

PCP5-1

Cu hyperfine coupling constants of $\text{HgBa}_2\text{CaCu}_2\text{O}_{6+\delta}$

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The ratios of ^{63}Cu hyperfine coupling constants in the double-layer high- T_c superconductor $\text{HgBa}_2\text{CaCu}_2\text{O}_{6+\delta}$ are estimated from the anisotropies in Cu nuclear spin-lattice relaxation rates and spin Knight shifts to study the nature of the ultraslow fluctuations causing the T_2 anomaly in the Cu nuclear spin-echo decay [1]. The ultraslow fluctuations may come from magnetic fluctuations spread over a region around the wave-vector $q = 0$, otherwise charge/phononic fluctuations.

[1] Y. Itoh, T. Machi, and A. Yamamoto: Phys. Rev. B **95**, 094501 (2017).

Keywords: Hg1212, NMR

PCP5-2

Rare-earth dependence of in-plane anisotropy of resistivity in Bi2201 series high temperature superconductors

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The presence of a pseudogap phase in the phase diagram of copper oxide high temperature superconductors is well established, where the density of states at the Fermi energy is partially depleted above the superconducting transition temperature. The origin of the pseudogap and its relation to superconductivity have been actively discussed. It was reported that the in-plane four-fold rotational (C_4) symmetry is spontaneously broken in the pseudogap phase. Measuring the resistivity anisotropy is one of the simplest ways to verify the electronic anisotropy arising from the broken C_4 symmetry. Indeed, it was reported that the in-plane resistivity is highly anisotropic for underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7+\delta}$ [1].

In $\text{Bi}_2\text{Sr}_2\text{CuO}_{6+\delta}$ (Bi2201), the superconducting transition temperature and pseudogap formation temperature (T^*) depend on the rare-earth element substituted for Sr even when the same amount is doped [2]. Hence, we can prepare samples that have the same hole concentration but different T^* , and it would be interesting to study the in-plane anisotropy and compare it with T^* . For this purpose, we measured the in-plane anisotropy of resistivity following the method reported in Ref. [3] on single crystals of Bi2201 doped with different rare earth elements. In a sample doped with 30% of La, we observed that the in-plane anisotropy increased with lowering the temperature, suggesting the breaking of C_4 symmetry. This is in a sharp contrast with a sample doped with 10% of La which showed no enhancement of the resistivity anisotropy. Based on our experimental results, we will discuss the relationship between T^* and the breaking of C_4 symmetry.

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[3] P. Walmsley and I. R. Fisher, *Rev. Sci. Instrum.* **88**, 043901 (2017).

Keywords: Cuprate, Pseudogap, Nematicity, Transport

PCP5-3

Effect of Ba-substitution for Sr in the Bi-2201 Phase of $\text{BiPb}(\text{Sr},\text{La})_2\text{CuO}_{6+\delta}$

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The highest value of the superconducting transition temperature T_c in the Bi-2201 phase is ~ 40 K for $(\text{Bi},\text{Pb})_2(\text{Sr},\text{La})_2\text{CuO}_{6+\delta}$, which is much lower than ~ 90 K in the Tl-2201 phase of $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$ [1,2]. Considering the empirical rule that T_c increases with an increase of the distance between Cu and O, the reason for the lower T_c of the Bi-2201 phase is because larger Ba^{2+} ions in the Tl-2201 phase are replaced by smaller Sr^{2+} ions in the Bi-2201 phase. In this research, we have attempted to increase T_c of the Bi-2201 phase by extending the Cu-O distance through the substitution of Ba for Sr in $(\text{Bi},\text{Pb})_2(\text{Sr},\text{La})_2\text{CuO}_{6+\delta}$.

Polycrystalline samples of $\text{BiPbSr}_{1.15-x}\text{Ba}_x\text{La}_{0.85}\text{CuO}_{6+\delta}$ were prepared by the solid-state reaction method. The iodometric titration was carried out to estimate the oxygen content. Magnetic susceptibility measurements were performed to determine T_c using a SQUID magnetometer. Measurements of the thermoelectric power at room temperature were carried out to estimate the hole-concentration in the CuO_2 plane [3].

The hole-concentration in the CuO_2 plane is almost constant despite the increase in the oxygen content $6+\delta$ with increasing $x(\text{Ba})$, as shown in Fig. 1. These results indicate that holes are supplied to Bi(Pb)-O layers rather than to the CuO_2 plane with increasing $x(\text{Ba})$. With increasing $x(\text{Ba})$, T_c does not increase but tends to decrease slightly. The decrease in T_c with increasing $x(\text{Ba})$ may be due to the local distortion caused by excess oxygen.

