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Systematic decrease of resistivity anisotropy in $Bi_2Sr_2CaCu_2O_y$ by Pb doping

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CuO_2 out-of-plane resistivity (ρ_c) and in-plane resistivity (ρ_a and ρ_b) measurements were carried out on $Bi_{2.1-x}Pb_xSr_{1.8}CaCu_2O_y$ single crystals with various Pb content, $0 \leqslant x \leqslant 0.6$. The absolute value of ρ_c in the normal state systematically decreased with increasing Pb content, whereas no appreciable change was observed in ρ_a and ρ_b by Pb doping. The resistivity anisotropy parameter $\gamma^2 \equiv \rho_c/\rho_{ab} = \rho_c/\sqrt{\rho_a\rho_b} = 8.5\times10^3$, 2.5×10^3, and 1.2×10^3 was obtained at 100 K for oxygen overdoped $x=0$, 0.3, and 0.6 crystals, respectively. These results suggest that the electrical conductivity of blocking layers is systematically enhanced by Pb doping, leading to a large reduction of the resistivity anisotropy in the present compound.

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I. INTRODUCTION

One of the most important characteristics of the high-temperature superconducting cuprates (HTSC) is the large electromagnetic anisotropy manifested in their transport and magnetic properties. This is caused by a highly two-dimensional crystal structure which consists of alternate stacking of superconducting CuO_2 and poorly conducting blocking layers. Generally, in all HTSC single crystals, the out-of-plane resistivity ρ_c, corresponding to the electrical conduction across the blocking layers, is always large with respect to the in-plane resistivity ρ_{ab} along the well-conducting CuO_2 planes. In representative compounds, such as $YBa_2Cu_3O_y$ (YBCO),[1] $(La_{1-x}Sr_x)_2CuO_4$ (LSCO),[2] and $Bi_2Sr_2CaCu_2O_y$ (Bi2212),[3] transport properties have been extensively studied on single crystals with various carrier concentrations. The important universal feature pointed out from these studies is that the anisotropy parameter $\gamma^2 \equiv \rho_c/\rho_{ab} = \rho_c/\sqrt{\rho_a\rho_b}$ in each system strongly depends on the carrier concentration.[4] The magnitude of ρ_{ab} and ρ_c always becomes smaller by increasing the carrier concentration, where ρ_c decreases much faster than ρ_{ab}, leading to the reduction of γ^2 by carrier doping. As the doping level of the crystals changes from carrier underdoped to optimal and further to overdoped states, γ^2 decreases monotonically, in contrast to the critical temperature T_c, which is a parabolic function of the doping level. The typical values of γ^2 obtained from the resistivity measurements are $10^2\sim3\times10^3$, $2\times10^2\sim5\times10^3$, and $3\times10^3\sim3\times10^4$ for YBCO (at 100 K),[1] LSCO (at 50 K),[2] and Bi2212 (at 100 K),[3] respectively. As mentioned in the later section, the absolute value of γ^2 is intimately related to the crystal structure of the materials.

Among the HTSC compounds, Bi2212 exhibits one of the largest electromagnetic anisotropy due to its thick blocking layers which consists of insulating Bi-O double layers. Although Bi2212 and its homologous compound $(Bi,Pb)_2Sr_2Ca_2Cu_3O_y$ (Bi2223) are the only materials currently utilized for wire applications at high temperatures up to liquid N_2 (77 K), their poor performance under magnetic fields still prohibits the extensive use for high-field applications. In order to overcome this problem, enormous efforts have been made for improving the critical current properties of Bi2212 and Bi2223 under magnetic fields. One of the most effective but not very practical methods has been the introduction of artificial pinning centers into the materials; drastic enhancements of the irreversibility field (H_{irr}) and the critical current density (J_{c}) were reported by heavy-ion irradiation, creating columnar defects.[5,6] However, even for such irradiated materials, degradation of pinning properties is still pronounced at higher temperatures. Substantial decreases of H_{irr} and J_{c} at high temperatures in the Bi-based superconductors are caused by the degradation of flux pinning due to their large electromagnetic anisotropy. It is thus considered that the reduction of the anisotropy is effective for improving the critical current properties of Bi2212 and Bi2223. We previously found that the irreversibility regions of Bi2212 single crystals can be systematically enhanced by reducing the electromagnetic anisotropy with increasing the carrier concentration, which can be controlled by oxygen annealing.[7] However, it has not been successful to enhance H_{irr} and J_{c} at high temperatures drastically only by the carrier doping, since the heavy carrier doping causes the decrease of T_{c} simultaneously.

