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# Vortex pinning properties of $(Y_{1-x}La_x)$ -Ba-Cu-O and $(Y_{1-x}Pr_x)$ -Ba-Cu-O superconducting bulks

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## Abstract

We have studied the effect of a small amount of Y-site substitution by La or Pr ions on the vortex pinning in the Y-Ba-Cu-O system.  $(Y_{1-x}La_x)$ -Ba-Cu-O and  $(Y_{1-x}Pr_x)$ -Ba-Cu-O bulks were fabricated by the melt-textured growth, in which xwas varied from 0 to 0.01. The critical current density  $J_c$  at 77 K is improved in magnetic fields parallel to the *c*-axis above 2-4.5 T and the corresponding irreversibility field,  $H_{irr}$ , shifts to the higher value in both bulks.

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 $(Y_{1-x}La_x)$ -Ba-Cu-O bulk;  $(Y_{1-x}Pr_x)$ -Ba-Cu-O bulk; critical current density; irreversibility field

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#### 1 Introduction

It is well known that RE-Ba-Cu-O (REBCO) superconducting bulks (RE=rare earth elements) demonstrate a high critical current density  $J_{\rm c}$  at 77 K and under high magnetic fields. REBCO bulk is a composite material which consists of the matrix phase of  $REBa_2Cu_3O_{7-\delta}$  (RE123) and secondary phase of  $RE_2BaCuO_5$  (RE211). Such a high  $J_c$  is brought about by various pinning centers, such as RE211 particles, oxygen vacancies in RE123, and local RErich regions. To achieve a higher  $J_{\rm c}$  in REBCO bulks, many studies relating with the introduction of other pinning centers have been performed. Zn substitution for Cu-site in RE123 is very effective for increasing  $J_{\rm c}$  [1]. However, at the same time the superconducting transition  $T_{\rm c}$  is suppressed by this substitution, because Zn is introduced to the superconducting  $CuO_2$  plane. Ishii et al. found that a very small amount of impurity (Fe, Co, Ga) substituted to Cu-site in the CuO chain gives very improved  $J_{\rm c}(H)$  properties in YBCO bulks [2]. Recently, in  $(Y_{1-x}Lu_x)BCO$  [3] and  $(Dy_{1-x}Tb_x)BCO$  [4] bulks the increase of  $J_{\rm c}(H)$  was found at a small amount of x, demonstrating that the impurity substituting to the RE-site is also effective for improving the pinning properties. These previous reports strongly suggest that the dilute impurity substituting to the RE-site and Cu-site in the CuO chain is a promising way to improve pinning properties of REBCO bulks.

In this paper, we report on the effect of the Y-site substitution by La or Pr ions on the vortex pinning properties such as  $J_{\rm c}(H)$  and the irreversibility field,  $H_{\rm irr}$ , at 77 K under the magnetic fields parallel to the *c*-axis in (Y<sub>1-x</sub>La<sub>x</sub>)BCO and (Y<sub>1-x</sub>Pr<sub>x</sub>)BCO bulks. These parameters were improved by the substitution and an origin of the improvement was discussed.

