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Anisotropic superconducting properties of aligned $(\mathrm{Bi,Pb})_2\mathrm{Ca}_{n-1}\mathrm{Sr}_2\mathrm{Cu}_n\mathrm{O}_{2n+4+\delta}$ powders ($n=1,2,3$) with $T_c=32$, 94, and 110 K

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The anisotropic superconducting properties of aligned powders embedded in epoxy for bismuth copper oxides with the compositions $(\mathrm{Bi}_{1.6}\mathrm{Pb}_{0.4})(\mathrm{Sr}_{1.5}\mathrm{La}_{0.5})\mathrm{CuO}_{6+\delta}$ ($n=1$, $T_c=32$ K), $\mathrm{Bi}_2\mathrm{Ca}_{1.2}\mathrm{Sr}_{1.8}\mathrm{Cu}_2\mathrm{O}_{8+\delta}$ ($n=2$, $T_c=94$ K), and $(\mathrm{Bi}_{1.85}\mathrm{Pb}_{0.15})\mathrm{Ca}_{2.2}\mathrm{Sr}_{1.8}\mathrm{Cu}_3\mathrm{O}_{10+\delta}$ ($n=3$, $T_c=110$ K) are reported. These c-axis-aligned samples were prepared in a 9.4-T applied magnetic field at room temperature. The temperature dependence of the anisotropy ratio $\chi_c(T)/\chi_{ab}(T)$ in the superconducting state is derived from both zero-field-cooled and field-cooled data using a small applied field H (less than or equal to the lower critical field H_{c1}) parallel and perpendicular to the orthorhombic c axis. A highly anisotropic χ_c/χ_{ab} ratio was observed for all three systems, the maximum value of $\chi_c/\chi_{ab}=9.8$ at 5 K and 10.8 at 85 K for the 2:2:2:3 compound $(\mathrm{Bi}_{1.85}\mathrm{Pb}_{0.15})\mathrm{Ca}_{2.2}\mathrm{Sr}_{1.8}\mathrm{Cu}_3\mathrm{O}_{10+\delta}$ was observed. The field dependence of the anisotropy ratio $\chi_c(H)/\chi_{ab}(H)$ for the 2:1:2:2 compound $\mathrm{Bi}_2\mathrm{Ca}_{1.2}\mathrm{Sr}_{1.8}\mathrm{Cu}_2\mathrm{O}_{8+\delta}$ up to 5 kG is discussed.

I. INTRODUCTION

High-temperature superconducting phases were reported in the bismuth copper-oxide family of the orthorhombic structure with the general formula $(\mathrm{Bi,Pb})_2\mathrm{Ca}_{n-1}\mathrm{Sr}_2\mathrm{Cu}_n\mathrm{O}_{2n+4+\delta}$ $(2,n-1,2,n)$.[1-21] The maximum superconducting transition temperature T_c(max) of 32 K was observed for the $\mathrm{Bi}_2\mathrm{Sr}_2\mathrm{CuO}_{6+\delta}$-type 2:0:2:1 structure,[1-2,4-13] a T_c(max) of 95 K was observed for the $\mathrm{Bi}_2\mathrm{CaSr}_2\mathrm{Cu}_2\mathrm{O}_{8+\delta}$ 2:1:2:2 structure and a T_c(max) of 110 K was observed for the $\mathrm{Bi}_2\mathrm{Ca}_2\mathrm{Sr}_2\mathrm{Cu}_3\mathrm{O}_{10+\delta}$ 2:2:2:3 structure.[3,14-21] Excess oxygen atoms ($\delta>0$) were commonly observed with the appearance of superstructure modulation along the orthorhombic b axis.

The superconductivity of this family is closely related to the presence of hole carriers in the quasi-two-dimensional Cu-O planes. These phases have units of CuO_x clusters where O coordinates Cu in different geometrical structures. For the 2:0:2:1 phase, CuO_6 forms an octahedron cluster; for the 2:1:2:2 phase, there are two CuO_5 pyramidal clusters separated by Ca; for the 2:2:2:3 phase, in addition to two CuO_5 pyramids, there is a CuO_4 planar cluster which is separated from the CuO_5 pyramids by Ca atoms. Anisotropies are expected for these high-T_c bismuth copper-oxide compounds which can only be studied using samples of single-crystal, c-axis-oriented thin film or c-axis-aligned powder. However, not many reports were found on the anisotropic superconducting properties of this family due to the difficulty of preparing a good single-phase sample. In this paper, we report the anisotropic superconducting properties of the highly oriented powders embedded in epoxy for the bismuth copper oxide with the compositions $(\mathrm{Bi}_{1.6}\mathrm{Pb}_{0.4})(\mathrm{Sr}_{1.5}\mathrm{La}_{0.5})\mathrm{CuO}_{6+\delta}$ ($n=1$, $T_c=32$ K), $\mathrm{Bi}_2\mathrm{Ca}_{1.2}\mathrm{Sr}_{1.8}\mathrm{Cu}_2\mathrm{O}_{8+\delta}$ ($n=2$, $T_c=94$ K), and $(\mathrm{Bi}_{1.85}\mathrm{Pb}_{0.15})\mathrm{Ca}_{2.2}\mathrm{Sr}_{1.8}\mathrm{Cu}_3\mathrm{O}_{10+\delta}$ ($n=3$, $T_c=110$ K).

