Influence of electromagnetic anisotropy on the flux-pinning strength of columnar defects in $Bi_{2.2-x}Pb_xSr_{1.8}CaCu_{2.0}O_y$

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Magnetization measurements have been performed on Bi(Pb)2212 single crystals with various electromagnetic anisotropies ($\gamma^2=1.2\times10^3\sim4.4\times10^4$) before and after introducing columnar defects by Ta ion irradiation. The irreversibility fields of these crystals increased systematically with decreasing γ^2 at various reduced temperatures $t=T/T_c$, indicating that the smaller anisotropy makes the columnar defects substantially more effective in flux pinning. The record-high irreversibility field and critical current density for single crystals of the Bi2212 phase was observed in our oxygen-overdoped Bi(Pb)2212 with columnar defects. This study experimentally reveals the reduction of the electromagnetic anisotropy and the introduction of pinning centers work cooperatively in high-T_c superconductor compounds.

I. INTRODUCTION

It is well known that the electromagnetic properties of high-T_c superconductors (HTSC's) are intrinsically anisotropic due to their two-dimensional crystal structures. Accordingly, in the case of HTSC's, Cooper pair condensation energy per coherence volume can be described as

$$\varepsilon_c=\frac{1}{2\mu_0}B_c^2\xi_{ab}^3\times\frac{1}{\gamma} \quad (1)$$

where $\gamma=\sqrt{m_c^*/m_{ab}^*}=\xi_{ab}/\xi_c$ is the electromagnetic anisotropy parameter. Since ε_c corresponds to the pair-breaking energy, Eq. (1) represents an inherent problem that superconductivity with larger anisotropy is easier to be destroyed by thermal fluctuation; ξ_{ab} is similar among HTSC's.[1] Moreover, expansion of interlayer distance of CuO_2 blocks, which usually increases together with the anisotropy,[2] reduce its condensation energy per unit volume, developing the influence of thermal fluctuation further.

The above discussion immediately explains the reason why the flux cannot be effectively pinned in $Bi_2Sr_2CaCu_2O_y$ (Bi2212) above 30 K under a magnetic field applied parallel to the c axis of the crystal, because it is extremely anisotropic material among HTSC's. Therefore, it is crucial for high-field and -temperature applications of Bi2212 to improve the flux-pinning properties, as it is the most developed material to date for superconducting wires and tapes due to its easily oriented nature and the superior coupling of its crystal grains.

One of the major approaches to overcome the poor flux pinning strength in HTSC's is to lower the electromagnetic anisotropy. Carrier doping by oxygen annealing has been reported to decrease the anisotropy of Bi2212, resulting in an increase of its irreversibility fields (B_{irr}).[3] In addition, we recently found that Pb doping essentially reduces the electromagnetic anisotropy of Bi2212, and dramatically improved flux-pinning properties were observed in heavily Pb-doped single crystals.[4–6] According to the resistivity measurements, the anisotropy parameter γ^2 of Bi(Pb)2212 was found to be lowered to $\gamma^2=\rho_c/\rho_{ab}$ (at 100 K) $\sim1.2\times10^3$ (Ref. 6).

Regarding irreversibility fields, a universal scaling law was proposed by Kishio[2] for typical HTSC's, such as YBCO, LSCO, and Bi2212, which describes irreversibility fields to be scaled as $B_{irr}\propto1/\gamma^2$ at the same reduced temperature ($t=T/T_c$) when t is larger than 0.7. Usually in the case of single crystals, structural defects or nonsuperconducting precipitates are the major pinning centers, and their effects on flux pinning are almost equivalent because they can be regarded as weak random pinning potentials. As a consequence, the observed universal scaling suggests that their smaller anisotropy makes the pinning centers more effective in flux pinning, which is a useful guiding principle in developing flux-pinning properties at high temperatures under magnetic fields.

