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Influence of random point defects introduced by proton irradiation on the flux creep rates and magnetic field dependence of the critical current density J_c of co-evaporated $GdBa_2Cu_3O_{7-\delta}$ coated conductors

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Abstract

We report the influence of random point defects introduced by 3 MeV proton irradiation (doses of 0.5×10^{16}, 1×10^{16}, 2×10^{16} and 6×10^{16} cm^{-2}) on the vortex dynamics of co-evaporated 1.3 μm thick, $GdBa_2Cu_3O_{7-\delta}$ coated conductors. Our results indicate that the inclusion of additional random point defects reduces the low field and enhances the in-field critical current densities J_c. The main in-field J_c enhancement takes place below 40 K, which is in agreement with the expectations for pinning by random point defects. In addition, our data show a slight though clear increase in flux creep rates as a function of irradiation fluence. Maley analysis indicates that this increment can be associated with a reduction in the exponent μ characterizing the glassy behavior.

Keywords: coated conductors, vortex dynamics, proton irradiation

(Some figures may appear in colour only in the online journal)

1. Introduction

During the last decade, an enormous effort has been made to enhance the superconducting properties of $RBa_2Cu_3O_{7-\delta}$ (RBCO; R: Sm, Y, Gd) coated superconductors for technical applications [1, 2]. Several strategies based on the optimization of the pinning landscape for specific requirements (temperature and magnetic field) have been used for the development of coated conductors. Usually, the vortex dynamics as well the suppression of J_c in magnetic fields, are strongly affected by the type and density of pinning centers [3]. The effectiveness of the pinning centers at different temperatures depends on their geometry and density [4]. Random point defects improve the critical currents density (J_c) mainly at temperatures below 40 K [5]. Large defects are effective pinning centers across the whole the temperature range [6]. Large pinning centers include normal secondary phases such as nanoparticles [7, 8], columnar defects [9, 10] or both [11]. Different pinning landscapes (including geometry and density) can be designed by modifying the synthesis process [12, 13]. In contrast, a noticeable improvement in $J_c(H)$ of coated conductors grown by metal–organic deposition (MOD) by proton irradiation has been recently reported [5]. Both proton and heavy ion irradiation enable effective artificial pinning centers to be generated in a controlled way. Proton irradiation is known for its ability to create between one and a few tens of atomic displacements, producing mainly random point defects and some small nanoclusters [14]. Heavy ion irradiation produces amorphous tracks [15]. Similar vortex dynamics is obtained from YBCO films with columnar defects generated by heavy ion irradiation and self-assembling of secondary phases [3].

The magnetic field–temperature (H–T) vortex phase diagram in high temperature superconductors (HTS) is strongly determined by the nature (i.e., the size and shape) and the density of the pinning sites [16]. At low temperatures the vortices are essentially frozen into their distorted configuration. As the temperature is raised, thermal fluctuation of the vortex-line positions become important and the pinning strength is reduced. In *R*BCO, the giant flux creep rate observed using magnetic relaxation of the persistent currents has been described according to the collective vortex creep model based on the elastic properties of the lattice. This model considers that every single-vortex-line is pinned by the collective action of many weak point-like pinning centers [16]. The pinning energy results from a competition between the pinning potential and the elastic deformation of the vortices. At low magnetic fields, in the so-called single-vortex regime (SVR), the vortex–vortex interaction is negligible compared to the vortex–defect interaction. At higher fields, vortex–vortex interactions become dominant, and the vortices are collectively trapped as bundles. According to vortex-glass theory and collective creep theory [16] the effective activation energy as a function of current density (J) is given by

$$U_{\mathrm{eff}} = \frac{U_0(T)}{\mu}\left[\left(\frac{J_{\mathrm{c}}}{J}\right)^{\mu} - 1\right] \tag{1}$$

where $U_0(T) = U_0 G(T)$ is the scale of the pinning energy, U_0 is the collective pinning barrier at $T = 0$ in the absence of a driving force, $G(T)$ contains the temperature (T) dependence of the superconducting parameters, and μ is the regime-dependent glassy exponent determined by the bundle size and vortex lattice elasticity. If $\mu > 0$, it corresponds to a glassy phase whose value depends on the bundle size. The model of the nucleation of vortex loops predicts for the three-dimensional case and for random point defects μ equal to 1/7, 3/2 or 5/2, and 7/9 for single-vortex creep, small-bundle creep, or large-bundle creep, respectively [16]. However, it has been reported that as H is increased, a gradual change of μ is observed (no discrete values) [17]. From equation (1), the temperature dependence of the creep rate (S) results in