II. EXPERIMENTAL DETAILS

Superconducting samples were synthesized using the solid-state reaction method. High-purity powders of $\mathrm{Bi}_2\mathrm{O}_3$, $\mathrm{Pb}_3\mathrm{O}_4$, CaCO_3, SrCO_3, $\mathrm{La}_2\mathrm{O}_3$, and CuO were used with the ratio

$$[\mathrm{Bi}+\mathrm{Pb}]:[\mathrm{Sr}+\mathrm{La}]:[\mathrm{Cu}]=(1.6+0.4):(1.5+0.5):1$$

for the 2:0:2:1 sample,

$$[\mathrm{Bi}]:[\mathrm{Ca}+\mathrm{Sr}]:[\mathrm{Cu}]=2:(1.2+1.8):2$$

for the 2:1:2:2 sample, and

$$[\mathrm{Bi}+\mathrm{Pb}]:[\mathrm{Ca}+\mathrm{Sr}]:\mathrm{Cu}=(1.85+0.15):(2.2+1.8):3$$

with excess PbO and CuO for the 2:2:2:3 sample. Well-mixed powders were calcined at 800 °C in air for 1 day with several intermediate regrindings. These powders were then pressed into pellets and sintered at 875 °C in air up to 3 days and then liquid-nitrogen quenched for the 2:0:2:1 sample, 850 °C up to 15 h and then liquid-nitrogen quenched for the 2:1:2:2 sample, and 859 °C up to 3 days and then furnace cooled for the 2:2:2:3 sample. Sintering conditions were determined from differential thermal analysis (DTA) data using an ULVAC model 7000 symmetrical thermomicrobalance. For anisotropic measurements, Farrel's method[22] was employed. Sintered single-phase superconducting pellets were grounded to powers with average microcrystalline grain size ≤ 1 μm, mixed with SPAR 5 minute epoxy and hardener in a quartz holder of diameter 8 mm with typical powder-epoxy ratio of 1:7, then aligned in a 9.4-tesla Bruker superconducting magnet at room temperature. The c axis of the orthorhombic microcrystallines are parallel to the applied magnetic field at room temperature which can be checked from x-ray-diffraction measurements. Powder x-ray-diffraction data for random oriented powders and epoxy-embedded aligned powders were obtained using a Rigaku D/MAX B diffractometer at a scanning rate of 0.25° in 2θ per min with a Si standard to eliminate any systematic errors. Structure identification, lattice parameters, and anisotropy were analyzed using the program Lazy Pulverix-PC (version 1).

Superconducting data were obtained by using a Quantum Design MPMS SQUID magnetometer from 2 to 300 K with applied magnetic field up to ± 5.5 T. For zero-field-cooled measurements, the "magnetic reset" option was used to quench the superconducting magnet and reduce the residual or remnant field to less than 1 G.

III. RESULTS AND DISCUSSION

A. $(\mathrm{Bi}_{1.6}\mathrm{Pb}_{0.4})(\mathrm{Sr}_{1.5}\mathrm{La}_{0.5})\mathrm{CuO}_{6+\delta}$ ($n=1$)

For the single Cu-O layer ($n=1$) system with the $\mathrm{Bi_2Sr_2CuO_{6+\delta}}$-type 2:0:2:1 structure, the $(\mathrm{Bi}_{1.6}\mathrm{Pb}_{0.4})(\mathrm{Sr}_{1.5}\mathrm{La}_{0.5})\mathrm{CuO}_{6+\delta}$ compound was chosen for its high T_c of 32 K.[11] The powder x-ray-diffraction patterns of $(\mathrm{Bi}_{1.6}\mathrm{Pb}_{0.4})(\mathrm{Sr}_{1.5}\mathrm{La}_{0.5})\mathrm{CuO}_{6+\delta}$ for both randomly oriented powders and c-axis-aligned powders embedded in epoxy are shown in Fig. 1. All lines can be indexed with the 2:0:2:1 orthorhombic structure with $a=5.372(5)$ Å, $b=5.375(5)$ Å, and $c=24.59(2)$ Å. The (00l) peaks are predominant in the aligned sample where the degree of alignment is better than 90% and confirms the anticipated c-axis orientation along the applied field at room temperature. Excess oxygen ($\delta > 0$) was observed from the iodometric titration measurement and indicates that the space group is probably the noncentric $A2aa$ with oxygen displacements in the (Bi,Pb)O plane.[13]

