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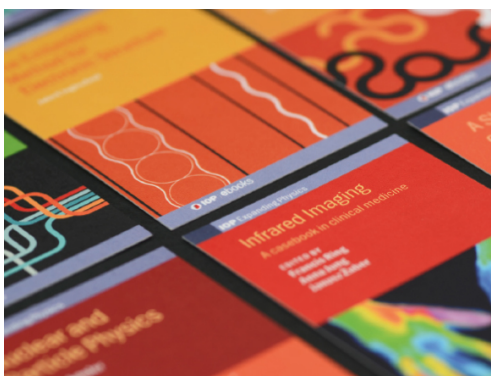
# Enhancement of critical current density in $\text{CaKFe}_4\text{As}_4$ single crystals through 3 MeV proton irradiation

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# Enhancement of critical current density in $\text{CaKFe}_4\text{As}_4$ single crystals through 3 MeV proton irradiation

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## Abstract

We study the influence of random point disorder on the vortex dynamics and critical current densities  $J_c$  of  $\text{CaKFe}_4\text{As}_4$  single crystals by performing magnetization measurements. Different samples were irradiated with a proton (p) beam at constant energy of 3 MeV to fluencies from  $2 \times 10^{15}$  p cm<sup>-2</sup> to  $4 \times 10^{16}$  p cm<sup>-2</sup>. The results show the addition of extrinsic random point disorder enhances the  $J_c$  values at low and intermediate temperatures over the entire range of magnetic fields applied. The optimum pinning enhancement is achieved with a proton fluence of  $3 \times 10^{16}$  p cm<sup>-2</sup>, increasing  $J_c$  at 5 K by factors  $\approx 5$  and 14 at self-field and  $\mu_0 H = 3$  T, respectively. We analyze the vortex dynamics using the collective creep theory. The enhancement in  $J_c$  matches with a systematic reduction in the flux creep relaxation rates as a consequence of a gradual increase in the collective pinning energy  $U_0$ . The substantial increment in  $J_c$  produced by random point disorder, reaching values of 9 MA cm<sup>-2</sup> at 5 K and self-field, makes  $\text{CaKFe}_4\text{As}_4$  a promising material for applications based on current carrying capacity at high magnetic fields.

Keywords: iron based superconductors, critical current densities, irradiation

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

## 1. Introduction

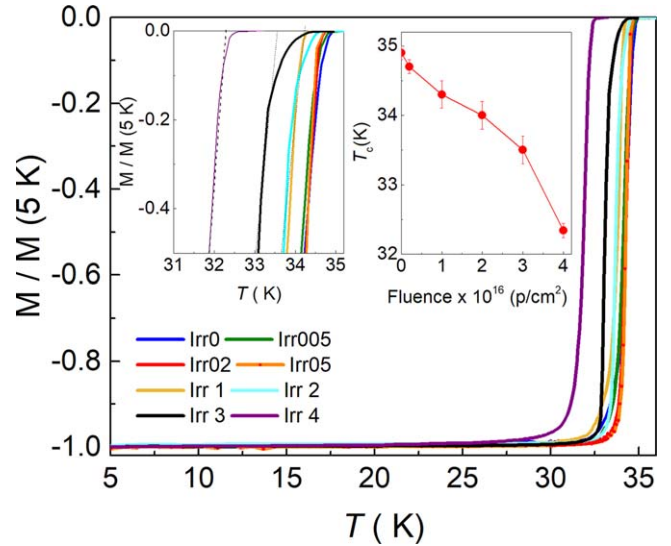
The current carrying capacity in superconducting materials depends on the intrinsic superconductor parameters and flux pinning. Technological applications such as superconductor magnets usually require an adequate operation temperature (related to the critical temperature,  $T_c$ ), large upper critical fields  $H_{c2}$ , and small upper critical field anisotropy  $\gamma$  [1]. Flux pinning refers to the ability to trap vortices on crystalline defects. The latter can be intrinsic to the material such as vacancies and twin boundaries, or added extrinsically such as random disorder and precipitates [1, 2]. The discovery of iron-based superconductors (IBS) allows for an expansion of