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Keywords: Bi-2201, Ba-substitution, Superconducting transition temperature

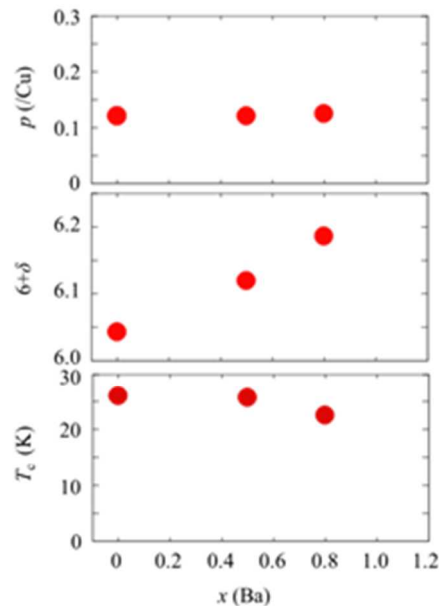


Fig. 1: Dependence of T_c , the oxygen content $6+\delta$ and the hole concentration in the CuO_2 plane p on the Ba content x in $\text{BiPbSr}_{1.15-x}\text{Ba}_x\text{La}_{0.85}\text{CuO}_{6+\delta}$.

PCP5-4

Superconductivity above 100 K in the Bi-2212 Phase of $(\text{Bi,Pb})_2\text{Sr}_2\text{CaCu}_2\text{O}_8$

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The high- T_c cuprate superconductor of the Bi-2212 phase, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$, with $T_c \sim 80$ K prepared in air is situated in the overdoped region, because excess oxygen atoms are easily introduced in the BiO plane. Therefore, its T_c increases up to about 95 K by the decrease of hole concentration through the reduction annealing or the substitution of Y^{3+} for Ca^{2+} [1,2]. In addition, the highest T_c of 98.5 K is observed in $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{CaCu}_2\text{O}_8$ annealed in flowing gas of N_2 [3]. This increase in T_c is because the excess oxygen atoms causing local distortion are removed through the Pb-substitution and N_2 -annealing. In this research, we have attempted to increase T_c further in Pb-substituted $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{CaCu}_2\text{O}_8$ without excess oxygen by optimizing the Pb content, namely, the hole concentration.

Polycrystalline samples of $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{CaCu}_2\text{O}_8$ ($0.32 \leq x(\text{Pb}) \leq 0.42$) were prepared by the conventional solid-state reaction method. The structural analysis was performed by the powder x-ray diffraction. Magnetic susceptibility measurements were carried out to determine T_c using a SQUID magnetometer.

Figure 1 shows the temperature dependence of the magnetic susceptibility for $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{CaCu}_2\text{O}_8$, or magnetic susceptibility rease in , in whic rconductors. or Sr has not succeeded yet, but their T_c . It has been found that the maximum value of T_c , defined as the onset temperature of the shielding effect, is 102 K for $x(\text{Pb})=0.36$. The present compound, or magnetic susceptibility rease in , in whic rconductors. or Sr has not succeeded yet, but their T_c is the first Bi-2212 phase with T_c above 100 K.

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[3] S. Kambe *et al.*, Phys. Rev. B **42** (1990) 2669.

