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CRITICAL CURRENTS AND VORTEX PINNING IN  $^{235}\text{U}$  DOPED Ag/Bi2223  
TAPES IRRADIATED WITH THERMAL NEUTRONS

EMIL BABIĆ<sup>a</sup>, IVICA KUŠEVIĆ<sup>a</sup>, DAMIEN MARINARO<sup>b</sup> and SHI XUE DOU<sup>b</sup>

<sup>a</sup>*Department of Physics, Faculty of Science, University of Zagreb, 10000 Zagreb, Croatia*

<sup>b</sup>*Centre for Superconducting and Electronic Materials, University of Wollongong,  
Wollongong, NSW 2522, Australia*

**Dedicated to Professor Kseno Ilakovac on the occasion of his 70<sup>th</sup> birthday**

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Systematic analysis of the transport critical current densities  $J_c$  at 77 K for  $^{235}\text{U}$ -oxide ( $\text{UO}_2 \cdot 2\text{H}_2\text{O}$ ) doped Ag-sheathed  $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  tapes (Bi2223) irradiated with thermal neutrons is presented. Within the explored range of U-oxide concentrations ( $c \leq 2$  wt%) and neutron fluences ( $\phi \leq 6 \cdot 10^{19}$  n/m<sup>2</sup>), the pinning of vortices in irradiated tapes increases approximately linearly with fission track density (proportional to  $c\phi$ ), as evidenced by progressively weaker magnetic field dependence of  $J_c$  on increasing  $c\phi$ . Accordingly, the crossover field  $B^*$  at which the pinning force density  $F_p$  reaches its maximum value  $F_{p\text{max}} = J_c B^*$  and the irreversibility field  $B_{\text{irr}}(F_p \rightarrow 0)$  increase approximately linearly with  $c\phi$ . Therefore,  $J_c - B$  and  $F_p - B$  variations for all tapes with vortex pinning dominated by fission tracks show universal scaling behaviour with the scaling parameter  $B^*$  (proportional to  $c\phi$ ). Taking into account the adverse effects of  $c$  and  $\phi$  on  $J_c$  of our tapes, we estimate a maximum  $B^*$  and  $B_{\text{irr}}$  that can be reached in  $^{235}\text{U}$ -oxide doped Bi2223 tapes at 77 K.

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## 1. Introduction

The progress in the methods for fabrication of Ag-sheathed Bi–Sr–Ca–Cu–O tapes resulted in sizable increase of their critical current densities  $J_c$  at elevated temperatures [1]. Although  $J_c(77\text{ K})$  for well prepared tape is still about twenty

times lower [2, 3] than that for corresponding epitaxial film [4, 5], it seems adequate for some large scale applications [1]. However, weak flux pinning at higher temperatures ( $T \geq 77$  K), which results in large decrease of  $J_c$  already in low fields ( $B < 0.1$  T), seems to rule out their applications at  $T \geq 77$  K in any field other than the self-field generated by current flow along the tape. (Moreover, it is possible that self-field limits  $J_c$  in tapes by causing percolative current flow through small fraction of the tape volume [6].) As a result, some promising applications of high-temperature superconductors (HTS), such as transformers and nuclear magnetic resonance magnets, are out of reach of the present day Bi–Sr–Ca–Cu–O tapes.

Since weak flux pinning at elevated temperatures is intrinsic to Bi–Sr–Ca–Cu–O compounds ( $J_c$  at 77 K of corresponding single crystals are much lower than those of films and tapes), it is clear that only the introduction of pinning centres can improve the  $J_c - B$  performance of tapes. The conventional methods for improvement of the flux pinning in superconductors (impurities, second phases, crystallographic defects, etc.) yielded modest enhancement of flux pinning in Bi–Sr–Ca–Cu–O compounds [7]. Much better results were obtained by using particle irradiation techniques [8]. In particular, the irradiation with high-energy heavy ions produces columnar defects in Bi–Sr–Ca–Cu–O compounds which at elevated temperatures cause strong uniaxial enhancement of flux pinning [8, 9]. The same technique makes possible the production of splayed columnar defects [10] which provide even stronger and more isotropic pinning of vortices [8]. However, this technique can only be used on short and thin samples (and is also too expensive for practical applications).

Splayed columnar defects can also be produced by fission fragments of heavy nuclei accommodated within the superconductor. Indeed, over a decade ago, sizable increase of vortex pinning has been observed in uranium-doped ceramic  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  samples irradiated with thermal neutrons [11]. We proposed [12] that the same “U/n” method is the best way for enhancing the flux pinning in Bi–Sr–Ca–Cu–O compounds, too. Splayed columnar defects have also been introduced in Bi–Sr–Ca–Cu–O and Hg–Ba–Ca–Cu–O compounds [13] by fissioning the Bi- or Hg-nuclei with high energy protons (0.8 GeV). Although effective for the improvement of the flux pinning, these Bi/p and Hg/p methods require high fluences of high energy protons [13] and are generally too expensive for practical applications. Therefore, the U/n method, using lower fluences of more easily available thermal neutrons, seems at present the best choice for the enhancement of flux pinning in HTS [12]. Indeed, already first results [14] for lightly  $^{235}\text{U}$ -doped Bi2223 tapes irradiated with neutrons confirmed our prediction [12]. In particular, sizable increase in the irreversibility field  $B_{\text{irr}}$ , reduction of the critical-current anisotropy and large enhancement of  $J_c(77\text{ K})$  at elevated magnetic fields (all compared to those of undoped tape) have been observed [14]. Further, more systematic studies [15, 16] of critical currents in  $^{235}\text{U}$ -oxide doped Bi2223 tapes, containing different amounts of  $^{235}\text{U}$  and irradiated to different neutron fluences, confirmed great potential of U/n technique, but also showed problems associated with high U-oxide contents (strong decrease of  $J_c$ ) [15] and high neutron fluences (radioactivity due to activation of Ag-sheathing [14]). This showed the necessity of finding the optimal combination of the neutron fluence ( $\phi$ ) and U-oxide content ( $c$ ) which gives the

largest increase in flux pinning capacity with the least decrease of  $J_c$  and tolerable induced radioactivity.