The temperature dependence of magnetization $M(T)$ for the aligned powder sample $(\mathrm{Bi}_{1.6}\mathrm{Pb}_{0.4})(\mathrm{Sr}_{1.5}\mathrm{La}_{0.5})\mathrm{CuO}_{6+\delta}$ field cooled and zero-field cooled with low applied field $H \equiv B_a = 20$ G parallel and perpendicular to c axis are shown in Fig. 2. Superconducting transition temperature T_c of 32 K was observed for this sample. Fairly high ZFC diamagnetic field shielding signal

$$-4\pi\chi_c \equiv -4\pi(M\cdot\rho)/(H\cdot m) \cong 0.59$$

for applied field parallel to c axis ($H \parallel c$) using the x-ray density $\rho = 7.28$ g/cm^3 and power mass m. The effect of very small epoxy diamagnetic signal with mass magnetic susceptibility $\chi_g \cong -6\times10^{-7}$ cm^3/g can be neglected for $T < T_c$. This diamagnetic signal $-4\pi\chi_c \cong 0.59$ is the highest value observed so far for the 2:0:2:1 structure.[1-14] High value is fully expected when the surface screening current of the microcrystalline grains is around the Cu-O superconducting a-b plane. For the FC (field-expulsion) data, a smaller value of $-4\pi\chi_c \cong 0.37$ is obtained for $H \parallel c$ due to the flux pinning inside the grain. For an applied field perpendicular to the c axis, a low diamagnetic signal for both ZFC and FC are expected for this highly anisotropic system.

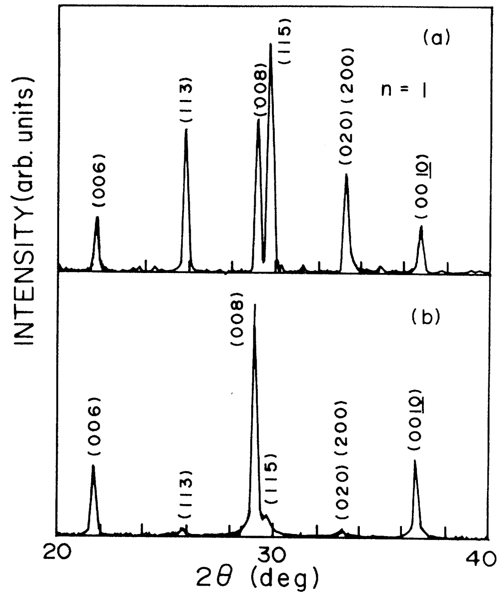

FIG. 1. Powder x-ray-diffraction patterns for the single Cu-O layer ($n=1$) compound $(\mathrm{Bi}_{1.6}\mathrm{Pb}_{0.4})(\mathrm{Sr}_{1.5}\mathrm{La}_{0.5})\mathrm{CuO}_{6+\delta}$: (a) randomly oriented, (b) c-axis aligned.

The temperature dependence of the anisotropy ratio $\chi_c(T)/\chi_{ab}(T)$ in 20 G, field cooled (open circles) and zero-field cooled (solid circles) for the aligned powder sample of $(\mathrm{Bi}_{1.6}\mathrm{Pb}_{0.4})(\mathrm{Sr}_{1.5}\mathrm{La}_{0.5})\mathrm{CuO}_{6+\delta}$, are shown in Fig. 3. The ZFC χ_c/χ_{ab} ratio is slightly larger than the FC ratio due to flux-pinning effect. As temperature increases, flux depinning due to thermal activation push the FC ratio up and close to the ZFC ratio. An anisotropic χ_c/χ_{ab} ratio of 6.9 was observed at 5 K. This value is much higher than the χ_c/χ_{ab} ratio of 2.5 observed for aligned $\mathrm{YBa_2Cu_3O_{7-x}}$ powders at low temperature[23,24] and indicates that the Bi copper-oxide family is a highly

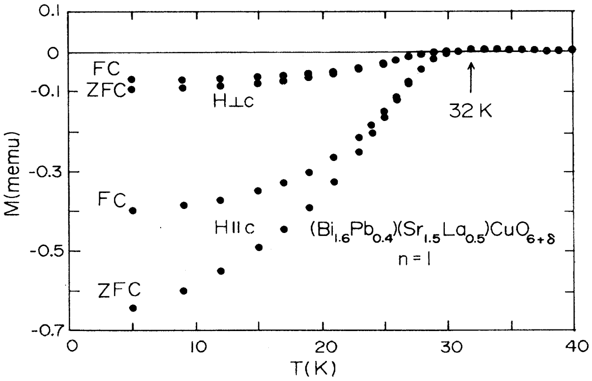

FIG. 2. Temperature dependence of magnetization $M(T)$ for the aligned powder sample $(\mathrm{Bi}_{1.6}\mathrm{Pb}_{0.4})(\mathrm{Sr}_{1.5}\mathrm{La}_{0.5})\mathrm{CuO}_{6+\delta}$ ($n=1$) field cooled (FC) and zero-field cooled (ZFC) with applied field $H=20$ G parallel and perpendicular to the c axis. $T_c=32$ K for this sample.

