Bulk YBCO with discontinuous irradiation defects: Bose-glass behaviour and very high critical current densities

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Abstract. Columnar defects were produced in melt-textured YBCO by irradiation with high-energy U238 ions at a constant matching field of Bφ = 10 T and for several energy losses between Se = 1.67 and 2.4 keV/Å. The influence of discontinuous or multiple-in-line-damage (MILD) columnar defects in melt-textured YBCO on flux pinning and the vortex matter phase diagram was investigated. The critical current density jc(H,T) was found to strongly increase with Se due to the increasing length of the MILD pins. Simultaneously, the irreversibility field Birr(T) for fields along the c-axis progressively shifts upwards reaching 9 T at 77 K. For Se = 2.4 keV/Å, a pronounced kink is observed in Birr(T) at 8 T which is a strong indication for Bose glass behaviour. The data for the irradiated YBCO are compared with data for bulk YBCO in which a periodic array of nanoscale twin boundaries was obtained by RuO2 additions.

1. Introduction
Much progress has been achieved in the last years to improve flux pinning in bulk melt-textured YBCO by chemical doping [1] and by irradiation methods [2,3]. Extended amorphous tracks with a diameter of 5-10 nm being highly suitable for flux pinning can be produced by irradiation with fast neutron irradiation [3] and heavy ions. The average separation dr between columnar defects produced by irradiation with heavy ions is determined by the ion fluence which is usually expressed in terms of the matching field Bφ = φ/dr2 producing an equivalent density of vortices in the superconductor. A columnar defect structure results in large pinning energies and high critical currents, particularly when the magnetic field is applied parallel to the defect as was demonstrated both for YBCO single crystals [4] and melt-textured YBCO [5].

The morphology of columnar defects is mainly determined by the energy loss Se =dE/dx transferred from the ion beam to the superconductor. In YBCO, continuous columns are formed at rates of Se ≥ 3.5 keV/Å [6], however, the columnar damage becomes discontinuous at lower Se [5]. For high irradiation doses or ion fluences, the critical current density jc becomes significantly higher for MILD pins than for CCPC [5,7]. The main reason is the strong reduction of the superconducting cross section.
in the presence of a high density of CCPC. The advantage of MILD pins is that much higher densities of columnar defects can be realized than for CCPC because a larger superconducting volume is available for current percolation. The properties of superconductors with correlated columnar defects are described by the Bose-glass model [8]. This model predicts the formation of a Bose-glass phase below a Bose-glass line which significantly shifts the irreversibility line \( B_{irr}(T) \) to higher fields. This is due to strong pinning of single vortices at columnar defects for \( \mu_o H < B_o \), where the pins outnumber the vortices. For fields \( \mu_o H > B_o \), the vortices outnumber the strong pins, so that the additional vortices are weakly pinned at interstitial sites between the strongly pinned vortices forming vortex bundles. In many cases, a kink in the temperature dependence of the irreversibility field \( B_{irr}(T) \) was observed near the matching field \( B_m \) [9].

Although the tracks introduced by irradiation can enhance the pinning properties significantly, large-scale applications are limited due to the short penetration length of heavy ions and by the high fabrication costs of irradiation techniques. Therefore, there is continued interest in finding simpler ways to generate new or to refine existing extended nanodefects in bulk YBCO. Twin boundaries (TBs) are an example for correlated disorder existing already in bulk YBCO. Because TBs are planar defects lying in the (100) or in the (110) plane, they are aligned with the c-axis and are expected to be effective pinning sites for vortices when the flux motion direction is transverse to the TB. Indeed, improved critical current densities were reported for melt-textured YBCO with refined twin structures [10]. Recently, it was demonstrated that a periodical array of nanoscale TBs can be formed within melt-processed YBCO by RuO₂ additions resulting in improved pinning properties. Clear signatures of a Bose-glass phase were found in this material [1].

In the present paper, the influence of MILD pins in YBCO on \( j_c, B_{irr}(T) \) and the vortex matter phase diagram and its anisotropy is studied for a series of samples irradiated by heavy \( ^{238}\text{U} \) ions at different energy losses \( S_e \). The main focus was to investigate the irreversibility line up to high applied fields in order to check typical features and peculiarities of the Bose glass behaviour in these samples.

2. Experiments

The investigated series of melt-textured YBCO samples was irradiated at the GSI Darmstadt by a beam of \( 60.2 \text{ GeV} \ ^{238}\text{U} \) ions at different energy losses \( S_e \) and at a fluence of about \( 2.5 \times 10^{11} \text{ ions/cm}^2 \) corresponding to a matching field of \( B_m = 10 \text{T} \). Whereas data for \( S_e \) up to \( 4 \text{ keV/Å} \) including samples with MILD pins and CCPC have been published elsewhere [5], the focus of the present paper is on MILD pins. In particular, data for three samples with \( S_e = 1.67, 2.0 \) and \( 2.4 \text{ keV/Å} \) will be presented. The obtained results are compared with those for a non-irradiated melt-textured YBCO sample.