Here we present main results of the systematic analysis of critical currents  $J_c$  and pinning force densities  $F_p = J_c B$  at 77 K for Bi2223 tapes doped with different amounts of  $^{235}\text{U}$ -oxide ( $c \leq 2$  wt%) and irradiated with different neutron fluences ( $\phi \leq 6 \cdot 10^{19}$  n/m<sup>2</sup>). This analysis reveals simple pattern for the enhancement of flux pinning at 77 K in the investigated system. Furthermore, it shows new features in  $J_c - B$  and  $F_p - B$  variations associated with dominant pinning of vortices by fission tracks. The analysis also enables one to estimate the optimal conditions (combination of  $c_0$  and  $\phi_0$ ) required to reach specified value of  $B_{\text{irr}}(77 \text{ K})$  with the lowest decrease of  $J_c(77 \text{ K})$ . Therefore, it also enables the estimate of the largest  $B_{\text{irr}}(77 \text{ K})$  which can be reached in the investigated system. Although we performed this analysis on the results for  $^{235}\text{U}$ -oxide doped tapes measured at 77 K, the same type of analysis should apply to all U/n treated Bi–Sr–Ca–Cu–O compounds (and probably other HTS) regardless of the type of  $^{235}\text{U}$  containing dopant [16] and should also work over an extended temperature range [10, 11, 17].

## 2. Experimental procedures

Ag-sheathed Bi(Pb)–Sr–Ca–Cu–O tapes doped with  $^{235}\text{U}$ -oxide were made using powder in tube technique [18]. The precursor powders with cation ratios Bi:Pb:Sr:Ca:Cu=1.83:0.35:1.95:2.05:3.05 were mixed with three weight percentages of  $^{235}\text{UO}_2 \cdot 2\text{H}_2\text{O}$  ( $c = 0.15, 0.6$  and  $2$  wt%, respectively) prior to processing. The corresponding atomic percentages of  $^{235}\text{U}$  in tapes were approximately 0.056, 0.23 and 0.75. Tapes were irradiated at the HIFAR reactor of Australian Nuclear Science and Technology Organisation to various fluences ( $\phi \leq 6 \cdot 10^{19}$  n/m<sup>2</sup>). This provided samples with eight different fission track (columnar defect) densities, nominally proportional to  $c\phi$ . In particular, assuming random distribution of  $^{235}\text{U}$ -nuclei within Bi2223 tape, one can estimate the density of columnar defects  $n_{\text{col}}$  from  $n_{\text{col}} = \phi\sigma_f n$ , where  $n$  is the density of U-atoms in tape and  $\sigma_f$  is the fission cross-section for  $^{235}\text{U}$ -nucleus. In addition to these U/n treated samples, several control samples (undoped–unirradiated, undoped–irradiated and doped–unirradiated) were prepared. The combinations of  $c$  and  $\phi$  for all samples are given in Table 1.

For all samples, the variation of transport critical current density  $J_c$  with magnetic field  $B$  was measured at 77 K. The  $J_c$  criterion was  $E_c = 1 \mu\text{V}/\text{cm}$ . The details concerning the measurement techniques were previously reported [3, 15]. Magnetic field was applied both perpendicular to the tape plane (thus parallel to the  $c$ -axis of Bi2223 grains,  $B_{\text{pc}}$ ) and parallel to it ( $B_{\text{oc}}$ ), but always perpendicular to  $J_c$ . From these measurements, we extracted the key parameters reflecting the vortex pinning [2]: the field  $B^*$  at which the pinning force density becomes maximum ( $F_{\text{p max}} = J_c B$ ) and the irreversibility field [19]  $B_{\text{irr}}$  ( $F_p$  and  $J_c \rightarrow 0$ ). For several samples, the transport measurements (including both  $J_c$  and magnetoresistance) were performed over an extended temperature range ( $T \geq 50 \text{ K}$ ) in order to see whether the results for 77 K adequately represent the behaviour of samples

at elevated temperatures or not [17]. These measurements will also be used for a more detailed study of vortex pinning by fission tracks [17] in Bi2223 tapes. Some data relevant to our samples are shown in Table 1.

TABLE 1. Data relevant to our Ag/Bi2223 tapes:  $c$  is the  $^{235}\text{U}$ -oxide content in wt%,  $\phi$  is the neutron fluence,  $J_c$  is the critical current density at 77 K in zero applied field,  $B^*$  and  $B_{\text{irr}}$  are the fields for maximum pinning force density ( $F_{\text{p max}} = J_c B^*$ ) and linear extrapolation of  $F_{\text{p}}$  to zero, respectively (all for field  $B$  perpendicular to the tape plane at  $T = 77$  K). For U/n treated tapes,  $B_{\text{irr}} = 2.3B^*$ .

$c(\text{wt}\%)$	$\phi(10^{19} \text{ n/m}^2)$	$J_c(\text{kA/cm}^2)$	$B^*(\text{T})$	$B_{\text{irr}}(\text{T})$
0	0	24.4	0.15	0.56
	2.25	22.9		
0.15	0	23.6		
	5.0	18.5	0.44	1.01
	6.0	17.8	0.40	0.92
0.6	0	22.4	0.08	
	1.25	19.7	0.31	0.72
	1.75	18.8	0.36	0.82
	2.25	18.1		
	3.0	16.6	0.56	1.29
	4.0	11.0	0.70	1.60
2	0.05	13	0.12	0.27
	0.2	13	0.24	0.54
	1.0	10	0.52	1.20