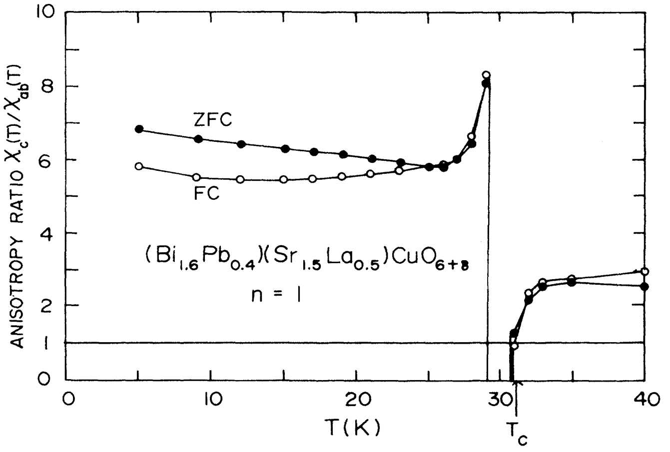

FIG. 3. Temperature dependence of the anisotropy ratio $\chi_c(T)/\chi_{ab}(T)$ of 20 G field-cooled (FC, open circles) and zero-field-cooled (ZFC, solid circles) $(Bi_{1.6}Pb_{0.4})(Sr_{1.5}La_{0.5})CuO_{6+\delta}$ ($n=1$).

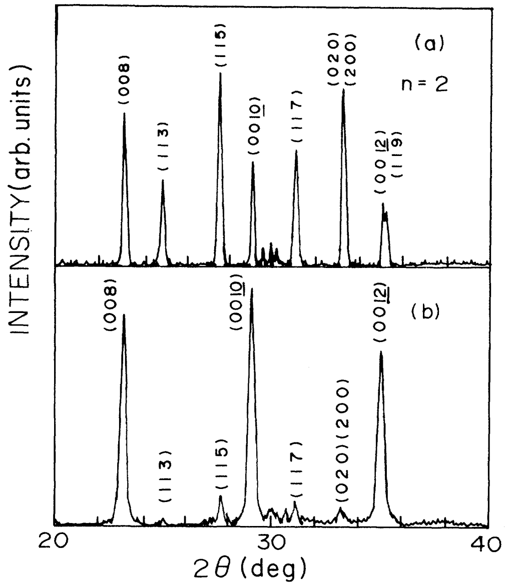

FIG. 4. Powder x-ray-diffraction patterns for the double Cu-O layer ($n=2$) compound $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$. (a) randomly oriented, (b) c-axis aligned. Superstructure modulation lines were not indexed (Ref. 19).

anisotropic superconductor. The χ_c/χ_{ab} ratio decreases from 6.9 at 5 K to 5.8 at 25 K and then increases sharply to a maximum value of 8.1 around 29 K before it drops sharply as the temperature approaches a T_c of 32 K. This anomaly was also observed for aligned $YBa_2Cu_3O_{7-x}$ powders at 50 G where the χ_c/χ_{ab} ratio increases from 2.5 at low temperature to a maximum value of 5.2 for T near a T_c of 91 K.[23] The anomaly is highly field sensitive and disappears using larger applied field. A detailed study of this anomaly is in progress and will be published in the near future.

For $T \cong T_c$, the true powder sample χ_c^p/χ_{ab}^p ratio should be expressed as

$$\chi_c/\chi_{ab} = [\chi_c^p + \chi(\text{epoxy})]/[\chi_{ab}^p + \chi(\text{epoxy})] .$$

As the temperature approaches T_c, a paramagnetic signal starts to appear in χ_{ab}^p while χ_c^p remains diamagnetic, which gives an effective negative χ_c/χ_{ab} ratio. A normal-state anisotropic χ_c/χ_{ab} ratio around 2.6 was observed for $(Bi_{1.6}Pb_{0.4})(Sr_{1.5}La_{0.5})CuO_{6+\delta}$.

B. $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ ($n=2$)

For the double Cu-O layer ($n=2$) system with the $Bi_2CaSr_2Cu_2O_{8+\delta}$-type 2:1:2:2 structure, the $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ compound was chosen for the location of the composition inside the single-phase line $Bi_2Ca_{1+x}Sr_{2-x}Cu_2O_{8+\delta}$ ($0 \le x \le 0.75$).[14–21] The powder x-ray-diffraction patterns of $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ for both randomly oriented powders and c-axis-aligned powders embedded in epoxy are shown in Fig. 4. All lines can be indexed with the 2:1:2:2 orthorhombic structure with $a=5.403(5)$ Å, $b=5.414(5)$ Å, and $c=30.79(3)$ Å. Incommensurate modulated superstructures lines were not indexed, which is due to extra oxygen ($\delta > 0$) with oxygen displacements in the BiO plane to accommodate excess oxygens.[19] The (001) peaks are predominant in the aligned samples; however, the degree of c-axis alignment is slightly worse compared with the 2:0:2:1 sample.

The temperature dependence of magnetization $M(T)$ for the aligned powder sample $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ field cooled and zero-field cooled with low applied field $H=30$ G parallel and perpendicular to the c axis is shown in Fig. 5. The superconducting transition temperature $T_c=94$ K is one of the highest observed in the 2:1:2:2 phase.[20] The ZFC diamagnetic signal $-4\pi\chi_c \cong 0.68$ for $H\|c$ using the x-ray density $\rho=6.48$ g/cm^3 was observed as compared with the FC value $-4\pi\chi_c \cong 0.50$.

