Effect of matrix composition on the flux pinning in a $(Nd, Eu, Gd)Ba_2Cu_3O_y$ superconductor

M. Muralidhar* and M. Murakami
Superconductivity Research Laboratory, ISTEC, Division III, 1-16-25, Shibaura, Minato-ku, Tokyo 105, Japan
(Received 5 June 2000)

In order to study the effect of matrix composition on flux pinning in the $(Nd, Eu, Gd)Ba_2Cu_3O_y$ (NEG-123) bulk superconductors, we prepared two groups of samples with various rare-earth (RE) ratios using the oxygen-controlled-melt-growth (OCMG) process. For the first group, we kept the Nd content constant and varied the Eu/Gd ratio, and for the second group the Gd content was kept constant and the Nd/Eu ratio was varied. All the samples exhibited strongly developed fishtail effects and high critical current densities. Magnetization measurements revealed that the irreversibility field and the peak position systematically varied with Nd and Gd content. Scaling analyses suggested that two different pinning mechanisms are active in the NEG-123 system depending on the RE ratio. Those are ΔT_c pinning and normal-type pinning, however, we found that field-induced pinning also behaves like normal-type pinning on scaling analyses when ΔT_c is large. In conclusion, a difference in the peak position on scaled curves can be explained in terms of a difference in ΔT_c of field-induced pinning centers.

Practical applications of high-T_c superconductors require large critical current density (J_c) and high irreversibility field at high temperatures. Recent experiments have demonstrated that $(Nd_{0.33}Eu_{0.33}Gd_{0.33})Ba_2Cu_3O_y$ bulk superconductors exhibit superior superconducting properties to $YBa_2Cu_3O_{7-\delta}$ (Y123) or $NdBa_2Cu_3O_{7-\delta}$ (Nd123).[1–4]

A scaling of the pinning force (F_p) versus the reduced field, $h=H_a/H_{irr}$ where H_a is the applied field and H_{irr} is the irreversibility field, provides useful information about the flux-pinning mechanism. Scaling studies have been performed on various high-T_c superconductors, and good scaling was found in many rare-earth (RE) 123 crystals. In Y-Ba-Cu-O, the peak in F_p-h curves is located at $h_0=0.33$, which is typical for normal point pinning.[5] Nd123 with enhanced fishtail effect exhibits the peak at $h_0=0.42$, which was the highest peak value amongst melt-processed RE123 and was a good proof of ΔT_c pinning.[6] However, recent studies showed that the peak was surprisingly high and located at $h_0=0.5$ for NEG-123 samples.[2] Such a high h_0 value has not been observed in other high-T_c superconductors. A similar scaling with $h_0=0.5$ was also observed for NEG-123 samples with second phase particles.[3] The scaled F_p-h curve of Nd123 shows a peak at $h=0.42$, which is an indication of ΔT_c-type pinning or $\Delta\kappa$ pinning provided by the compositional fluctuation in the matrix.[6,7] In contrast, Nd123 composite with 15 mol % Nd422 particles showed a peak at $h=0.33$, showing that normal defects dominate the pinning mechanism in this material.[8]

The peak effect in the RE123 materials originates either from field-induced pinning by RE-Ba chemical fluctuation or oxygen deficiency in the superconducting RE123 matrix. The presence of RE-rich $R_{1+x}Ba_{2-x}Cu_3O_y$ (RE123*ss*) clusters of 10–50 nm diameter has been confirmed both by scanning tunneling microscopy[9] and TEM (Ref. 10) observations. Furthermore, Chikumoto *et al.*[11] have confirmed that the peak effect disappears in Nd123 after high-temperature annealing, by which RE123*ss* clusters are annealed out. In general, ΔT_c pinning provided by chemical fluctuation is stronger than that provided by oxygen vacancies.[12,13] This is due to the fact that the presence of oxygen defects deteriorates the superconducting properties of the parent phase. Since the enhancement of the peak effect requires the introduction of a large amount of oxygen vacancies, there is a limit in the enhancement of the peak effect without deteriorating T_c.

