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Absence of correlated flux pinning by columnar defects in irradiated epitaxial Bi₂Sr₂CaCu₂O₈ thin films

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Abstract

Using heavy-ion irradiation, we produced columnar defects of different density and orientation in epitaxial $Bi_2Sr_2CaCu_2O_8$ thin films. Although this increases the normal state resistivity and the critical temperature is reduced proportionally to the volume fraction of damaged material, pinning-related quantities like critical current density, activation energy and depinning field are enhanced in external magnetic fields. Transport measurements in dependence of the magnetic field and its orientation consistently indicate *two-dimensional* pinning of pancake vortices at the columnar defects. We observe the absence of correlated flux pinning by columnar defects and compare to heavy-ion-irradiated single crystals and tapes, where line-like correlations have been observed. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Experimental [1,2] and theoretical [3,4] investigations have shown that columnar defects created by heavy-ion irradiation are the most effective form of artificial disorder for the pinning of magnetic flux in high- T_c cuprate superconductors. These defects enhance the critical current density by orders of magnitude [5,6] and considerably shift the irreversibility line to higher fields and temperatures [7–9]. Best results are obtained by a splay configuration of the amorphous tracks because this forces vortex entanglement which even more prevents flux lines from moving [10–14]. Due to the strong interaction between flux lines and columnar defects [15], heavy-ion irradiation also offers a tool to analyze pinning mechanisms and vortex dynamics. In compounds like YBa₂Cu₃O₇ (YBCO) [16] and Tl₂Ba₂CaCu₂O₈ [17], which are less two-dimensional than Bi₂Sr₂CaCu₂O₈ (Bi-2212), heavy-ion irradiation leads to strongly anisotropic pinning with a maximum for the applied magnetic field aligned to the columns. In Bi-2212, however, where vortex dynamics are dominated by weak Josephson coupling between the layers, this effect should be much smaller.

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Nevertheless, some experiments on Bi-2212 single crystals [18–21] and tapes [22] show strongest pinning for the magnetic field oriented parallelly to the columnar defects. Although these effects are very small, they indicate line correlations of vortices after the introduction of correlated disorder, even in this highly anisotropic material.

In this paper, we investigate vortex dynamics in epitaxial Bi-2212 thin films exposed to heavy-ion irradiations of different doses and directions. By means of detailed transport measurements in external magnetic fields and in dependence of its orientation, conclusions can be drawn on the vortex behavior in the presence of columnar defects. Our studies establish the absence of correlated flux pinning by columnar defects which is an essential deviation from some results obtained on irradiated crystals and tapes. This paper is an extension of a previous work [23]. It includes analysis and results in a final and complete form.

2. Experimental details

Epitaxial Bi-2212 films with a thickness of 4000 Å were prepared on $9 \times 9 \times 1 \text{ mm}^3$ (100) SrTiO₃ by dc sputtering in flowing oxygen atmosphere. Details of preparation and structural characterization of the *c*-axis oriented films are described elsewhere [24]. In order to examine the effect of different irradiations, several specimens of the same quality have to be prepared which is not a trivial task for Bi-2212 films. Therefore we patterned four identical striplines on the same film (Fig. 1). Subjecting the striplines of each film separately to different irradiation procedures, the analysis is thus unaffected by varying superconducting properties one would have in the case of different samples. For the presented experiments, this plays a basic role.

Irradiations with ¹⁹⁷Au ions of ~ 2.2 GeV kinetic energy were carried out at the UNILAC of the Gesellschaft für Schwerionenforschung (GSI) in Darmstadt. After similar irradiations, the production of continuous amorphous tracks of approximately 8 nm in diameter along the linear path of each ion extending through the whole thickness of the films was confirmed by transmission electron microscopy [25]. Two series of irradiations of different types



Fig. 1. Patterning of the films with four identical microbridges of $2 \text{ mm} \times 50 \mu \text{m}$ on the same substrate. By particular combinations of four of the ten annealed silver contact pads each stripline can be measured in the standard four-point technique.

were performed at room temperature on two different films, respectively. Series A are irradiations on three striplines of the first sample parallel to the film's *c* axis with different doses n_{cd} corresponding to dose equivalent fields $B_{\phi} = n_{cd}\Phi_0$ (Φ_0 is the flux quantum) of 0.3, 1 and 2 T. Series B are irradiations on three striplines of the second sample perpendicular to the long axis of the striplines but under different angles φ of 45°, 30° and 15° with respect to the film's *c* axis and each with a dose corresponding to $B_{\phi} = 1$ T.