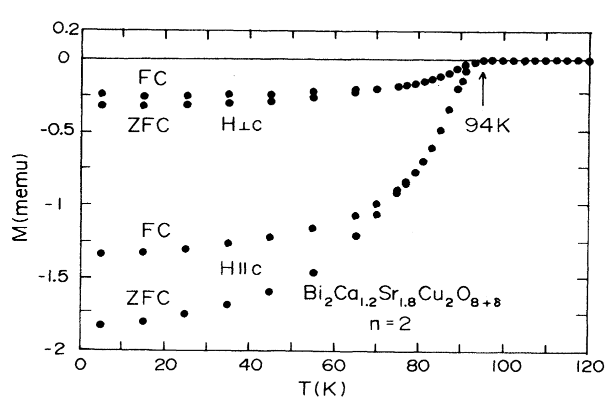

FIG. 5. Temperature dependence of magnetization $M(T)$ for the aligned powder sample $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ ($n=2$), field cooled (FC) and zero-field cooled (ZFC) with applied field $H=30$ G parallel and perpendicular to the c axis. $T_c=94$ K for this sample.

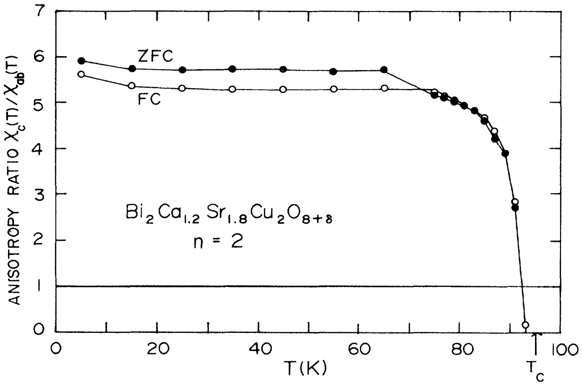

FIG. 6. Temperature dependence of the anisotropy ratio $\chi_c(T)/\chi_{ab}(T)$ of 30-G field-cooled (FC, open circles) and zero-field-cooled (ZFC, solid circles) $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ ($n=2$).

The temperature dependence of the anisotropy ratio $\chi_c(T)/\chi_{ab}(T)$ of 30 G FC (open circles) and ZFC (solid circles) for the aligned powder sample $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ ($n=2$) are shown in Fig. 6. The flux-pinning effect for the FC χ_c/χ_{ab} ratio was also observed; depinning due to thermal activation was achieved only for temperatures above 70 K for this 94-K superconductor. An anisotropic χ_c/χ_{ab} ratio of 5.9 was observed at 5 K which decreases steadily to 3.9 at 89 K and then drops sharply as the temperature approaches a T_c of 94 K. No anomaly near T_c was observed for this sample in a 30-G applied field. A negative normal-state anisotropic χ_c/χ_{ab} ratio was observed for the 2:1:2:2 sample $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$, which was also reported in the single crystal $Bi_2Ca_1Sr_2Cu_2O_{8+\delta}$ with a weak normal-state diamagnetic signal $\chi_{ab} < 0$.[23]

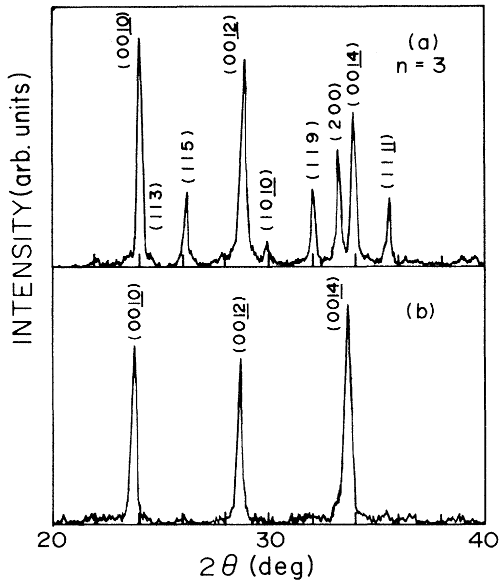

FIG. 7. Powder x-ray-diffraction patterns for the three Cu-O layer ($n=3$) compound $(Bi_{1.85}Pb_{0.15})Ca_2Sr_2Cu_3O_{10+\delta}$: (a) randomly oriented, (b) c-axis aligned.

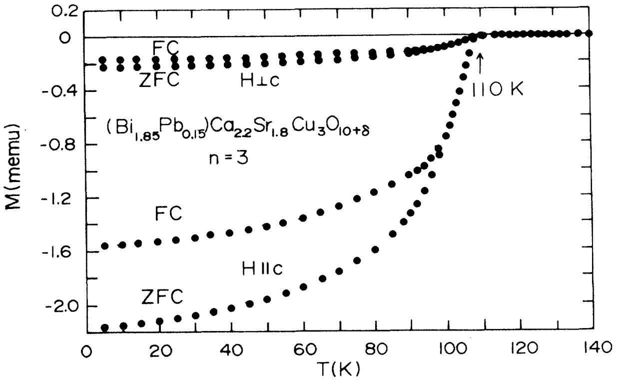

FIG. 8. Temperature dependence of magnetization $M(T)$ for the aligned powder sample $(Bi_{1.85}Pb_{0.15})Ca_2Sr_2Cu_3O_{10+\delta}$ ($n=3$), field cooled (FC) and zero-field cooled (ZFC) with applied field $H=30$ G parallel and perpendicular to the c axis. $T_c=110$ K for this sample.