Keywords: Bi-2212, Pb-substitution, Superconducting transition temperature

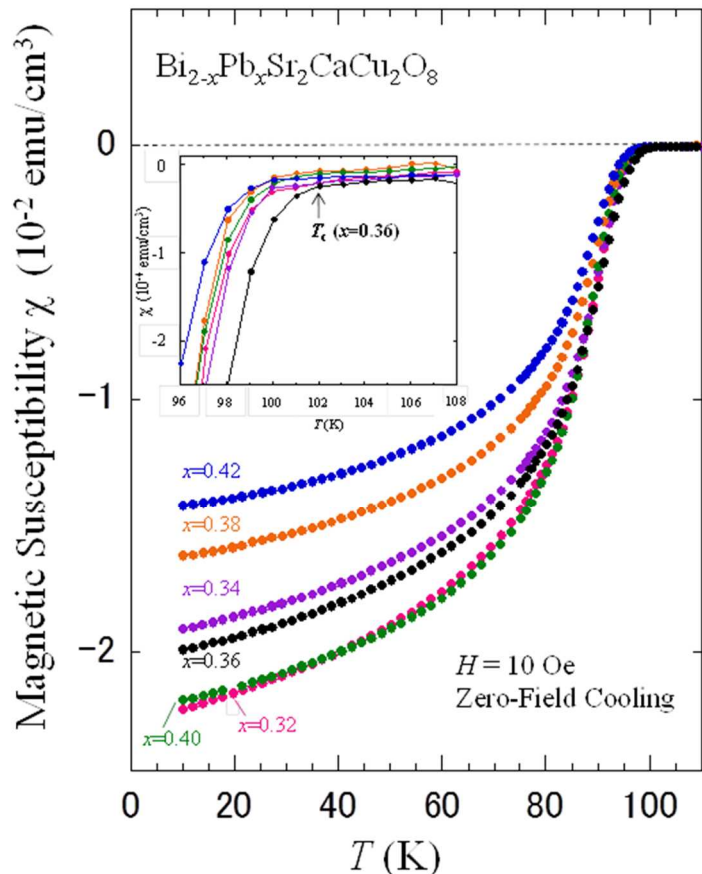


Fig. 1. Temperature dependence of the magnetic susceptibility for $\text{Bi}_{2-x}\text{Pb}_x\text{Sr}_2\text{CaCu}_2\text{O}_8$.

PCP5-5

Preparation of (11*n*) Oriented Bi₂Sr₂CaCu₂O_{8+x} Thin Films by Solution Methods using NdGaO₃ (100) Substrates

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Bi₂Sr₂CaCu₂O_{8+x} (Bi2212) has a layered perovskite structure and forms an intrinsic Josephson junction (IJJ). This is considered to be a promising device that fills a frequency domain called the "terahertz gap". It has already been proved that a μ W class continuous coherent terahertz wave can be oscillated [1]. The major preparation methods thus far have been reported on *c*-axis oriented Bi2212 which requires complicated procedures and dry etching, such as a precisely controlled etching process in depth direction in order to form the *c*-axis current paths. If a non-*c*-axis oriented thin film of which the *c*-axis parallel or incline to the substrate can be prepared, a planar type IJJ device can be fabricate simply by forming current paths parallel to the substrate [2]. The reason is that the *c*-axis current component is included in the current paths. In preparing such non-*c*-axis oriented thin films, it is important that selection of substrates focusing on lattice matching. If it is possible to prepare a non-*c*-axis oriented thin film by a solution method, planar type IJJ device can be fabricated by a simple process such as heat treatment after applying a solution by a printing method. According to our previous reports [3], (117) oriented Bi2212 thin films were formed by using SrTiO₃ (110) substrates metal-organic decomposition (MOD) method. In this study, we will report fabrication of (11*n*) oriented Bi2212 thin film using NdGaO₃ (100) substrate by the MOD method and flux method. Evaluation of the thin film was carried out through X-ray diffraction pattern and pole figure, observation of surface morphology by scanning electron microscope image, measurement of resistivity temperature dependency.

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Keywords: BSCCO, Solution Method, Non-*c*-axis Oriented

PCP5-6

Intermediate Phase Evolution of YBCO Superconducting Film Fabricated by Fluorine Free MOD Method

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The second generation of high temperature superconducting tape, which use ReBCO film as superconducting carrier layer, has high superconducting current carrying capacity under strong field and good mechanical properties relative to other practical superconducting wire / tape, is considered to have Good application prospects. In its preparation process, Fluorine Free-Metal Organic Deposition has been studied in recent years because of its no need of vacuum environment, pyrolysis and crystallization growth rate, and environmental friendliness. In this paper, YBCO films were prepared on the single crystal and CeO₂ / IBAD-MgO / Y₂O₃ / Al₂O₃ / Hastelloy C276 technical substrate by FF-MOD technique. The samples were quenched and characterized in to systematically study the process of its phase evolution, it is presumed that BaCO₃ does not react directly with the oxides of Y and Cu elements at the time of phase formation, but undergoes a more complicated intermediate process. The YBCO phase crystals are randomly oriented, and then gradually converted into a biaxially textured YBCO crystal.