All transport measurements presented in this work were performed in a superconducting magnet of a low-temperature cryostat using the standard fourpoint technique. In order to change the magnetic field orientation relative to the film's *c* axis, the samples were mounted onto a rotatable copper block which includes a heater and a Pt resistive thermometer. Using pulsed currents $\leq 100 \ \mu$ A, resistive measurements were well in the ohmic regime [26] with thermal voltages eliminated by current reversal and averaging the absolute voltage drops. From the linear part of resistive transitions in Arrhenius representation, the activation energy for thermal flux movement could be derived. The transport critical current

160

140

120

100

80

B = 0 T

stripline 1

stripline 2

--- stripline 3

- stripline 4

densities were determined with the aid of a 0.5 μ V/mm voltage criterion. Variation of the criterion only shows small influence on the absolute values. The applied magnetic field at which the current density of 500 A/cm² gives a voltage drop of 60 nV/mm is defined as the depinning field B_{dep} .

3. Results and discussion

3.1. Transport measurements

Before irradiation the nearly identical behavior of the four striplines on a substrate was checked with regard to normal and superconducting properties. As an example, in Fig. 2a the resistive transitions of four striplines nearly coincide and T_c^{zero} is the same. In external fields parallel to the film's c axis (B||c), the low part of the resistivity follows $\rho =$ $\rho_0 \exp(-U(T,B)/k_{\rm B}T)$ [26,27] with an activation energy $U(T,B) \sim (1/\sqrt{B})(1-T/T_c)$ for plastic double-vortex kink formation [26,28-30]. The values of the activation energy of the four striplines are found to be equal.

After irradiation series A (Fig. 2b), we observe an increase of the normal state resistivity and a reduction of T_c^{zero} . The decrease of T_c^{zero} depends linearly on the ion dose. After irradiation series B (Fig. 2c), this behavior is qualitatively the same but shows no dependence on the direction of irradiation. With increasing angle φ , the amorphous tracks in the films become longer but at the same time their density in the *ab* plane decreases by the same factor. Therefore the damaged volume is independent of φ . From this, we conclude a reduction of T_c proportional to the volume fraction of damaged material, which also seems to be the relevant quantity for the change of the normal state resistivity. Furthermore, derived from resistive transitions after irradiation series A Fig. 3 shows the field dependence of the activation energy. As a consequence of the matching effect between the vortex density and the density of columnar defects, the values of the irradiated striplines deviate from $U \sim 1/\sqrt{B}$ in such a way that strongest enhancement is found for fields close to B_{ϕ} .

A test for the correlated nature of vortex pinning by columnar defects is the observation of alignment



on the same substrate (a) without irradiation, (b) after irradiation series A and (c) after irradiation series B (see text). In (b) the dose equivalent field and in (c) the angle φ of the irradiation is specified for each stripline. The resistivity in (b) is normalized with respect to the resistivity at 120 K of each stripline before irradiation.

effects when the external magnetic field is continuously rotated by an angle ϑ with respect to the film's c axis. Except if noted, all the following measurements are performed after irradiation series B, while magnetic field and columnar defects are always parallel to a plane being perpendicular to the fixed direction of the transport current. The accuracy of the orientations are $\sim 2^{\circ}$ for the magnetic field and $< 1^{\circ}$ for the columnar defects. Fig. 4 shows the transport critical current density in dependence of the



Fig. 3. Activation energy of four identical striplines on the same substrate after irradiation series A (see text). For each stripline the dose equivalent field of the irradiation is specified.

field orientation. $J_c(\vartheta)$ exhibits strong intrinsic pinning peaks at $\pm 90^\circ$ which are slightly smaller for the irradiated striplines due to T_c -reduction and degradation of the material. In contrast to similar experiments on YBCO-thin films [16], Fig. 4 gives no indication of irradiation-induced directional pinning because $J_c(\vartheta)$ is a symmetric function with no additional maxima for the field direction aligned parallelly to the columnar defects, i.e., for $\vartheta = \varphi$. Nevertheless, the heavy-ion irradiation has a definite influence on $J_c(\vartheta)$. Close to $\vartheta = 0^\circ$, there is a clear enhancement of J_c which seems to be independent of the direction of irradiation. This behavior is seen by a measurement of J_c in dependence of the field



Fig. 4. Transport critical current density J_c (criterion: 0.5 μ V/mm) of four identical striplines on the same substrate after irradiation series B (see text). For each stripline the angle φ of irradiation is specified and indicated by arrows.