#### 2 Experimental procedure

 $(Y_{1-x}La_x)BCO \text{ and } (Y_{1-x}Pr_x)BCO \text{ bulks } (x=0, 0.0005, 0.001, 0.003, \text{ and } 0.01)$ were grown by a melt-texture technique. (Y,La)123, (Y,Pr)123 and Y211 powders were prepared by a solid state reaction method. Raw powders of  $RE_2O_3$  $(RE=Y, La), Pr_6O_{11}, BaCO_3 and CuO were weighted with proper molar ra$ tios and ground by an agate mortar and pestle for 1 h. For the RE123 phase, the mixed powder was calcined at 900 °C for 24 h in air. The calcined powder was pulverized and ground, and was re-sintered at 920 °C for 72 h in air. For the Y211 phase, the mixture was sintered at 940 °C for 24 h in air. The RE123 powder mixed with the RE211 powder with a molar ratio of 1:0.3, was pressed into pellets with 20 mm in diameter and about 10 mm in thickness. The 0.5wt%  $CeO_2$  powder was added in all samples. We used a Nd123 single crystal as the seed crystal, which was set on the center of top surface of each pellet before the heating up, i.e., the cold seeding. The pellet was heated up to  $1010 \,^{\circ}$ C, kept for 2 h, and slowly cooled down from 1005  $^{\circ}$ C to 970  $^{\circ}$ C at a rate of 0.5  $^{\circ}$ C/h in air. The rectangular shaped samples with typical dimensions of  $2 \times 2 \times 1$  $mm^3$  were cut from an as-grown bulk at 1 mm and 3 mm just from the seed crystal, as schematically shown in the inset of Fig. 1(a). We call these samples as RE-1mm and RE-3mm or 1mm-sample and 3mm-sample, respectively. The amount of oxygen deficiency,  $\delta$ , in the RE123 phase is varied by controlling the annealing temperature [5]. In this study, all the samples were annealed at 400 °C for 168 h in 0.1 MPa flowing oxygen gas. The magnetization was measured in magnetic fields parallel to the c-axis up to 5 T using a commercial SQUID magnetometer (MPMS-5S and MPMS-XL, Quantum Design Inc.). The critical current density,  $J_c$ , was estimated from the M(H) hysteresis loop using the extended Bean model,  $J_{\rm c} = 20\Delta M/a(1-a/3b)$ , where  $\Delta M$  is the width of the M(H) loop, and a and b (a < b) are the cross-sectional dimensions of the sample perpendicular to the applied magnetic field.

#### 3 Results

Figs. 1(a)-1(d) show the temperature dependence of the normalized magnetization, M(T), under the applied field of  $\mu_0 H=0.4$  mT parallel to the *c*-axis after the zero-field cooling (ZFC) for (Y,La)BCO and (Y,Pr)BCO bulks. The onset of the superconducting transition temperature,  $T_c$ , is about 90.5-91.2 K for all the samples, meaning that the dilute La- and Pr-substitution up to 1% slightly reduces the superconductivity.

Figs. 2(a)-2(d) show the magnetic field dependence of the critical current density,  $J_c(H)$ , at 77 K for  $H \parallel c$ -axis for (Y,La)BCO and (Y,Pr)BCO bulks. For the La-1mm samples,  $J_c(H)$  of (Y<sub>0.999</sub>La<sub>0.001</sub>)BCO is slightly higher than that of the non-substituted YBCO above 4.5 T and the corresponding irreversibility field  $H_{irr}$  defined at  $J_c=4$  A/cm<sup>2</sup> exceeds 5 T. However,  $J_c$  of other La-1mm samples are quite smaller than that of YBCO. For the Pr-1mm samples, both  $J_c$  and  $H_{irr}$  are improved for the samples with x=0.001 and 0.003.  $J_c$  of the La-3mm samples with x=0.0005 and 0.001 and the Pr-3mm samples with x=0.0005-0.003 is higher than that of YBCO above approximately 2 T. In addition,  $H_{irr}$  of these samples is also higher than 5 T. The x dependence of  $H_{irr}$  is summarized in Fig. 3.

## 4 Discussion

The obtained results indicate that the substitution of Y-site by La or Pr ions improves both  $J_c$  and  $H_{irr}$  under relatively high magnetic field. To discuss an effect of the substitution, the role of the La or Pr ions in the matrix phase should be considered, i.e., whether La123 or Pr 123 phases are created in the matrix of bulks or the La or Pr ions form other compounds.  $T_c$  of La123 is much higher than that of Y123 [6], on the other hand, Pr123 phase is believed not

to show the superconductivity [7]. As seen in Fig. 1, the  $T_c$ 's of both La- and Pr-substituted samples are basically lower than that of YBCO, meaning that at least La123 phase is not created. In addition, as described in the preceding paragraph, the starting materials of (Y,La)123 and (Y,Pr)123 were fired at 920 °C, which might be too low to create La123 or Pr123 phases. Consequently, we consider that La123 and Pr123 phases are absent in the samples.