Recently, Chong *et al.*[8] and Shimoyama *et al.*[9] found that the heavily Pb doped Bi2212 (nominal composition of $Bi_{1.6}Pb_{0.6}Sr_{1.8}CaCu_2O_y$) single crystals show large H_{irr} and J_{c} at high temperatures compared to Pb-free Bi2212. An increase in J_{c} by a few orders of magnitude resulting from heavy Pb doping in Bi2212 single crystals have also been reported in the previous studies.[10,11] As a possible origin of the observed superior performance, a reduced electromagnetic anisotropy caused by Pb doping has been suggested. Prior to these studies, the effect of Pb doping on the electromagnetic anisotropy of Bi2212 had been also reported in literatures. Régi *et al.* reported that ρ_c of $Bi_{1.6}Pb_{0.4}Sr_2CaCu_2O_y$ was reduced by approximately two orders of magnitude and changes the temperature dependence from semiconductive to nearly metallic.[12] Kleiner and Müller[13] and Winkeler *et al.*[14] also reported similar results. However, the results were not in a quantitative agreement, probably due to the differences in the carrier doping state between Pb-free and Pb-doped crystals. The Pb doping may also change the carrier doping state. Therefore it has not

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TABLE I. Growth conditions and chemical compositions of Bi(Pb)2212 single crystals prepared in the present study.

Sample	Starting composition Bi : Pb : Sr : Ca : Cu	Analyzed composition Bi : Pb : Sr : Ca : Cu	Pb/(Bi+Pb)	Growth rate ($\mathrm{mm\,h^{-1}}$)
$x=0$	2.1 : — : 1.8 : 1.0 : 2.0	2.1_6 : — : 1.7_0 : 0.9_4 : 2.00	0	0.2
$x=0.1$	2.0 : 0.1 : 1.8 : 1.0 : 2.0	1.9_2 : 0.0_4 : 1.7_5 : 0.9_5 : 2.00	0.018	0.5
$x=0.2$	1.9 : 0.2 : 1.8 : 1.0 : 2.0	2.0_2 : 0.1_2 : 1.8_4 : 0.9_7 : 2.00	0.056	0.5
$x=0.3$	1.8 : 0.3 : 1.8 : 1.0 : 2.0	1.8_2 : 0.2_2 : 1.8_3 : 0.9_8 : 2.00	0.108	0.5
$x=0.6$	1.6 : 0.6 : 1.8 : 1.0 : 2.0	1.6_5 : 0.4_4 : 1.8_5 : 0.9_6 : 2.00	0.211	1.0

been possible to know which is more effective for the reduction of anisotropy by Pb doping; either change in the electronic structure, or an increase in the carrier concentration.

In the present study, we have measured ρ_a, ρ_b, and ρ_c of $\mathrm{Bi_{2.1-x}Pb_xSr_{1.8}CaCu_2O_y}$ [Bi(Pb)2212] single crystals by systematically varying Pb content up to $x=0.6$. It has been found that the anisotropy parameter γ^2 of the oxygen overdoped Bi(Pb)2212 drastically decreases with increasing x. In a heavily Pb-doped crystal ($x=0.6$), γ^2 as low as 1.2×10^3 has been obtained at 100 K. This value was approximately one order of magnitude smaller than that of the oxygen overdoped Pb-free crystal ($\gamma^2=8.5\times10^3$). From the systematic resistivity measurements in the present study, it has been clarified that Pb doping in Bi2212 brings about both an increase in the carrier concentration and the enhancement of the electrical conductivity of blocking layers, leading to a large decrease in the resistivity anisotropy. It is hence strongly suggested that the reduced anisotropy leads to the observed significant improvement of the critical current properties in the heavily Pb-doped Bi2212.[8–11]

II. EXPERIMENT

Bi(Pb)2212 single crystals with various Pb contents x were grown from feed rods with nominal cation ratios of Bi : Pb : Sr : Ca : Cu = $2.1-x$: x : 1.8 : 1.0 : 2.0 by the floating zone technique using an image furnace equipped with a double ellipsoidal mirror. Details of the crystal growth are described elsewhere.[8] For Pb-rich compositions, sublimation of PbO was observed during the growth. It was therefore necessary to add some excess amount of PbO in starting compositions to compensate such losses. We hence adopted a starting composition Bi : Pb : Sr : Ca : Cu = $2.2-x$: x : 1.8 : 1.0 : 2.0 for the heavily Pb-doped crystal ($x=0.6$). The Pb-doped crystals were grown with relatively higher growth rate to suppress the sublimation loss of PbO. Starting compositions and growth rates in the present study are summarized in Table I. Cation compositions of the grown crystals were determined by ICP (inductively coupled plasma) analysis as shown in Table I. Although analyzed Pb-content Pb/(Bi+Pb) monotonically increased with increasing starting Pb composition, these values were slightly smaller compared to that of the starting compositions. Thin plate crystals with c axis along the shortest dimension (~0.1 mm) were cut from the grown boules and were cleaved to obtain fresh surfaces. Since the crystal structure of Bi2212 is also anisotropic within the ab plane, existence of the in-plane anisotropy was expected in the resistivity measurements. In order to perform the ρ_a and ρ_b measurements independently, a and b axes were determined by the x-ray back reflection Laue technique as well as a polarizing microscope. In the diffraction patterns, we could easily determine the a and b directions from superspots due to the modulated structure of Bi2212 along the b axis.[15]