Chemical fluctuation or the distribution of RE123*ss* clusters in the RE123 matrix is affected by the kind of RE elements, and therefore, the control of the matrix composition is important for improving the flux pinning. It may be possible to further enhance flux pinning of NEG-123 simply by controlling the ratio of RE elements in the matrix. Previous investigations of NEG-123 samples showed that the Nd and Gd contents greatly affected the peak J_c and the peak position.[2–4]

With this in mind, we prepared two sets of NEG-123 samples to explore the effect of chemical ratio on the flux-pinning properties. In the first set we fixed the amount of Nd and changed the concentration of Eu and Gd. In the second set, the amount of Gd was kept constant and the ratio of Nd/Eu was varied. We then performed scaling analyses of the normalized volume pinning forces, F_p, versus a reduced field $h=H_a/H_{irr}$ for NEG-123 samples.

High-purity commercial powders ($5N$) of Nd_2O_3, Eu_2O_3, Gd_2O_3, $BaCO_3$, and CuO were pre-dried and weighed to have nominal compositions of $(Nd_{0.33}, Eu_x, Gd_{0.66-x})Ba_2Cu_3O_y$, and $(Nd_{0.66-x}, Eu_x, Gd_{0.33})Ba_2Cu_3O_y$ in which x ranged from 0 to 0.33. The details of the sample preparation and the oxygen-controlled-melt-growth (OCMG) process were described elsewhere.[14] The OCMG process was performed under controlled oxygen partial pressure of 0.1% O_2 with a gas flow rate of about 300 ml/min. For oxygen annealing, the samples with dimensions $(a\times b\times c)=1.5\times1.5\times0.5\,mm^3$ were cut from as-grown crystals and annealed in flowing O_2 gas in the temperature range of 300–600 °C. Magnetization hysteresis loops were measured mainly at 77 K in applied fields up to

0163-1829/2000/62(21)/13911(4)/$15.00

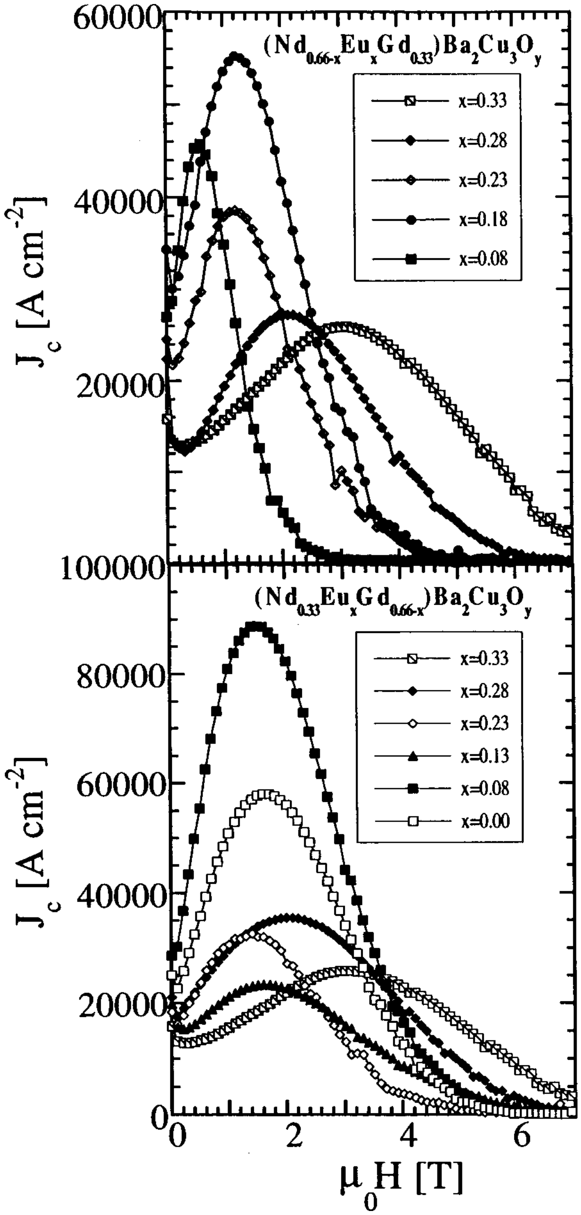

FIG. 1. Field dependence of the critical current density ($T = 77\,\mathrm{K}$, H_a parallel to the c axis) for various NEG-123 superconductors. Note that record high critical current density around 90 000 A cm^{-2} at the peak field 1.7 T. All samples are prepared by Ar 0.1% partial pressure of O_2.