parallel to the film's c axis (B||c) which is shown in Fig. 5a presenting its typical characteristic: compared to the unirradiated stripline in small fields, J_c of the irradiated striplines is reduced due to T_c -reduction and degradation of the material, while in higher fields the prevailing pinning of the columnar defects leads to an enhancement of J_c . From the values $J_{c}(B)$ in Fig. 5a, we calculate $J_{c}^{*}(\vartheta) = J_{c}(B||c)$ $B_0 |\cos\vartheta|$). With $B_0 = 0.5$ T, $J_c^*(\vartheta)$ coincides with $J_{c}(\vartheta)$ in Fig. 4 for all ϑ in all striplines. That means in spite of the irradiation, $J_{\alpha}(\vartheta)$ is determined only by $B \| c$, the component of the magnetic field in the direction of the film's c axis. Therefore in Fig. 4, the enhancement of J_c near $\vartheta = 0^\circ$ becomes clear since in this angular range $B \parallel c$ is close to 0.5 T for which the enhancement is given in Fig. 5a. For all other angles, $B \parallel c$ is less than 0.5 T which, in accordance with Fig. 5a has the consequence of a smaller enhancement and in the vicinity of $\vartheta = +90^{\circ}$, the



Fig. 5. (a) Transport critical current density J_c (criterion: 0.5 μ V/mm) of four identical striplines on the same substrate after irradiation series B (see text) for fixed orientation B||c. In (b) the enhancement factors are calculated from (a). For each stripline the angle φ of irradiation is specified.

reduction of J_{a} . Additionally this result is confirmed by a measurement similar to that of Fig. 4 using an applied field of 3 T instead of 0.5 T. Again with $B_0 = 3$ T, we find $J_c^*(\vartheta) = J_c(\vartheta)$, but the most enhanced values of the irradiated striplines now appear at $\vartheta \approx +75^\circ$, $+105^\circ$ where $B \parallel c \approx 0.75$ T corresponding to the best improvement of J_{c} in Fig. 5a. At $\vartheta = 0^\circ$ ($\Rightarrow B \parallel c = 3$ T), there is no difference for the values of the four striplines as is also expected from Fig. 5a at B = 3 T. Altogether nine measurements, analogous to those of Figs. 4 and 5a at temperatures from 40 to 82 K and in fields between 5 mT and 3 T. qualitatively lead to the same results discussed above. In order to investigate pinning by columnar defects at smaller matching fields $B \approx B_{\phi}$ and therefore with a lower ratio of the vortex-vortex in-plane interaction to the coupling in c direction, a further sample with $B_{\phi} = 0.28$ T was analyzed. In this case irradiation was performed under an angle of $\varphi = 45^{\circ}$ and columnar defects were created by 238 U ions of ~ 1.4 GeV kinetic energy. The same behavior as discussed above was observed.

Supposing the existence of pancake vortices with very weak Josephson coupling between the layers in Bi-2212 thin films, the pinning of magnetic flux by columnar defects then is reduced to an individual pinning of pancakes at the correlated disk-like defects created by the irradiation in each CuO₂-plane. If every pancake only interacts with those defects in that plane it is contained in, it is imaginable that for an enhanced pinning the presence of the disks is important but not the direction of the ion beam from which they are created. This explains that in Fig. 4 the curves $J_{\alpha}(\vartheta)$ of the irradiated striplines are nearly identical implying the independence on the direction of irradiation. However, if decoupled pancakes exist there should be small but significant differences depending on the disk density in the planes which varies proportionally to $\cos \varphi$. Because highest pinning should occur for roughly equal numbers of pancakes and disks (matching effect), this can be observed by evaluating the enhancement factors of $J_c(B)$ in Fig. 5a. The result in Fig. 5b displays maxima at fields just below $B_{\phi} = 1$ T. With increasing angle φ of irradiation, the position of the maxima is found at smaller fields which is an indication for the matching between pancakes and disks. Moreover, the locations of the maxima scale with $\cos \varphi$. That means, with B_{max} being the position of the respective maxima in Fig. 5b and with $B_{\phi}^{\text{eff}} = B_{\phi} \cos \varphi$ defined as the effective dose equivalent field in the planes, the data points plotted as B_{max} vs. B_{ϕ}^{eff} lie almost on a straight line.

In order to exclude directional pinning by columnar defects in the Bi-2212 thin films as well in low current transport measurements, the activation energy $U(T = 0, B = 1 \text{ T}, \vartheta)$ was derived from resistive transitions measured in an applied field of 1 T at various angles ϑ . Resulting from the strong pinning by columnar defects, Fig. 6a clearly shows an improvement in wide regions of the angle ϑ . Since the curves $U(\vartheta)$ are symmetric with respect to $\vartheta = 90^{\circ}$ without any maxima for $\vartheta = \varphi$, no irradiation-induced directional pinning can be observed.