Unfortunately, however, there has been no report that directly substantiates this suggestion. In order to elucidate the relationship between electromagnetic anisotropy and flux-pinning strength, it is necessary to study the pinning properties without changing any other factors, such as the size, shape, and density of predominant pinning centers, as well as the crystal structure.

There is a valuable method to produce artificial pinning centers in HTSC's, which is well known as another approach to improve the flux pinning. Irradiation by various particles such as neutrons and ions, produces amorphous defects in the HTSC crystals, which act as effective pinning centers and improve the flux-pinning properties depending on their shapes and densities. Particularly, columnar defects are the most prominent ones, which are produced by heavy-ion irradiation.[7–14]

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TABLE I. T_c, γ^2, and annealing conditions of each Bi(Pb)2212 single crystal.

Crystals x in $Bi_{2.2-x}Pb_x$ (carrier doping state)	Annealing Conditions Temp./°C	$P(O_2)$/atm	T_c/K	γ^2
$x=0$ (UD)	600	3.9×10^{-4}	77.7	44000 [a]
$x=0$ (LOV)	400	2.3×10^{-3}	81.8	17000 [a]
$x=0$ (HOV)	400	2.1	77.2	8700 [a]
$x=0.6$ (LOV)	600	3.9×10^{-4}	80.7	3000 [b]
$x=0.6$ (HOV)	400	2.1	63.5	1200 [b]

[a]From second peak field.
[b]From resistivity measurements.

In the present study, magnetic properties of Bi2212 and Bi(Pb)2212 single crystals with widely and systematically changed anisotropies have been investigated before and after Ta ion irradiation, which produced perfectly penetrated columnar defects in the crystals. After the introduction of these columnar defects, the highest irreversibility field and the largest critical current density as Bi(Pb)2212 single crystals were experimentally observed. Based on the experimental results, the influence of electromagnetic anisotropy on flux pinning strength will be discussed. It was found that their smaller anisotropy makes the columnar defects more effective in flux pinning, which directly proved the efficiency of lowering the anisotropy.

II. EXPERIMENT

Single-crystal growth of Pb-free Bi2212 and heavily Pb-doped Bi2212 was performed using the floating-zone method. The nominal Pb compositions in a chemical formula $Bi_{2.2-x}Pb_xSr_{1.8}CaCu_{2.0}O_y$ were $x=0$ and 0.6, respectively. Cation compositions of the grown crystals were determined to be Bi:Pb:Sr:Ca:Cu$=2.1_5:0:1.8_7:0.9_6:2.0_0(x=0)$, and $1.6_5:0.4_4:1.8_5:0.9_6:2.0_0(x=0.6)$ by inductively coupled plasma analysis. This indicates that a small amount of Pb evaporated during the growth.

The grown boules were cut and cleaved, and platelike crystals with a typical size of $1\times1\times0.1$ mm^3 were obtained. Carrier doping levels were controlled by post-annealing at various temperatures and partial pressure of oxygen. Crystals were sealed into quartz ampoules and annealed for 72 h at 400–600 °C at an effective pressure of $P(O_2)=3.9\times10^{-4}\sim2.1$ atm, and quenched to room temperature. After these procedures, five types of crystals were prepared: Pb-free ($x=0$) underdoped (UD), lightly overdoped (LOV), heavily overdoped (HOV), and heavily Pb-doped ($x=0.6$), LOV and HOV.

In order to introduce columnar defects, Ta ion irradiation was performed at GANIL (Caen, France) with an incident energy of 7.2 GeV. The Ta ions were irradiated parallel to the c axis of the crystals. Since five crystals with different anisotropy were irradiated at the same time, the real density and shape of the produced columnar defects are identical among these crystals. Typical diameter of the defects is approximately 7 nm,[9,15] and a dose equivalent field B_Φ, calculated from the total fluence 2×10^{11} ions/cm^2, is 2 T. No appreciable decrease of T_c was observed after the irradiation.

