

**Bose glass state in bulk (Nd, Eu, Gd)Ba<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> with a high irreversibility field**

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(Nd, Eu, Gd)Ba<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> (NEG123) bulk shows the high irreversibility properties. In order to clarify the origin of high irreversibility fields, the transport properties of NEG123 bulk with the high irreversibility field were investigated against high magnetic fields, temperatures, and field angles. We found that a dip in the angular dependent resistivity appears at  $B//c$ , originated to the correlated disorder along the  $c$  axis. In addition, the Bose glass phase was observed around  $B//c$  by the detailed analysis of the glass transition temperature and exponent. Therefore the Bose glass state plays an important role for realization of the high irreversibility field. It is suggested that new effective pinning centers correlated along the  $c$  axis are responsible for the high irreversibility field.

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It is well known that the REBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (RE123, RE=rare earth) system shows high irreversibility fields,  $B_i$ , even at high temperatures such as the boiling point of liquid nitrogen, 77.3 K. The typical  $B_i$  value of RE123 is about 8 T at 77.3 K for  $B//c$  and is much higher than those of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> (Bi2212) and Bi<sub>2</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> (Bi2223) systems.<sup>1</sup> Hence RE123 is expected as a promising high- $T_c$  material for high field applications at high temperatures. In fact, much effort has been made for the development of coated conductors of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$</sub> .<sup>2,3</sup> However, the improvement of the  $B_i$  value as well as an increase in the critical current density,  $J_c$ , are still important issues. It has recently been reported that a high irreversibility field over 14 T is achieved at 77.3 K for  $B//c$  in RE123 bulk where the RE site is compounded with Nd, Eu, and Gd in an appropriate ratio. According to Muralidhar *et al.*,<sup>4</sup> nanolamellae structure is observed in the samples with extremely high irreversibility fields. It is well known that the disorders correlated along the  $c$  axis can enhance both the  $J_c$  and  $B_i$  values through the vortex phase transition from the vortex glass to the Bose glass state. The examples of such  $c$ -axis correlated disorders are columnar defects and twin boundaries.<sup>5-7</sup> The columnar defects introduced by heavy ion irradiation are effective in improving  $J_c$  even when  $T_c$  is degraded. However, the enhancement of  $B_i$  is relatively small. Thus if one can introduce enough density of  $c$ -axis correlated disorders without any degradation of critical temperature, a large enhancement of  $B_i$  is expected. The origin of high irreversibility fields in NEG123 may be ascribed to the defects correlated along the  $c$  axis. With this in mind, we investigate the transport prop-

erties of the NEG123 bulk with focus placed on the vortex state.

The sample used in this study is (Nd<sub>0.33</sub>, Eu<sub>0.38</sub>, Gd<sub>0.28</sub>)Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$</sub>  with 3 mol % NEG211, 10 wt % Ag, and 0.5 mol % Pt prepared by the top-seeded melt-growth method under oxygen partial pressure of 0.1% O<sub>2</sub>,<sup>4</sup> the detail of which is described elsewhere.<sup>8</sup> The sample was cut into a bar with dimensions of about 0.15 × 0.2 × 2 mm<sup>3</sup>. This bar shape sample was mounted on the rotated sample holder with cernox and capacitance thermometers and a heater. The sample temperature was controlled by both He gas flow in a temperature variable cryostat and the heater placed on the sample holder. Magnetic fields were applied using a 20 T superconducting magnet at the High Field Laboratory for Superconducting Materials (HFLSM), Institute for Materials Research (IMR), Tohoku University. Magnetic field angle was defined such that the  $B//c$  axis was  $\theta = 0^\circ$  and transport currents were always perpendicular to the field and  $c$  axis. Transport current density was fixed to be 13 A/cm<sup>2</sup> in the present study.

The inset of Fig. 1 shows the angular dependence of the resistivity at 77.3 K. One should notice that a dip is observed around  $B//c$  and becomes small with increasing magnetic field. In order to discuss the behavior of the dip, the resistivity at  $\theta = 0^\circ$  and  $12^\circ$  are plotted as a function of temperature in Fig. 1, where  $\theta = 0^\circ$  and  $12^\circ$  correspond to the onset and offset angles of the dip, respectively. The kinks in the temperature dependent resistivity for  $\theta = 0^\circ$  and the resistivity rapidly decreases with lowering temperature below the characteristic temperature of the kink,  $T_k$ . However, the resistiv-

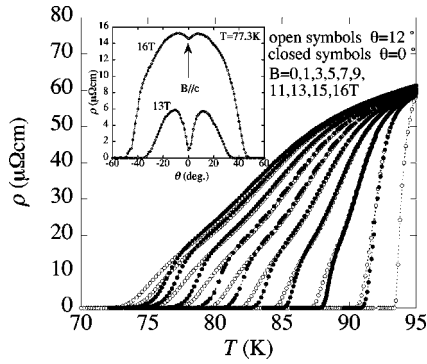


FIG. 1. Temperature dependence of resistivity at  $\theta=0^\circ$  and  $12^\circ$ . The inset shows the angular dependence of resistivity at 77.3 K.