Carrier concentration was controlled by post annealing the grown crystals in sealed quartz tubes under prescribed oxygen partial pressures and temperatures. Crystals with nominal compositions of $x=0$, 0.1, 0.2, 0.3, and 0.6 were treated at 400 °C, $P(\mathrm{O_2})=2.1$ atm, where $P(\mathrm{O_2})$ corresponds to the oxygen partial pressure in quartz tubes at annealing temperatures. This annealing procedure produced oxygen overdoped Bi(Pb)2212 samples. Hereafter we denote these samples ‘‘OV’’ (overdoped). For crystals with $x=0$ (Pb-free) and $x=0.6$ (the largest Pb content in the present study), we also prepared samples with lower carrier concentrations by oxygen reduced annealing. Annealing conditions 450 °C, $P(\mathrm{O_2})=2.0\times10^{-2}$ atm for $x=0$ and 600 °C, $P(\mathrm{O_2})=3.9\times10^{-4}$ atm for $x=0.6$ were selected for these two samples to produce nearly same T_c's ~85 K. From the reported relationship between annealing conditions and T_c,[9,16] these samples are considered to be in the lightly oxygen overdoped state. We denote these samples ‘‘LOV’’ (lightly overdoped).

In-plane resistivities (ρ_a and ρ_b) were measured by a standard dc four-probe technique. Figure 1(a) shows the typical geometry of the sample and electrodes. Out-of-plane resistivity (ρ_c) measurements were carried out by a dc four-probe method with current electrodes consisting of annular rings as shown in Fig. 1(b). Prior to post annealing in quartz tubes, these electrodes were set using gold paste and fired at 800 °C for 15 min to obtain good electrical contacts with the crystals. After the post annealing, gold wires of 0.05 mm ϕ were attached on these electrodes with silver paste. Following such a procedure, the contact resistance of each electrode could be suppressed to smaller than 2 Ω. Applied current used in the resistivity measurements was fixed at 10 mA in all the measurements, corresponding to the current density $j\sim10^2$ A/cm^2 for ρ_{ab} and ~1 A/cm^2 for ρ_c measurements. We have checked the reproducibility of the temperature depencences and the absolute values of ρ_a, ρ_b, and ρ_c by measuring several crystals for each composition.

III. RESULTS AND DISCUSSION

A. Out-of-plane resistivity ρ_c

Figure 2 shows the temperature dependence of ρ_c of the Bi(Pb)2212 single crystals. For the oxygen overdoped (OV)

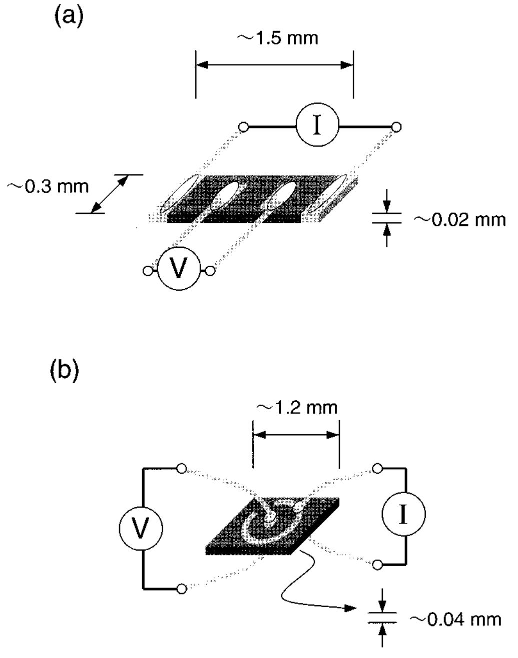

FIG. 1. Schematic drawings of the electrode configurations for (a) ρ_{ab} and (b) ρ_c measurements.

samples, shown with solid curves, the critical temperature T_c, which is defined at a midpoint of resistivity transition, systematically decreases from 79.1 to 65.1 K with increasing Pb content from $x=0$ to 0.6. The magnitude of ρ_c also systematically decreases with increasing Pb content. In the heavily Pb-doped sample $x=0.6$, the value of ρ_c is 0.21 Ω cm at 100 K, corresponding to more than one order of magnitude smaller value compared to that of $x=0$: 2.67 Ω cm at the same temperature. As mentioned later, the magnitudes of ρ_a and ρ_b were found to be nearly independent of Pb content. Therefore the present result clearly suggests that the resistivity anisotropy ($\gamma^2 \equiv \rho_c/\rho_{ab} = \rho_c/\sqrt{\rho_a \rho_b}$) systematically decreases by Pb doping. In this figure, ρ_c for samples annealed in the oxygen reduced atmosphere, $x=0$(LOV) and $x=0.6$(LOV), are also shown as broken curves. T_c values of $x=0$(LOV) and $x=0.6$(LOV) are 85.0 and 85.5 K, respectively. These LOV samples show larger ρ_c compared to the OV samples of the same Pb content. However, it should be noted that small ρ_c is exhibited in $x=0.6$(LOV): $\rho_c=1.20$ Ω cm at 100 K. This value is by a factor of ~0.2 smaller than that of the Pb-free LOV sample which has nearly the same T_c [$x=0$(LOV)] and, furthermore, smaller than that of the Pb-free OV sample [$x=0$(OV)].