$$S = -\frac{\mathrm{d}(\ln J)}{\mathrm{d}(\ln t)} = \frac{T}{U_0 + \mu T\ \ln\left(t/t_0\right)} = \frac{T}{U_0}\left(\frac{J}{J_{\mathrm{c}}}\right)^{\mu} \tag{2}$$

where U_0 is the collective barrier in the absence of a driving force and t_0 is an effective hopping attempt time. In *R*BCO superconductors (usually at intermediates temperatures) $U_0 \ll \mu T\ \ln t/t_0$, and the S (T) dependences presents a plateau with $S \approx \frac{1}{\mu\ \ln {}^{t}/_{t_0}}$. Magnetic relaxation measurements in *R*BCO films show that, for all types of pinning centers, the collective vortex creep based on the elastic motion of the vortex lattice has a crossover to fast creep, with a dramatic drop in the J_{c} values [18]. This crossover is determined by intrinsic vortex fluctuations and by the geometry and density of the pinning site [3, 19].

The effects of random point defects introduced by proton irradiation (p-irradiation) on the absolute values of J_{c} and the vortex dynamics of 1.3 μm thick GBCO thin films grown by co-evaporation are reported in this work. This technique allows one to make ~1 km length conductors in only 2–5 h [20, 21], with critical current density above 3 MA cm^{-2} at 77 K. The pinning landscape in the as-grown films presents a dispersion of $\mathrm{Gd_2O_3}$ nanoparticles [22]. The angular (θ) dependence of J_{c} indicates that correlated pinning by twin boundaries (TBs) also contributes to pinning [22]. Our results show that the superconducting critical temperature (T_{c}) is gradually reduced from 93.4 K (pristine sample) to 89.5 K for p-irradiated samples with doses equal to 6×10^{16} cm^{-2}. The J_{c} values are affected by the irradiation and the best effects are observed at temperatures lower than 40 K. The increment in the proton doses produce a gradual suppression of the J_{c} values at small field and smoother J_{c} (H) dependences, which is manifested as an increment of the J_{c} values at high magnetic fields. In addition, our data show a slight though clear increase in flux creep rates as a function of irradiation fluence, which can be associated with a decrease of the glassy exponent μ.

2. Experiment

The GBCO tape was grown by the co-evaporation technique previously described in [20, 22]. The magnetization (**M**) measurements were performed by using a superconducting quantum interference device (SQUID) magnetometer with the applied magnetic field (**H**) parallel to the c-axis (**H**||c). The T_{c} values used in this work (based on magnetization data) were determined from M (T) at $\mu_0 H = 0.5$ mT applied after zero field cooling. The J_{c} values were calculated from the magnetization data using the appropriate geometrical factor in the Bean model, $J_{\mathrm{c}} = \frac{20\Delta M}{w\left(l - {}^{w}/_{3}l\right)}$, where ΔM is the difference in magnetization between the top and bottom branches of the hysteresis loop, and l and w are the length and the width of the film ($l > w$), respectively. The creep rate measurements were recorded for more than 1 h. The initial time was adjusted considering the best correlation factor in the log–log fitting of the $\boldsymbol{J}_{\mathbf{c}}(t)$ dependence. The initial critical state for each creep measurement was generated using $\Delta H \sim 4H^{*}$, where H^{*} is the field for full-flux penetration [23]. It is important to mention that a good correlation between J_{c} values obtained by magnetization and by electrical transport in coated conductors has been reported [5].

Irradiation with 3 MeV protons (p-irradiation) produces mostly Frenkel pairs, i.e. random point defects [24]. Table 1 shows the cumulative amount of displacement damage (displacements per atom, DPA) after each dose (as estimated using the SRIM code [25]). The irradiation was performed on pieces with typical area 1.5 × 1.5 mm. Wherever used, the notation IRRx indicates a GBCO film without irradiation ($x = 0$), and $x = 0.5$, 1, 2 and 6 corresponding to films irradiated with proton dose $0.5 \times 10^{16}\,\text{cm}^{-2}$, $1 \times 10^{16}\,\text{cm}^{-2}$, $2 \times 10^{16}\,\text{cm}^{-2}$ and $6 \times 10^{16}\,\text{cm}^{-2}$, respectively. All samples included a 0.6 μm thick Ag-layer which protected the superconducting film.

