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Flux Pinning in Melt Grown LRE123 Superconductors

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Unlike Y123 which forms only a stoichiometric compound, the light rare earth elements (LRE: La, Nd, Sm, Eu, Gd) form a solid solution $LRE_{1+x}Ba_{2-x}Cu_3O_y$. When LRE123 superconductors are melt-processed in a reduced oxygen atmosphere, they exhibit high T_c's and large J_c values in high magnetic fields associated with a second peak effect. We believe that such a behavior is closely related to the presence of a LRE-Ba solid solution. In this paper we propose the flux pinning mechanism of LRE 123, and then discuss the source of the peak effect.

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1. INTRODUCTION

While reported J_c's of melt-processed Y-Ba-Cu-O superconductors [1-5] have already surpassed the lower limit for some practical applications, further J_c enhancement to the level $>10^5$ A/cm^2 and its less severe degradation in high magnetic fields at 77 K are desired for both better performance and a safety margin, which definitely requires more effective flux pinning centers. To date, most investigations employing the melt process have been focused on the Y-Ba-Cu-O system. A few reports on the rare earth (RE)-Ba-Cu-O systems only show that the most suitable material for the bulk type applications is still Y-Ba-Cu-O superconductor [6]. This is mainly due to the fact that for the light rare earths (LRE) of La, Nd and Sm, it has been unsuccessful to fabricate melt-processed $LREBa_2Cu_3O_y$ (LRE123) superconductors with high T_c (> 90K) and a sharp transition.

As a promising process for overcoming the present limitations of Y-Ba-Cu-O, we have recently developed the oxygen-controlled-melt-growth (OCMG) process for the fabrication of Nd-Ba-Cu-O superconductors with high T_c and a sharp transition, using a reduced oxygen atmosphere during the melt growth [7]. We could find that the Po_2 is a critical processing parameter for the successful melt process of Nd-Ba-Cu-O superconductors. They also exhibit J_c values larger than that of a good quality melt-processed Y-Ba-Cu-O

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superconductor [7-10]. The OCMG process was also confirmed to be effective in obtaining high T_c and strong flux pinning for other LRE-Ba-Cu-O superconductors including Sm [9,11] and Eu [12] in addition to Nd, and even for any combinations between these three elements [13].

In this paper, we first report the resutls of magentization studies of the OCMG processed LRE123 superconductors and then propose possible flux pinning mechanism.

2. SAMPLE PREPARATION - THE OCMG PROCESS

The samples were prepared with the OCMG process, which is basically identical to other melt-processes for the Y-system except the Po_2 control in the range of 10^{-2} - 10^{-3} atm during the melt growth stage, where the solidification of the LRE123 phase takes place. The thermal processing conditions for the melt growth need to be rescheduled in accordance with the Po_2 values since the peritectic decomposition temperatures of LRE123 family are different from each other [14] and also very sensitive to the oxygen partial pressure [15-17].

Here, it should be born in mind that although the LRE123 samples are melt-processed in a reduced oxygen atmosphere, oxygen-annealing must be performed in order to achieve high T_c after the melt-growth stage. The optimum annealing temperature was 300℃ which was lower than that of Y123.

Fundamentally, the OCMG process is to preferentially stabilize high T_c phase among the $LRE_{1+x}Ba_{2-x}Cu_3O_y$ solid solution utilizing a significant difference in their melting points in reduced oxygen atmospheres [15]. During solidification in low Po_2, the high T_c phase of suppressed LRE substitution for Ba site could preferentially nucleate and grow while a low T_c phase with a heavy substitution was still depressed. LRE123 single crystals with good T_c and J_c can also be fabricated by a flux method in a reduced oxygen atmosphere. The details of which is described elsewhere [18].

3. FLUX PINNING PROPERTIES IN OCMG-PROCESSED LRE123

3.1. Mangetization measurements

Figure 1 shows the M-H loops of Nd-Ba-Cu-O samples melt-processed in Po_2= 10^{-2} and 10^{-3} atm for both H//c and H⊥c (H//ab plane) at 77K. The measurements were performed with a SQUID magnetometer after zero field cooling. Like usual melt-textured Y-Ba-Cu-O superconductors, large conduction anisotropies between along ab-plane and c direction were observed from the M-H loops. It is interesting to note that anomalous second peak effects are observed in the M-H loops. However, the peak effect of the sample processed in Po_2= 10^{-3} atm is somewhat depressed and the maximum peak position shifted to a higher field compared with the sample processsed in Po_2= 10^{-2} atm. Unlike the case in oxygen deficient Y123 single crystal, additional long-term oxygenation at various temperatures did not affect the peak