### 3. Results and discussion

Clearly, both intrinsic (critical temperature  $T_c$ , and fields  $H_{c1}$  and  $H_{c2}$ ) and extrinsic ( $J_c$ ,  $B^*$ ,  $B_{\text{irr}}$ , etc.) parameters of doped and/or irradiated type II superconductor depend in principle on dopant ( $c$ ) and/or irradiation ( $\phi$ ) levels. Since the change in intrinsic parameters of U/n treated Bi2223 tapes seems small [14, 15, 20], we shall ignore probably small effects of these changes on their extrinsic parameters (directly related to flux pinning). In general, the flux pinning capacity of U/n treated tape will depend on the interplay of pinning inherent to Bi2223 tapes (associated with their preparation and processing [21]) with contributions to pinning due to dopant ( $c$ ), irradiation ( $\phi$ ) and fission tracks ( $c\phi$ ). The  $J_c - B$  curves for control samples did not show any significant contribution to vortex pinning associated with either dopant level or irradiation fluence. Therefore, the pinning capacity of U/n treated tapes should mainly arise from the interplay of pinning inherent to tapes (often referred as uncorrelated [22]) and that due to fission tracks (correlated [22]). This interplay is not well understood [22], so we assume (and results seem to confirm) that these pinning contributions are additive. Since fission tracks (amorphous columns 2–4  $\mu\text{m}$  long with about 5 nm diameter [20]) form strong individual pinning centres, their contribution to the pinning capacity should be proportional

to their density, i.e., to  $c\phi$  (providing that the areal density of vortices, proportional to  $B$ , is not sizably larger than that of fission tracks). Therefore, for not too high fields  $B$  and at temperatures sufficiently below  $T_c$  (pinning of vortices by fission tracks/columnar defects is not efficient close to  $T_c$  [10, 22], whereas at low temperatures the inherent pinning dominates [19]), we expect that all parameters directly related to flux pinning capacity ( $B^*$ ,  $B_{\text{irr}}$ ) increase linearly with  $c\phi$  in U/n treated tapes.

In what follows, we mainly analyze the results for the field  $B$  perpendicular to the tape plane ( $B_{\text{pc}}$ ). The advantages of this geometry for Bi–Sr–Ca–Cu–O compounds are simpler vortex structure and a large change of  $J_c$  with  $B_{\text{pc}}$  (almost consistent with 2D anisotropy [22]). Accordingly, the improvement of flux pinning in these materials depends on the results for  $B_{\text{pc}}$ . The  $J_c - B$  curves at 77 K for all U/n treated tapes showed clear self-similarity [15, 17]. In particular, they showed asymptotic power-law variations [2, 3] (proportional to  $B^\alpha$ ) both at low and high  $B$  with the crossover field increasing with increasing  $c\phi$ . As shown earlier for undoped Bi–Sr–Ca–Cu–O tapes [2], such self-similar  $J_c - B$  curves can be scaled to a single one by using field  $B^*$  (at which pinning force density reaches its maximum value  $F_{\text{p max}} = J_c B^*$ ) as a scaling parameter. A rather good scaling of  $J_c - B$  curves (i.e.  $J_c(B)/J_c(B^*)$  vs.  $B/B^*$  plot) for our tapes at 77 K and for perpendicular field ( $B_{\text{pc}}$ ) is shown in Fig. 1. The scaling of  $J_c - B$  curves for parallel field ( $B_{\text{oc}}$ ) was similarly good. Scaling of  $J_c - B$  curves implies the scaling of  $F_p - B$  ones (Fig. 2), too. However,  $J_c$  and  $F_p$  scaling is almost perfect only for tapes with  $c\phi \geq 0.4$  ( $10^{19} \text{ m}^{-2} \text{ wt}\% \text{ } ^{235}\text{U}$ -oxide). Bi2223 tapes with lower  $c\phi$  (including the undoped ones) show clear deviation from this scaling (slower decrease of  $J_c$  and  $F_p$  with  $B$  for  $B > B^*$ ,

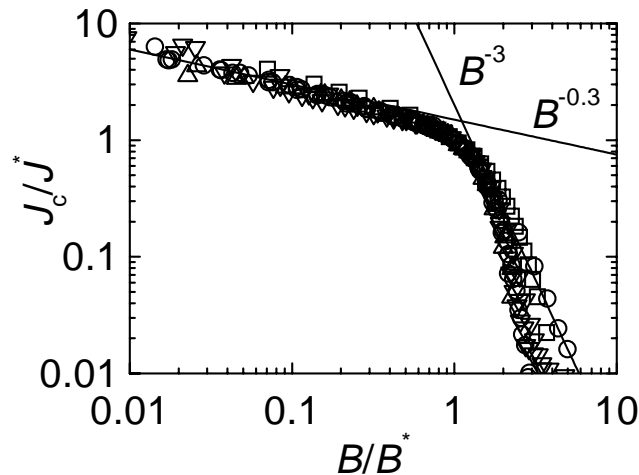


Fig. 1. Variation of normalized critical current density  $J_c(B)/J_c(B^*)$  with normalized field  $B/B^*$  ( $B^*$  is the field at which pinning force density becomes maximum,  $F_{\text{p max}} = J_c B^*$ ) for our Ag/Bi2223 tapes at 77 K with field perpendicular to the tape plane. Undoped and unirradiated tape is denoted by  $\square$ .

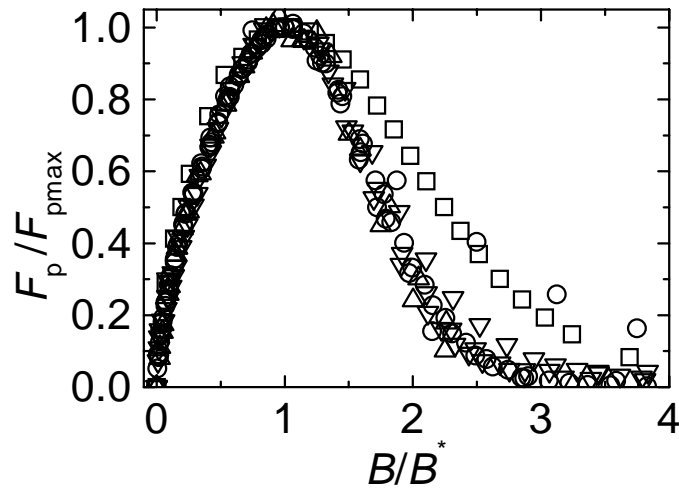


Fig. 2. Variation of normalized pinning force density  $F_p/F_{p\max}$  with normalized field  $B/B^*$  for Ag/Bi2223 tapes from Fig. 1 at 77 K. Note different variation of undoped and unirradiated sample ( $\square$ ) for  $B/B^* > 1$ .

see Figs. 1 and 2). Therefore, the increase in track density ( $c\phi$ ) simultaneously enhances the pinning capacity and changes the mechanism of flux pinning. The pinning mechanism in type II superconductor can in principle be deduced from the scaling analysis of  $F_p - B$  curves [23]. For our tapes with  $c\phi \geq 0.4 \cdot (10^{19} \text{ m}^{-2} \text{ wt}\% \text{ } ^{235}\text{U}\text{-oxide})$  the position of  $F_{p\max}$ ,  $B^*$ , in respect to that of  $F_p(B > B^*) \rightarrow 0$ , seems to indicate  $\Delta T_c$ -pinning [22]. This conjecture seems consistent with the expected normal-metal behaviour ( $T_c \approx 0$ ) of columnar defects. However, the straightforward application of scaling analysis for conventional superconductors (low  $T_c$ ) to HTS ones is questionable [2], because it ignores the thermal effects which are large in HTS [24].