The observed field-induced enhancement of pinning properties indicates that the weak superconducting regions are introduced. One possible origin is the appearance of the RE-rich region, because it is generally accepted that the RE-rich region in REBCO suppresses  $T_c$  of the matrix phase. It is well known that in RE123 bulks with RE=La, Nd, Sm a part of the RE ions is substituted for a Ba-site [8], which gives rise to form the RE<sub>1+x</sub>Ba<sub>2-x</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>-type solid solution, because both ionic radii are comparable. This migration increases the weak superconducting regions. This RE-rich region acts as a field-induced pinning center. In usual, such the solid solution cannot occur in the Y123 bulks, because the ionic radius of Y<sup>3+</sup> is quite smaller than that of Ba<sup>2+</sup>. The La- or Pr-substitution enable us to make the field-induced pinning sites in YBCO bulks.

Recently, Shimoyama *et al.* [3] reported that the Lu-substitution for Y-site in the YBCO bulk is effective for the increase of  $J_c(H)$  at 77 K in relatively high magnetic field, which is basically consistent with our results. Tazaki *et al.* [4] reported that in  $(Dy_{1-x}Tb_x)BCO$  bulks (x=0, 0.1) the  $J_c(H)$  values at 77 K are enhanced by the substitution for Dy-site by Tb. This result was interpreted as follows. First, the BaTbO<sub>3</sub> particles are created by the Tbsubstitution, which causes the substitution of Dy for Ba-site. Second, Tb is substituted for Dy-site. These ideas are also consistent with our interpretation, although the mechanisms causing the substitution between RE and Ba sites is different from each other.

We note that the relation between  $J_{\rm c}$  and the size and distribution of the Y211

parts evaluated from the SEM images (not shown here) was not observed, which supports that the improvement of  $J_c$  originates from the effect of the La- or Pr-substitution.

#### 5 Summary

To study the effect of the impurity substitution for the Y-site in YBCO bulk on the vortex pinning properties, we have evaluated the critical current density,  $J_c(H)$ , and the irreversibility field,  $H_{irr}$ , at 77 K from measuring the magnetization using the  $(Y_{1-x}La_x)BCO$  and  $(Y_{1-x}Pr_x)BCO$  bulks (x=0-0.01). The low level of RE-substitution tends to improve  $J_c(H)$  and the corresponding  $H_{irr}$  in the relatively high field. This improvement of the vortex pinning properties is expected to originate from introducing the field-induced pinning centers such as the RE-rich region caused by the  $RE_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ -type solid solution (RE=La, Pr).

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#### Figure captions

## Fig. 1

The temperature dependence of the magnetization, M(T), after the zero-field cooling in  $\mu_0 H(\parallel c)=0.4$  mT for  $(Y_{1-x}La_x)BCO$  (a,b) and  $(Y_{1-x}Pr_x)BCO$ (c,d) bulks (x=0-0.01). The RE-1mm and RE-3mm samples are cut from the position 1 mm and 3 mm below the bottom of the seed crystal, respectively, as shown schematically in the inset of (a).

## Fig. 2

The magnetic field dependence of the critical current density,  $J_c(H)$ , at 77 K and  $H \parallel c$  for  $(Y_{1-x}La_x)BCO$  (a,b) and  $(Y_{1-x}Pr_x)BCO$  (c,d) bulks (x=0-0.01). The dotted lines represent  $J_c(H)=4$  A/cm<sup>2</sup> which is the criterion for the irreversibility field,  $H_{irr}$ .

## Fig. 3

x dependence of the irreversibility field,  $H_{irr}(x)$ , at 77 K and  $H \parallel c$  for  $(Y_{1-x}La_x)BCO$  and  $(Y_{1-x}Pr_x)BCO$  bulks (x=0-0.01).

Correction

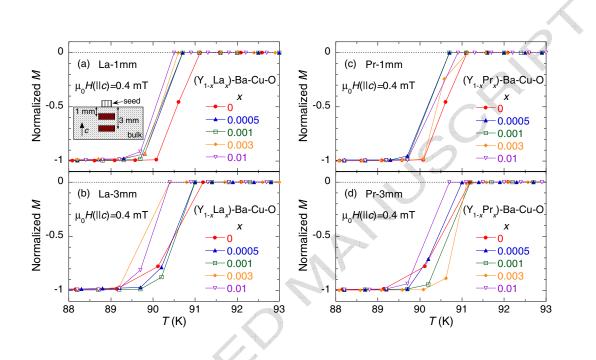


Fig. 1. T. Naito et al.