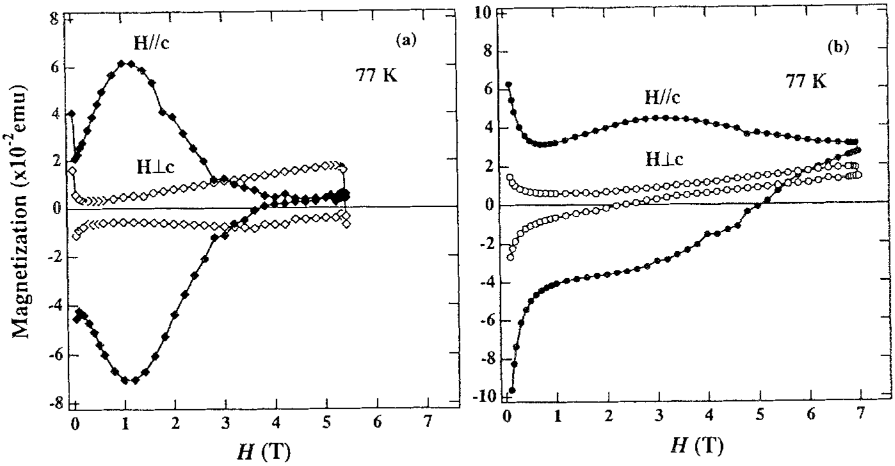

Fig. 1. The M-H loops of Nd-Ba-Cu-O samples OCMG-processed in 1% O_2 in Ar (a) and 0.1% O_2 in Ar (b) at 77K for the field parallel (H//c) and perpendicular (H⊥c) to the c-axis of samples.

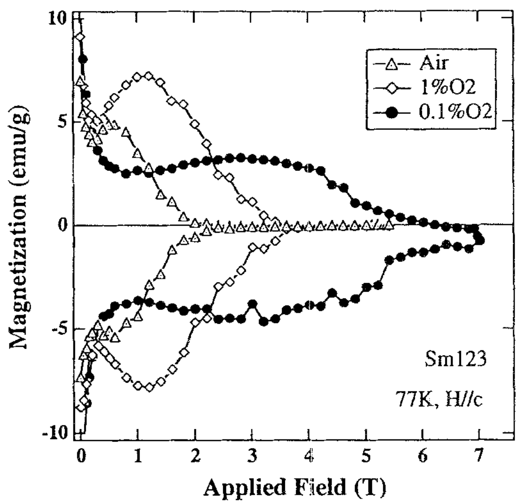

Fig. 2. The M-H loops for OCMG-processed Sm-Ba-Cu-O samples at 77 K and for H//c.

effect itself, although the maximum peak position shifted to a higher field with increasing T_c.

It should be noted that in the sample melt-processed in Po_2= 10^{-3} atm, even for H//c, the M-H loop is kept open to 7 T. Independent M-H loop measurement up to 9 T with a vibrating sample magnetometer revealed that the irreversibility field exceeded 8 T at 77 K [10]. Stronger flux pinning of this sample in a high field region probably originates from both enhanced T_c and the introduction of a new type of pinning center [8], the details of which will be described later.

The peak effects are commonly observed in the LRE123 systems when they are melt-processed in a reduced oxygen atmosphere (see Fig. 2). These results suggest that the source of the peak effect is attributable to the same origin in the LRE123 systems. We have also confirmed that the peak effect is even more clearly observed in Nd123 single crystals grown by the flux method in a reduced oxygen atmosphere, which strongly suggests that the defects responsible for the peak effect are present in the matrix [19].

It is also important to notice that the peak field (H_{pk}) and the H_{irr} are dependent on the Po_2 during the melt growth, which implies that the H_{pK} and H_{irr} can be controlled by monitoring the Po_2 under a fixed melt growth condition.

3.2. Temperature dependence of ΔM-H properties

Figures 3 and 4 show temperature dependence of ΔM-H properties for H//c and H⊥c (or H//ab plane) in a Nd123 single crystal grown by the flux method. Second peak effects are clearly observed in both directions. However, for both cases, the peak becomes less clear as the measuring temperature is lowered. It is probable that the peak field shifts to a higher field with decreasing temperature, and therefore, the peak is not observable within a limited experimental field range (0 - 9T). It is also true that other defects may become effective in pinning at lower temperatures, and thus the contribution of the defect resposible for the peak effect becomes small as the temperature is decreased.