the knowledge on the interplay between intrinsic thermal fluctuations and vortex pinning [2–5]. Moreover, the span of different properties makes them attractive for applications. The recently discovered  $\text{CaKFe}_4\text{As}_4$  displays unusual features in the vortex pinning as a consequence of a particular defect structure [6, 7].  $\text{CaKFe}_4\text{As}_4$  crystallizes in a tetragonal structure. It has  $T_c = 35$  K, an extrapolated  $H_{c2}(0) > 70$  T with the in-plane coherence length  $\xi_{GL}^{ab}(0) \approx 1.4$  nm and the penetration depth  $\lambda^{ab}(0) = 208$  (4) nm [8, 9]. The anisotropy parameter  $\gamma = H_{c2}^{ab}/H_{c2}^c$  reduces with the temperature being  $\approx 1.5$  at 25 K and 2.5 near  $T_c$  [9]. Despite being a superconductor with small  $\gamma$ , the magnetic field dependence  $\mathbf{H}$  of the critical current density  $J_c$  exhibits high anisotropy.

Indeed,  $J_c^{H||ab}$  is up to one order of magnitude higher than  $J_c^{H||c}$  [7, 10, 11]. Moreover,  $J_c^{H||c}$  displays an unusual peak in temperature [10–12]. The sources of pinning in CaKFe<sub>4</sub>As<sub>4</sub> single crystals are stacking faults [11], planar CaFe<sub>2</sub>As<sub>2</sub> intergrowth [7], random point disorder [13], and regions in which the superconducting order parameter suppress (size comparable to the coherence length,  $\xi$ ) [14]. The strong vortex pinning for  $\mathbf{H}||ab$  originates in the planar defects [7, 11]. However, for  $\mathbf{H}||c$ , the vortex pinning at low temperatures is mostly weak. The latter is evident in large vortex creep rates and the small critical current-density ratio  $J_c/J_0 \approx 1.4\%$  (with  $J_0 = cH_c/3\sqrt{6}\pi\lambda$  the depairing critical current,  $c$  the speed of light, and  $H_c$  the thermodynamic critical field) [13]. The peak in the  $J_c^{H||c}(T)$  appears as a consequence of the emergence of new pinning centers in a regime dominated mainly by random point disorder [15]. The low pinning in pristine crystals agrees with the fact that substitutional disorder produced by Ni-substitution masks the peak in  $J_c^{H||c}(T)$  [13].

It is usually accepted that the best way to improve pinning in superconductor materials is by combining different types of pinning centers [1]. These include second phases (nanorods or nanoparticles), point defects, and structural disorder such as twin boundaries and stacking faults [16–19]. The particular microstructure of CaKFe<sub>4</sub>As<sub>4</sub> with stacking faults and CaFe<sub>2</sub>As<sub>2</sub> intergrowths makes it promising for the analysis of the influence of mixed pinning landscapes on its resulting  $J_c$  values. The standard method to introduce defects in a controlled way is particle irradiation. For any superconductor, the maximum electrical current density is determined by the vortex pinning defects that can be tailored into the material without degrading the superconductor properties. Depending on the mass and energy of the ions and the properties of the superconducting material, irradiation allows for the production of defects such as points, clusters, or tracks [1]. Whereas the pinning produced by random disorder and clusters is isotropic, pinning produced by tracks can be highly anisotropic. The effectiveness of random point disorder enhancing  $J_c$  has been verified in different IBS such as FeSe [20], Co-doped BaFe<sub>2</sub>As<sub>2</sub> [21, 22] and Ba<sub>1-x</sub>K<sub>x</sub>Fe<sub>2</sub>As<sub>2</sub> [23], among others. On the other hand, the vortex pinning in these materials is also largely improved by amorphous tracks [24–27].

Here, we show the influence of add random point disorder on the pinning and vortex dynamics of CaKFe<sub>4</sub>As<sub>4</sub> single crystals. Different samples were irradiated with 3 MeV protons to fluencies up to  $4 \times 10^{16}$  p cm<sup>-2</sup>. The  $J_c$  values at low and intermediate temperatures increase in the entire range of magnetic fields. For instance, at the optimal fluence of  $3 \times 10^{16}$  p cm<sup>-2</sup>,  $J_c$  at 5 K increases by factors  $\approx 5$  and 14 at self-field and  $\mu_0 H = 3$  T, respectively. The significant and systematic enhancement in the  $J_c$  values at low and intermediate temperatures correlates with a substantial reduction in the flux creep relaxation rates. The analysis of the vortex dynamics in the framework of the collective creep theory indicates that irradiation produces gradual increment in the pinning energy  $U_0$ .