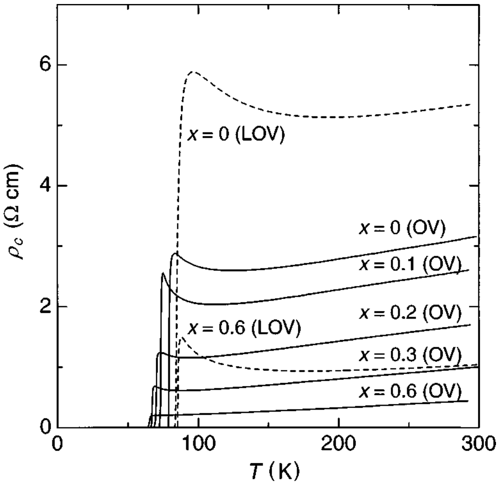

FIG. 2. Temperature dependence of ρ_c of Bi(Pb)2212. ρ_c of the oxygen overdoped (OV) and the lightly overdoped (LOV) samples are represented by solid and broken curves, respectively.

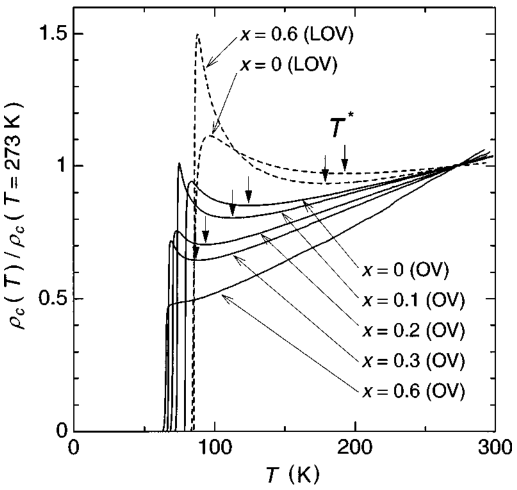

FIG. 3. Temperature dependence of normalized ρ_c of the oxygen overdoped (solid curves) and the lightly overdoped (broken curves) Bi(Pb)2212. Normalization was done with respect to $\rho_c(T=273\text{ K})$ of each crystal. The temperatures where ρ_c shows the minimum value (defined as T^*) are marked by short arrows for each crystal.

Figure 3 shows the temperature dependence of ρ_c normalized by ρ_c ($T=273$ K) of each crystal. In $x=0$(OV), the temperature dependence of ρ_c changes from metallic ($d\rho_c/dT>0$) to semiconductive ($d\rho_c/dT<0$) at $T=124.2$ K, where ρ_c shows the minimum value. We defined this temperature as T^*. A similar feature has also been reported in the previous studies of oxygen overdoped Pb-free Bi2212 single crystals.[3,17] In the present study, as the Pb content increases, the ρ_c-T curve of the OV samples becomes more metallic and T^* systematically decreases from 124.2 K ($x=0$) to 86.6 K ($x=0.3$). Finally, the sample with $x=0.6$(OV), which has the largest Pb content in the present study, shows a metallic behavior over the whole temperature range in the normal state. At temperatures higher than 150 K, ρ_c exhibits a nearly linear T dependence with a positive slope in all these oxygen overdoped samples. The temperature derivative of ρ_c at 273 K normalized by ρ_c(273 K), $(d\rho_c/dT)/\rho_c$(273 K), monotonically increases with increasing Pb content.

With decreasing the oxygen content in the crystals from OV to LOV, the ρ_c-T curve of $x=0$ becomes predominantly semiconductive. It is known that ρ_c's of both optimally carrier doped and underdoped Pb-free Bi2212 show semiconductive temperature dependence up to room temperature.[3,16] In contrast, the lightly oxygen overdoped sample $x=0$(LOV) in the present study exhibits a metallic behavior down to $T^*=192.7$ K. This result, including the absolute values of ρ_c, is in good agreement with the previous study.[17] A similar feature can be seen in the heavily Pb-doped crys-

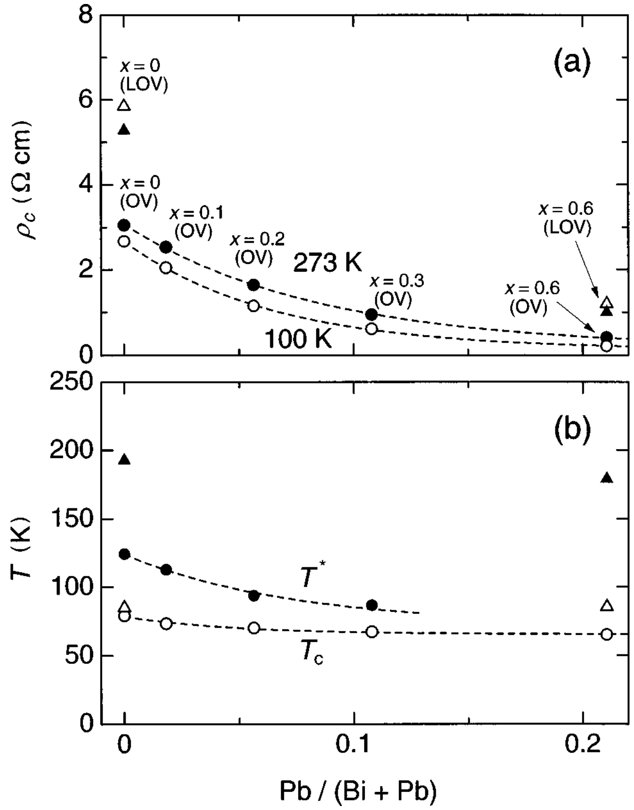

FIG. 4. Observed ρ_c values, T^*, and T_c of the oxygen overdoped (circles) and the lightly overdoped (triangles) Bi(Pb)2212 as a function of the analyzed Pb-concentration Pb/(Bi+Pb). (a) ρ_c values at 100 K (open symbols) and 273 K (closed symbols), (b) T^* (closed symbols) and T_c (open symbols). Broken curves in this figure are guides to eye.