C. $(Bi_{1.85}Pb_{0.15})Ca_{2.2}Sr_{1.8}Cu_3O_{10+\delta}$ ($n=3$)

For the three Cu-O layers ($n=3$) system with the $Bi_2Ca_2Sr_2Cu_3O_{10+\delta}$-type 2:2:2:3 structure, the compound $(Bi_{1.85}Pb_{0.15})Ca_{2.2}Sr_{1.8}Cu_3O_{10+\delta}$ is chosen to ensure the single-phase property of the 2:2:2:3 structure.[21] Excess PbO and CuO are necessary in order to ensure the prevention of the formation of 2:0:2:1 or 2:1:2:2 phases. The powder x-ray-diffraction patterns of $(Bi_{1.85}Pb_{0.15})Ca_{2.2}Sr_{1.8}Cu_3O_{10+\delta}$ for both randomly oriented powders and c-axis-aligned powders embedded in epoxy are shown in Fig. 7. All lines can be indexed with the 2:2:2:3 orthorhombic structure with $a=5.409(5)$ Å, $b=5.411(5)$ Å, and $c=37.09(3)$. No 2:0:2:1 or 2:1:2:2

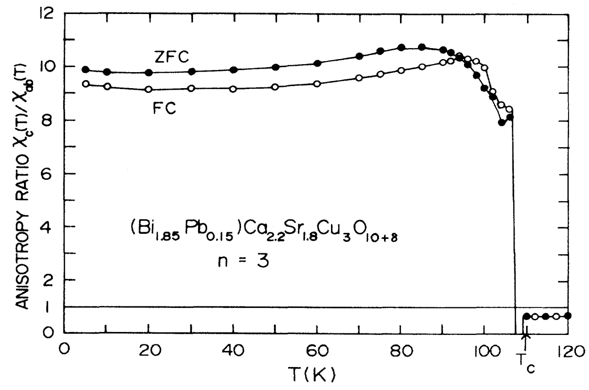

FIG. 9. Temperature dependence of the anisotropy ratio $\chi_c(T)/\chi_{ab}(T)$ of 30-G field-cooled (FC, open circles) and zero-field cooled (ZFC, solid circles) $(Bi_{1.85}Pb_{0.15})Ca_2Sr_2Cu_3O_{10+\delta}$ ($n=3$).

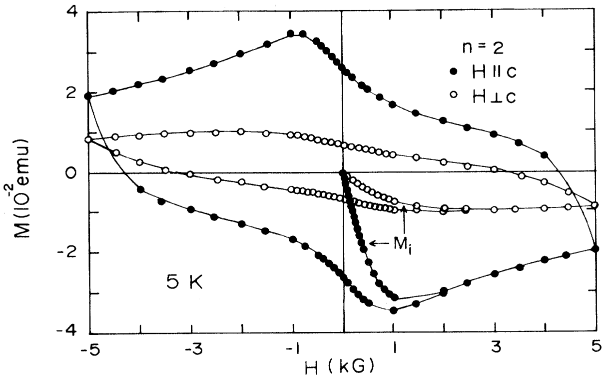

FIG. 10. Initial magnetization curve $M_i(H)$ and magnetic hysteresis loop $M(H)$ for the aligned powder sample $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ ($n=2$) with applied field parallel and perpendicular to the c axis.

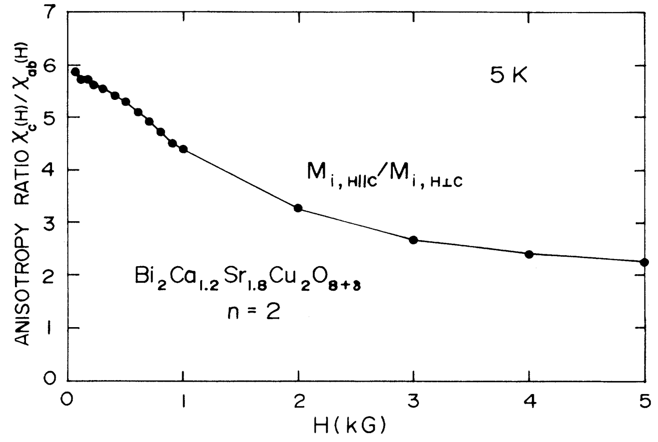

FIG. 11. Field dependence of anisotropy ratio $\chi_c(H)/\chi_{ab}(H)$ of $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ ($n=2$) from initial magnetization curve $M_i(H)$.

phase lines can be observed in the diffraction patterns. The (001) peaks are the only lines observed in the aligned samples, this indicates excellent c-axis alignment. The orthorhombic c parameter is consistent with the approximately c-axis rule of $c=18.4+6.2n$ Å.