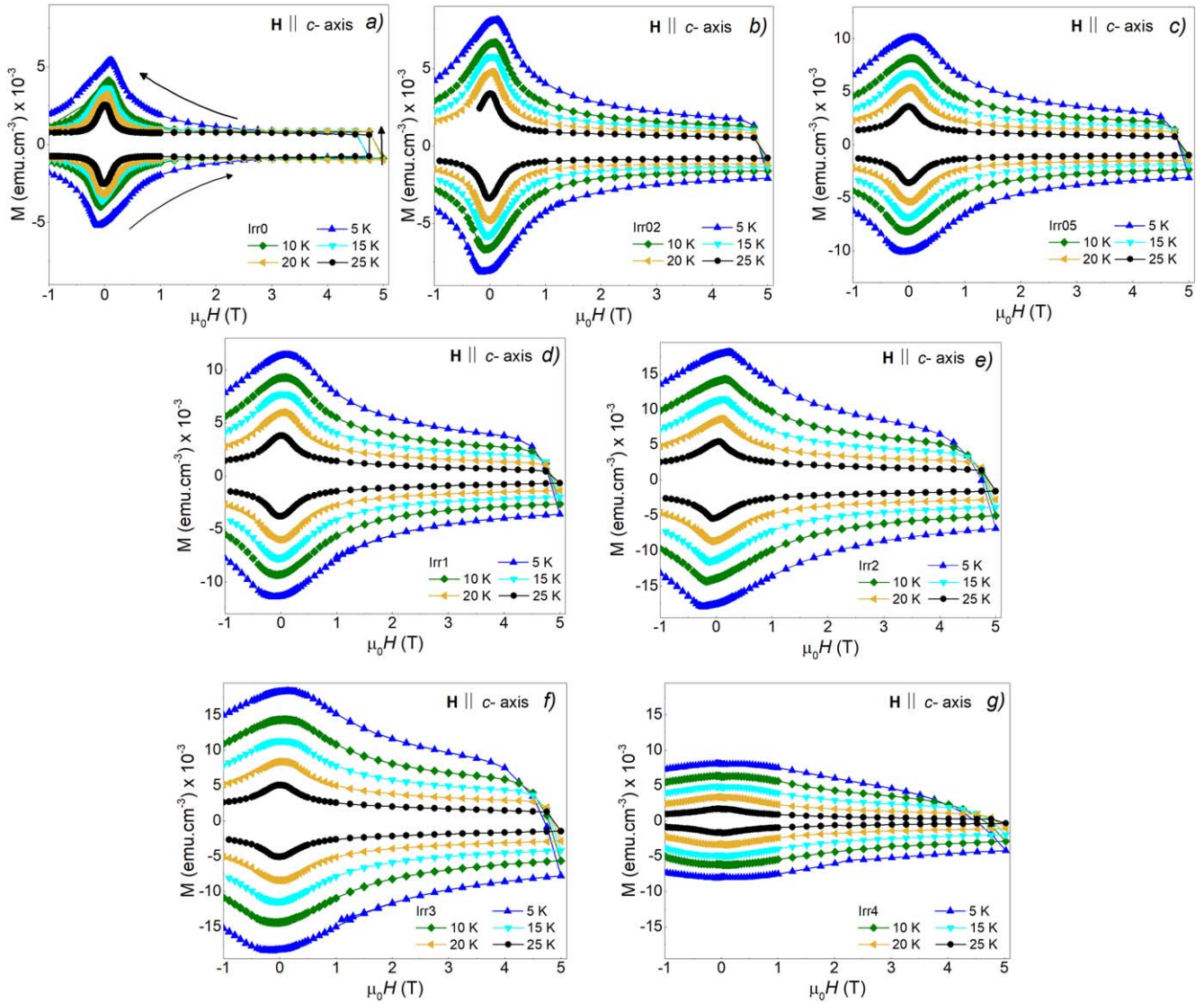


**Figure 1.** Normalized magnetization versus temperature for CaKFe<sub>4</sub>As<sub>4</sub> single crystals irradiated with different proton doses. The data corresponds to  $\mu_0 H = 1$  mT applied parallel to the  $c$ -axis. Inset at the right shows the  $T_c$  as function of the proton fluence. Inset at the left shows the criterion used to determine  $T_c$  applied to the Irr3 sample.

