Pinning Performance of $(Nd_{0.33}Eu_{0.2}Gd_{0.47})Ba_2Cu_3O_y$ Single Crystal

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***Abstract* - The critical current density J_c, the pinning force density $F(=BJ_c)$, and the relaxation rate Q were determined from magnetic hysteresis loops (MHL) measured from 65 K to 90 K on a twinned $(Nd_{0.33}Eu_{0.2}Gd_{0.47})Ba_2Cu_3O_y$ single crystal with a strip-like surface structure. The strong second peak observed on the MHL at 65 K continuously decreased with increasing temperature but persisted up to 84 K. None of the $J_c(B)$ and $F(B)$ dependences scaled, let alone in a narrow range of T. A strong effect of twin channeling was observed but no special pinning effect due to the strip-like surface structure was recognized.**

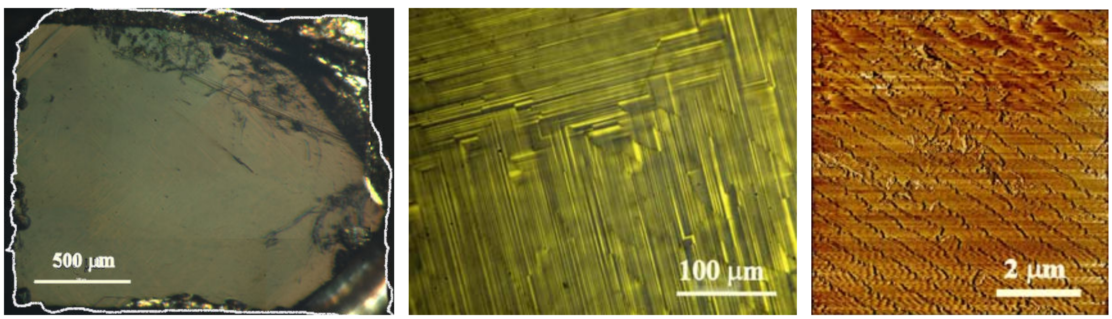
Manuscript received November 29, 2007; accepted December 19, 2007. Reference No. ST13, Category 2
Paper submitted to Proceedings of EUCAS 2007; published in JPCS **98** (2008), paper # 012191

I. INTRODUCTION

Bulk high-T_c superconductors possess a big potential for application as quasi-permanent magnets capable of trapping magnetic field by order of magnitude higher than the best conventional magnetic materials. Even with the moderate electromagnetic performance of $YBa_2Cu_3O_y$ (Y-123) it was possible to trap magnetic field as high as 17 Tesla in a pellet of only 26.5 mm in diameter and 12 mm thick [1]. LRE-123 compounds (LRE=light rare earth, Nd, Sm, Eu, Gd) can carry significantly higher engineering currents than Y-123 [2-4]. Thus, in an LRE-123 bulk a similarly high magnetic field could be trapped at a significantly higher temperature than in the case of Y-123 (29 K) [1]. Binary and ternary LRE-123 compounds offer for pinning structure tailoring one more degree of freedom, in the variation of the LRE ions ratio. To date, such materials have been studied mostly on melt-textured (MT) samples. Although the MT superconductors are a great promise for applications, as they can be fabricated in rather large blocks and a relatively low cost, their structure suffers from many pores, cracks, and imperfections appearing mainly during the oxygenation process. These do not only deteriorate the mechanical strength of the compound but also make the current flow not well defined. Both from physical and technological point of view, studying of the more perfect single-crystalline form of the material should be of interest. Recently, a new kind of nanoscale pinning structure, mediated by the LRE elements ratio, was discovered in twinned MT samples of (Nd,Eu,Gd)-123 [5]. It was a fine lamellar structure with period of 3-5 nm, aligned with the twin planes and filling the channels between them. As the thickness and period of the lamellas was comparable with the coherence length and thus also the vortex core size, vortex motion along the regular twin boundaries (channeling effect) was prevented. Consequently, vortex pinning was enhanced, especially in high magnetic fields, and the irreversibility field increased nearly twice. A fine strip-like structure with period of a several tens of nm has been also observed by atomic force microscopy (AFM) and scanning tunneling microscopy (STM) on surfaces of various LRE-123 single crystals [6,7]. This was also the case of the present single crystal sample (Figure 1). The question arises, what is the origin of this strip-like structure and whether it contributes to the pinning performance or not. In this paper we present magnetic study of the critical current density, J_c, the pinning force, F, and the logarithmic relaxation rate, Q, on a twinned $(Nd_{0.33}Eu_{0.2}Gd_{0.47})Ba_2Cu_3O_y$ single crystal with a special focus onto the role of the observed stripes.