Magnetic properties were investigated using a superconducting quantum interference device magnetometer (Quantum Design MPMS XL-5S), before and after the irradiation. In all measurements, magnetic fields were applied parallel to the c axis. The critical temperature (T_c) was determined from a midpoint of the zero-field-cooled (ZFC) magnetization transition measured under 1 G. Magnetic hysteresis loops were measured at various temperatures between 20 K and T_c, under cyclic fields of ±5 T. The critical current density (J_c) was calculated from the width of magnetic hysteresis loops (ΔM), using the extended Bean model.[16] An irreversibility field (B_{irr}) at each temperature was defined with a criterion of $J_c=100$ A/cm^2 in this study.

Anisotropy parameters γ^2 of Pb-free crystals ($x=0$) were estimated from the dimensional crossover field, which can be observed as the second peak field B_{pk} in the magnetic hysteresis loops, using an equation $\gamma^2=\Phi_0/(B_{pk}s^2)$.[6,17] In this formula, Φ_0 and s represent the flux quantum and the distance between superconducting layers, respectively, where we use $s=1.54$ nm as a usual value of Bi2212. In the case of heavily Pb-doped crystals ($x=0.6$), this formula may be no longer applicable because B_{pk} becomes strongly temperature dependent.[5] Therefore γ^2 were estimated from the resistivity measurements as $\gamma^2=\rho_c/(\rho_a\rho_b)^{1/2}$.[6]

T_c and γ^2 of all the crystals are summarized in Table I together with each annealing condition. It is noteworthy that γ^2 was controlled over 40 times from $x=0.6$ (HOV) to $x=0$ (UD), while the crystal structure remained essentially unchanged.

III. RESULTS AND DISCUSSION

A. Irreversibility line

In all the crystals, dramatic changes were observed in their magnetization hysteresis measurements after Ta ion irradiation. As a typical result, magnetic hysteresis loops of the crystals $x=0.6$ (LOV), taken at 60 K before and after the irradiation, are shown in Fig. 1. The irradiated crystals exhibited much larger loops than those of the pristine ones, indicating that introduction of columnar defects considerably improved the flux pinning properties.

Irreversibility fields of Bi(Pb)2212 single crystals before and after irradiation are shown in Fig. 2 as a function of temperature, which represents the so-called irreversibility lines (IL's). The first aspect to be noted in this figure is the expansion of each irreversible region via introduction of columnar defects. Comparing IL's between pristine and irradi-

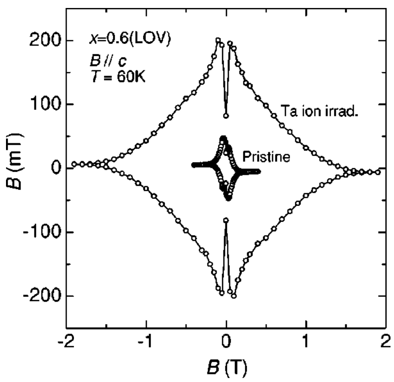

FIG. 1. Magnetic hysteresis loops of crystal $x=0.6$ (LOV) before (pristine) and after Ta ion irradiation ($B_\Phi=2$ T), measured at 60 K.

ated crystals, one can clearly find that IL's for irradiated crystals are all located in the reversible regimes of the pristine crystals. It is the columnar defects that dominate the flux pinning in these expanded regimes, which allow us to investigate the flux-pinning properties in terms of electromagnetic anisotropy.

It is especially noteworthy that the $x=0.6$ (LOV) crystal with columnar defects shows the highest IL as a Bi2212 single crystal. The reported highest irreversibility field at $T=60$ K was $B_{\mathrm{irr}}\approx 1.2$ T, which was observed in Sn-ion irradiated[7] and Pb-ion irradiated Bi2212,[12] whereas our crystal here recorded $B_{\mathrm{irr}}=1.8$ T at the same temperature. It will be revealed later that the small anisotropy caused this record, as well as the relatively high T_c of this crystal. Note, however, that the most steeply rising IL is that of the $x=0.6$ (HOV) crystal with columnar defects, whose anisotropy is the smallest in this study while its T_c is lower than that of the $x=0.6$ (LOV) crystal, approximately by 17 K.