In Bi-2212 single crystals, anisotropic pinning due to columnar defects was demonstrated by devia-



Fig. 6. (a) Activation energy and (b) scaled depinning field of four identical striplines on the same substrate after irradiation series B (see text). For each stripline the angle φ of irradiation is specified and indicated by arrows. In (b) the decrease of $B_{dep}|\cos\vartheta|$ with increasing ϑ is based on a systematic behavior of the experimental setup. The raw data shown are not corrected for this effect.

tions from scaling rules for the irreversibility field $B_{inv}(T = \text{const}, \vartheta)$ [21]. The scaling approach of Blatter et al. [31] only works for isotropic disorder and a breakdown is a sensitive proof of correlated pinning arising from the columnar defects. For Bi-2212 with high anisotropy ($\gamma > 150$), the scaling relation $B_{\rm irr}(T,\vartheta) = B_{\rm irr}(T,\vartheta=0^\circ)/\varepsilon_\vartheta$ with $\varepsilon_\vartheta^2 = \cos^2\vartheta + \frac{1}{2}$ $1/\gamma^2 \sin^2 \vartheta$ yields $B_{irr}(T,\vartheta) \cos \vartheta$ to be independent of ϑ as long as the applied field is not too close to the *ab* plane. Based on $B_{irr} \approx B_{dep}$, Fig. 6b shows that in our measurements for a Bi-2212 film, the scaling remains valid even after the introduction of columnar defects. $B_{dep}(T = \text{const}, \vartheta) |\cos \vartheta|$ of the irradiated striplines is improved but the independence of ϑ is conserved which confirms the absence of directional pinning due to the columnar defects.

Finally we exclude that the nonobservation of irradiation-induced directional pinning could be possibly due to the fact that vortices are locked to the columns over a very broad angular range and directional pinning in fact would dominate the whole experiment. Because then it would be impossible that all presented transport measurements in dependence of the magnetic field orientation show such an exact symmetric behavior and that in spite of the irradiation, the transport critical current density is determined only by the component of the magnetic field in the direction of the film's c axis. Moreover, for the depinning field the scaling approach of Blatter et al. should not fit so well because this only works for isotropic disorder.

3.2. Comparison with single crystals and tapes

Comparing our results of thin films with magnetization measurements of Bi-2212 single crystals [18– 21] and tapes [22] differences in the dimensionality of pinning by columnar defects are obvious. While in thin films vortices behave as decoupled two-dimensional pancakes, in crystals and tapes their line-like correlation was established by unidirectional pinning along the ion tracks, even though much less pronounced than in YBCO (compare with Ref. [32]). Since also transport measurements on irradiated tapes of (Bi,Pb)₂Sr₂Ca₂Cu₃O₁₀ [33] and in the flux transformer geometry performed on Bi-2212 single crystals [34,35] show three-dimensional pinning and give evidence to an irradiation-induced line correlation of pancakes, we think that the different methods of measurement (transport/magnetization) are not be the reason for this discrepancy.

To our understanding, the most appropriate interpretation of this puzzle is due to the different microstructures of films and crystals. In the latter the three-dimensional behavior may be explained by a theory by Koshelev et al. [36] who found that in layered superconductors, columnar defects effectively increase interlayer Josephson coupling by suppressing thermal fluctuations of pancakes. But in films there is a much higher concentration of natural pointlike defects. These are in competition with the columnar pinning by presenting additional, although weaker, pinning sites and reduce the width and height of the energy barrier vortices have to overcome for a detachment from the columns. Therefore the probability of finding vortex segments outside the columnar defects is increased, having the consequence of a reduced localization of vortices at the columns which finally defeats the effect of vortex stabilization described by Koshelev et al. Finally, the irradiation-induced enhancements of the critical current density, observable in c axis textured tapes. show that the critical current density is not limited by the weak coupling between grains. Therefore, for the observation of directional pinning by columnar defects introduced in tapes, the argument of the different microstructure can be applied as well, since in relevant aspects the grains of a tape rather resemble single crystals than the structure of thin films.

A further reason for the discrepancy could be the very different thicknesses of films ($\sim 0.4 \mu m$) and crystals (~ 50 μ m). Shnerb [37] has proposed a critical length (sample thickness) above which the tilt of the external magnetic field is irrelevant to the hopping mechanism of flux lines. The theory predicts a surface roughness extension in which vortices are completely delocalized from the columnar defects while remaining pinned only deep in the bulk. Although our experiment deals with the opposite extreme, looking for some length below in which pinning is tilt-independent, one may raise the question as to whether in thick samples directional pinning is generated by the dominating bulk region whereas thin films mainly consist of surface regions for which no alignment effects are expected. However, a dominating surface region is not compatible with the increased (nondirectional) pinning, which is still observed after irradiation in our experiments.

4. Summary

Using heavy-ion irradiation in three of four identical striplines on the same substrate of epitaxial Bi-2212 thin films, columnar defects of varying density and orientation were produced. As a result, the films suffered a reduction of T_c proportional to the volume fraction of damaged material. With regard to measurements of the transport critical current density, activation energy and depinning field in dependence of the field orientation, no irradiation-induced directional pinning could be observed. The analysis consistently shows enhanced but two-dimensional pinning of pancake vortices at columnar defects which is in contrast to some investigations on single crystals and tapes. Possible explanations for this discrepancy were discussed.

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