3. Results and discussion

The T_c in the as-grown film is 93.3 K and is gradually suppressed with the doses. All values are consistent with those previously reported in single crystals (i.e. $\Delta T_c \approx 1.7$ K for $2 \times 10^{16}\,\text{cm}^{-2}$) [26]. Table 1 shows the T_c values after irradiation. The reduction in T_c of our samples with p-irradiation doses are larger than those reported in films with nanoparticles grown by MOD (i.e. $\Delta T_c \approx 1.5$ K for $8 \times 10^{16}\,\text{cm}^{-2}$) [5]. A wider superconducting transition is observed, possibly due to a more inhomogeneous irradiation and higher disorder, when the doses are increased to $6 \times 10^{16}\,\text{cm}^{-2}$.

Figure 1 shows a summary of the log–log J_c (H) and S (H) at 5 K, 27 K, 40 K, 65 K and 77 K for IRR0, IRR2 and IRR6. The J_c (H) dependences present three clear regimes: (I) the low-field regime ($B < B^*$), which can be associated with single-vortex pinning [27] but can also be affected by self-field effects and geometrical barriers [28]; (II) a power-law regime, $J_c\,(H) \propto H^{-\alpha}$, usually associated with strong pinning centers [27, 29]; and (III) a fast drop of J_c (H) (only evident in figures 1(d) and (e), which is related to a high creep rate. The increment of the p-irradiation fluence systematically suppresses the J_c ($H \rightarrow 0$) values for all the temperatures (see figure 2(a)). For example, J_c ($H = 0$, 5 K) drops from 29 MA cm^{-2} to 25 MA cm^{-2}, and J_c ($H = 0$, 77 K) drops from 3.6 MA cm^{-2} to 2.2 MA cm^{-2}, for IRR0 and IRR6, respectively. The extension of the regime (I) is weakly affected by the irradiations. The crossovers to the regime (II) are in good agreement with the expectations by self-field effects estimated as J_cx thickness. In all the samples in regime II, the α exponent increases with temperature. This can be attributed to an increment in the vortex–vortex interactions (see figure 2(b)) [3]. On the other hand, the α values (at the same T) are reduced as a consequence of the addition of random point defects by p-irradiation. The effect of this addition changes above 40 K [4]. Even for 40 K, the α exponent decreases systematically with the p-irradiations doses (see figure 2(b)) and the smooth J_c (H) dependences produce a huge increment of the J_c values at high magnetic fields. For comparison, at 27 K and $\mu_0 H = 3$ T the inclusion of random point defects produces an increment of J_c from 1.9 MA cm^{-2} to 3.2 MA cm^{-2} (≈1.7 times). Although the increment ratio is similar to that reported, our absolute J_c values are close to half the values reported in [5]. Above 40 K ($T = 65$ K and 77 K), the increment of J_c is not noticeably altered by the addition of random points. At 65 K, the irradiated samples present higher J_c values than IRR0 above $\mu_0 H \approx 0.7$ T. At 77 K, the irradiated samples present smaller J_c values than IRR0 at the regimes (I) and (II). Slightly higher J_c values are predicted at 65 K and 77 K by the Bean model in the field range dominated by fast creep rates. It is important to mention that the crossover field to the regime (III) (only evident for 65 K and 77 K) is weakly affected by irradiation.