7 T using a commercial superconducting quantum interference device magnetometer (Quantum Design, model MPMS7). To minimize field inhomogeneities, the scan length was restricted to 1 cm. The external magnetic field was applied parallel to the c axis of the samples. The magnetic J_c values were estimated based on the extended Bean's critical-state model using the following relation:

$$J_c = 2\Delta m / a^2 d(b - a/3), \tag{1}$$

where d is the thickness of the sample and a and b are cross sectional dimensions, $b \geqslant a$.[15]

Figure 1 shows the critical current density characteristics of the NEG-123 samples measured at 77 K for fields applied parallel to the c axis. It is evident that all the samples show a strongly developed secondary peak effect. The peak position and the irreversibility field systematically shift to lower fields with increasing either Nd or Gd content, which shows that the NEG-123 sample with Nd:Eu:Gd=1:1:1 exhibits the best pinning at the highest fields. However, the highest J_c of 90 000 A cm^{-2} was achieved in the NEG-123 sample with Gd=0.58 at 77 K and 1.7 T. These results show that the flux-pinning properties of NEG-123 can be modified by changing the matrix ratio and may be controllable to suit J_c-H properties for the end use applications.

With the aim of further studying the effect of RE concentration on the flux pinning in the NEG-123 system, we performed scaling analyses of normalized volume pinning forces, F_p, versus reduced field $h = H_a / H_{\mathrm{irr}}$ for two sets of NEG-123 samples at 77 K (Fig. 2). H_{irr} is obtained from magnetization loops with a criterion of 100 A cm^{-2}. It was found that the peak field in the scaling curves shifted from $h_0 = 0.50$ to lower values with either increasing Gd or Nd content, which suggests that ΔT_c pinning is the most prominent for the NEG-123 sample with Nd:Eu:Gd=1:1:1.

The NEG-123 material with the matrix mixing ratio close to 1:1:1 show the peak at $h_0 = 0.5$, which is a strong indication of T_c variation throughout the sample. The samples with Nd=0.43 and 0.48 samples show the maximum peak at 0.33, which suggests that the normal-type pinning centers are present. The Gd-rich sample shows a peak around $h_0 = 0.4$, which is an indication of ΔT_c and normal-type pinning centers. Although the background normal-type pinning is usually present in the sample due to crystal defects, we believe that the main pinning centers are still RE-rich RE123*ss* clusters in all the NEG-123 samples with different RE ratios. This is supported by the fact that all samples exhibit clear secondary peak effect in that the peak position is dependent on ΔT_c of the clusters. When the clusters are driven normal at low fields, that is ΔT_c is large, they behave like normal conducting pinning centers on scaling analyses, so that the peak field lies at $h = 0.33$.

The pinning mechanism is more extensively analyzed with the scaling curve by fundamental pinning components as projected by Dew-Hughes.[16] The formula is based on the variation of $F_p / F_{p\,\max}$ for each pinning component as a function of reduced field and is described by

$$F_p / F_{p,\,\max} = C h^p (1 - h)^q, \tag{2}$$

where h is a reduced field, p and q are the parameters describing the pinning mechanism and $h_{\mathrm{peak}} = p/(p+q)$, and C is a numerical parameter. The solid lines in Fig. 2 are fitting curves with various p and q parameters. The best fit was found to be $p = q = 2$ for NEG-123 with Nd:Eu:Gd=1:1:1, and $p = 1$ and $q = 2$ for the NEG-123 with Nd=0.48. This again suggests that different pinning centers are active in the NEG-123 samples depending on the chemical ratio. One is associated with a local variation in superconducting transition temperature due to the formation of RE-rich RE123*ss* clusters with depressed T_c, which is ΔT_c (Refs. 16 and 17) or $\Delta\kappa$ pinning.[18] The other pinning center is normal conducting.