Here, it is interesting to note that the clear peak effect is observed even for H⊥c, which has not been so clearly observed in the Y123 system with oxygen defects. We believe that this fact indicates that the source of the peak effect in LRE123 is not oxygen deficiency. It is also important to note that the peak field is strongly tempearture dependent, therefore the peak effect is not attributable to the matching effect.

Figure 5 shows plots of the reduced loop width $\Delta M/\Delta M_{pk}$ versus the reduced field H/H_{pk} in the temperature range of 82-90 K for H//c axis and H//ab plane. All the curves can be scaled in a single master curve. This success in scaling at various temperatures strongly suggests that the magnetization behavior is dominated by a single type of pinning center in a high temperature region [20].

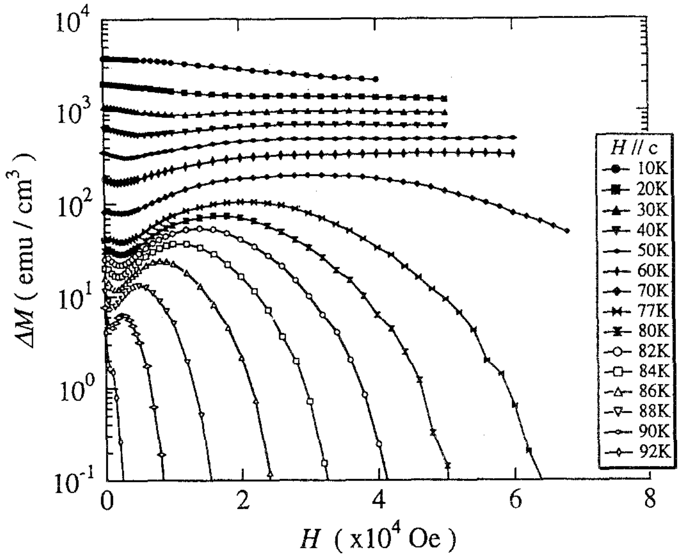

Fig. 3. Temperature dependence of ΔM-H properties for Nd123 single crystal for H//c axis.

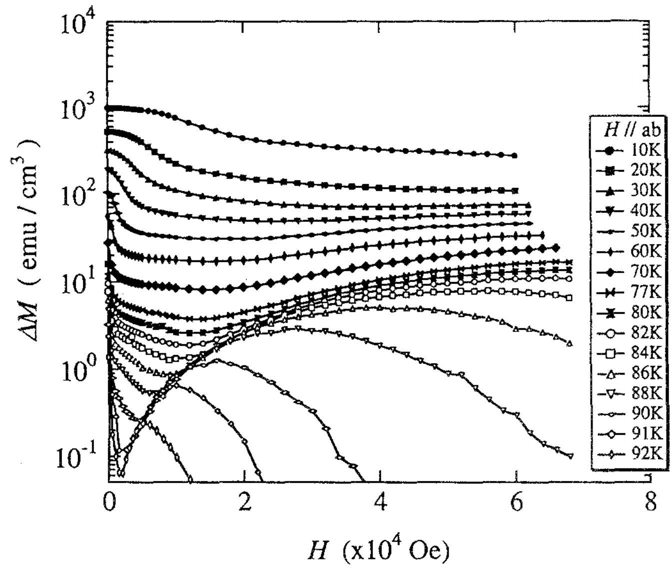

Fig. 4. Temperature dependence of ΔM-H properties for Nd123 single crystal for H//ab plane (or H⊥c).

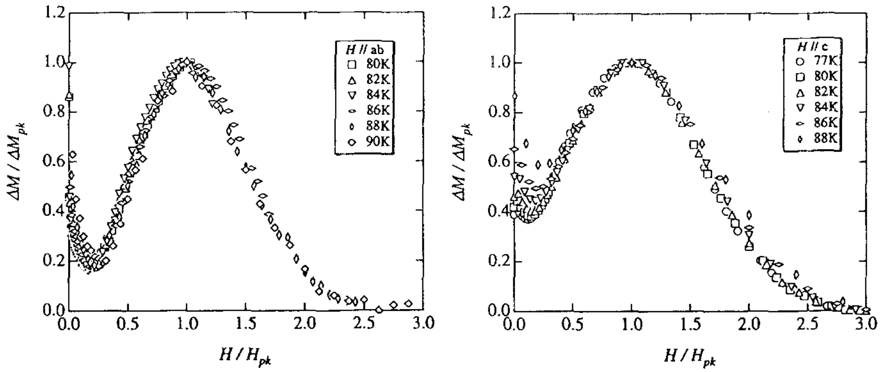

Fig. 5. Plots of $\Delta M/\Delta M_{pk}$ versus H/H_{pk} for Nd123 single crystal with fields parallel and perpendicular to the c axis.