II. EXPERIMENTAL DETAILS

High purity commercial powders of Nd_2O_3, Eu_2O_3, Gd_2O_3, and $BaCO_3$ were mixed in the nominal composition $(Nd_{0.33}Eu_{0.2}Gd_{0.47})Ba_2Cu_3O_y$. The single crystal was grown from the self-melt in air. The form, twin structure, and the stripes observed on the investigated sample are shown in Figure 1. The crystal was 0.5 mm thick and approximately 2 x 1.5 mm^2 in size.



a **b** **c**

Fig.1. (a) Optical microscopic image of the investigated NEG-123 single crystal with contours marked by the white line; (b) the twin structure observed in polarized light; (c) STM image of the crystal surface at RT.

Magnetization hysteresis loops (MHLs) were measured by means of a vibrating sample magnetometer (VSM) in the PPMS (Quantum Design) equipped with 9 Tesla magnet. The MHLs were measured with magnetic field applied parallel to the *c*-axis, in the temperature range 65 K to 90 K. Most curves between 65 K and 77 K were measured with the field sweep rate 0.72 T/min; the dynamic relaxation was studied using two additional sweep rates, 0.36 T/min and 0.12 T/min. The MHL curves at temperatures 80 K and higher were measured with the field sweep rates 0.36 T/min and 0.12 T/min. Critical current density, J_c, values were estimated using the Bean critical state formula for a sample with rectangular cross-section [8], $J_c=2\Delta M/[a^2c\ (b-a/3)]$, where M is the total magnetic moment and ΔM the vertical difference between the MHL branches (in units of magnetic moment), c is the sample dimension along the *c*-axis and a and b are the lateral sample dimensions, $a\leq b$. Note that this formula does not involve demagnetisation effects and thus applies strictly only for high magnetic fields. For a thin sample ($c < a$, b) the J_c resulting from this expression for low magnetic fields is somewhat underestimated due to the demagnetization effect. It is, however, a common practice to use this formula for the whole field range and for sake of compatibility we will do the same.

The crystal was oxygenated at 410° C, a temperature rather high for the optimum oxygenation of LRE-123 materials, however the critical temperature was still high (93.7 K).

III. RESULTS AND DISCUSSION

The magnetic hysteresis loops detected at temperatures between 65 K and 90 K are shown in Figure 2, where *M* is magnetization and *B* magnetic field (induction). Moreover, at temperatures 65 K, 77 K, and 84 K dynamic relaxation was also measured. All the measurements were carried out with magnetic field applied along *c*-axis. The curves for 65 K exhibited the typical fishtail form, with a strong secondary peak slightly depressed at its summit. Such a depression has been previously attributed to the twin plane

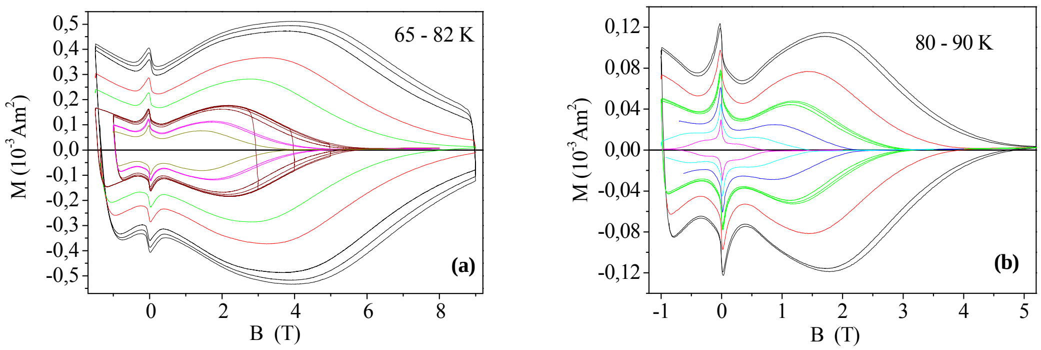


Fig. 2. Magnetic hysteresis loops of the NEG–123 sample for temperatures indicated in the figures (65 to 90 K). While the first (central) peak close to zero field is rather narrow, the most significant feature is the secondary peak at intermediate fields that shifts with increasing temperature to lower fields.

