln $(1-T/T_c)$ plot are shown in Fig. 4. Linear behavior in the low field region (≤200 G) indicates that both lines can be accurately fitted by the simple power law

$$H_r = a\cdot(1-T/T_c)^n$$

with $n=2.19$, $a=3.68$ T (36.8 kG) for $H \perp c$ and $n=2.99$, $a=2.23$ T for $H /\!/ c$. The power value $n=2.99$ ($H /\!/ c$) for the (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ Bi(2223) compound is the largest value observed thus far for all high-T_c superconductors and is larger than the value $n=1.5$ predicted by

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*J. B. Shi and H. C. Ku: unpublished.

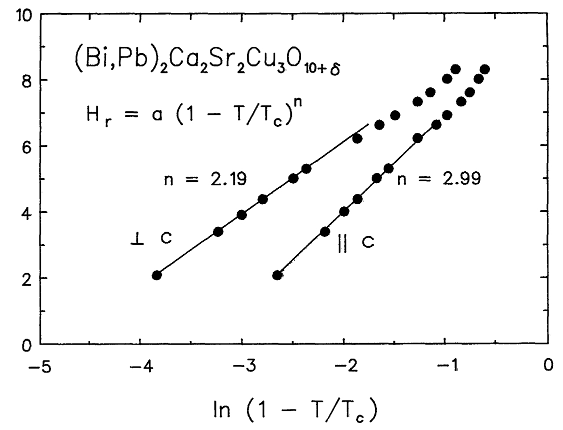

Fig. 4. ln H_r versus ln $(1-T/T_c)$ of the c-axis-aligned powder sample. Linear behavior was observed in the low field range up to 200 G.

the standard flux creep model and $n=2$ by the vortex lattice melting model.[10,16] The large reversible region in the H-T plane indicates very low flux pinning for this c-axis-aligned powder sample.

In the higher field region (>200 G), no simple power law can be found. However, using the ln H_r versus T plot (Fig. 5), the irreversibility lines can be fitted by the exponential function

$$H_r=b\cdot\exp\left(-T/T_0\right)$$

with $T_0=14.2$ K, $b=36.2$ T for $H\perp c$ and $T_0=13.7$ K, $b=14.0$ T for $H /\!/ c$. The $H_r(/\!/ c)=14.0\cdot\exp(-T/13.7)$ T for $H_r\geq 500$ G ($T<80$ K) is compatible with the $H_r=14.7\cdot\exp(-T/13.3)$ T value reported for the bulk but possibly preferentially oriented sample below 80 K.[7)]

The (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ Bi(2223) orthorhombic ($a=5.409$ Å, $b=5.411$ Å, $c=37.09$ Å) phase has a structure in which alternating conduction layers (with three Cu-O planes, CuO-Ca-CuO-Ca-CuO) of thickness $d\approx 9$ Å alternate with charge reservoir layers (SrO-BiO-BiO-SrO) of thickness $d'\approx 9.5$ Å. Interlayer and/or intralayer Josephson coupling are necessary due to small coherence length along the c-axis $\xi_c\approx 2$ Å. In the low-field region, a 3D-like power law for $H_r(T)$ line is expected. With higher applied field in the vortex state region, interlayer and/or interlayer coupling will be broken and a crossover from 3D-like to 2D-like exponential behavior is expected.[7,17)]

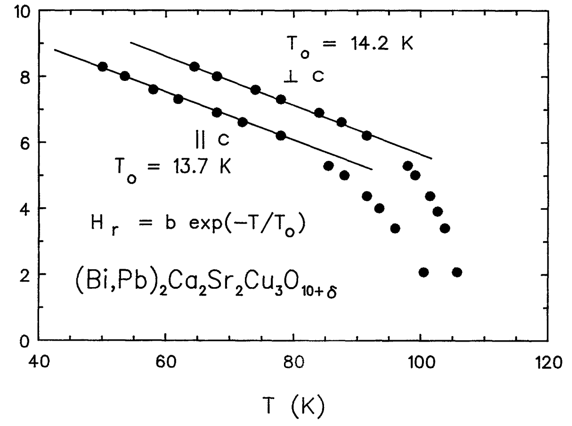

Fig. 5. ln H_r versus T of the c-axis-aligned powder sample. Exponential dependence of the irreversibility lines for $H>200$ G was observed.

The H-T phase diagram of this quasi-two-dimensional type II superconductor with strong thermal fluctuation indicates a possible phase transition along the anisotropic irreversibility lines $H_r(T)$ from the low-temperature vortex glass phase to the vortex liquid phase for $T>T_r$.[11-12)] The vortex glass long-range order phase is due to the presence of random local pinning centers for samples with the complex nominal composition (Bi$_{1.85}$Pb$_{0.15}$)Ca$_{2.2}$Sr$_{1.8}$Cu$_3$O$_{10+\delta}$. The vortex fluid phase is a fully disordered phase with only local pairing where the pairing field is strongly fluctuating with only a finite but large correlation length.

Anisotropic irreversibility lines $H_r(T)$'s were observed for the c-axis-aligned powder sample (Bi, Pb)$_2$Ca$_2$Sr$_2$Cu$_3$O$_{10+\delta}$ with the nominal composition (Bi$_{1.85}$Pb$_{0.15}$)Ca$_{2.2}$Sr$_{1.8}$Cu$_3$O$_{10+\delta}$. The anisotropic ratio $H_r(\perp c)/H_r(/\!/ c)$ decreases sharply from 13.6 at 100 K to low values of 3.2 at 80 K and 3.1 at 70 K. The simple power law $H_r=a\cdot(1-T/T_c)^n$ was observed only in the low-field region (≤ 200 G) with $n=2.19$ for $H\perp c$ and 2.99 for $H/\!/ c$. In the higher field region up to 4 kG, $H_r(T)$ lines can be fitted with the exponential law $H_r=b\cdot\exp(-T/T_0)$ with $T_0=14.2$ K for $H\perp c$ and 13.7 K for $H/\!/ c$.

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