In order to subtract the contribution of difference in T_c, IL's of the irradiated crystals are now shown in logarithmic scales as a function of the reduced temperature $t=T/T_c$ in Fig. 3. It is immediately found that IL's shift upwards systematically in the order of UD, LOV, and HOV ($x=0$), and LOV and HOV ($x=0.6$), in which the anisotropy parameter γ^2 decreases monotonically.

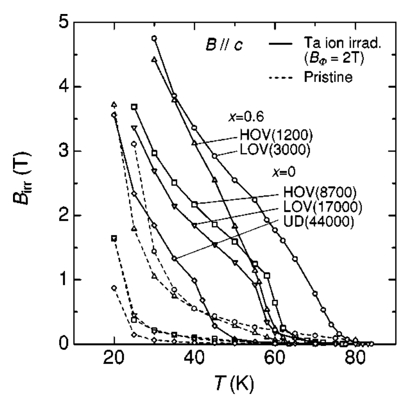

FIG. 2. Irreversibility lines (IL's) of Bi(Pb)2212 single crystals before (dashed lines) and after (solid lines) Ta ion irradiation ($B_\Phi=2$ T) with various electromagnetic anisotropy. Parenthesized values represent $\gamma^2=m_c^*/m_{ab}^*$ of each crystal. Crystal $x=0.6$ (LOV) with columnar defects shows the highest IL as a Bi2212 single crystal.

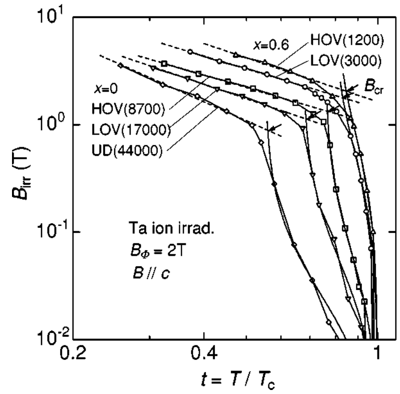

FIG. 3. IL's of Bi(Pb)2212 single crystals after Ta ion irradiation as a function of reduced temperature $t=T/T_c$. For all t, B_{irr} systematically increases with decreasing γ^2. Each IL is fitted by both the power law (dashed line: $B>B_{\mathrm{cr}}$) and Eq. (2) (solid line: $B<B_{\mathrm{cr}}$), where B_{cr} are determined by their intersections (marked by arrows). Note that even if $B>B_{\mathrm{cr}}$, lowering γ^2 still enhances B_{irr}.

Since flux pinning is ruled by columnar defects in the expanded irreversible regime, B_{irr} represents the maximum field where columnar defects are effective in each crystal. In other words, Fig. 3 suggests the fact that in a crystal with smaller anisotropy, columnar defects are effective up to higher temperatures and higher fields, which demonstrates that *smaller anisotropy makes the columnar defects more effective*.

Another important aspect to be noted is that the slopes of each IL show the rapid changes around the field B_Φ slightly below 2 T. Similar behaviors have been observed in YBCO and Bi2212 single crystals with columnar defects, and explained that they are due to a crossover from strong single vortex pinning to weaker collective pinning regime by columnar defects.[9,13,18–20]

Krusin-Elbaum *et al.*[18] found that, below the crossover field (B_{cr}), the IL showed the same temperature dependence as that of the Bose-glass transition line (BGL). The BGL can be described as the first-order transition line of a flux line lattice modified by the pinning energy of columnar defects.[21] Taking into account the decoupling theory of the flux line lattice,[22] one can express the temperature dependence of the BGL, using the proportional constant α, as

$$B_{\mathrm{BG}}=\alpha\left(\frac{A}{t_{\mathrm{BG}}+A-1}-1\right), \qquad (2)$$

$$A=\left(1+\frac{1}{16c_L\sqrt{\mathrm{Gi}}}\,\frac{r^2}{\xi(0)d}\right)^{-1}, \qquad (3)$$

where $t_{\mathrm{BG}}=T_{\mathrm{BG}}/T_c$. Here c_L is the Lindemann criterion, Gi is the Ginzburg number, r is the geometrical radius of the columnar defects, and d is average spacing between defects.[21]