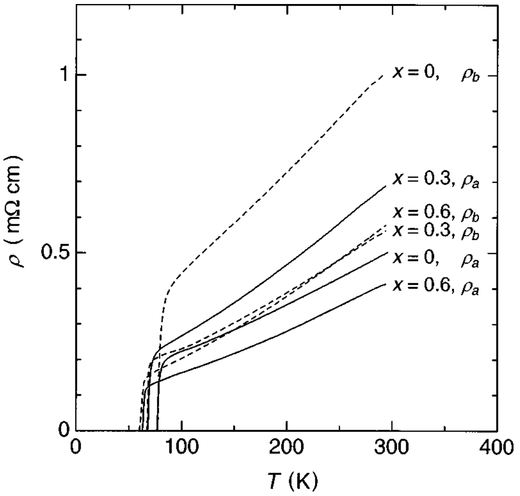

FIG. 5. Temperature dependences of ρ_a and ρ_b of the oxygen overdoped $x=0$, $x=0.3$, and $x=0.6$. Solid and broken curves represent ρ_a and ρ_b, respectively.

tals $x=0.6$(LOV), which exhibits relatively higher $T^*=178.8$ K despite its small ρ_c. Such a strongly semconductive behavior has also been reported in other HTSC compounds such as LSCO and YBCO which have much smaller ρ_c values.[18] The temperature dependence of ρ_c changes from metallic to semiconductive by decreasing the carrier concentration in both systems of LSCO ($\rho_c=10^{-2}\sim10^{0}$ Ω cm at 100 K) and YBCO ($\rho_c=10^{-3}\sim10^{-2}$ Ω cm at 100 K). The ρ_c behavior with temperature is strongly affected by the carrier concentration, and seems to be almost independent of the absolute value of ρ_c. Note that the ρ_c-T curves of $x=0$(LOV) and $x=0.6$(LOV) show a similar tendency despite a large difference in the magnitude of ρ_c by a factor of 5. In Fig. 4, ρ_c values, T^*, and T_c of each sample in the present study are plotted as a function of the analyzed Pb-concentration Pb/(Bi+Pb). All these values systematically change with increasing Pb concentration. Observed ρ_c values, T^*, and T_c are summarized in Table II.

B. In-plane resistivities ρ_a and ρ_b

Figure 5 shows the temperature dependence of ρ_a and ρ_b of the oxygen overdoped $x=0$, $x=0.3$, and $x=0.6$ crystals, where solid and broken curves represent ρ_a and ρ_b, respectively. In all the crystals examined, the values of ρ_a and ρ_b are all in the order of 10^{-4} Ω cm at 100 K; no appreciable change was observed by Pb doping less than a factor of 2. This feature is very contrastive to that of ρ_c, where resistivity at 100 K is reduced by more than one order of magnitude by Pb doping. To emphasize this difference, logarithmic plots of ρ_a, ρ_b, and ρ_c for OV samples are shown in Fig. 6. A drastic change by Pb doping can be seen only in the ρ_c-T curves.

For Pb-free Bi2212, the distinct difference between ρ_a and ρ_b has been reported previously,[3,19–21] whereas there is no report concerning the in-plane anisotropy of Pb-doped Bi2212. In the present study, Pb-free crystal $x=0$ shows larger ρ_b than ρ_a; e.g., $\rho_b/\rho_a=2.0$ at 100 K, clearly suggesting the existence of the in-plane anisotropy. Such a

TABLE II. Observed ρ_c values at 100 and 273 K, T^* and T_c of Bi(Pb)2212.

Sample	ρ_c(273 K) / Ω cm	ρ_c(100 K) / Ω cm	T^* / K	T_c / K
$x=0$ (OV)	3.05	2.67	124.2	79.1
$x=0.1$ (OV)	2.53	2.05	112.7	73.3
$x=0.2$ (OV)	1.64	1.15	93.6	70.0
$x=0.3$ (OV)	0.946	0.615	86.6	67.2
$x=0.6$ (OV)	0.416	0.214	—	65.1
$x=0$ (LOV)	5.28	5.85	192.7	85.0
$x=0.6$ (LOV)	1.00	1.20	178.8	85.5