## 2. Material and methods

Single crystals of CaKFe<sub>4</sub>As<sub>4</sub> were grown using the procedure described in [9] and [28]. We use plate-like single crystals with length  $l$ , width,  $w$ , and thickness  $d$  around 0.15 cm, 0.1 cm and 0.002–0.003 cm, respectively. Magnetization measurements were collected using a Quantum Design SQUID magnetometer. The thicknesses  $d$  were calculated from Meissner slopes with  $\mathbf{H}||ab$  using the area ( $l \times w$ ) and the superconductor volume [13]. We extracted  $J_c(T)$  from the magnetization data using the Bean Model [29, 30]. For  $\mathbf{H}||c$ ,  $J_c = \frac{20\Delta M}{w(1-w/(3l))}$ , where  $\Delta M$  is the difference in magnetization between the top and bottom branches of the hysteresis loop. The flux creep rate,  $S = -\delta \ln J / \ln t$  was recorded as a function of time,  $t$ , over periods of 1 h using a scan length of 3 cm. The magnetization of the sample holder was measured and subtracted from the data by averaging the initial points of the time relaxation for the lower and upper magnetic branches.

The single crystals were irradiated with 3 MeV protons along the normal of the crystal plane (parallel to the  $c$ -axis) using a 1.7 MV tandem accelerator. Typical beam currents of  $\approx 30$  nA were used. The crystals were fixed to the holder with silver paint. The irradiations were performed with the ion beam positioned at the center of the sample (size spot  $\approx 1.8$  mm). The 3 MeV protons produce mainly random point defects and also some nanoclusters of a few nanometers in size [17, 31]. Simulations using the SRIM-TRIM software package [32] indicate that irradiation with 3 MeV protons produces a damage profile in which most of the ion's cross the samples. The so-called Bragg peak (energy loss of ionizing radiation during its travel through matter) for CaKFe<sub>4</sub>As<sub>4</sub> is at around 40  $\mu\text{m}$ , being a distance larger than the thickness of the single



**Figure 2.** (a)–(g) Magnetization loops under  $\mathbf{H}||c$  for pristine and irradiated  $\text{CaKFe}_4\text{As}_4$  crystals. The curves correspond to magnetic fields between  $\mu_0 H = -1$  T and 5 T. Data for pristine crystal in panel (a) are taken from the [13].

