



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 x was 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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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_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 RE-rich regions. To achieve a higher J_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_c [1]. However, at the same time the superconducting transition T_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 $J_c(H)$ 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 $J_c(H)$ 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 $J_c(H)$ and the irreversibility field, H_{irr} , at 77 K under the magnetic fields parallel to the c -axis in $(Y_{1-x}La_x)BCO$ and $(Y_{1-x}Pr_x)BCO$ bulks. These parameters were improved by the substitution and an origin of the improvement was discussed.

2. Experimental procedure

$(Y_{1-x}La_x)BCO$ and $(Y_{1-x}Pr_x)BCO$ 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 ratios 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 0.5 wt% 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 °C, kept for 2 h, and slowly cooled down from 1005 °C to 970 °C at a rate of 0.5 °C/h in air. The rectangular shaped samples with typical

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dimensions of $2 \times 2 \times 1 \text{ mm}^3$ 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. 1a. We call these samples as RE-1 mm and RE-3 mm or 1 mm-sample and 3 mm-sample, respectively. The amount of oxygen deficiency, δ , 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, $J_c = 20\Delta M/a(1 - a/3b)$, where ΔM 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

Fig. 1a–d shows the temperature dependence of the normalized magnetization, $M(T)$, under the applied field of $\mu_0 H = 0.4 \text{ mT}$ 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.

Fig. 2a–d shows 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-1 mm samples, $J_c(H)$ of $(Y_{0.999}La_{0.001})$ BCO is slightly higher than that of the non-substituted YBCO above 4.5 T and the corresponding irreversibility field H_{irr} defined at $J_c = 4 \text{ A/cm}^2$ exceeds 5 T. However, J_c of other La-1 mm samples are quite smaller than that of YBCO. For the Pr-1 mm samples, both J_c and H_{irr} are improved for the samples with $x = 0.001$ and 0.003. J_c of the La-3 mm samples with $x = 0.0005$ and 0.001 and the Pr-3 mm 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 Pr123 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 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 $RE_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ -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 a solid solution cannot occur in the Y123 bulks, because the ionic radius of Y^{3+} is quite smaller than that of Ba^{2+} . 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 $BaTbO_3$ particles are created by the Tb-substitution, which causes the substitution of Dy for Ba-site. Second, Tb is substituted for Dy-site. These ideas are also consistent with our

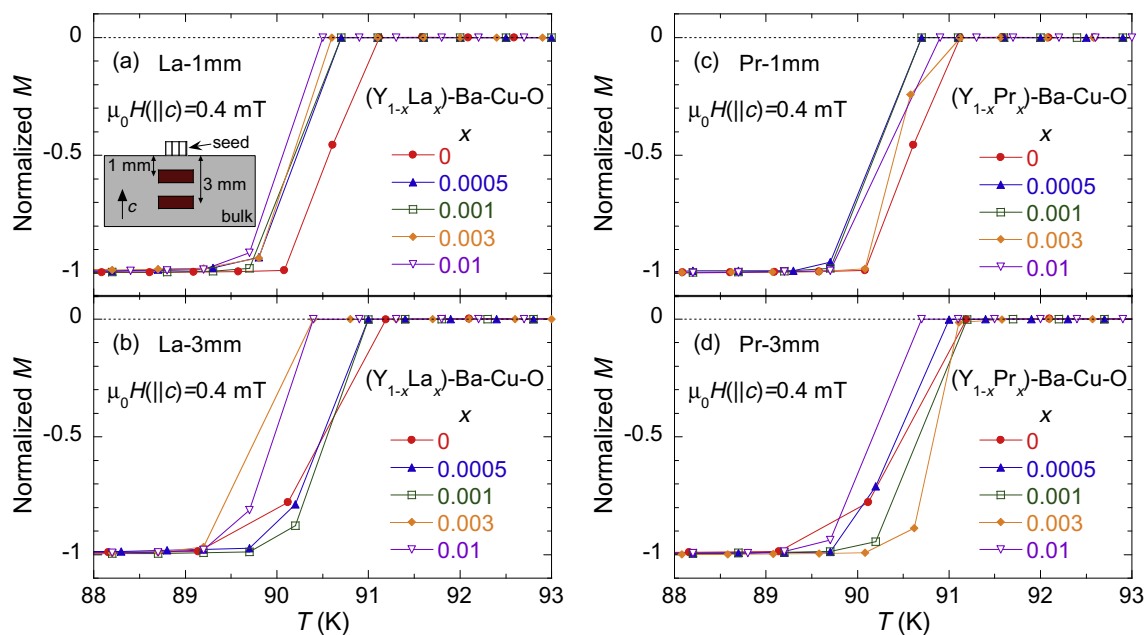


Fig. 1. The temperature dependence of the magnetization, $M(T)$, after the zero-field cooling in $\mu_0 H(\parallel c) = 0.4 \text{ mT}$ for $(Y_{1-x}La_x)$ BCO (a and b) and $(Y_{1-x}Pr_x)$ BCO (c and d) bulks ($x = 0 - 0.01$). The RE-1 mm and RE-3 mm samples are cut from the position 1 mm and 3 mm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