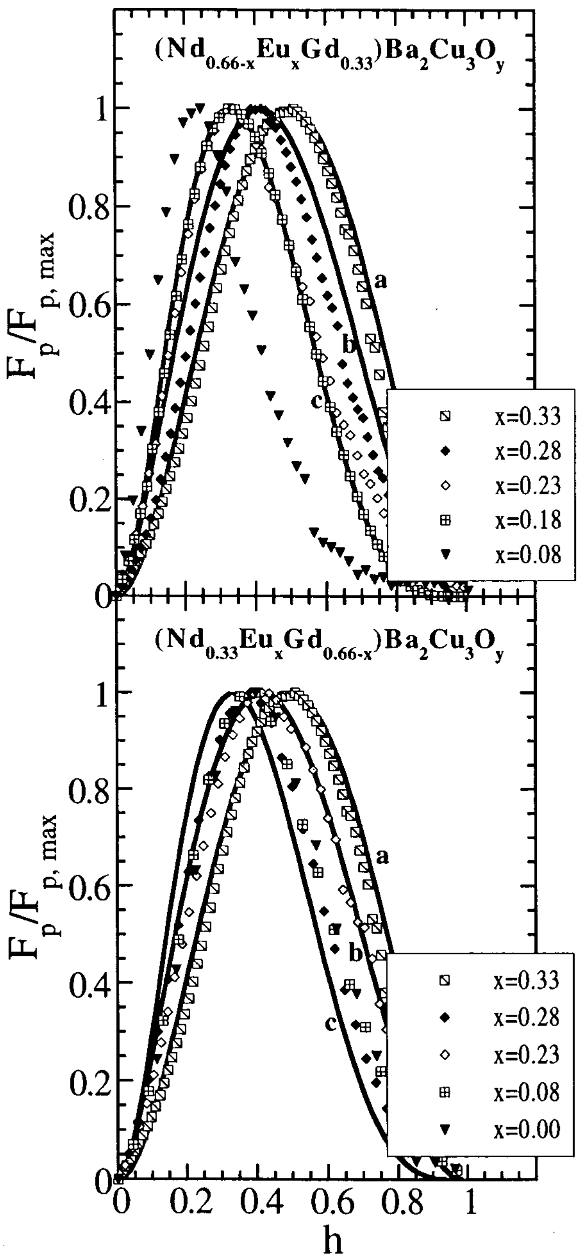

FIG. 2. Normalized pinning force as a function of reduced field of various NEG-123 superconductors at 77 K. The notations of the scaled curves (solid line) are a for $p=q=2$, b for $p=1.45$, $q=2$, and c for $p=1$, $q=2$.

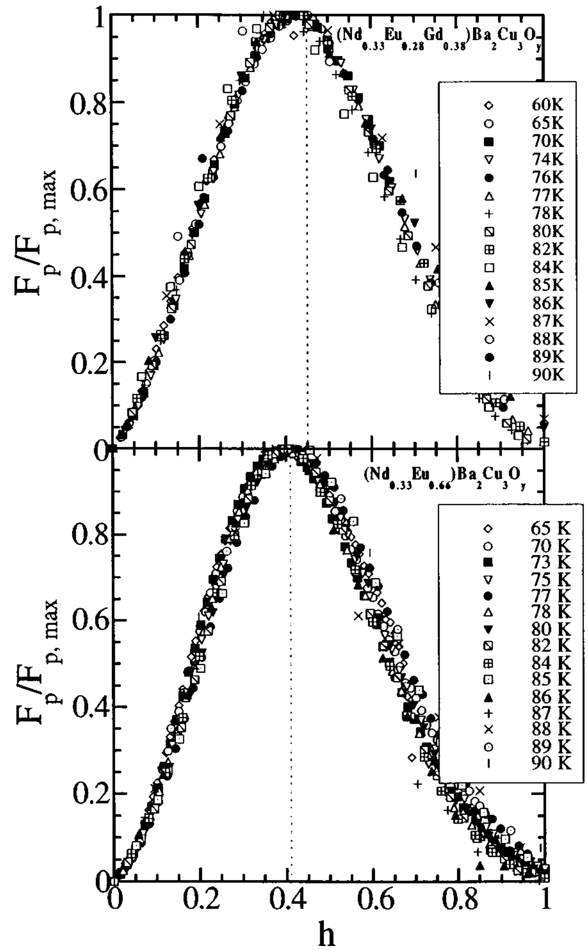

FIG. 3. $F_p/F_{p,\max}$ scaling of NEG-123 (Gd=0.38 and Gd=0.66) samples in a temperature range of 65–90 K. The scaling is found to be excellent; yielding peaks at $h_0=0.45$ for Gd=0.38 and 0.40 for Gd=0.66.

However, as already mentioned above, the strongly developed peak effect is observed in the J_c-H curve even in the NEG-123 sample with Nd=0.48, for which the scaling behaves like that of normal conducing pinning. Thus one can conclude that the type of dominant pinning center is identical and field-induced pinning type even though h_0 varies from 0.5 to 0.3.