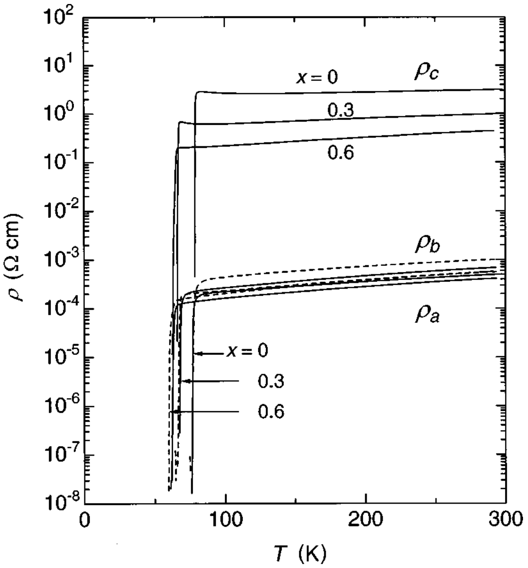

FIG. 6. Temperature dependences of ρ_a, ρ_b, and ρ_c of the oxygen overdoped $x=0$, $x=0.3$, and $x=0.6$. The top three curves represent ρ_c-T. ρ_a-T (solid curves) and ρ_b-T (broken curves) are shown at the lower part of the figure.

larger resistivity along the b axis has also been reported by several groups.[20–22] In contrast to this Pb-free Bi2212, observed difference between ρ_a and ρ_b is rather small in the Pb-doped crystals; $\rho_b/\rho_a(100\ \mathrm{K})=0.9$ and 1.3 for $x=0.3$ and $x=0.6$, respectively. Therefore the in-plane resistivity of Pb-doped Bi2212 is more isotropic compared to that of the Pb-free Bi2212. Note that only ρ_b of $x=0$ shows large values among ρ_{ab}-T curves in Fig. 5.

Pb-free Bi2212 exhibits a modulated crystal structure along the b axis with a modulation period $q_b \sim 4.8b$, where b is the lattice parameter (~ 5.4 Å).[15] The anisotropic in-plane resistivity of Bi2212 may be related to the modulated structure in this compound, because the corrugated CuO_2 planes along the b axis would give a larger effective mass of the carrier, resulting in a larger resistivity along this direction. It is also known that the modulation period q_b is increased by Pb doping, for example, $q_b=8.2b$ and $9.3b$ when analyzed Pb concentration Pb/(Bi+Pb) are 0.18 and 0.20, respectively,[23] i.e., leading to less corrugated CuO_2 planes by Pb doping. Such structural change would be an origin of the smaller in-plane anisotropy ρ_b/ρ_a observed in the Pb-doped Bi2212 crystals.

C. Electromagnetic anisotropy

The anisotropy parameter $\gamma^2 \equiv \rho_c/\rho_{ab} = \rho_c/\sqrt{\rho_a \rho_b}$ of the oxygen overdoped $x=0$, $x=0.3$, and $x=0.6$ crystals can be calculated using the measured ρ_a, ρ_b, and ρ_c values. In Table III, calculated γ^2 at 100 and 273 K are summarized. γ^2 of $x=0.6$ is approximately one order of magnitude smaller than that of $x=0$. Since the magnitude of ρ_{ab} is not very sensitive to the compositional change, γ^2 is nearly proportional to the ρ_c value. Figure 7 shows the temperature dependence of the calculated anisotropy parameter γ^2. Short arrows are marked at T_c of each sample. The magnitude of γ^2 decreases with increasing Pb content over the whole temperature range in the normal state. $x=0$ and $x=0.3$ crystals exhibit a strong upturn of γ^2 near T_c which is due to the semiconductive ρ_c behavior. In contrast, γ^2 of $x=0.6$ is almost independent of temperature, reflecting a nearly metallic behavior along the c axis even near T_c. In this figure, γ^2 of Pb-free and Pb-doped Bi2212 by Régi *et al.*[12] are also shown with broken curves. From the reported T_c, absolute value of ρ_c, and temperature dependence of ρ_c, we believe that the Pb-free crystal in Ref. 12 is slightly carrier underdoped, while the Pb-doped one, which has a similar analyzed Pb concentration of the $x=0.6$ crystal in the present study, is carrier overdoped. It is considered that drastic reduction of γ^2 by Pb doping in Ref. 12 was caused by the large difference in the carrier doping level between ''carrier underdoped'' Pb-free and ''carrier overdoped'' Pb-doped crystals. These results are thus quite consistent with the present result taking the carrier doping state of each crystal into account.

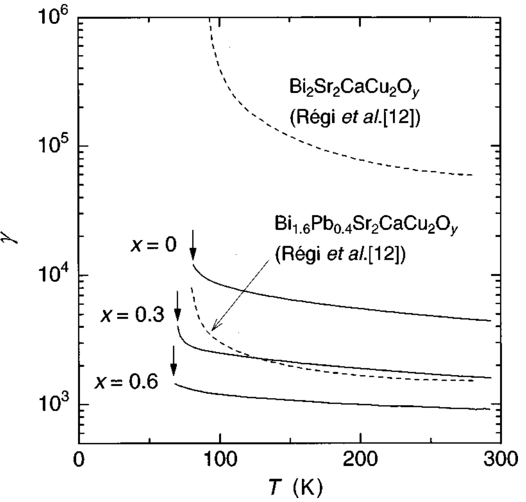

FIG. 7. Temperature dependence of γ^2 of the oxygen overdoped $x=0$, $x=0.3$, and $x=0.6$ (solid curves). Short arrows are marked at T_c of each sample. γ^2 of Pb-free and Pb-doped Bi2212 by Régi *et al.* (Ref. 12) are also shown as broken curves.