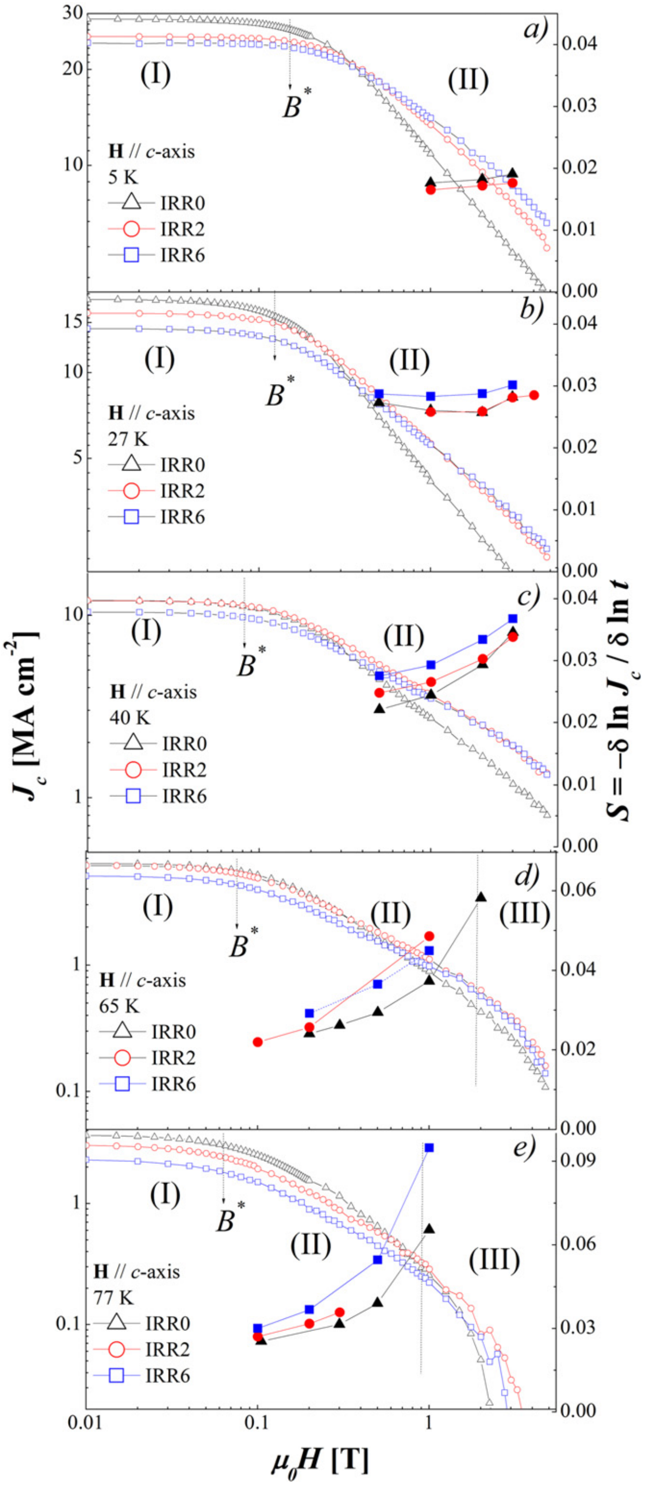

Figure 1. Magnetic field dependence of the critical current density (left) and flux creep rates (right) for IRR0, IRR2 and IRR6 at different temperatures. (a) 5 K; (b) 27 K; (c) 40 K; (d) 65 K; and (e) 77 K. All measurements were performed with **H**//c-axis. The lines between the crossovers are guided by the eye.

Table 1. Summary of proton irradiation dose, displacements per atom (dpa), and superconducting critical temperature (T_c). The T_c values were obtained from magnetization in 5 Oe with **H**//*c*-axis after zero field cooling.

Film	3 MeV Proton dose [cm^{-2}]	*Dpa*	T_c [K]
IRR0	—	0	93.4 (0.2)
IRR05	5×10^{15}	3.5×10^{-3}	92.6 (0.2)
IRR1	1×10^{16}	7×10^{-3}	92.4 (0.2)
IRR2	2×10^{16}	1.4×10^{-2}	91.7 (0.2)
IRR6	6×10^{16}	4.2×10^{-2}	89.5 (0.5)