We also analyzed the scaling of the NEG-123 samples with Gd=0.38 and 0.66 over a temperatures range of 65–90 K. The scaling curves for these samples are presented in Fig. 3. The peaks are $h_0=0.45$ and 0.40 for Gd=0.38 and 0.66, respectively. In our previous work, scaling worked very well for the sample with Nd:Eu:Gd=1:1:1 in the entire temperature range of 65–90 K, and the peak position was at $h_0=0.50$[2].

Like Nd-rich NEG-123 samples, the peak position shifts to lower h value as the Gd content is increased. The present NEG-123 samples do not contain RE211 inclusions, so that normal conducting particles are not responsible for a decrease in h value. These results also support the fact that a difference in ΔT_c of RE-rich RE123*ss* clusters.

In summary, magnetic measurements of NEG-123 samples with various RE mixtures suggested that flux-pinning behavior is dependent on the chemical ratio in the matrix. From scaling analyses two types of pinning mechanisms seem to be active in NEG-123 material, since the peak position shifted from $h_0=0.50$ down to 0.27, however, this is simply due to a difference in ΔT_c of RE-rich RE123*ss* clusters that act as field-induced pinning centers.

This work was supported by New Energy and Industrial Technology Development Organization (NEDO) as a part of its Research and Development of Fundamental Technology for Superconductor applications project under the sunshine program administrated by the Agency of Industrial Science and Technology M.I.T.I of Japan. M.M.D. is thankful to the NEDO Govt. of Japan for providing financial support.

*Email address: Miryala1@istec.or.jp

[1] M. Muralidhar, M. R. Koblischka, T. Saitoh, and M. Murakami, Supercond. Sci. Technol. **11**, 1349 (1998).
[2] M. R. Koblischka, M. Muralidhar, and M. Murakami, Appl. Phys. Lett. **73**, 2351 (1998).
[3] A. K. Pradhan, M. Muralidhar, M. R. Koblischka, M. Murakami, K. Nakao, and N. Koshizuka, Appl. Phys. Lett. **75**, 253 (1999).
[4] M. Muralidhar, M. R. Koblischka, P. Diko, and M. Murakami, Appl. Phys. Lett. **76**, 91 (2000).
[5] Hai-hu Wen and Zhong-xian Zhao, Appl. Phys. Lett. **68**, 856 (1996).
[6] M. R. Koblishka, A. J. J. van Dalen, T. Higuchi, S. I. Yoo, and M. Murakami, Phys. Rev. B **58**, 2863 (1998).
[7] A. K. Pradhan, K. Kuroda, B. Chen, and N. Koshizuka, Phys. Rev. B **58**, 9498 (1998).
[8] N. Hari Babu, D. A. Cardwell, W. Lo, and A. M. Campbell, Phys. Rev. B **61**, 735 (2000).
[9] W. Ting, T. Egi, K. Kurada, N. Koshizuka, and S. Tanaka, Appl. Phys. Lett. **70**, 770 (1997).
[10] T. Egi, J. G. Wen, K. Kuroda, H. Unoki, and N. Koshizuka, Appl. Phys. Lett. **67**, 2406 (1995).
[11] N. Chikumoto, J. Yoshioka, M. Otsuka, N. Hayashi, and M. Murakami, Physica C **281**, 253 (1997).
[12] A. Erb, J. Y. Genound, M. Dhalle, E. Walker, and R. Flukiger, Physica C **282-287**, 2145 (1997).
[13] Th. Wolf, A. C. Bornarel, H. Kupfer, R. Meier Hirmer, and B. Obst, Phys. Rev. B **56**, 6308 (1997).
[14] M. Muralidhar, M. R. Koblischka, and M. Murakami, *Advances in Superconductivity XII* (Springer-Verlag, Tokyo, 2000), p. 443.
[15] C. P. Bean, Rev. Mod. Phys. **36**, 2489 (1964); D. X. Chen and R. B. Goldfarb, J. Appl. Phys. **66**, 2489 (1989).
[16] D. Dew-Hughes, Philos. Mag. **30**, 293 (1974).
[17] K. Yamafuji and F. Irie, Phys. Lett. **25A**, 387 (1967); E. J. Kramer, J. Appl. Phys. **44**, 1360 (1974).
[18] R. Griessen, Phys. Rev. Lett. **72**, 1910 (1994).