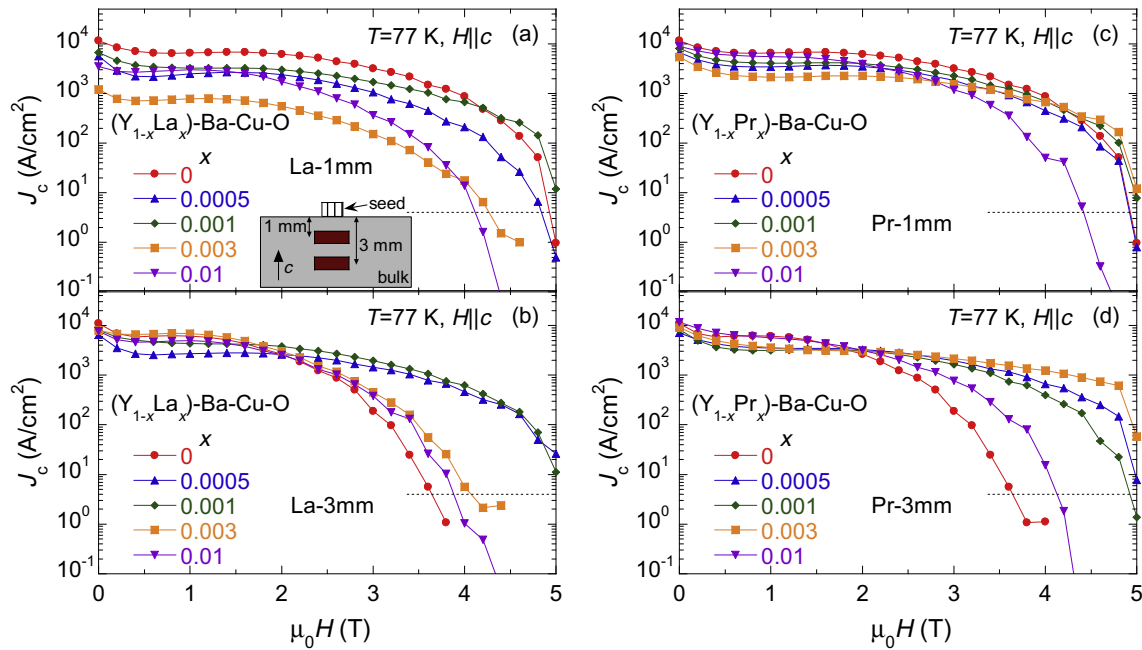


Fig. 2. The magnetic field dependence of the critical current density, $J_c(H)$, at 77 K and $H||c$ for $(Y_{1-x}La_x)BCO$ (a and b) and $(Y_{1-x}Pr_x)BCO$ (c and d) bulks ($x = 0 - 0.01$). The dotted lines represent $J_c(H) = 4 \text{ A/cm}^2$ which is the criterion for the irreversibility field, H_{irr}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

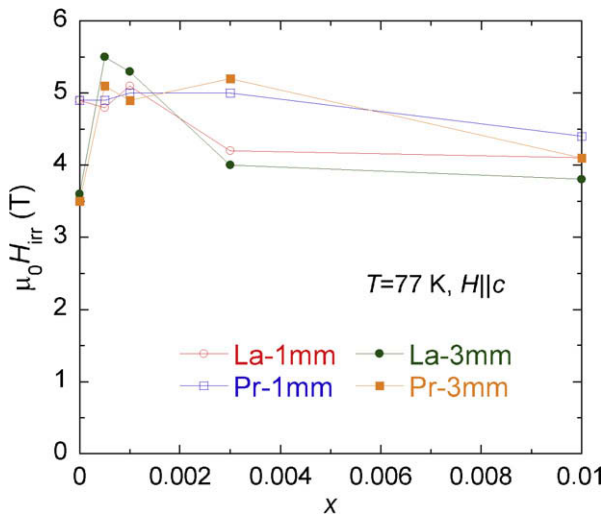


Fig. 3. x dependence of the irreversibility field, $H_{irr}(x)$, at 77 K and $H||c$ for $(Y_{1-x}La_x)BCO$ and $(Y_{1-x}Pr_x)BCO$ bulks ($x = 0 - 0.01$).

interpretation, although the mechanisms causing the substitution between RE and Ba sites is different from each other.

We note that the relation between J_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 evalu-

ated 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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