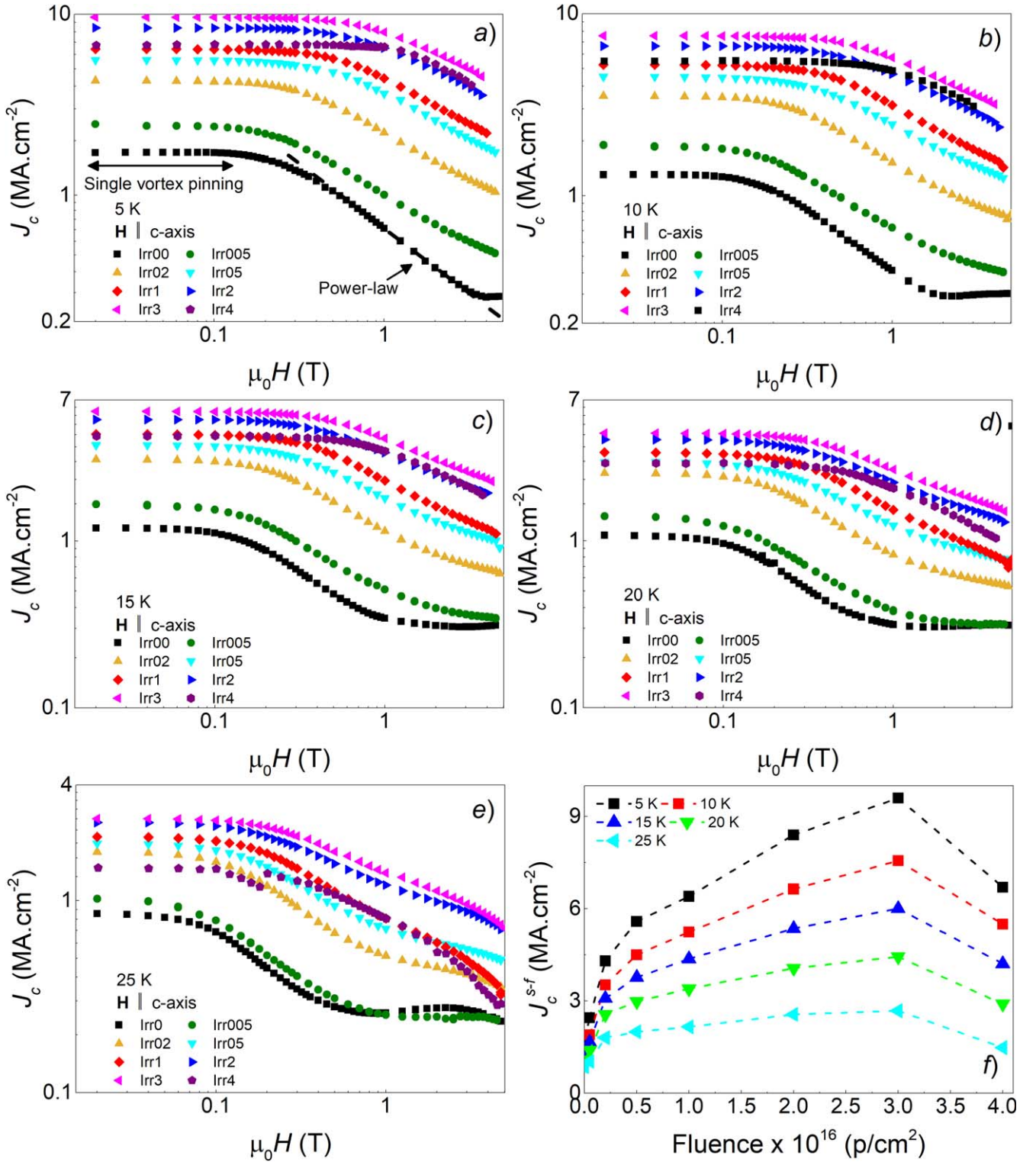
crystals used ( $\leq 30 \mu\text{m}$ ). Wherever used, the notation Irr $x$  indicates a  $\text{CaKFe}_4\text{As}_4$  single crystal without irradiation ( $x = 0$ ), and  $x = 02$  ( $2 \times 10^{15} \text{ p cm}^{-2}$ ), 05 ( $5 \times 10^{15} \text{ p cm}^{-2}$ ), 1 ( $1 \times 10^{16} \text{ p cm}^{-2}$ ), 2 ( $2 \times 10^{16} \text{ p cm}^{-2}$ ), 3 ( $3 \times 10^{16} \text{ p cm}^{-2}$ ) and 4 ( $4 \times 10^{16} \text{ p cm}^{-2}$ ).

### 3. Results and discussion

Figure 1 shows the temperature dependence of the normalized magnetization ( $M(T)/M(5 \text{ K})$ ) for the studied single crystals. The measurements were performed with  $\mathbf{H}||c$  under zero-field cooling with an applied magnetic field of 1 mT. The criterion for  $T_c$  determination, maximum slope of  $M(T)$  data extrapolated up to the  $M(T)/M(5 \text{ K}) = 0$  line, is shown in left inset in figure 1. The tails at the onsets and offsets of the curves may be related with non-uniform irradiation at the corners. The  $T_c = 34.8 \text{ K}$  for pristine crystal is gradually suppressed as irradiation fluence increases. For Irr4 (the

maximum fluence),  $T_c$  reduces to  $\approx 32.4 \text{ K}$  (see right inset in figure 1). The suppression of  $T_c$  with random disorder in  $\text{CaKFe}_4\text{As}_4$  was previously discussed in [33]. The changes in  $T_c$  and the variations  $\lambda(T)$  at low-temperatures agree with a multi-band superconductor with  $s_{\pm}$  pairing [34].