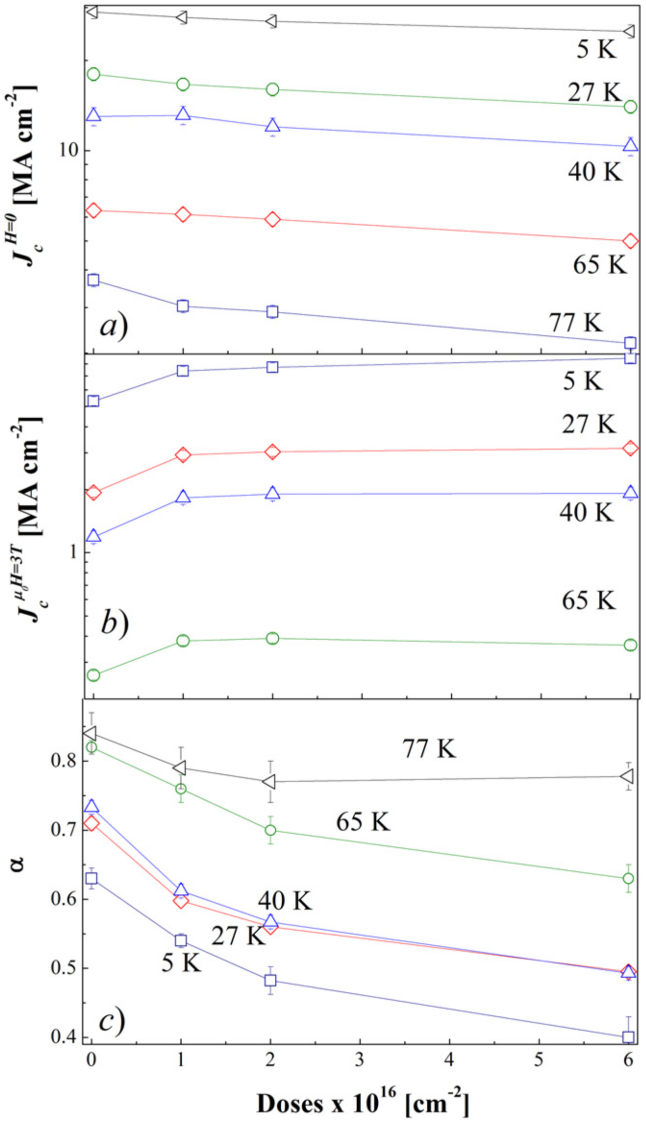

Figure 2. (a) Critical current density versus proton doses at $\mu_0 H \rightarrow 0$. (b) Critical current density versus proton doses at $\mu_0 H = 3$ T. (c) α versus proton doses obtained from $J_c\,(H) \propto H^{-\alpha}$. In all cases the values are included for 5 K, 27 K, 40 K, 65 K and 77 K (except in (b)) for Irr0, Irr1, Irr2 and Irr6.

Another issue to be discussed in our data is the influence of the *p*-irradiation on the *S* (*H*) values presented in figure 1. At low temperatures (5 K and 27 K) the *S* (*H*) dependences remain approximately constant inside the regime determined by the power-law dependence [3]. At temperatures above 40 K the *S* (*H*) dependences present a gradual increment as the field is increased. An evolution in the *S* (*H*) is expected by considering changes in the vortex bundle size [17] and a gradual increment in the vortex–vortex interactions [3]. We note that *p*-irradiation produces (except for 5 K) small increments in the *S* values independently of the J_c values. Intuitively, for similar film thickness, smaller *S* values are expected for higher J_c values [3]. On the other hand, a reduction in the *S* values and a large increment in the values of J_c have been reported in *p*-irradiated YBCO single crystals [24].