The temperature dependence of magnetization $M(T)$ for the aligned powder sample $(Bi_{1.85}Pb_{0.15})Ca_{2.2}Sr_{1.8}Cu_3O_{10+\delta}$ field cooled and zero-field cooled with a low applied field $H=30$ G parallel and perpendicular to c axis is shown in Fig. 8. The superconducting transition temperature of $T_c=110$ K indicates the formation of the 2:2:2:3 phase while a smooth $M(T)$ indicates the successful prevention of the 2:1:2:2 phase. The excellent ZFC diamagnetic signal $-4\pi\chi_c\cong0.77$ for $H\parallel c$ using the x-ray density $\rho=6.20$ g/cm^3 was observed as compared with the FC value $-4\pi\chi_c\cong0.55$.

The temperature dependence of the anisotropy ratio $\chi_c(T)/\chi_{ab}(T)$ of 30 G FC (open circles) and ZFC (solid circles) of the aligned powder sample $(Bi_{1.85}Pb_{0.15})Ca_{2.2}Sr_{1.8}Cu_3O_{10+\delta}$ ($n=3$) is shown in Fig. 9. The flux-pinning effect for the FC χ_c/χ_{ab} ratio was also observed; the depinning due to thermal activation was achieved only for temperatures above 95 K for this 110-K superconductor. A very high anisotropic χ_c/χ_{ab} ratio of 9.8 was observed at 5 K which increases steadily to a maximum value of 10.8 at 85 K and then drops sharply as the temperature approaches a T_c of 110 K. No anomaly was observed for this sample. A normal-state anisotropic χ_c/χ_{ab} ratio around 0.7 was observed for $(Bi_{1.85}Pb_{0.15})Ca_{2.2}Sr_{1.8}Cu_3O_{10+\delta}$.

These low-field ($H\le H_{c1}$) temperature-dependent anisotropic $\chi_c(T)/\chi_{ab}(T)$ data for all three superconducting samples studied are listed together in Table I for comparison. The effect of a higher applied field can be seen from the field dependence of the initial magnetization curve $M_i(H)$ and the magnetic hysteresis loop $M(H)$ with a magnetic field up to ±5 kG for the aligned powder sample $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ ($n=2$) with an applied field parallel and perpendicular to the c axis (Fig. 10). A lower critical field H_{c1} was obtained from the breakaway from the linearity of the initial magnetization curve $M_i(H)$ with $H_{c1}^{c}=42$ G and $H_{cl}^{ab}=30$ G at 5 K. The field dependence of the anisotropy ratio $\chi_c(H)/\chi_{ab}(H)$ at 5 K extracted from the initial magnetization curve $M_i(H)$ is shown in Fig. 11. The $\chi_c(H)/\chi_{ab}(H)$ ratio decreases steadily from 5.9 in low field to 4.2 at 11 kG and 2.2 at 5 kG.

TABLE I. Crystallographic and superconducting data for $(Bi_{1.6}Pb_{0.4})(Sr_{1.5}La_{0.5})CuO_{6+\delta}$ ($n=1$), $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ ($n=2$), and $(Bi_{1.85}Pb_{0.15})Ca_2Sr_2Cu_3O_{10+\delta}$ ($n=3$). (ZFC is zero-field cooled and FC is field cooled.)

	$n=1$	$n=2$	$n=3$
T_c (K)	32	94	110
Lattice parameter c (Å)	24.59	30.79	37.07
X-ray density ρ (g/cm^3)	7.28	6.48	6.20
Susceptibility $-4\pi\chi_c$ (5 K, ZFC)	0.59	0.68	0.77
Susceptibility $-4\pi\chi_c$ (5 K, FC)	0.37	0.50	0.55
χ_c/χ_{ab} (5 K, ZFC)	6.9	5.9	9.8
χ_c/χ_{ab} (max)	8.1(29 K)	5.9(5 K)	10.8(85 K)

IV. CONCLUSIONS

In conclusion, high-T_c superconducting aligned powders embedded in epoxy for the bismuth copper oxide with the compositions $(Bi_{1.6}Pb_{0.4})(Sr_{1.5}La_{0.5})CuO_{6+\delta}$ ($n=1$, $T_c=32$ K), $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ ($n=2$, $T_c=94$ K), and $(Bi_{1.85}Pb_{0.15})Ca_{2.2}Sr_{1.8}Cu_3O_{10+\delta}$ ($n=3$, $T_c=110$ K) are prepared in a 9.4-tesla applied field at room temperature. The temperature dependence of anisotropy ratio $\chi_c(T)/\chi_{ab}(T)$ in the superconducting state were derived from both zero-field-cooled and field-cooled data using small applied field. A high anisotropy ratio of $\chi_c/\chi_{ab}=9.8$ was observed for the 2:2:2:3 compound $(Bi_{1.85}Pb_{0.15})Ca_{2.2}Sr_{1.8}Cu_3O_{10+\delta}$ at 5 K. The field dependence of the anisotropy ratio $\chi_c(H)/\chi_{ab}(H)$ for the 2:1:2:2 compound $Bi_2Ca_{1.2}Sr_{1.8}Cu_2O_{8+\delta}$ at 5 K decreases from 5.9 in low field to 2.2 at 5 kG.

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[1] C. Michel, M. Hervieu, M. M. Borel, A. Grandin, F. Deslandes, J. Provost, and B. Raveau, Z. Phys. B **68**, 421 (1987).