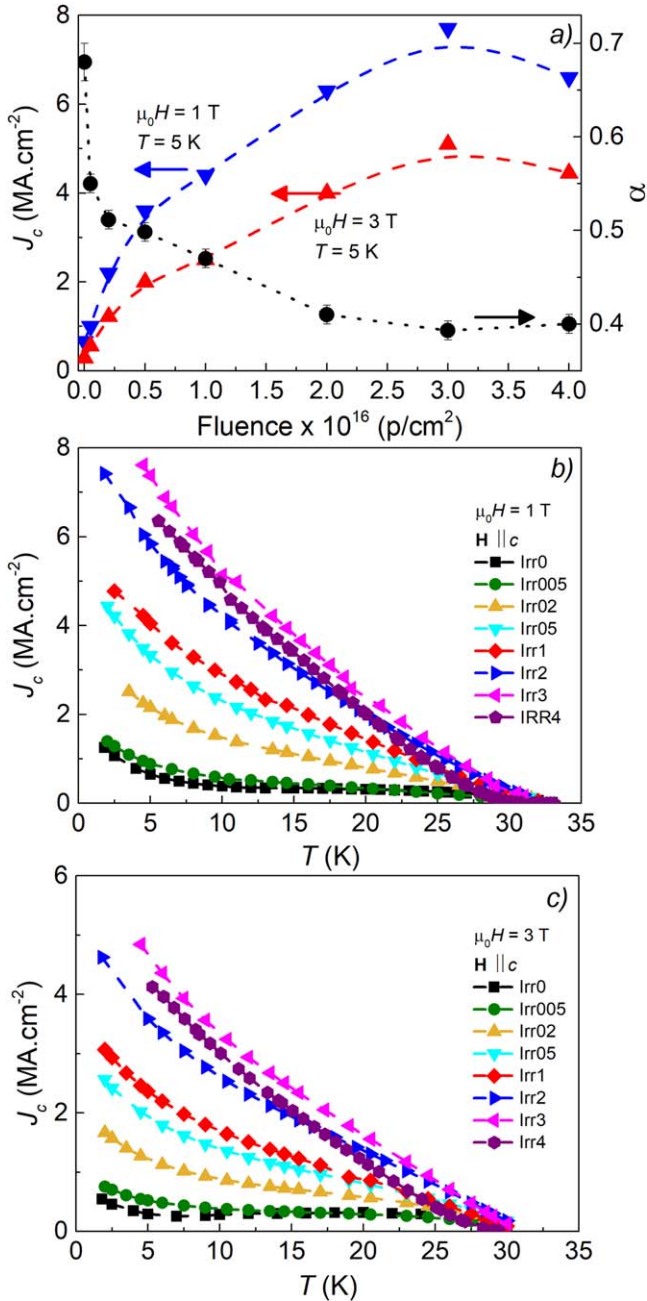
Figure 2 shows hysteresis loops for the pristine and irradiated crystals under  $\mathbf{H}||c$  at several temperatures. The loops are almost symmetric with respect to both  $H$ - and  $M$ -axes, indicating that bulk pinning dominates. The evolution of the  $J_c$  under magnetic field as a function of the irradiation fluence for temperatures between 5 and 25 K is summarized in figures 3(a)–(e). The results show that  $J_c(H)$  values enhance systematically as irradiation fluence increases up to  $3 \times 10^{16} \text{ p cm}^{-2}$  and decrease for Irr4. Upon irradiation to a dose of  $3 \times 10^{16} \text{ p cm}^{-2}$ ,  $J_c$  systematically enhances at low and intermediate temperatures across the whole range of fields. An exception is observed at  $T \geq 20 \text{ K}$  for Irr1, which may be related to differences in  $J_c(H)$  at high magnetic fields and temperatures in the pristine samples (usually related to a



**Figure 3.** (a)–(e) Magnetic field dependence of the critical current densities  $J_c$  in for  $\text{CaKFe}_4\text{As}_4$  single crystals irradiated with different proton fluencies at 5 K, 10 K, 15 K, 20 K and 25 K, respectively. (f) Summary of the self-field critical current densities  $J_c^{sf}$  as function of the proton fluence for several temperatures. Criterion for  $\alpha$  determination is included in panel (a).

modulation in  $J_c(H)$  due to a second peak in the magnetization) [13]. The comparison of the self-field critical current  $J_c^{sf}$  as a function of irradiation fluence is shown in figure 3(f). As we mention above,  $J_c^{sf}$  values enhance systematically as irradiation fluence increases up to  $3 \times 10^{16} \text{ p cm}^{-2}$  and

decreases for Irr4. For instance, at 5 K the  $J_c^{sf}$  value rises from  $\approx 1.7 \text{ MA cm}^{-2}$  for Irr0 to  $\approx 9 \text{ MA cm}^{-2}$  for Irr3. In addition to the substantial enhancement of  $J_c$  at low magnetic fields, the  $J_c(H)$  dependences improve with the irradiation for higher fields. The pristine single-crystal displays different regimes