There is an alternative way for estimating the electromagnetic anisotropy of Bi2212. In magnetization measurements

TABLE III. Resistivity values and the calculated γ^2 of the oxygen overdoped Bi(Pb)2212 with $x=0$, $x=0.3$, and $x=0.6$.

	100 K				273 K			
Sample	ρ_a (mΩ cm)	ρ_b (mΩ cm)	ρ_c (Ω cm)	γ^2	ρ_a (mΩ cm)	ρ_b (mΩ cm)	ρ_c (Ω cm)	γ^2
$x=0$(OV)	0.224	0.442	2.67	8.5×10^3	0.462	0.950	3.05	4.6×10^3
$x=0.3$(OV)	0.267	0.230	0.615	2.5×10^3	0.641	0.523	0.946	1.6×10^3
$x=0.6$(OV)	0.163	0.203	0.214	1.2×10^3	0.383	0.533	0.416	9.2×10^2

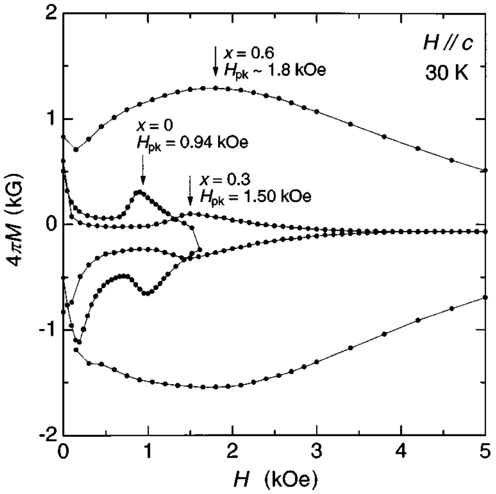

FIG. 8. M-H hysteresis loops of the oxygen overdoped $x=0$, $x=0.3$, and $x=0.6$ at 30 K. The second peak fields H_{pk} of each crystal are marked by arrows.

with magnetic fields parallel to the c axis, a characteristic second peak is known to appear in the M-H hysteresis loops at low temperatures. As one of the scenarios for the origin of this peak effect, the dimensional crossover of the vortex lattice has been proposed.[24] In this mechanism, the crossover field $H_{\text{2D-3D}}$ is related to the anisotropy parameter $\gamma^2 \equiv m_c^*/m_{ab}^*$ in the superconducting state; $\mu_0 H_{\text{2D-3D}} = \Phi_0/\gamma^2 s^2$, where Φ_0 and s are the flux quantum and the superconducting layer spacing, respectively. In our previous study, magnetization and resistivity measurements were systematically carried out on Pb-free Bi2212 single crystals with various oxygen content obtained from the same grown boule.[3,25] The γ^2 values estimated from the second peak fields were semiqualitatively in good agreement with the resistivity anisotropy parameters caluculated from the ρ_a, ρ_b, and ρ_c values.[4] Figure 8 shows the M-H hysteresis loops of our oxygen overdoped $x=0$, $x=0.3$, and $x=0.6$ crystals at 30 K. From these loops, $\mu_0 H_{\mathrm{pk}} = 0.094$ T for $x=0$ and 0.150 T for $x=0.3$ are obtained. Since the observed H_{pk}'s of these crystals are almost independent of temperature, we can calculate $\gamma^2(H_{\mathrm{pk}}) \equiv \Phi_0/(s^2 \mu_0 H_{\mathrm{pk}}) = 9.3\times 10^3$ and 5.8×10^3 for $x=0$ and $x=0.3$, respectively, assuming H_{pk} to correspond to $H_{\text{2D-3D}}$, where $s=15.4$ Å was used. The estimated $\gamma^2(H_{\mathrm{pk}})$ of $x=0$ is in good agreement with that obtained from the resistivity data at 100 K ($\gamma^2 = 8.5\times 10^3$). On the other hand, $\gamma^2(H_{\mathrm{pk}})$ of $x=0.3$ is rather larger than γ^2 from the resistivity measurements at 100 K, but well agrees with that near T_{c} ($\gamma^2 = 4\times 10^3 \sim 5\times 10^3$ around 70 K, see Fig. 7). For $x=0.6$, $\gamma^2(H_{\mathrm{pk}}) = 4.8\times 10^3$ was also obtained from $\mu_0 H_{\mathrm{pk}} = 0.18$ T at 30 K. However, H_{pk} strongly depends on temperature in whole temperature range below T_{c}, suggesting a different mechanism for the origin of the second peak effect in such heavily Pb-doped crystals.[9,26] Therefore, for $x=0.6$, there would be large ambiguity in the estimated $\gamma^2(H_{\mathrm{pk}})$.