Keywords: 2G-HTS, YBCO, FF-MOD, phase evolution

PCP5-7

Superconductivity and magnetism in lanthanoid-substituted $\text{FeSr}_2\text{YCu}_2\text{O}_{6+\delta}$

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$\text{FeSr}_2\text{YCu}_2\text{O}_{6+\delta}$ has tetragonal $\text{Ba}_2\text{YCu}_3\text{O}_{6+\delta}$ -type structure and exhibits superconductivity on the CuO_2 layer at 60 K and antiferromagnetic order of Fe at 20 K [1]. The lanthanoid atoms from Nd to Yb can be substituted for Y in $\text{FeSr}_2\text{YCu}_2\text{O}_{6+\delta}$ similarly to $\text{Ba}_2\text{YCu}_3\text{O}_{6+\delta}$, and additionally Tb can be substituted for Y in $\text{FeSr}_2\text{YCu}_2\text{O}_{6+\delta}$ while $\text{Ba}_2\text{TbCu}_3\text{O}_{6+\delta}$ is not formed. T_c decreases with increasing the ionic size of the lanthanoid atoms, and $\text{FeSr}_2\text{NdCu}_2\text{O}_{6+\delta}$ does not exhibit superconductivity although the oxygen content, $6+\delta$, is not mostly changed by the substitution of the lanthanoid atoms. The magnetic order of Fe is independent of the substitution of the lanthanoid atoms except for Gd, Tb and Dy. $\text{FeSr}_2\text{TbCu}_2\text{O}_{6+\delta}$ exhibit the magnetic order of Tb at 7 K.

[1] T. Mochiku *et al.*, J. Phys. Soc. Jpn. 71 (2002) 790.

Keywords: $\text{FeSr}_2\text{YCu}_2\text{O}_{6+\delta}$, lanthanoid substitution, magnetic order

PCP5-8

Dependence of T_c on the RE -ion Size in $(RE,Ca)Ba_2Cu_3O_6$

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According to the empirical rule of high- T_c for cuprate superconductors, the superconducting transition temperature T_c increases with an increase of the distance between Cu in the CuO_2 plane and the so-called apical-oxygen O_{ap} . The $Cu(2)$ - O_{ap} distance in $REBa_2Cu_3O_6$ (RE : rare-earth elements) without the oxygen O_{chain} in the $Cu(1)O$ chain is longer than that in $REBa_2Cu_3O_7$ with $T_c \sim 90$ K [1], so that its T_c is expected to increase up to more than 90 K. However, $REBa_2Cu_3O_6$ is an insulator because of the lack of O_{chain} which supplies holes to the $Cu(2)O_2$ plane. Therefore, we have carried out the hole-doping through the partial substitution of Ca^{2+} for RE^{3+} in insulating $REBa_2Cu_3O_6$.

Polycrystalline samples of $Gd_{0.6}Ca_{0.4}Ba_2Cu_3O_{6+\delta}$, $Yb_{0.6}Ca_{0.4}Ba_{1.6}Sr_{0.4}Cu_3O_{6+\delta}$ and $Lu_{0.6}Ca_{0.4}Ba_{1.6}Sr_{0.4}Cu_3O_{6+\delta}$ were prepared by the solid-state reaction method. The partial substitution of Sr^{2+} for Ba^{2+} was done in order to extend the solid solubility limit of Ca^{2+} for $RE = Yb^{3+}$ and Lu^{3+} . The iodometric titration was carried out to estimate the oxygen content. Magnetic susceptibility measurements were performed to determine T_c using a SQUID magnetometer. The dependence of T_c on the oxygen content $6+\delta$ is shown in Fig. 1. At $6+\delta \sim 6.0$, it is found that T_c tends to increase as the ionic radius of RE decreases. T_c is 55 K in $Lu_{0.6}Ca_{0.4}Ba_{1.6}Sr_{0.4}Cu_3O_{6+\delta}$ with $6+\delta \sim 6.0$, while no superconductivity appears in $Gd_{0.6}Ca_{0.4}Ba_2Cu_3O_{6+\delta}$ with $6+\delta \leq 6.3$. This is presumably because holes are more supplied to $Cu(1)$ than to the $Cu(2)O_2$ plane with increasing ionic radius of RE . In the present work, the maximum T_c of $(RE,Ca)Ba_2Cu_3O_6$ is considerably lower than 90 K. The hopeful way to increase T_c further is discussed.

[1] M. Guillaume *et al.*, Z. Phys. B **6** (1994) 7963.