**Figure 4.** (a) Dependence of the critical current density  $J_c$  for  $\mu_0 H = 1$  T and 3 T at 5 K (left axis) and  $\alpha$  exponent (right axis) with the proton fluence for CaKFe<sub>4</sub>As<sub>4</sub> single crystals. (b), (c) Temperature dependence of the critical current densities  $J_c$  for the different studied single crystals with  $\mu_0 H = 1$  T and  $\mu_0 H = 3$  T, respectively.

previously discussed in [13]: (I) at low field  $J_c \approx$  constant that could be associated to single vortex pinning; (II) a power-law dependence  $J_c \propto H^{-\alpha}$  related to strong pinning centers; (III) a third regime (at the end of the power-law) related to random disorder with  $J_c(H) \approx$  constant or a modulation with a peak originated from the second peak in the magnetization (SPM); and (IV) a high-field regime which is characterized by a fast drop in  $J_c(H)$  and is usually related to a crossover from elastic to plastic relaxation of the vortex lattice [34]. The regime (I) extends to higher fields for Irr4 than for the other

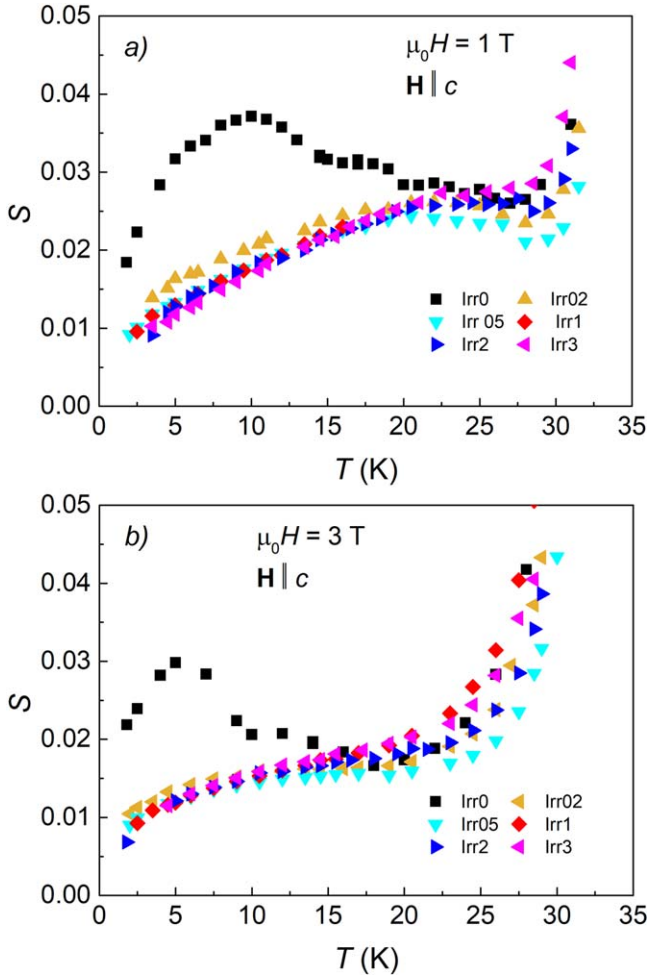
samples (see figure 3(a)). This fact can be related to the influence of the thickness and the self-field in the  $J_c$  values at low fields [35]. Indeed, Irr4 has a thickness of 30  $\mu\text{m}$  instead of the 20  $\mu\text{m}$  for the rest of the single crystals, which reduces  $J_c$  at the regime (I) without affecting their values at regime (II). Considering that the  $J_c$  values at the regime (II) for Irr4 are smaller than for Irr3, the resulting  $J_c^{sf}$  should be smaller in Irr4 than in Irr3 when corrected for equal thickness [35].