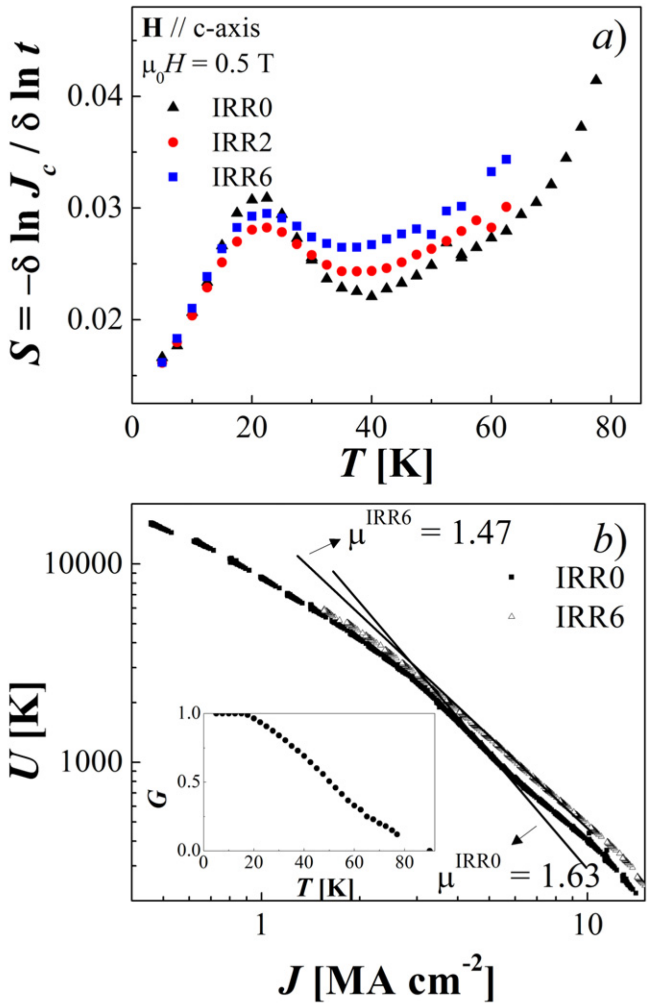

Figure 3. (a) Temperature dependence of the creep flux rate (*S*) at $\mu_0 H = 0.5$ T for IRR0, IRR2 and IRR6. (b) Maley analysis at $\mu_0 H = 0.5$ T for IRR0 and IRR6. The inset shows the *G*(*T*) dependence used for the Maley analysis. All measurements were performed with **H**//*c*-axis.

In order to analyze the increment of the flux creep rates with the *p*-irradiation doses in detail, we performed *S* (*T*) measurements at $\mu_0 H = 0.5$ T (above self-field). Figure 3(a) shows the results for IRR0, IRR2 and IRR6. The initial increment of *S* (*T*) at low temperatures can be ascribed to an Anderson–Kim-like mechanism with $S \approx T/U$. Non-negligible *S* values are expected at $T = 0$ from quantum creep [16]. Below 20 K both, irradiated and IRR0 samples present similar S values. This behavior is opposite to the one observed in iron-based superconductors [30, 31] where the irradiation enhances U and reduces the absolute S values. It should be noted that the S (T) dependences at $\mu_0 H = 0.5$ T present a small peak around 23 K. This peak can be related to double kink excitations due to the presence of correlated pinning, originated by the formation of twin boundaries [32]. The temperature peak maximum is weakly affected by p-irradiation. However, the peak height is suppressed by the addition of random point defects. This fact can be associated with the suppression of the double kink excitations by the presence of random point defects and nanoclusters [33]. The plateau that appears at intermediate temperatures (i. e 40 K) in figure 3(a) can be associated with glassy relaxation $\left(S \approx \frac{1}{\mu \ln t / t_0}\right)$ [16, 23]. Finally, when the temperature is increased, the creep rates are faster due to an increment in the thermal fluctuations and a decrease in the effective pinning energy [3]. It is worth mentioning that the systematic increment in the S values previously observed in figures 1(b)–(e) appears above the peak associated with double kink excitations (≈23 K). Motivated by the origin of this behavior, we decided to further investigate the influence of the p-irradiation on the μ values. According to Maley analysis [34], the effective activation energy $U_{\text{eff}}(J)$ can be experimentally obtained considering the approximation in which the current density decays as $\frac{\mathrm{d}J}{\mathrm{d}t} = -\left(\frac{J_c}{\tau}\right)\mathrm{e}^{-\frac{U_{\text{eff}}(J)}{T}}$. The final equation for the pinning energy is

$$U_{\text{eff}} = -T\left[\ln\left|\frac{\mathrm{d}J}{\mathrm{d}t}\right| - C\right] \tag{3}$$

where $C = \ln(J_c/\tau)$ is a nominally constant factor. For an overall analysis, it is necessary to consider the function G (T), which results in $U_{\text{eff}}(J,\ T = 0) \approx U_{\text{eff}}\ (J,\ T)/G(T)$ [35]. Figure 3(b) shows the Maley analyses for IRR0 and IRR6 at $\mu_0 H = 0.5$ T, where $C = 15$ and a G (T) were used (see figure 3(b) inset). In the glassy regime and above the peak corresponding to double kink expansion (≈23 K), $J \ll J_c$, and thus the μ exponent can be estimated as $\Delta \ln U(J)/\Delta \ln J$ [36]. The slopes corresponds to $\mu^{\text{IRR0}} = 1.63$ (0.02) and $\mu^{\text{IRR6}} = 1.47$ (0.02). On the other hand, by using the equation (2) and the S values obtained at the plateau, we have ln $(t/t_0) \approx 27$, which is close to the value previously reported in YBCO single crystals [36]. With ln $(t/t_0) = 27$ and equation (2), we obtain $\mu^{\text{IRR2}} = 1.52$ (0.02). These μ values are within those predicted by the collective creep theory for small vortex bundles due to random point defects [16]. Although theoretical models provide a small set of discrete μ for different vortex bundle size [16], experimental studies on YBCO usually present a gradual evolution of μ from small to large bundles as H is increased [17]. As shown in figure 2, the S (H) values for the same T are systematically higher when the p-irradiation dose is increased. These results suggest a gradual reduction in the μ values for the same magnetic field when the p-irradiation dose is increased [16, 17]. Although no-predictions of μ for mix landscapes (usually presented in films) have been theoretically reported, samples with strong pinning centers usually present smaller flux creep rates when the J_c is increased [3]. The relationship between the J_c and S values in coated conductors is an open question of relevant impact for some technological applications [37]. Systematic analyses of samples of similar thickness [1] and with different pinning landscape (films and clean single crystals) are expected to contribute to clarifying this issue.