The superconducting transition temperature was determined from \( ac \) susceptibility using the onset of superconductivity as criterion. The upper critical field \( B_{c2} \) and the irreversibility field \( B_{irr} \) were obtained from \( dc \) magnetization curves \( M(T) \) of the YBCO samples which were measured in magnetic fields up to \( 7 \text{T} \). The critical current density \( j_c \) was determined from \( M(H) \) magnetization loops using the relation \( j_c(H) = 30 \Delta M(H)/d \) where \( \Delta M \) is the difference of the magnetization between descending and ascending field branches of the magnetic hysteretic loop measured in emu/cm², \( d \) is the diameter of the cylindrical samples measured in cm and \( j_c \) is measured in A/cm².

3. Morphology of columnar defects

The diameter of the damaged region, \( d_d \), of columnar irradiation defects was studied for many ion species at different energies [6]. Using these data, the simple empirical relation

\[
d_d = 4.8 \text{ nm} \ (S_e - 0.7)
\]

was established for YBCO [5], where \( S_e \) is given in \( \text{keV/Å} \). For \( S_e \), just above \( 0.7 \text{ keV/Å} \), spaced beads of damage are created which elongate and increase in diameter as \( S_e \) increases forming short columnar defects separated by large superconducting gaps. Equation (1) applies to irradiation with heavy ions including \( ^{238}\text{U} \) ions. For \( S_e > 3.5 \text{ keV/Å} \), the damage in YBCO was found to become continuous [6].
Using the rather sparse data available for the pinning-effective fraction $f$ in YBCO, the relation between $f$ and $S_e$ has been approximated by [5]

$$f = 0.213 (S_e - 0.7)^{1.5}$$  \hspace{1cm} (2)

where $S_e$ is given in keV/Å. According to equation (2), discontinuous or MILD pins are expected to form in the range of $S_e$ values between 0.7 and 3.5 keV/Å. Data for the morphology of the columnar defects in the investigated samples are listed in Table 1.

4. Results and discussion

4.1. Superconducting transition temperature
The transition temperatures $T_c$ of the investigated samples are collected in Table 1 together with data for the morphology of the columnar irradiation defects. $T_c$ is found to slightly decrease with increasing $S_e$ which is due to enhanced disorder introduced by ion irradiation.

Table 1. Energy loss $S_e$, $T_c$, diameter $d$ and pinning-effective fraction $f$ of the columnar defects.

| $S_e$ (keV/Å) | $T_c$ (K) | $d$ (nm) | $f$  |
|--------------|-----------|---------|------|
| 1.67         | 90.6      | 4.7     | 0.20 |
| 2.0          | 90.0      | 6.2     | 0.32 |
| 2.4          | 89.8      | 8.2     | 0.47 |

4.2. Critical current density
In figure 1(a), the field dependence of the critical current density at 77 K is shown for three irradiated samples with $S_e = 1.67$, 2.0 and 2.4 keV/Å. The high $j_c$ of 275 kA/cm$^2$ achieved for $S_e = 2.0$ keV/Å and 2.4 keV/Å at low applied fields is a record value for bulk YBCO. In figure 1(b), $j_c(H)$ data of the sample with $S_e = 2.4$ keV/Å are shown for several temperatures between 77 and 60 K. At 60 K, $j_c$ increases up to 850 kA/cm$^2$.

Note that $j_c$ was reported to decrease for $S_e$ exceeding 2.4 keV/Å, i.e. $j_c$ vs. $S_e$ exhibits a peak [7]. This decrease of $j_c$ is caused by (i) the strong reduction of the superconducting area in the presence of

Figure 1. (a) Field dependence of the critical current density of three irradiated YBCO samples measured at 77 K for applied fields $H || c$. The energy losses of the ion beam are given in keV/Å. (b) Field dependence of $j_c$ for the YBCO sample with $S_e = 2.4$ keV/Å measured for several temperatures at $H || c$. 

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a high density of continuous columnar defects and (ii) the enhanced damage and the consequent reduction of $T_c$ of the remaining superconducting area.

4.3. Vortex matter phase diagram

In figure 2, the determination of the upper critical field $B_{c2}$ and of the irreversibility field $B_{irr}$ is exemplified. Shown is the magnetization $M(T)$ after zero-field (ZFC) and after field cooling (FC) measured at an applied field of 6 T. The temperature $T_{c2}$ (with $B_{c2}(T_{c2}) = 6$ T) is obtained by linear extrapolation of the reversible $M(T)$ curve to $M$ in the normal state, whereas $T_{irr}$ (with $B_{irr}(T_{irr}) = 6$ T) is taken at the transition from the irreversible to the reversible part of the magnetization curve.

The vortex matter phase diagram of the samples with $S_e = 1.67$ and 2.4 keV/Å is compared in figure 3 for $H\parallel c$. The data in figure 3 reveal a significant enhancement of $B_{irr}(T)$ for $H\parallel c$ due to the higher energy transferred from the ion beam to the bulk YBCO, whereas $B_{c2}(T)$ for $H\parallel ab$ and the upper critical fields $B_{c2}(T)$ for both field directions remain nearly unchanged after the stronger irradiation.