Faster decrease of  $J_c$  and  $F_p$  with  $B$  for  $B > B^*$  in tapes with higher density of fission tracks ( $c\phi$ ), compared to that for undoped tape, seems to indicate enhanced flux creep rate in the former case. This is at variance with predicted reduction of flux creep rate by splayed defects [10, 22]. However, the theory considered small splay angles and rather uniform distribution of columnar defects [10]. Therefore, the randomness in directions and positions of fission tracks in our tapes can promote flux creep, which may explain this discrepancy. Finally, we note the difference between the  $J_c$  and  $F_p$  scaling with  $B^*(T)$ , which is usually reported for HTS samples containing no fission tracks or other type of correlated disorder [2], and that of U/n treated tapes reported here (Figs. 1 and 2). In the former case [2], scaling shows that the same pinning mechanism (not well understood for Bi-Sr-Ca-Cu-O tapes [7, 21]) dominates over an extended temperature range, whereas here scaling shows that for a broad range of  $c\phi$ , the fission track pinning dominates at 77 K. In particular, we do not expect that the  $J_c - B$  and  $F_p - B$  curves for

our samples with different  $c\phi$  will scale with  $B^*(T)$  over a very broad temperature range.

Providing that our assumptions regarding pinning capacity of U/n treated tapes (see beginning of this chapter) are fulfilled, we expect  $B^*(77\text{ K})$  for our tapes to increase linearly with  $c\phi$ . Figure 3 shows that within the explored range of  $c$  and  $\phi$ ,  $B^*$  (perpendicular field) indeed increases approximately linearly with  $c\phi$ . Moreover, for  $c\phi = 0$ ,  $B^*$  extrapolates to that of undoped and unirradiated tape,  $B_0^*$ . Therefore,  $B^*(77\text{ K})$  for our tapes roughly obeys the expression

$$B^* = B_0^* + Ac\phi, \quad (1)$$

where  $A \simeq 0.23\text{ T}/(10^{19}\text{ m}^{-2}\text{wt}\% \text{ U-oxide})$  is the average slope of variation of  $B^*$  with  $c\phi$  for perpendicular field ( $B_{\text{pc}}$ ) at 77 K. Clearly, this slope will depend on temperature ( $T_0$ ) and field direction ( $B_{\text{pc}}$  or  $B_{\text{oc}}$ , in general on the angle between  $B$  and the average direction of the  $c$ -axis of Bi2223 grains within the tape). Indeed, the data for  $B^*(77\text{ K})$  for parallel field ( $B_{\text{oc}}$ ) also roughly obeyed Eq. (1), but with almost four times larger  $A \approx 0.85\text{ T}/(10^{19}\text{ m}^{-2}\text{wt}\% \text{ U-oxide})$ . However, the relative rates of increase  $A/B_0^*$  of  $B^*$  with  $c\phi$  for  $B_{\text{pc}}$  and  $B_{\text{oc}}$  were quite similar, about 1.5 and 1.2/( $10^{19}\text{ m}^{-2}\text{wt}\% \text{ U-oxide}$ ), respectively. Similar values of  $A/B_0^*$  for  $B_{\text{pc}}$  and  $B_{\text{oc}}$  seem consistent with random orientations of fission tracks, whereas somewhat larger  $A/B_0^*$  for  $B_{\text{pc}}$  than that for  $B_{\text{oc}}$  shows larger enhancement of flux pinning due to fission tracks for this field direction. This enhances the reduction  $J_c(B_{\text{oc}})/J_c(B_{\text{pc}})$  in the critical current anisotropy of U/n treated tapes, which is

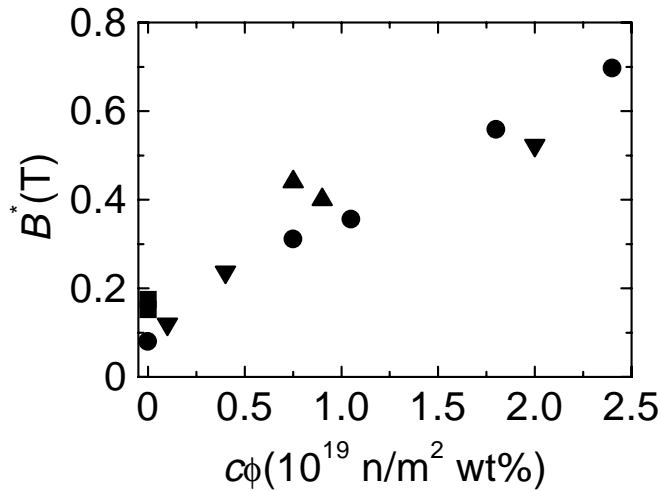


Fig. 3. Variation of field  $B^*$  (defined by  $F_{\text{pmax}} = J_c B$ ) with product of  $^{235}\text{U}$ -oxide content  $c$  (in wt%) and neutron irradiation fluence  $\phi$  ( $c\phi$  is proportional to fission track density within the tape) for Ag/Bi2223 tapes from Fig. 1 at 77 K. Symbols  $\square$ ,  $\triangle$ ,  $\circ$  and  $\nabla$  denote  $^{235}\text{U}$ -oxide contents of 0, 0.15, 0.6 and 2 wt%, respectively.