ity for  $\theta=12^\circ$  monotonically decreases with decreasing temperature without kink and is almost the same as that at the onset angle of the dip above  $T_k$ . The difference in the resistivity between  $\theta=0^\circ$  and  $\theta=12^\circ$  grows large below  $T_k$ . These results indicate that the pinning due to the correlated disorders along the  $c$  axis works effectively below  $T_k$ . Details of the high field transport property have been published elsewhere.<sup>9</sup> The temperature dependence and the field angle dependence of resistivity for the NEG123 bulk are very similar to those for twinned Y123 single crystals.<sup>7</sup> In twinned YBCO crystals, the twin boundaries act as the  $c$ -axis correlated disorders in the vortex state and lead to the Bose glass state. The scaling theory of the Bose glass state predicts that when the temperature approaches the Bose glass temperature, the linear resistivity  $\rho$  at a small current density vanishes as  $\rho \sim (T - T_g)^s$ , where  $T_g$  is a glass transition temperature and  $s = \nu'(z' - 2)$  is a critical exponent related with a static exponent  $\nu'$  and dynamic one  $z'$  for the Bose glass.<sup>5</sup> While the scaling theory of the vortex glass state gives a similar expression of  $\rho \sim (T - T_g)^s$  with  $s = \nu(z - 1)$ , where  $\nu$  and  $z$  are critical exponents for the vortex glass.<sup>10</sup> Here we use the same characteristic temperature  $T_g$  as the glass transition temperature for both the Bose glass and the vortex glass states in this study. If  $(d \ln \rho / dT)^{-1}$  is plotted as a function of temperature, we can obtain  $s$  and  $T_g$  values from the linear temperature dependent region. The inset of Fig. 2 shows an example of the determination of  $T_g$  and  $s$ . A solid line is a fitted line obtained using a least-squares method. A small  $s$  value of 3.4 is obviously reasonable from this figure at 7 T. The linear temperature dependence of  $(d \ln \rho / dT)^{-1}$  means that the linear resistivity is associated with the relationship of  $\rho \sim (T - T_g)^s$  and the critical state of the Bose glass or the vortex glass exists in this region. The  $s$  values for various magnetic fields for  $B//c$  are shown in Fig. 2. The  $s$  value in high magnetic fields above 13 T is 7 to 8, and decreases down to about 4 below 9 T and tends to increase below 0.2 T. It is well known that the  $s$  values in the Bose glass phase are smaller than that in the vortex glass phase. In heavy ion irradiated samples, the  $s$  values in the Bose glass state are about 1.3 for the Y123 single crystal, about 3.5 for the Bi2212 single crystal, and 3.6–4.5 for the  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  films.<sup>6,11,12</sup> In addition,  $s$  increases above the matching field  $B_\phi$ , where the number of the vortices is equal to that of the  $c$ -axis correlated defects. Therefore it is sug-

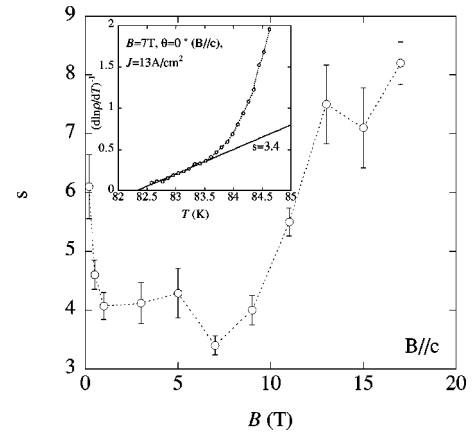


FIG. 2. Critical exponent  $s$  at various magnetic fields for  $B//c$  (onset angle of the dip). The inset is an example of the least-squares fit for determination of  $s$  and  $T_g$ .

gested that the Bose glass state is realized in this sample with the effective matching field of about 9 T.

The temperature dependent resistivities for field angles from  $0^\circ$  to  $-20^\circ$  are shown in Fig. 3. The inset of Fig. 3 indicates the angular dependence of  $s$  and  $T_g$  at 0.5 T. Both  $s$  and  $T_g$  are almost constant for  $-20^\circ < \theta < -6^\circ$  but vary with the field angle above  $-6^\circ$ . The  $s$  values below  $-6^\circ$  are 6 to 7 and correspond to those at high magnetic fields above 13 T for  $B//c$ . The Bose glass theory predicts that the angular dependence of  $T_g$  shows a cusplike peak around  $B//c$  for the  $c$ -axis correlated disorder, which was also confirmed empirically.<sup>5,13</sup> The angular dependence of  $T_g$  and  $s$  also suggests the presence of the Bose glass phase. There are only a few studies on the vortex dynamics such as the vortex glass for the melt textured RE123 bulk, although many reports on the transport and flux pinning properties have been published.<sup>14,15</sup> In particular, no detailed experiment on the critical exponent in the glassy state of the vortex was reported up to now. Hence the Bose glass behavior of the RE123 bulk system without columnar defects was confirmed for the first time by the detailed evaluation of the critical exponents in this study.