channelling effect* [9,10]. This depression became less evident with increasing temperature and at the first glance it seemed to disappear at all. However, our attempt to fit $J_c(B)$ curves (Figure 3a) with the phenomenological formula for bulk twin-free samples, $J_c(B) = J_{c1} \exp(-B/B_L) + J_{c2}\, b \exp[(1-b^n)/n]$, $b=B/B_{max}$ (the dashed lines), failed up to 84 K. In this function J_{c1} is the magnitude of the central peak, B_L is a characteristic field range [7], J_{c2} and B_{max} are the magnitude and position of the secondary peak, and *n* is the scaling factor, related to the shape of the secondary peak [11,12]. In Figure 3b the corresponding plots of the pinning force density, $F(B)=BJ_c$, normalized to its maximum, are displayed. At the first glance, one could expect a good scaling of the $J_c(B)$ and $F(B)$ curves from Figures 3 a,b with temperature. However, as shown in Figure 4, no temperature range could be found, where the curves would collapse onto a universal curve. One possible explanation might be a complex pinning structure with an effective distribution continuously varying with temperature. However, we believe that the reason lies in the effect of twin plane channeling, which caused $J_c(B)$ curves deformation hiding the real secondary peak summit and preventing a correct curve normalization [9,10]. This conclusion is also supported by the shape of the field dependence of the logarithmic dynamic relaxation rate, $Q=\partial \ln J_c/\partial \ln(dB/dt) \approx \Delta\ln\Delta m/\Delta\ln(dB/dt)$, shown in Figure 4 c for three different temperatures. While a smooth power-law increase of $Q(B)$ would be expected in a twin-free sample [11], here we observe a

* As twin planes represent a strong obstacle for vortex motion, vortices are forced to move along the channels formed by twin boundaries usually oriented approximately at 45^o towards the sample edges. Due to finite sample dimensions, critical currents distribution is irregular and does not follow Bean's model. An analytical formula for a relationship between J_c and the magnetic moment does not exist for such a case. The effect of twin channelling on the magnetization curve shape can be, however, estimated by comparison with the analytical $J_c(B)$ function for a bulk twin-free sample (see Figure 3 and the corresponding discussion).

multiple linear dependence, similar to that reported for twinned single crystals, e.g., in Ref. [13]. Concerning the stripes or the layers observed by both AFM and STM on this crystal (see Figure 1 (c)), it was difficult to differentiate between their effect on the crystal's pinning performance and that of the regular twin planes. The periodicity of both structures was similar, in the range of hundreds nm. Note that these two structures were not aligned.

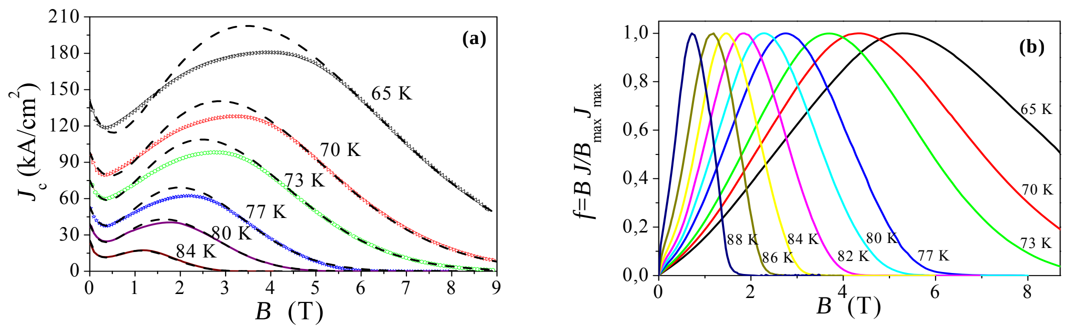


Fig. 3. Temperature scans of (a) the $J_c(B)$ and (b) the normalized pinning force, $f(B)$, dependences. The dashed curves in Fig. 3 a show the best fits by the phenomenological model function for twin-free RE-123 samples.