favourable for their applications [20, 25]. We wish to emphasize rapid increase of  $B^*$  (77 K) with  $c\phi$  for both field directions ( $B_{pc}$  and  $B_{oc}$ ). In particular, for  $c\phi = 2.4 \cdot (10^{19} \text{ n/m}^2 \text{ wt}\% \text{ } ^{235}\text{U-oxide})$  ( $\phi = 4 \cdot 10^{19} \text{ n/m}^2$ )  $B^* \simeq 5B_0^*$  in perpendicular field (Fig. 3). The corresponding increase of  $B^*$ ,  $\Delta B^* = B^* - B_0^*$ , is practically the same as that obtained at 75 K by fissioning Bi-nuclei in a similar tape [26]. However, in the later case [26], twenty times higher fluence ( $8 \cdot 10^{20} \text{ p/m}^2$ ) of high energy ( $E = 0.8 \text{ GeV}$ ) protons was used. This example clearly shows the advantage and large potential of the U/n method.

Although a sizable part of the data scattering in Fig. 3 arises from the uncertainty in the determination of  $B^*$  from rather broad maxima of  $F_p$  vs.  $B$  curves (Fig. 2), few data for the samples with the lowest (0.15 wt%) and highest (2 wt%)  $^{235}\text{U-oxide}$  contents seem to show that in addition to strong increase with  $c\phi$  (Eq. (1)),  $B^*$  also exhibits sizably weaker increase with  $\phi$ . In order to verify this hypothesis, we plot the variation of  $B^*$  with  $\phi$  for all irradiated samples in Fig. 4. As expected, the slopes of the  $B^*$  vs.  $\phi$  variations for U/n treated samples increase linearly with U-oxide content (inset to Fig. 4), but their extrapolation to  $c = 0$  yields a non-zero intercept. This confirms the presence of linear in  $\phi$  contribution to  $B^*$  of U/n treated samples. Therefore, a more accurate expression for  $B^*$  (77 K) for our U/n treated samples is

$$B^* = B_0^* + A'c\phi + D\phi, \quad (2)$$

where  $D$  is the intercept of  $\Delta B^*/\Delta\phi$  vs.  $c$  variation (inset to Fig. 4) for  $c = 0$ . For the perpendicular field,  $D \simeq 0.02\text{T}/(10^{19} \text{ m}^{-2})$  and  $A' \simeq 0.18\text{T}/(10^{19} \text{ m}^{-2}\text{wt}\%$

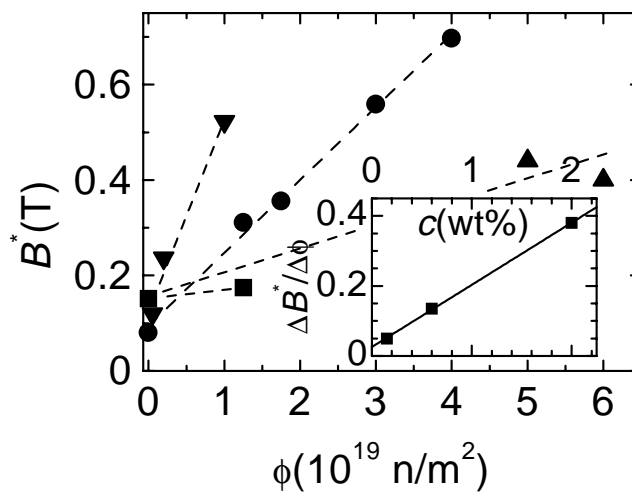


Fig. 4. Variation of the field  $B^*$  with neutron fluence  $\phi$  for our Ag/Bi2223 tapes at 77 K. Symbols for different  $^{235}\text{U-oxide}$  content are the same as in Fig. 3. The inset: variation of the slopes  $\Delta B^*/\Delta\phi$  (in units  $\text{T}/(10^{19} \text{ n/m}^2)$ , with the  $^{235}\text{U-oxide}$  content in wt%.

$^{235}\text{U}$ -oxide). Clearly, this  $D\phi$  term is quite small (inset to Fig. 4) and contributes sizably to  $B^*$  only for samples with low  $c$  (0.15 wt%) irradiated to high fluences  $\phi$ . Indeed, this  $D\phi$  term explains seemingly paradoxical result [25] that from four Bi2223 tapes with different  $c$ , irradiated to nominally the same fission track density ( $c\phi$ ), one with the lowest  $c$  showed the highest enhancement of  $J_c$  at elevated field. This  $D\phi$  term is associated with fission of  $^{235}\text{U}$ -nuclei and not with direct influence of neutron fluence on Bi2223 matrix (see data for undoped sample in Fig. 4). Several effects can yield this term [11], the simplest being the nonuniform distribution of U-atoms within the sample. Such distribution has been observed [16, 20] in samples with higher  $^{235}\text{U}$ -oxide contents ( $c > 1$  wt%). Therefore, better ways [16] of  $^{235}\text{U}$ -doping of Bi2223 tapes may erase this term and lead to a higher enhancement of  $B^*$  at lower neutron fluences.