Figure 4 shows the  $T_g$  value in a  $B$ - $T$  plane. The  $T_g$  value is about 10.5 T at 77.3 K and is larger than that for twinned Y123 single crystals.<sup>16</sup> The normalized  $T_g$  obtained in this study is compared with that of the twinned Y123 single crystal in the inset of Fig. 4. The  $T_g$  of the single crystal shows the kink at  $B_\phi$  and is smaller than the extrapolated values from the low field region above  $B_\phi$ . Whereas, there is no anomaly on  $T_g$  around 9 T where the  $s$  value increases for the NEG123 bulk. In addition, the  $T_g$  value agrees with that of the single crystal below 9 T but is larger above 9 T. The Monte Carlo simulation shows that the curvature of  $T_g$  changes at  $B_\phi$  and  $B_\phi/3$  due to the discontinuous change of the averaging trapping rate to the columnar defects in case of coexistence of the columnar and weak random point pinning sites in Bi2212.<sup>17</sup> However, in the film samples with randomly distributed strong pinning defects, the anomaly of  $T_g$  is not observed at  $B_\phi$  and the  $T_g$  value becomes large above  $B_\phi$ , compared with those of the single crystals.<sup>18</sup> Hence we can conclude that the coexistence of the strong random pin-

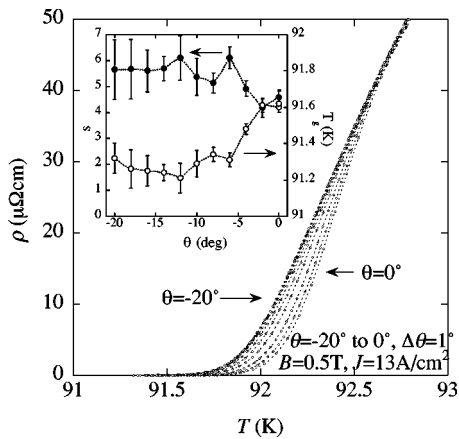


FIG. 3. Temperature dependence of resistivity at 0.5 T and various field angles from  $-20^\circ$  to  $0^\circ$ . The inset shows the angular dependence of the obtained  $s$  and  $T_g$  values at 0.5 T.

ning and the  $c$ -axis correlated disorder are responsible for the high irreversibility field for NEG123 bulk. A theoretical model on the Bose glass state in the competition system of the correlated and the random pinning centers will be necessary to fully understand the nature of the NEG123 bulk system in detail.

The average distance between the  $c$ -axis correlated disorders is estimated at about 10 nm from the effective matching field of 9 T. This distance is in agreement with the average distance among the nanolamellae in the NEG123 bulk with the high irreversibility field over 14 T at 77.3 K.<sup>4</sup> Therefore it is considered that the nanolamellae has a  $c$ -axis correlation and becomes an origin of the high irreversibility fields. This is consistent with the fact that the nanolamellae are observed only in NEG123 bulk samples with the high irreversibility field.<sup>8</sup> However, the possibility that the twin boundaries may be effective as the  $c$ -axis correlated disorder still remained because the effective  $B_\phi$  is very analogous to that of the twinned Y123 single crystal and the normal resistance of the NEG123 bulk is as small as that of the single crystal. The more detailed microstructural observations are necessary.

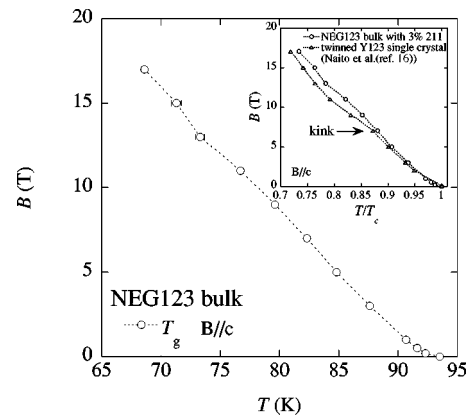


FIG. 4. Obtained  $T_g$  at various magnetic field for  $B//c$ . Inset shows the comparison of  $T_g$  between the NEG123 bulk and the twinned Y123 single crystal of Ref. 16.

On the other hand, an increase of  $s$  at low magnetic fields less than 0.2 T may also be related with the low field anomaly around  $B_\phi/3$  estimated by the Monte Carlo simulation, although 0.2 T is smaller than  $B_\phi/3$ .<sup>17</sup> An increase of the critical exponent below  $B_\phi/3$  was already observed experimentally for the Y123 films.<sup>18</sup>

In summary, transport measurements were carried out for the NEG123 bulk sample with the high irreversibility field in order to study its origin. We found that the Bose glass nature for the NEG123 bulk plays an important role for the increase of the irreversibility field and the introduction of the correlated disorder surely improves the irreversibility field as well as the critical current density.

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