To identify the crossover field B_{cr} in our study, we fit Eq. (2) to the data of low-field region where slopes of IL's are steep, and fit the power law to those of high-field region, where slopes are gentle, with the power $n=-1.5\pm0.3$. From the fitting parameter A in Eq. (2), we could obtain $\mathrm{Gi}=10^{-2}(x=0,\mathrm{UD})\sim10^{-3}(x=0.6,\mathrm{HOV})$ by Eq. (3) using

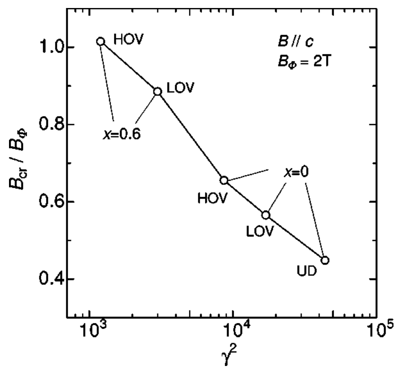

FIG. 4. γ^2 dependence of the crossover field B_{cr} normalized by $B_\Phi = 2$ T. Each B_{cr} was determined in Fig. 3 as an intersection of the fitting functions. The magnitude of B_{cr}/B_Φ increases with decreasing γ^2, suggesting that reduction of anisotropy results in an enlargement of the pinning energy.

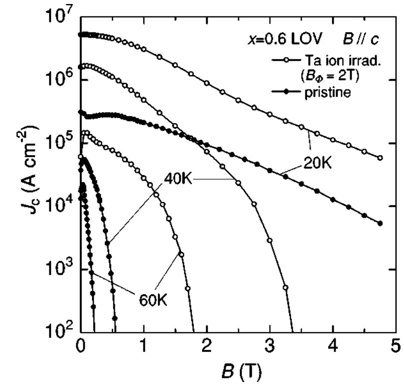

FIG. 5. Magnetic-field dependence of J_c of a crystal $x=0.6$ (LOV) measured at $T=20$, 40, and 60 K before (pristine) and after Ta ion irradiation ($B_\Phi = 2$ T). Crystal $x=0.6$ (LOV) with columnar defects shows the largest J_c values above 40 K as a Bi2212 single crystal.

$c_L = 0.1$, $r = 3.5$ nm, and $d = 35$ nm, which systematically ascends together with the anisotropy parameter γ^2, in accordance with a theoretical prediction.[23] Here we define B_{cr} as an intersection field of two fitting functions, which are marked by arrows in Fig. 3.

Figure 4 shows B_{cr} normalized by $B_\Phi = 2$ T as a function of γ^2. Obviously B_{cr} increases with decreasing γ^2, which indicates that in materials with smaller anisotropy, columnar defects maintain a strong single-vortex pinning up to higher magnetic field. From this result, we can derive a natural conclusion that a *smaller anisotropy makes the pinning energy of columnar defects larger*.

When the repulsive force of vortex interaction surpasses the columnar defects' pinning, crossover from strong single-vortex pinning to weaker collective pinning occurs. Therefore, B_{cr} corresponds to a field where the shear energy of the flux line lattice becomes equal to the columnar pinning energy. Theoretically, shear energy increases with magnetic field irrespective of γ^2 when the field is applied parallel to the c axis.[21] Consequently, the increase of B_{cr} observed here is brought about by the increase of the shear energy, which is required to balance itself with the columnar pinning energy enhanced via decreased γ^2.

In addition, it should be pointed out that B_{irr} still increases with decreasing γ^2 above B_{cr}. A smaller anisotropy is effective in flux pinning by columnar defects even in the weaker collective pinning regime.