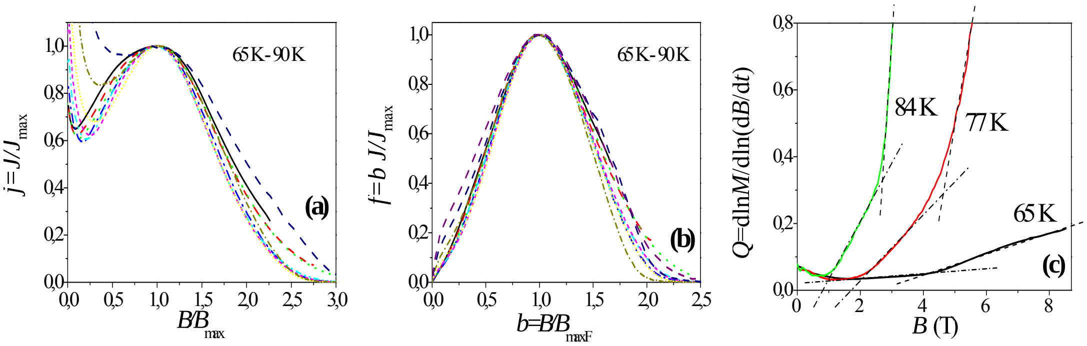


Fig. 4. (a) $J_c(B)$ and (b) $f(B)$ curves normalized with respect to the peak positions; (c) $Q(B)$ dependences at 65 K, 77 K, and 84 K.

From the magnetic studies, we were able to recognize a rather strong point-like pinning disorder, in which LRE/Ba solid solution and oxygen vacancies most probably participate, and a correlated disorder of twin planes (and potentially also the stripes or layers observed by STM). The central peak (lying close to zero field), was comparable in height with the secondary peak in the intermediate fields. With increasing temperature the (central) peak decreased more slowly than the secondary peak, similarly as in MT materials. This indicated a certain amount of "large" uncorrelated defects that enhanced the peak like the secondary phase precipitates in melt-textured samples. The X-ray diffraction analysis identified $BaCuO_2$ impurity particles, which might be the origin of such pinning centers. Note that the central peak in Dy-123, Y-123, or Tm-123 single crystals measured before decreased with increasing temperature faster than the secondary peak.

At the secondary peak position, 2.2 T, the J_c(77 K) value of 62 kA/cm^2 is comparable to the values obtained in good-quality ternary LRE-123 melt-textured materials, implying a nearly optimal random point-like disorder. The study of the individual contributions of oxygen deficiency and LRE/Ba solid solution to the point-like pinning structure are under way. No evidence of a nanoscale correlated (lamellar) substructure that would cause an additional enhancement of pinning at high magnetic fields [5] was recognized in the present magnetic study. Correspondingly, the irreversibility field was rather moderate, reaching about 6 T at 77 K. The minimum value of the logarithmic relaxation rate was rather high at all three investigated temperatures, reaching 0.03 to 0.04 as seen in Figure 4 c. Position of the $Q(B)$ minimum or of the minimum plateau on the $Q(B)$ curve (65 K) was always below the secondary peak position on the corresponding $J_c(B)$ dependence, supporting our understanding that the minimum relaxation is associated with a magnetic field value close to the inflection point between the $J_c(B)$ minimum and the secondary peak maximum [11].

IV. CONCLUSIONS

By means of magnetic studies of the twinned (Nd,Eu,Gd)-123 single crystal we found a close similarity of electromagnetic properties with melt-textured samples of a similar composition. Three sources of pinning could be recognized, namely, a random point-like disorder, twin structure, and a pinning agent effective at low magnetic fields, probably precipitates of $BaCuO_2$. The latter particles contributed to the rather strong central peak on $J_c(B)$ that decreased slowly with increasing temperature and became dominant above 84 K. on the other hand, at lower temperatures the secondary peak originating from contribution of point-like pins of LRE/Ba solid solution and oxygen vacancies was dominant feature. It was, however, depressed due to twin plane channeling effect and none of the normalized $J_c(B)$ and $f(B)$ curves scaled with temperature. We attributed this behavior also to the channeling effect. The significant role of twins was also recognized in the shape of the $Q(B)$ dependence with the multiple linear character similar to that observed in *twinned* RE-123 crystals by other authors. The minimum value of Q was rather high, ranging between 0.03 and 0.04, implying a relatively weak intrinsic pinning barrier. The stripes observed on the crystal surface by STM or AFM had rather large periodicity, comparable to that of the regular twin structure. It was therefore difficult to distinguish their effect from that of regular twins. The observed periodicity was much larger than coherence length; therefore, one could hardly expect a strong influence of these stripes on vortex motion. A careful angle-resolved magnetic experiment, taking care of the mutual orientation of the surface stripes and twin planes might help to recognize a potential pinning effect of the stripes if they represent two-dimensional objects. More work is needed, however, to elucidate the stripes origin.

Acknowledgements
This work was accomplished under the support of Grant Agency of the Czech Republic (GACR 202/05/0173), and MEYS of the Czech Republic (1P05ME728).

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