The analysis of the J_c (H) with **H**// c-axis indicates that the initial pinning landscape presents high values at low-field (i.e. 27 MA cm^{-2} at 5 K and 3.2 MA cm^{-2} at 77 K) and a poor in-field dependence (see α values in figure 2(c)). Our results show that it is possible to improve the in-field dependences by adding random point defects by p-irradiation. The best changes in J_c (H) are observed at intermediate and high fields and below 40 K. Isotropic pinning is expected from random point defects [38]. The smooth J_c (H) dependences by p-irradiation are in agreement with data previously published by other authors [5]. However, the doses that we use in our study are similar to those used in YBCO single crystals [24] and smaller than those reported in [5] for coated conductors. In our study, p-irradiation doses around 2×10^{16} cm^{-2} improve the in-field J_c without further detriment of the J_c values at small magnetic fields. A larger dose reduces the T_c and the J_c ($H \to 0$) without significant improvement in the J_c values at high fields compared to 2×10^{16} cm^{-2}. The suppression of the self-field J_c increasing p-irradiation doses suggests that the superconducting properties are intrinsically affected by the irradiation beyond the large improvement in the in-field pinning. Usually the p-irradiation in clean YBCO single crystal produces a large increment of the J_c at both low and high magnetic fields [24], which masks any influence on the superconducting properties. In d-wave superconductors with short ξ, the order parameter is suppressed by defects (impurities) and recovers its bulk value at few atomic lattice constants a [39–41]. It has been reported that irradiation in YBCO single crystals contributes to reducing the critical temperature of the normalized superfluid density $\rho_s \approx \left(\frac{1}{\lambda(T)}\right)^2$ [42]. The distortion of the lattice in the environment of strong pinning centers usually contributes to the pinning [43]. However, the correlation between pinning and the influence of disorder in the penetration length (λ) has not been extensively discussed. The local suppression of the superconductivity around the defects could locally increase the penetration length (λ), affecting properties depairing critical currents and locally increasing the vortex fluctuations [16]. For example, an increment in $\lambda = 140$ nm (optimal T_c) [16] to $\lambda \approx 155$ nm (a reduction in $T_c \approx 4$ K) [42] should reduce the depairing current density (J_0) by approximately 20%. To estimate, $J_0(T = 0K) = cH_c/3\sqrt{6\pi\lambda}$ was used, where c is the speed of light and $H_c = \frac{\Phi_0}{2\sqrt{2}\pi\lambda(0)\xi(0)}$ is the thermodynamic critical field ($\xi(0) = 1.6$ nm). Therefore, an improvement in the J_c values produced by better pinning and a degradation in the intrinsic superconducting properties (increment in λ) is expected from the resulting J_c (H) dependences. More studies, including a systematic analysis of local suppression of the superfluid density by the addition of strong and weak pinning centers, are necessary. These experiments should contribute to a better understanding of the influence of the disorder on the superconducting properties of d-wave superconductors with short ξ, which should contribute to the optimization of J_c in coated conductors for technological applications.

4. Conclusions

In summary, we have studied the influence of additional random point defects in the critical current densities and vortex dynamics of co-evaporated 1.3 μm thick GBCO coated conductors. The p-irradiation reduces J_c at small fields but significantly improves the in-field dependences. The main improvements took place at temperatures below 40 K. Proton doses of around $2 \times 10^{16}\,\mathrm{cm}^{-2}$ enhance the J_c values at intermediate and high magnetic fields with a small reduction in the J_c ($H \rightarrow 0$). In addition, our data show a slight though clear increase in flux creep rates as a function of irradiation fluence. The strong reduction in the J_c ($H \rightarrow 0$) at high temperature (i.e. 77 K) suggests that intrinsic vortex fluctuations are increased by the p-irradiations. This fact could be associated with a local reduction of the superfluid density originated by the presence of random disorder. Futures studies should aim to clarify this point.

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