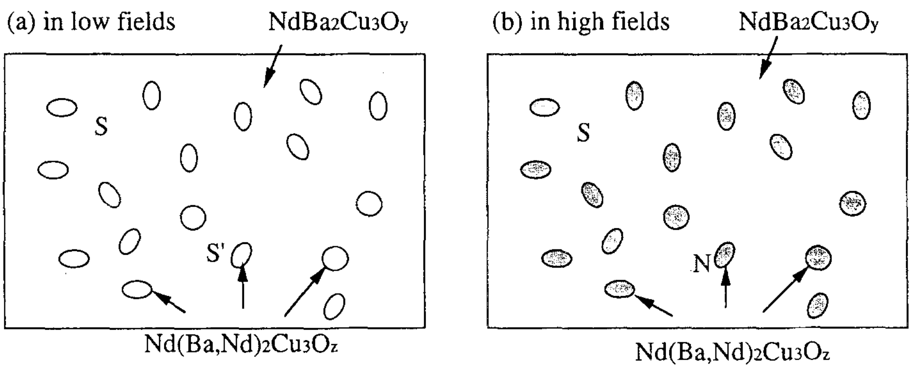

Fig. 6. Schematic illustration of field-induced flux pinning by finely distributed Nd rich regions.

4. FlUX PINNING IN OCMG-PROCESSED LRE123

4.1. Flux pinning mechanism

An explanation for the peak effect in Y123 single crystal was first offered by Dauemling et al. [21]. In their model, the increasing portion in the J_c-B curve is attributed to local oxygen-deficient regions of depressed T_c in the good superconducting matrix of high T_c. When the applied field is increased, local oxygen-deficient regions are driven normal since an upper critical field, H_{c2}, of those regions is considered to be lower than that of the matrix. Thus, these oxygen deficient regions, driven normal, act as effective pinning sites causing the peak effect.

We believe that the idea of field induced pinning also applies to the Nd123 system, however, the defects responsible for the peak effect are different. As mentioned previously, there is a wide range of composition in the Nd-Ba solid solution. Thus, the chemical composition of the Nd123 system can be described as $Nd(Ba_{1-x}Nd_x)_2Cu_3O_y$. If we synthesize this system in air, x lies in the range of 0.0 to 0.3, and therefore T_c also varies over a wide range, and zero resistivity temperature decreases to lower than 90 K. When this compound is synthesized in a reduced oxygen atmosphere, x shifts to lower values, which means that the chemical composition becomes close to 123, and thereby T_c is remarkably improved. However, even in such a case, we believe that there exists a slight chemical variation and thus Nd-substituted Ba regions of low T_c phases are distributed in the high T_c matrix. As schematically shown in Fig. 6, these substituted regions are superconducting in low fields and may not contribute to flux pinning. As the strength of magnetic field is increased, some of these regions will become normal, and can then contribute to flux pinning. The field for these regions to be driven normal depends on chemical composition. This is the origin for the anomalous peaks. The present interpretation also corresponds to other LRE-systems.

Recently, Nd substituted Ba regions have been detected by Egi *et al.* through TEM [22]. For such observations, they used the dark-field diffraction contrast image technique of TEM, which is believed to be very sensitive to the defect structure. Ting *et al.* [23] have also succeeded in observing such Nd-rich regions in a Nd123 single crystal by scanning tunneling microscopy. Oval clusters 10 - 30 nm in diameter with different contrast are dispersed in the matrix, which is similar to the regions observed by TEM. We believe that these observations confirmed the presence of fine Nd-rich regions which are responsible for field-induced pinning and thus the second peak effect.