In Bi–Sr–Ca–Cu–O tapes,  $B^*$  is proportional to the irreversibility field [2, 3]  $B_{\text{irr}}$ . The irreversibility line  $B_{\text{irr}}(T)$  is particularly important parameter in HTS, since it forms the borderline between the vortex-solid (nondissipative) and vortex-liquid (dissipative) regions in  $B - T$  diagrams of HTS [22]. Therefore, the enhancement of  $B_{\text{irr}}$  at given temperature expands the nondissipative (useful) region to higher fields [19]. We can use either our  $J_c - B$  or  $F_p - B$  data in order to estimate  $B_{\text{irr}}(77\text{ K})$  for our U/n treated tapes. Usually [25],  $B_{\text{irr}}$  is defined as the field at which  $J_c$  reduces to some specified low value. For Ag-sheathed tapes, this method is questionable, unless the correction for the conduction via Ag-sheathing is performed [27]. Therefore, we estimate  $B_{\text{irr}}$  from  $F_p - B$  curves (Fig. 2) using linear extrapolation of  $F_p$  vs.  $B$  data for  $B > B^*$  to zero [2, 28]. This extrapolation of data for  $0.2 \leq F_p/F_{p\text{max}} \leq 0.8$  (Fig. 2) of U/n treated tapes with  $c\phi \geq 0.4 \cdot (10^{19} \text{ n/m}^2 \text{ wt}\% \text{ } ^{235}\text{U}\text{-oxide})$  yields  $B_{\text{irr}}(77\text{ K}) \approx 2.3B^*(77\text{ K})$ . For parallel field ( $B_{\text{oc}}$ ), corresponding extrapolation for tapes within the same range of  $c\phi$  yields  $B_{\text{irr}}(77\text{ K}) \approx 3B^*(77\text{ K})$ . Therefore, in order to show variation of  $B_{\text{irr}}(77\text{ K, perpendicular field})$  with  $c\phi$  for U/n treated tapes, one should simply multiply the scale on the ordinate axis of Fig. 3 by a factor 2.3. For undoped tape, the ratio  $B_{\text{irr}}(77\text{ K})/B^*(77\text{ K})$  was larger (about 3.6 times for both field directions) which is consistent with the observed slower decrease of  $J_c(77\text{ K})$  with  $B$  for  $B > B^*$  (Fig. 1) and with previous results for undoped and unirradiated Bi–Sr–Ca–Cu–O tapes [2, 3]. In spite of this, the enhancement of  $B_{\text{irr}}$  (thus expansion of the non-dissipative region) in our U/n treated tapes is very large. In particular, for the tape with maximum  $c\phi = 2.4 \cdot (10^{19} \text{ m}^{-2} \text{ wt}\% \text{ } ^{235}\text{U}\text{-oxide})$ , we find  $B_{\text{irr}}(77\text{ K, perpendicular field}) \approx 1.6\text{ T}$ , which is three times larger than that for non-irradiated sample (0.55 T).

In order to verify whether the same linear relationship between  $B_{\text{irr}}$  and  $B^*$  holds over an extended temperature range (hence, whether it is intrinsic to fission track pinning or not), we measured [17] independently  $B_{\text{irr}}(T)$  from the resistive transitions in applied field [19] ( $R \rightarrow 0$ ) and  $B^*(T)$  from  $I - V$  curves ( $F_p(B^*) = F_{p\text{max}}$ ) for several U/n treated samples over and extended temperature range ( $T \geq 70\text{ K}$ ). As illustrated in Fig. 5 for the tape with  $c\phi = 0.75 \cdot (10^{19} \text{ m}^{-2} \text{ wt}\% \text{ } ^{235}\text{U}\text{-oxide})$  in perpendicular field,  $B_{\text{irr}}(T) = 2.3B^*(T)$  within the explored temperature range. Therefore, the description of vortex pinning by fission tracks presented above for  $T = 77\text{ K}$  seems to hold over an extended temperature range for U/n treated

Bi2223 tapes.

Since  $B^*$  and  $B_{\text{irr}}$  of our samples were limited by the explored  $c\phi$  range (Fig. 3), it is interesting to estimate what are the maximum values of these quantities that can be reached in  $^{235}\text{U}$ -oxide doped U/n treated Bi2223 tapes at 77 K. For this purpose, one has to take into account the decrease of  $J_c(77\text{ K})$  due to U-oxide doping level  $c$  [15] and neutron irradiation fluence  $\phi$ , i.e. to find maximum allowed  $c\phi$  giving nonvanishing  $J_c(77\text{ K})$ . Clearly, at fixed field  $B_0$  and temperature  $T_0$ , the critical current density  $J_c(T_0, B_0)$  of our tapes is a function of  $c$  and  $\phi$  only ( $J_c(c, \phi)$ ). Therefore, small change of  $J_c$  (total differential at  $T_0$  and  $B_0$ ) can be written as  $dJ_c = fdc + g d\phi$ , where  $f$  and  $g$  are the functions of  $c$  and  $\phi$ . Furthermore,

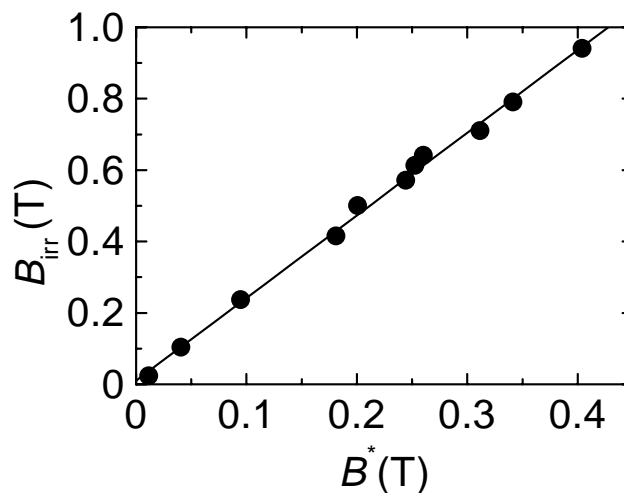


Fig. 5. Variation of the irreversibility field  $B_{\text{irr}}(\text{T})$  (deduced from magnetoresistance measurements,  $R(B_{\text{irr}}, \text{T}) \rightarrow 0$ ) with  $B^*(\text{T})$  (deduced from  $I - V$  curves) for U/n treated Ag/Bi2223 sample with  $c\phi = 0.75 \cdot (10^{19} \text{ n/m}^2 \text{ wt}\% \text{ } ^{235}\text{U}\text{-oxide})$ . Note the linear variation  $B_{\text{irr}}(\text{T}) \simeq 2.3B^*(\text{T})$ . Field  $B$  was perpendicular to the tape plane.