B. Critical current density

To see the flux pinning in the irreversible region, J_c's of pristine and irradiated crystals $x=0.6$ (LOV) measured at $T = 20$, 40, and 60 K are shown as functions of the magnetic field in Fig. 5. Improvement of J_c by the introduction of columnar defects becomes larger and larger with increasing temperature, so that the crystal $x=0.6$ (LOV) with columnar defects showed the largest J_c value as reported for Bi2212 single crystal above 40 K.[7,11]

Figure 6 shows the magnetic-field dependence of J_c for all irradiated crystals at the same reduced temperature $t = 0.7$, which allows a comparison of J_c in terms of γ^2. Concerning the Pb-free crystals ($x=0$), a systematic increase of J_c with decreasing γ^2 has been observed, indicating that a *smaller anisotropy makes columnar defects more effective in the irreversible region.*

However, concerning the heavily Pb-doped Bi2212 ($x = 0.6$), the LOV crystal shows a larger J_c value than that of the HOV crystal above $J_c = 10^4$ A/cm^2. In this study, nonsystematic behaviors with respect to anisotropy were sometimes observed in the irreversible region $x=0.6$ of the crystals in large J_c conditions at low temperatures. Because similar behaviors have been observed even in pristine crystals, it is clear that this does not originate in the introduction of columnar defects, as well as their subsequent effects. We think these behaviors may be caused by the micro-phase-segregation observed in heavily Pb-doped Bi2212 single crystals, which consists of a modulated phase with a lower Pb content and a modulation-free phase with a higher Pb content.[4,24] Our Pb-doped Bi2212 single crystals ($x=0.6$) also have similar microstructures,[25] and, for some reasons, they restrict the large current flowing along the b axis. Moreover, this restriction could be enhanced by carrier ''overdoing'' with oxygen annealing, which results in the inversion

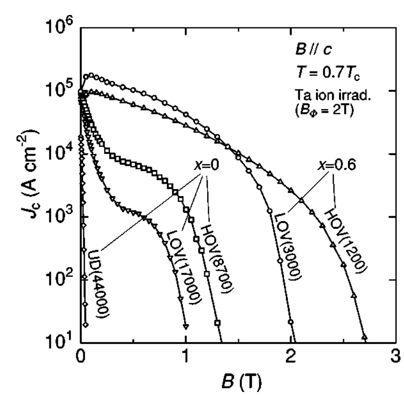

FIG. 6. Magnetic-field dependence of J_c for Ta-ion-irradiated crystals with various anisotropy measured at a reduced temperature $t=0.7$. Parenthesized values represent γ^2 of each crystal. Pb-free crystals ($x=0$) show a systematic increase in J_c with decreasing γ^2, whereas heavily Pb-doped crystals ($x=0.6$) represent an inversion above $J_c = 10^4$ A/cm^2.

of J_c observed in Fig. 6. Here we suspect that these microstructures sometimes act as effective pinning centers, particularly in HOV crystals.[5]

IV. SUMMARY

Using Bi(Pb)2212 single crystals with columnar defects, the influence of electromagnetic anisotropy on flux pinning has been studied. From the systematic dependence of IL's on γ^2, it has been revealed that a smaller anisotropy makes the columnar defects effective up to a higher temperature and magnetic field. From the anisotropy dependence of a crossover field in flux pinning by columnar defects, an enhancement of the pinning energy due to the reduction of anisotropy is suggested. Moreover, J_c increases with decreasing γ^2, indicating that anisotropy has a strong influence on flux pinning by columnar defects in the irreversible regime. The present study demonstrated that electromagnetic anisotropy has an extremely large influence on flux pinning not only in relatively clean crystals but also in crystals with strong pinning centers. Finally it was found that a Bi(Pb)2212 single crystal with columnar defects, lightly overdoped with oxygen showed the highest irreversibility field and the largest critical current density among Bi2212-phase compounds, which is a practical demonstration of the above conclusions.

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