4.2. Temperature dependence of H_{irr} and H_{pk}

Figure 7 shows the temperature dependence of H_{irr} and H_{pk} for both fields parallel and perpendicular to the c axis. The obtained values for the anisotropy in H_{irr} ($\gamma_{irr}=H_{irr}^{ab}/H_{irr}^{c}$) and in H_{pk} ($\gamma_{pk}=H_{pk}^{ab}/H_{pk}^{c}$) are also shown in the inset. The value of γ_{irr} is about 4 and the value of γ_{pk} is slightly higher. The both γ's are almost temperature independent over the experimental temperature range. We believe that the γ_{irr} reflects the γ in H_{c2} of the matrix, while γ_{pk} reflects that of low T_c phase with Nd rich compsotion. The c axis lattice parameter

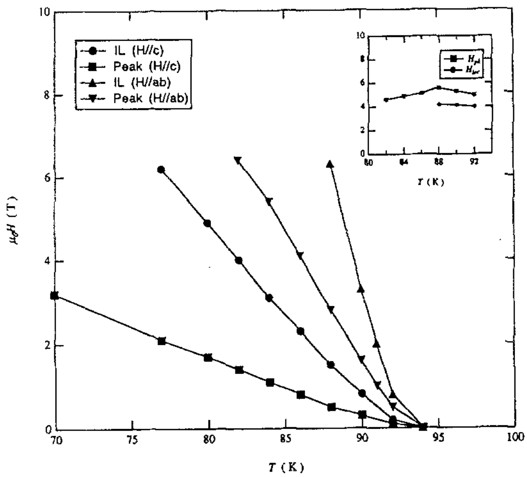

Fig. 7. Temperature dependence of H_{irr} and H_{pk} for H//c and H//ab plane. The inset shows temperature dependence of the anisotropy in H_{irr} and H_{pk}.

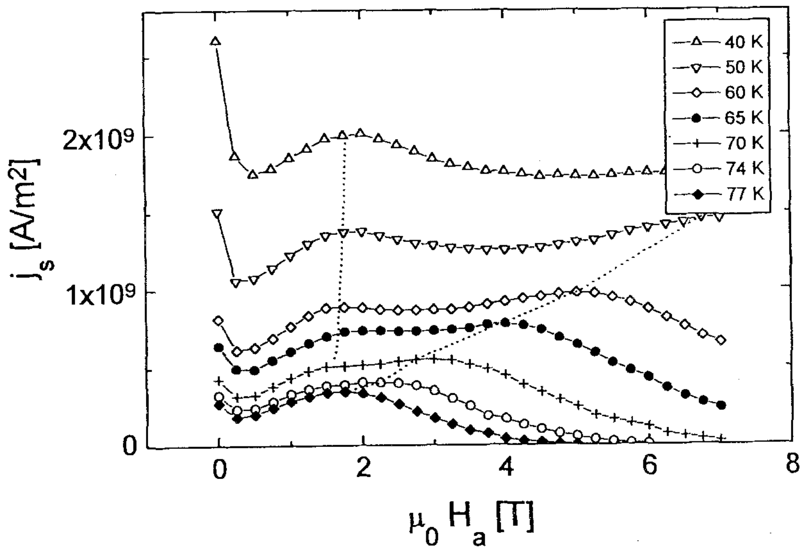

Fig. 8. Temperature dependence of the peak fields in Nd123 single crystal for H//c axis. Note that the temperature independent peak fields are observed. Such peaks are not observed for H//ab plane.

of Nd rich phase is longer than that of the Nd123 phase, and thus the anisotropy of H_{pk} is somewhat larger. Therefore, the temperature dependence of H_{irr} and H_{pk} is consistent with our picture that Nd rich low T_c phase is dispersed in the high T_c matrix, and the former is responsible for the peak effect.

4. 3. Temperature independent peaks

Recently, several groups have observed the presence of temperature independent peak in the M-H loops of Nd123 single crystals [24,25]. Figure 8 shows the temperature dependence of M-H loops and J_c-B curves for a flux-grown Nd123 single crystal for H//c. There are two peaks, one temperature dependent and the other temperature independent. In this figure, the matching field is around 2T, while the matching field of 4T is reported in a different sample [24].

Nakamura et al. [24] proposed the idea that Nd-Ba ordered structre caused by spinodal decomposition at around 500℃ may be responsible for the matching effect. Considering the fact that the peaks are not observed for H⊥c, which should be observed in the case of pinning by Nd rich clusters as described in section 4. 1, it is more plausible that the peak effect is related to some oxygen ordered structure. Further study will be necessary to fully understand the source of such temperature independent peak effect.

5. SUMMARY

The characteristic strong flux pinning in the OCMG-processed LRE-Ba-Cu-O superconductors originates from the LRE123 matrix having a chemical modulation by local LRE-substitution on Ba site. The regions exhibit lower Tc and will be driven normal in the presence of magnetic field, and thus can act as field-induced pinning centers.

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