As mentioned in the Introduction, the possibility of the decrease in the anisotropy of Bi2212 by Pb doping was reported in the previous literatures.[12–14] However, the mechanism of reducing anisotropy has not been well understood. Two possible explanations for the decrease of the anisotropy in the Pb-doped Bi2212 are considered, i.e., (1) change in the electronic structure, and (2) increase of the carrier concentration by Pb doping. The systematic investigation in the present study revealed that the magnitude of ρ_c (or anisotropy parameter γ^2) decreases with increasing Pb content, and T_{c} decreases simultaneously after annealing under the same condition (Fig. 4 and Table II). This result suggests that the substitution of Bi^{3+} for Pb^{2+} in Bi-O double layers contributes to the increase in the carrier (hole) concentration of the $\mathrm{CuO_2}$ planes. This is consistent with the fact that the heavily Pb-doped Bi2212 crystals require strong oxygen reduced conditions for obtaining the carrier lightly overdoped samples having the same $T_{\mathrm{c}} \sim 85$ K compared to Pb-free crystals. The anisotropy of Bi2212 systematically decreases with increasing the carrier concentration by oxygen annealing.[3,7] Therefore the decrease of the resistivity anisotropy in Pb-doped Bi2212 may be partly caused by the increase of the carrier concentration through Pb doping. However, the heavily Pb-doped crystal with $T_{\mathrm{c}} = 85.5$ K [$x=0.6$(LOV)] shows much smaller ρ_c compared to the Pb-free one with $T_{\mathrm{c}} = 85.0$ K [$x=0$(LOV)]. From the similarity of T_{c} between these crystals, they are thought to have nearly the same effective carrier concentration. Therefore it is strongly suggested that Pb doping substantially reduces the resistivity anisotropy of Bi2212 even with the same carrier concentration of the $\mathrm{CuO_2}$ planes. We believe Pb doping into Bi-O double layers of Bi2212 enhances the electrical conductivity of these layers, resulting in the reduction of the resistivity anisotropy. It should be noted that the absolute value of ρ_c of Pb-doped Bi2212 is strongly affected by the oxygen content similarly as in the Pb-free Bi2212. The quantitative disagreement concerning the anisotropy of Bi(Pb)2212 in the previous literatures[12–14] would have been simply caused by the difference in the carrier doping state of the samples.

One of the present authors has previously found that the anisotropy of HTSC is strongly associated with the distance between $\mathrm{CuO_2}$ planes [$d(\mathrm{CuO_2}\text{-}\mathrm{CuO_2})$]; a material with larger $d(\mathrm{CuO_2}\text{-}\mathrm{CuO_2})$ shows larger γ^2.[4] For example, the electromagnetic anisotropy of carrier optimally doped Bi2212 ($\gamma^2 \sim 2\times 10^4$), $\mathrm{HgBa_2Ca_2Cu_3O_y}$ ($\sim 2.5\times 10^3$), and LSCO ($\sim 2.5\times 10^2$) is systematically reduced by decreasing $d(\mathrm{CuO_2}\text{-}\mathrm{CuO_2})$, 12.1, 9.5, and 6.6 Å, respectively. From this relationship, an extensive control of the anisotropy seems to be rather difficult because $d(\mathrm{CuO_2}\text{-}\mathrm{CuO_2})$ is essentially governed by the basic crystal structure. Therefore we emphasize that the observed $\gamma^2 = 1.2\times 10^3$ of $x=0.6$(OV) is an extraordinary small value for the Bi-based superconductors which contain thick blocking layers. X-ray-diffraction analyses revealed that all Bi(Pb)2212 single crystals used in the present study have nearly the same lattice parameters $c_0 = 30.65 \sim 30.70$ Å, suggesting that the structural change by Pb doping is negligible. Thus Pb doping reduces the anisotropy of Bi2212 without changing the superconducting layer spacing appreciably.

IV. CONCLUSION

In order to elucidate the effect of Pb doping on the anisotropy of $\mathrm{Bi_2Sr_2CaCu_2O_y}$ (Bi2212), out-of-plane resistivity (ρ_c), and in-plane resistivity (ρ_a and ρ_b) of

$Bi_{2.1-x}Pb_xSr_{1.8}CaCu_2O_y$ single crystals with various Pb content ($0 \leqslant x \leqslant 0.6$) have been measured. The absolute value of ρ_c in the normal state systematically decreased with increasing Pb content. On the other hand, no appreciable change was observed in ρ_a and ρ_b by Pb doping. Anisotropy parameter $\gamma^2 \equiv \rho_c/\rho_{ab} = \rho_c/\sqrt{\rho_a\rho_b} = 8.5\times10^3$, 2.5×10^3, and 1.2×10^3 was obtained at 100 K for the oxygen overdoped $x = 0$, 0.3, and 0.6 crystals, respectively. The present results suggest that the electrical conductivity of the blocking layers in Bi2212 is systematically enhanced by Pb doping, leading to a large reduction of the resistivity anisotropy. We believe this study shows that the anisotropy can be systematically reduced by chemical substitutions into blocking layers of the Bi2212 compound. The present result is of particular importance for material developments because it has shown the possibility that the critical current properties can be drastically improved by chemical substitutions into blocking layers. A smaller in-plane resistivity anisotropy $\rho_b/\rho_a \sim 1$ was observed in Pb-doped crystals compared to Pb-free one ($\rho_b/\rho_a = 2.0$). The change in the anisotropic value of the in-plane resistivity by Pb doping would be explained by less anisotropic in-plane crystal structure of Pb-doped Bi2212.

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