[2] J. Akimitsu, A. Yamazaki, H. Sawa, and H. Fujiki, Jpn. J. Appl. Phys. **26**, L2080 (1987).

[3] H. Maeda, Y. Tanaka, M. Fukitomi, and T. Asano, Jpn. J. Appl. Phys. **27**, L209 (1988).

[4] L. C. Porter, E. Appelman, M. A. Beno, C. S. Cariss, K. D. Carlson, H. Cohen, U. Geiser, R. J. Thorn, and J. M. Williams, Solid State Commun. **66**, 1105 (1988).

[5] H. Sawa, H. Fujiki, K. Tomimoto, and J. Akimitsu, Jpn. J. Appl. Phys. **27**, L830 (1988).

[6] G. Van Tendeloo, H. W. Zandbergen, and S. Amenlinckx, Solid State Commun. **66**, 927 (1988).

[7] C. C. Torardi, M. A. Subramanian, J. C. Calabrese, J. Gopalakrishnan, E. M. McCarron, K. J. Morrissey, T. R. Askew, R. B. Flippen, U. Chowdhry, and A. W. Sleight, Phys. Rev. B **38**, 225 (1988).

[8] J. M. Tarascon, W. R. McKinnon, P. Barboux, D. M. Hwang, B. G. Bagley, L. H. Greene, G. W. Hull, Y. LePage, N. Stoffel, and M. Giroud, Phys. Rev. B **38**, 8885 (1988).

[9] G. H. Hwang, J. B. Shi, and H. C. Ku, Chin. J. Phys. **27**, 273 (1989).

[10] H. C. Ku, J. B. Shi, G. H. Hwang, M. F. Tai, S. W. Hsu, D. C. Ling, M. J. Shieh, T. Y. Lin, and T. J. Watson-Yang, in *Progress in High Temperature Superconductivity,* edited by P. T. Wu, H. C. Ku, W. H. Lee, and R. S. Liu, (World Scientific, Singapore, 1989), Vol. 19, p. 96.

[11] H. C. Ku, S. J. Sun, Y. T. Peng, G. H. Hwang, J. B. Shi, and B. S. Chiou, *Proceedings of the ROC-Japan Symposium for High Temperature Superconductor* (Japan Interchange Association, Tokyo, 1990), p. 25.

[12] H. C. Ku, J. B. Shi, C. C. Lai, and S. J. Sun, Physica B **165&166**, 1153 (1990).

[13] J. B. Shi, B. S. Chiou, Y. T. Peng, and H. C. Ku, Chin. J. Phys. **28**, 339 (1990).

[14] R. M. Hazen, C. T. Prewitt, R. J. Angel, N. l. Ross, L. W. Finger, C. G. Hadidiacos, D. R. Veblen, P. J. Heaney, P. H. Hor, R. L. Meng, Y. Y. Sum, Y. Q. Wang, Y. Y. Xue, Z. J. Huang, L. Gao, J. Bechtold, and C. W. Chu, Phys. Rev. Lett. **60**, 1174 (1988).

[15] S. A. Sunshine, T. Siegrist, L. F. Schneemeyer, D. W. Murphy, R. J. Cava, B. Batlogg, R. B. van Dover, R. M. Fleming, S. H. Glarum, S. Nakahara, R. Farrow, J. J. Krajewski, S. M. Zahurak, J. V. Waszczak, J. H. Marshall, P. Marsh, L. W. Rupp, Jr., and W. F. Peck, Phys. Rev. B **38**, 893 (1988).

[16] J. M. Tarascon, Y. Le Page, P. Barboux, B. G. Bagley, L. H. Greene, W. R. McKinnon, G. W. Hull, M. Giroud, and D. M. Hwang, Phys. Rev. B **37**, 9382 (1988).

[17] M. A. Subramanian, C. C. Torardi, J. C. Calabrese, J. Gopalakrishnan, K. J. Morrissey, T. R. Askew, R. B. Flippen, U. Chowdhry, and A. W. Sleight, Science **239**, 1015 (1988).

[18] G. S. Grader, E. M. Gyorgy, P. K. Gallagher, H. M. O'Bryan, D. W. Johnson, Jr., S. Sunshine, S. M. Zahurak, S. Jin, and R. C. Sherwood, Phys. Rev. B **38**, 757 (1988).

[19] M. Onoda, A. Yamamoto, E. Takayama-Muromachi, and S. Takehawa, Jpn. J. Appl. Phys. **27**, L833 (1988).

[20] N. H. Wang, C. M. Wang, H. C. I. Kao, D. C. Ling, H. C. Ku, and K. H. Lii, Jpn. J. Appl. Phys. **28**, L1505 (1989).

[21] A. Maeda, M. Hase, I. Tsukada, K. Noda, S. Takebayashi, and K. Uchinokura, Phys. Rev. B **41**, 6418 (1990).

[22] D. E. Farrell and B. S. Chandrasekhar, Phys. Rev. B **36**, 4025 (1987).

[23] W. C. Lee and D. C. Johnston, Phys. Rev. B **41**, 1904 (1990).

[24] D. C. Johnston and J. H. Cho, Phys. Rev. B **42**, 8710 (1990).