From figure 3, one finds a $B_{c2}$ anisotropy $\gamma_c = B_{c2}^{ab}/B_{c2}^c$ of about 4.1 at 88.5 K both for $S_e = 1.67$ and 2.4 keV/Å. In contrast, the anisotropy $\gamma_l = B_{irr}^{ab}/B_{irr}^c$ of $B_{irr}(T)$ strongly reduces from $\gamma_l = 4.1$ for $S_e = 1.67$ to 1.67 keV/Å and $S_e = 2.4$ keV/Å. The irreversibility line $B_{irr}(T)$ for $H\parallel c$ strongly shifts upwards by irradiation with $S_e = 2.4$ keV/Å. In contrast, $B_{c2}^{ab}(T)$, $B_{c2}^{ab}(T)$ and $B_{c2}^{ab}(T)$ remain nearly unchanged.
1.67 keV/Å to $\gamma = 2.5$ after irradiation with $S_e = 2.4$ keV/Å. For comparison, the non-irradiated reference sample exhibits a relatively large $B_{c2}$ anisotropy of $\gamma_c \approx 7$ which is within the range of $\gamma_c$ values between 5 and 7 reported for YBa$_2$Cu$_3$O$_{7-\delta}$ single crystals.

In figure 4, the evolution of $B_{irr}$ (for $H||c$) with increasing irradiation is shown together with $B_{c2}$ data. Additionally, $B_{irr}(t)$ and $B_{c2}(t)$ data for the non-irradiated YBCO reference sample are included in this figure. The $B_{c2}(t)$ data of this sample which has a $T_c$ of 91.5 K perfectly coincide with those of the irradiated samples (see figure 4). The irreversibility lines compared in figure 4 follow a power law

$$B_{irr}(T) = B_0 (1 - T / T_c)^n$$

with $n = 1.2 \ldots 1.4$. These values of $n$ are close to $n = 4/3 = 1.33$ predicted by Fisher et al. [11] for vortex glass melting.

### 4.4. Bose-glass behaviour of YBCO after irradiation and by RuO$_2$ addition

The temperature dependence of the irreversibility line for $H||c$ was analyzed in an extended temperature and field range by including $B_{irr}$ data derived from the critical current density $j_c(H)$. $B_{irr}^*$ was taken from the $j_c(H)$ dependence at a fixed value of $j_c = 100$ A/cm$^2$. At this criterion, $B_{irr}^*$ was found to coincide with the $B_{irr}$ value obtained from the reversible $M(T)$ dependence.

The obtained irreversibility lines presented in figure 5 cover a field range up to 14 T. For $S_e = 1.67$ and 2.0 keV/Å, the $B_{irr}(T)$ data were found to follow at high fields the same power law established for fields up to 7 T (not shown in figure 4). In contrast, a kink in the $B_{irr}(T)$ dependence is found for $S_e = 2.4$ keV/Å, i.e. for that sample which was exposed to the strongest irradiation. This kink which appears at $B_k = 8.0$ T close to the switching field $B_s = 10$T is a strong indication for Bose glass behaviour and the presence of correlated disorder in this sample [8].

In figure 5, $B_{irr}(T)$ data for a YBCO sample with RuO$_2$ additions are included. The kink in $B_{irr}(T)$ observed at about $B_k = 2.8$ T is due to a periodical array of nanoscale twin boundaries (TBs) which was reported to form in this material [1]. An average defect separation of about 30 nm estimated from this field is comparable with the spacing of nanotwins which was observed by TEM investigations in this YBCO sample. In spite of the enhanced irreversibility fields within the Bose-glass phase below 2.8 T, the critical current density of this YBCO sample at 1T is only by about 50% higher than $j_c$ of the reference sample measured at the same applied field. The reason might be that flux pinning by TBs
is strongly anisotropic, i.e. TBs act as strong pinning centers only when the flux motion direction is transverse to the TB.

5. Conclusions

A high concentration of discontinuous columnar defects was created in melt-textured YBCO by irradiation with high-energy U^{238} ions at \(B_{\phi} \approx 10\) T and an energy loss of \(S_e \approx 2.4\) keV/Å. The irreversibility line of this sample is shifted to high magnetic fields and exhibits a pronounced kink at 8 T close to the matching field of \(B_{\phi} \approx 10\) T indicative of correlated disorder and Bose glass behaviour. In YBCO with a periodical array of nanoscale twin boundaries along the \(c\)-axis, a very similar kink in \(B_{irr}(T)\) was found at a field of 2.8 T, i.e. the density of nanotwins in this superconductor is about three times smaller than the density of columnar defects in the irradiated YBCO sample. Whereas extremely strong flux pinning is achieved by the discontinuous columnar irradiation defects, the question remains open to what extent the flux pinning properties of YBCO with the nanotwins can be further improved.

6. References

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