Vortex pinning properties of \((Y_{1-x}\text{La}_x)\)-Ba–Cu–O and \((Y_{1-x}\text{Pr}_x)\)-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. \((Y_{1-x}\text{La}_x)\)-Ba–Cu–O and \((Y_{1-x}\text{Pr}_x)\)-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 \(\text{RE}_2\text{Ba}_2\text{Cu}_3\text{O}_7\) \(\text{(RE211)}\) and secondary phase of \(\text{RE}_2\text{BaCuO}_5\) \(\text{(RE123)}\). Such a high \(J_c\) is brought about by various pinning centers, such as \(\text{RE211}\) particles, oxygen vacancies in \(\text{RE123}\), and local \(\text{RE}\)-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 \(\text{RE123}\) 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 CuO2 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 \(J_c\) properties in YBCO bulks [2]. Recently, in \((Y_{1-x}\text{Lu}_x)\)BCO [3] and \((\text{Dy}_{1-x}\text{ Tb}_x)\)BCO [4] bulks the increase of \(J_c\) 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 \((Y_{1-x}\text{La}_x)\)BCO and \((Y_{1-x}\text{Pr}_x)\)BCO bulks. These parameters were improved by the substitution and an origin of the improvement was discussed.

2. Experimental procedure

\((Y_{1-x}\text{La}_x)\)BCO and \((Y_{1-x}\text{Pr}_x)\)BCO bulks \((x = 0, 0.0005, 0.001, 0.003, \text{and} 0.01)\) were grown by a melt-texture technique. \((Y\text{La})123, (Y\text{Pr})123\) and \(\text{Y211}\) powders were prepared by a solid state reaction method. Raw powders of \(\text{RE}_2\text{O}_3\) \((\text{RE} = \text{Y, La, Pr})\), \(\text{BaCO}_3\) and \(\text{CuO}\) were weighted with proper molar ratios and ground by an agate mortar and pestle for 1 h. For the \(\text{RE123}\) 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 \(\text{Y211}\) phase, the mixture was sintered at \(940\) °C for \(24\) h in air. The \(\text{RE123}\) powder mixed with the \(\text{RE211}\) 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% \(\text{CeO}_2\) 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...
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, $\delta$, 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-SS 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, $J_c = 20\Delta M/\Delta t (1 - a/b^3)$, 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

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, $J_c$, at 77 K for $H||c$-axis for (Y,La)BCO and (Y,Pr)BCO bulks. For the La-1 mm samples, $J_c$ of (Y,La)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 RE1$_{1-x}$Ba$_x$Cu$_2$O$_y$-$\gamma$-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 enables 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 $J_c$ at 77 K in relatively high magnetic field, which is basically consistent with our results. Tazaki et al. [4] reported that in (Y$_{1-x}$Tb$_x$)-Ba$_2$Cu$_3$O$_7$ bulks ($x = 0.1$) the $J_c(77 \text{ K})$ 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
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 evaluated the critical current density, \( J_c(H) \), and the irreversibility field, \( H_{\text{irr}} \), at 77 K from measuring the magnetization using the \((Y_{1-x}La_x)-Ba-Cu-O\) and \((Y_{1-x}Pr_x)-Ba-Cu-O\) bulks \((x = 0 - 0.01)\). The low level of RE-substitution tends to improve \( J_c(H) \) and the corresponding \( H_{\text{irr}} \) 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-rich region caused by the RE\(_{1-x}\)Ba\(_{2-x}\)Cu\(_3\)O\(_7\)\(_{1-x}\)-type solid solution \((\text{RE} = \text{La}, \text{Pr})\).

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