for a rather narrow (but finite) range of  $c$  and  $\phi$  values, we expect  $f$  and  $g$  to be constants or linear functions. Figure 6 shows the  $J_c(77\text{ K}, B = 0)$  data for all our samples (Table 1) as function of  $\phi$ . It can be seen that for  $\phi = 0$ ,  $J_c$  decreases approximately linearly with  $c$  (therefore  $f = \text{const.}$ ), and that for all  $^{235}\text{U}$ -oxide contents (including  $c = 0$ ),  $J_c$  decreases with  $\phi$ . Our (limited) data suggest linear decrease of  $J_c$  with  $\phi$ , with the rate increasing with  $c$ . As shown in the inset to Fig. 6, the relative rates of decrease of  $J_c(c, \phi)$  with  $\phi$ ,  $J_c(c, 0)^{-1} \Delta J_c / \Delta \phi$  (where  $\Delta J_c = J_c(c, 0) - J_c(c, \phi)$ ), increase approximately linearly with  $c$  (therefore,  $g$  is a linear function of  $c$ ). We note the non-zero intercept for  $c = 0$  in the inset to Fig. 6. This shows that neutron irradiation is detrimental to  $J_c$  of Ag/Bi2223 tapes (regardless whether U-doped or not) and that this effect is linear in  $\phi$  and not related to fission events (the same for doped and undoped tapes). According to

the above, the approximate expression for the relative change of  $J_c(77\text{ K}, B = 0)$ ,  $\Delta J_c/J_{c0}$  (where  $\Delta J_c = J_c(0, 0) - J_c(c, \phi)$  and  $J_{c0} = J_c(0, 0)$ ), of our tapes is

$$\Delta J_c/J_{c0} = Ec + F\phi + Gc\phi \quad (3)$$

where  $E$ ,  $F$  and  $G$  are constants for the explored range of  $c$  and  $\phi$ . In particular, Eq. (3) with  $E \simeq 0.19/(\text{wt}\% \text{ } ^{235}\text{U-oxide})$ ,  $F \simeq 0.027/(10^{19} \text{ n/m}^2)$  and  $G \simeq 0.12/(10^{19} \text{ n/m}^2 \text{ wt}\%)$ , describes quite well the data in Fig. 6.

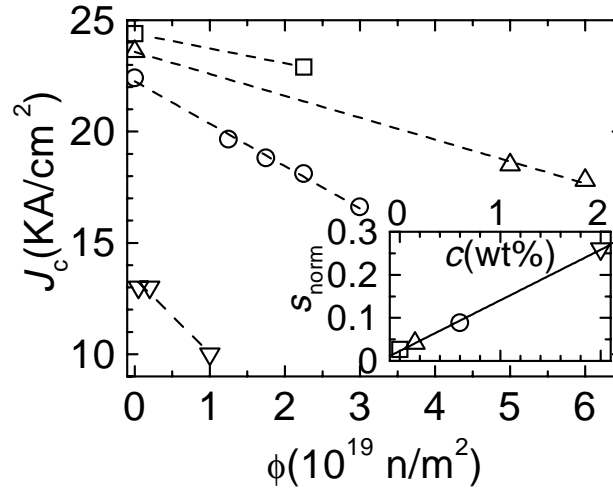


Fig. 6. Variation of critical current density  $J_c$  with neutron fluence  $\phi$  of our Ag/Bi2223 tapes at 77 K in zero applied field. The inset: variation of the normalized slopes  $s_{\text{norm}} = J_c^{-1}(\Delta J_c/\Delta\phi)$  (in units  $(10^{19} \text{ n/m}^2)^{-1}$ , where  $J_c = J_c(c, \phi = 0)$  and  $\Delta J_c = J_c(c, 0) - J_c(c, \phi)$ ) with  $^{235}\text{U}$ -oxide content  $c$  (in wt%) for Ag/Bi2223 tapes from the same figure. Symbols for different  $^{235}\text{U}$ -oxide contents are the same as in Fig. 3.

Since only the third term on the right side of Eq. (3) is related to fission events, the reduction in coefficient  $E$  (through a less harmful substitution of  $^{235}\text{U}$  in Bi2223 tape [16]) is vital, because, providing that less harmful substitution also leads to more uniform distribution [16] of  $^{235}\text{U}$ , it will reduce both  $Ec$  and  $F\phi$  terms (the same  $c\phi$  will be achieved with lower  $\phi$ ) and therefore lead to sizably reduced decrease of  $J_c(77\text{ K}, B = 0)$  in U/n treated samples. This will also reduce the radioactivity of U/n treated tapes, which is caused by activation of Ag-sheathing [25].

One can use Eq. (3) in order to deduce the optimal combination of  $c_0$  and  $\phi_0$ , which causes the smallest decrease of  $J_c(77\text{ K}, B = 0)$  for a specified value  $c\phi = K$ . We find  $c_0 = (FK/E)^{1/2}$  and  $c_0/\phi_0 = F/E$ . In particular, for  $K = 2.4 \cdot (10^{19} \text{ n/m}^2 \text{ wt}\% \text{ } ^{235}\text{U-oxide})$ , we find  $c_0 = 0.58 \text{ wt}\% \text{ U-oxide}$  and  $\phi_0 = 4.1 \cdot 10^{19} \text{ n/m}^2$ , which are very close to the actual  $c$  and  $\phi$  values (Table 1) for our sample with the highest  $B^*(77\text{ K})$  (Fig. 3). Assuming that Eq. (3) holds (with the same coefficients  $E$ ,  $F$  and

G) also for  $c\phi$  values outside the explored  $c\phi$  range, and letting  $\Delta J_c/J_{c0} \rightarrow 1$ , we can estimate  $K_{\max} = c_{0\max}\phi_{0\max}$  which limits the maximum enhancement of  $B^*(77\text{ K})$  and  $B_{\text{irr}}(77\text{ K})$  in the investigated system. We find  $K_{0\max} \leq 6.2 \cdot (10^{19}\text{ n/m}^2\text{ wt}\% \text{ }^{235}\text{U-oxide})$  ( $c_0 \simeq 0.89\text{ wt}\% \text{ }^{235}\text{U-oxide}$ ), which for perpendicular field yields  $B^*(77\text{ K}) \leq 1.6\text{ T}$  and  $B_{\text{irr}}(77\text{ K}) \leq 3.7\text{ T}$ . For the parallel field ( $B_{\text{oc}}$ ), we find about four times larger values of  $B^*(77\text{ K})$  and  $B_{\text{irr}}(77\text{ K})$ . These field values are comparable to those observed for novel forms of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  film samples [29] exhibiting strong pinning of vortices at 77 K. Since in the investigated system the maximum enhancement of pinning capacity is controlled by the detrimental influence of  $^{235}\text{U}$ -oxide ( $c$ ) on  $J_c$  (Eq. (3)), it is clear that the use of other  $^{235}\text{U}$  containing dopants [16] (which affect less  $J_c$ ) can yield U/n treated Bi2223 tapes with vortex pinning at elevated temperatures exceeding that in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  samples.