Keywords: $REBa_2Cu_3O_6$, Ca-Substitution, hole doping

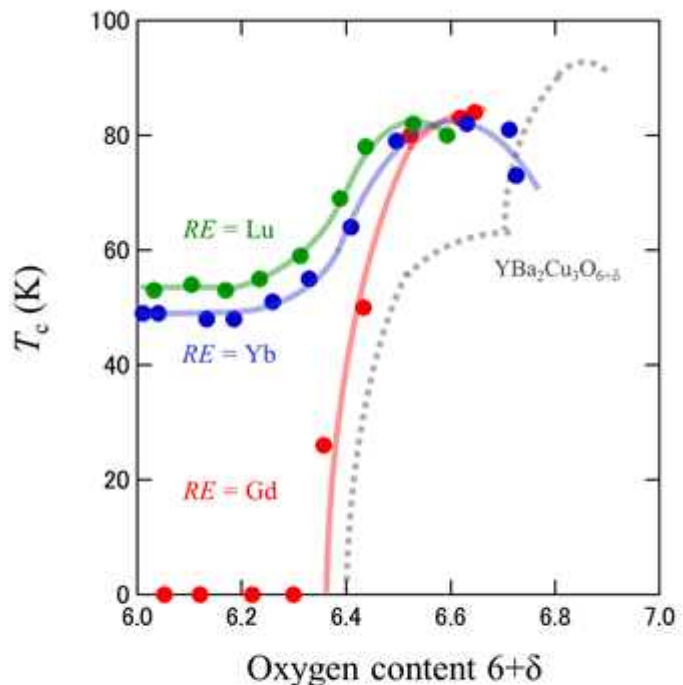


Fig. 1. Dependence of T_c on the oxygen content $6+\delta$ in $RE_{0.6}Ca_{0.4}(Ba,Sr)_2Cu_3O_{6+\delta}$.

PCP5-9

Dependence of critical temperature on chemical composition in $Y(\text{Sr},\text{Ba})_2(\text{Cu},\text{Mo})_3\text{O}_z$ ($z \sim 7$)

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Sr-substitution for Ba in $\text{YBa}_2\text{Cu}_3\text{O}_z$ ($z \sim 7$; Y-"1-2-3") has been widely investigated and its superconductivity transition temperature (T_c) has proved to lower as Sr-content increases. Solubility of Sr is about 60 at.% [1] and fully substituted compounds, $\text{YSr}_2\text{Cu}_3\text{O}_z$ has been synthesized only under high-pressure [2]. Den and Kobayashi [3] have reported that partial substitution of Mo for Cu stabilizes the Sr-based Y-"1-2-3", *i.e.*, $\text{YSr}_2(\text{Cu}_{2.7}\text{Mo}_{0.3})\text{O}_z$ is obtained as a single "1-2-3" phase. It is interesting that, on the other hand, effort of Mo substitution for Cu in the Ba-based one has been unsuccessful regardless of the chemical similarity between Ba and Sr. In this study, we report the effect of Sr/Ba ratio on the "1-2-3" formation in $Y(\text{Sr},\text{Ba})_2(\text{Cu},\text{Mo})_3\text{O}_z$ and composition dependence on T_c . Results are discussed mainly based on a high valency of Mo and effective ionic radius of Sr/Ba site.

Samples were prepared by a solid-state reaction of oxides and carbonates of each metallic element. Nominal compositions of $Y(\text{Sr}_{1-y}\text{Ba}_y)_2(\text{Cu}_{1-x}\text{Mo}_x)_3\text{O}_z$ ($x=0 \sim 0.5$, $y=0 \sim 1$) was used. Samples were evaluated by means of powder X-ray diffractometry (XRD) and four-probe method down to ~ 20 K respectively for phase identification and for a measurement of temperature dependence of electrical resistivity.

For Mo-free compositions ($x=0$), single "1-2-3" were not obtained for $x < 0.5$ as described in [1], but it was found that Mo-substitution for Cu ($x < 0.4$) moved the single-phase compositional range to Sr-rich region, *i.e.*, y values giving single-"1-2-3" samples were decreased as x increased up to ~ 0.3 . This suggested that an effective ionic radius of Sr/Ba site and high valence-state of Mo (6+; confirmed by X-ray photoemission spectroscopy) played a crucial role for the stability of the Sr-based "1-2-3". Dependence of T_c on Ba content y was clearly different for a range of Mo content x of $x \leq 0.1$ ($T_c > \sim 60$ K) and $x \geq 0.2$ ($T_c < \sim 50$ K). This will be discussed on the stand point of oxygen configuration in the "charge reservoir" layer.

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Keywords: Sr-based "1-2-3", Mo substitutin for Cu, oxgen configuration