#### 4. Conclusions

We have performed a systematic analysis of the variation of critical current densities ( $J_c$ ) at 77 K with magnetic field ( $B$ ) for thirteen  $^{235}\text{U}$ -oxide doped ( $c$ ) Bi2223/Ag tapes irradiated with thermal neutrons ( $\phi$ ). This analysis revealed simple pattern which governs the enhancement of vortex pinning capacity and the associated change in the dominant pinning mechanism with the increasing fission track densities (proportional to  $c\phi$ ) in U/n treated Bi2223 samples.

In particular, the pinning capacity ( $B^*, B_{\text{irr}}$ ) of U/n treated tapes increases rapidly (approximately linearly) with  $c\phi$ , which results in large enhancement (up to threefold) of the irreversibility field  $B_{\text{irr}}(77\text{ K})$  already for low fission track densities ( $n_{\text{f.t.}} \leq 9 \cdot 10^{19}\text{ m}^{-3}$ ). Accordingly, the nondissipative vortex solid region in the  $B - T$  phase diagram has expanded at 77 K by the factor of three compared to that of untreated tape. This result proves the predicted efficiency of U/n technique as applied to Bi2223 tapes [12]. The dominant pinning mechanism also changes, as evidenced by the universal scaling of field variations of critical currents and pinning force densities ( $F_p$ ), with a single scaling parameter  $B^*$  ( $F_p(B^*) = F_{p\max}$ ) proportional to  $c\phi$ . These scaling relationships provide unique opportunity for detailed investigation of the nature and properties of vortex pinning with randomly splayed columnar defects [10, 22]. Particularly promising seems the investigation of the U/n treated bulk (tapes, current leads) and single-crystal forms of Bi2212 compound, because of its very weak inherent flux pinning (no interplay of different pinning mechanisms), very good intergranular connectivity in tapes [2, 30] and the availability of high-quality single crystals. We also propose parallel investigation of the samples with the controlled splay of columnar defects (obtained with heavy-ion irradiation) and U/n treated ones, both with the same defect densities.

Both U-doping and neutron irradiation decrease  $J_c$  of Bi2223 tapes, which ultimately limits the maximum achievable enhancement of vortex pinning at 77 K. The analysis of  $J_c$  at 77 K and  $B = 0$  for all (control and U/n treated) samples enables the separation of the effects of U-doping ( $c$ ), neutron irradiation ( $\phi$ ) and fission events ( $c\phi$ ) on the reduction of  $J_c(77\text{ K}, B = 0)$ . The resulting relationship

(Eq. (3)) enables the evaluation of optimal combination of  $c_0$  and  $\phi_0$  required to reach specified value of  $c\phi$  (hence  $B^*$  and  $B_{\text{irr}}$ ) with the smallest decrease of  $J_c$ . This result in turn enables the estimation of the maximum pinning enhancement ( $B^*$ ,  $B_{\text{irr}}$ ) that can be reached in the investigated system at 77 K. The estimated (perpendicular field)  $B^*(77\text{ K}) \leq 1.6\text{ T}$  and  $B_{\text{irr}}(77\text{ K}) \leq 3.7\text{ T}$  are close to those for novel  $\text{YBa}_2\text{Cu}_3\text{O}_7$  thick films [29] and also show that a better way of incorporating [16]  $^{235}\text{U}$  in Bi2223 samples will lead to tapes with flux pinning at 77 K exceeding that in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  thick films.

Although the analyses presented in this paper were performed on  $^{235}\text{U}$ -oxide doped Bi2223 tapes only, they were based on few simple assumptions and yielded all salient features of fission effects in bulk type II superconductors. Therefore, we believe that their applicability will neither depend on the type of fissioning nuclei nor on the type of superconducting compound.

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#### KRITIČNE STRUJE I ZAPINJANJE MAGNETSKIH VRTLOGA U $\text{Ag/Bi2223}$ VRPCAMA DOPIRANIM S $^{235}\text{U}$ I OZRAČENIM TERMIČKIM NEUTRONIMA

Predstavljamo sustavnu analizu transportnih gustoća kritične struje  $J_c$  na 77 K sa  $^{235}\text{U}$ -oksidom ( $\text{UO}_2 \cdot 2\text{H}_2\text{O}$ ) dopiranih, srebrom obloženih  $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  vrpce ozračenih termičkim neutronima. U istraživanom području koncentracija U-oksida ( $c \leq 2$  tež.%) i toka neutrona ( $\phi \leq 6 \cdot 10^{19}$  n/m<sup>2</sup>), zapinjanje magnetskih vrtloga u vrpcama raste približno linearno s gustoćom fizijskih tragova (razmjerno sa  $c\phi$ ), što se ogleda u sve slabijoj ovisnosti  $J_c$  o magnetskom polju pri porastu  $c\phi$ . U skladu s time, prijelazno polje  $B^*$  na kojem gustoća sile zapinjanja  $F_p$  postigne najveću vrijednost  $F_{p\max} = J_c B^*$  i ireverzibilno polje  $B_{\text{irr}}(F_p \rightarrow 0)$  rastu približno linearno s  $c\phi$ . Zbog toga,  $J_c - B$  i  $F_p - B$  ovisnosti za sve vrpce u kojima je zapinjanje vrtloga pretežito zbog fizijskih tragova pokazuju univerzalno skaliranje s parametrom skaliranja  $B^*$  (razmjerno  $c\phi$ ). Uzimajući u obzir negativni utjecaj  $c$  i  $\phi$  na  $J_c$  naših vrpce, procijenili smo najveća polja  $B^*$  i  $B_{\text{irr}}$  koja se mogu postići u srebrom obloženim  $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  vrpcama dopiranim s  $^{235}\text{U}$ -oksidom na 77 K.