The irradiation reduces the  $\alpha$  exponent and masks the modulation in  $J_c(H)$  at low temperatures. Features related to a SPM remain at intermediate and high temperatures, even for high irradiation fluencies. Figure 4(a) (right y-axis) shows the evolution of the  $\alpha$  exponent with irradiation fluence for  $J_c(H)$  at 5 K. Similar  $\alpha$  exponents than for 5 K are observed up to 25 K, indicating that the vortex pinning at the power-law regime is provided by mixing of extrinsic random disorder and other defects present in the pristine crystals. The thermal fluctuations manifest as a shift to lower fields in the vortex crossovers between the different regimes. The result shows a fast decrease in  $\alpha$  from  $\approx 0.68$  in the pristine crystal to  $\approx 0.51$  for Irr02, which is in agreement with a low defect density in the former [13]. As irradiation fluence increases  $\alpha$  reduces, reaching a value of  $\approx 0.4$  for Irr3 and Irr4. A similar evolution of  $\alpha$  reaching a minimum value of approximately 0.4, has been previously observed in cuprate thin films [17]. The value of  $\alpha$  depends on the pinning landscape and usually decreases with a mixing of random disorder with other sources of pinning. Typical values ranges from  $\approx 0.5$  to 0.6 for nanoparticles [36] to  $\approx 0.2$  for mixed pinning landscapes with correlated disorder [37]. Intermediate values are typically observed in mixed pinning landscapes including strong and weak pinning centers [17, 22].

For CaKFe<sub>4</sub>As<sub>4</sub>, the introduction of small random defects via irradiation and the change in  $\alpha$  improve significantly the in-field dependences of  $J_c$  at low and intermediate temperatures. For instance, at 5 K for  $\mu_0 H = 1$  T and  $\mu_0 H = 3$  T, the  $J_c$  values in Irr3 increases in factors  $\approx 9.5$  and 14 with respect to the pristine crystal. It is important to note that the  $J_c^{H \parallel c}(T)$  peak usually observed in pristine crystals [7, 10–12] is masked for irradiation fluencies higher than  $2 \times 10^{15}$  p cm<sup>-2</sup>. Moreover, the enhancement of  $J_c$  at high magnetic fields takes place at  $T < 25$  K, where random disorder produces more effective vortex pinning (see figures 4(b) and (c)) [17]. In addition to the low effectiveness of small pinning centers to trap vortices close  $T_c$ , the  $J_c$  values at high temperatures in the irradiated crystals may also be affected by an increment in the vortex fluctuations due to variations in the superfluid density as a consequence of high disorder at the nanoscale [38].

To analyze in detail the correlation between the enhancement in  $J_c$  and the resulting vortex dynamics, we performed magnetic relaxation measurements of the persistent currents for proton fluencies up to  $3 \times 10^{16}$  p cm<sup>-2</sup>. The collective creep theory predicts a temperature dependence of the normalized flux creep rates as:

$$S = -\frac{d(\ln J)}{d(\ln t)} = \frac{T}{U_0 + \mu T \ln(t/t_0)} = \frac{T}{U_0} \left( \frac{J}{J_c} \right)^\mu, \quad (1)$$



**Figure 5.** (a), (b) Temperature dependence of the flux creep rates  $S$  for  $\text{CaKFe}_4\text{As}_4$  single crystals irradiated with different proton fluencies for  $\mu_0 H = 1$  T and  $\mu_0 H = 3$  T, respectively.

where  $t_0$  is a vortex hopping attempt time,  $U_0$  is the collective pinning barrier at  $T = 0$  in the absence of a driving force, and  $\mu > 0$  is the regime-dependent glassy exponent determined by the bundle size and vortex lattice elasticity [39]. Based on the model of nucleation of vortex loops, for random point defects in the three-dimensional case,  $\mu$  is  $1/7$ ,  $3/2$ – $5/2$ , and  $7/9$  for single vortex regime (SVR), small-bundle, and large-bundle creep, respectively. It is important to note that, despite specific numbers for the bundle size theoretically predicted [39], the experimental  $\mu$  values usually evolve gradually with magnetic fields [40]. As we have shown in [13], pristine  $\text{CaKFe}_4\text{As}_4$  crystals display high  $S$  values even for low temperatures, which is a signature of low  $U_0$  values. A distinctive feature in its vortex dynamics is a systematic increment in  $U_0$  with temperature due to the emergence of new pinning centers [7, 11, 12]. Figures 5(a) and (b) show  $S(T)$  dependences for  $\mu_0 H = 1$  T and  $\mu_0 H = 3$  T, respectively. The results show that in comparison with the pristine crystals, the damage produced by the irradiation provides a substantial reduction in the  $S$  values at low and intermediate

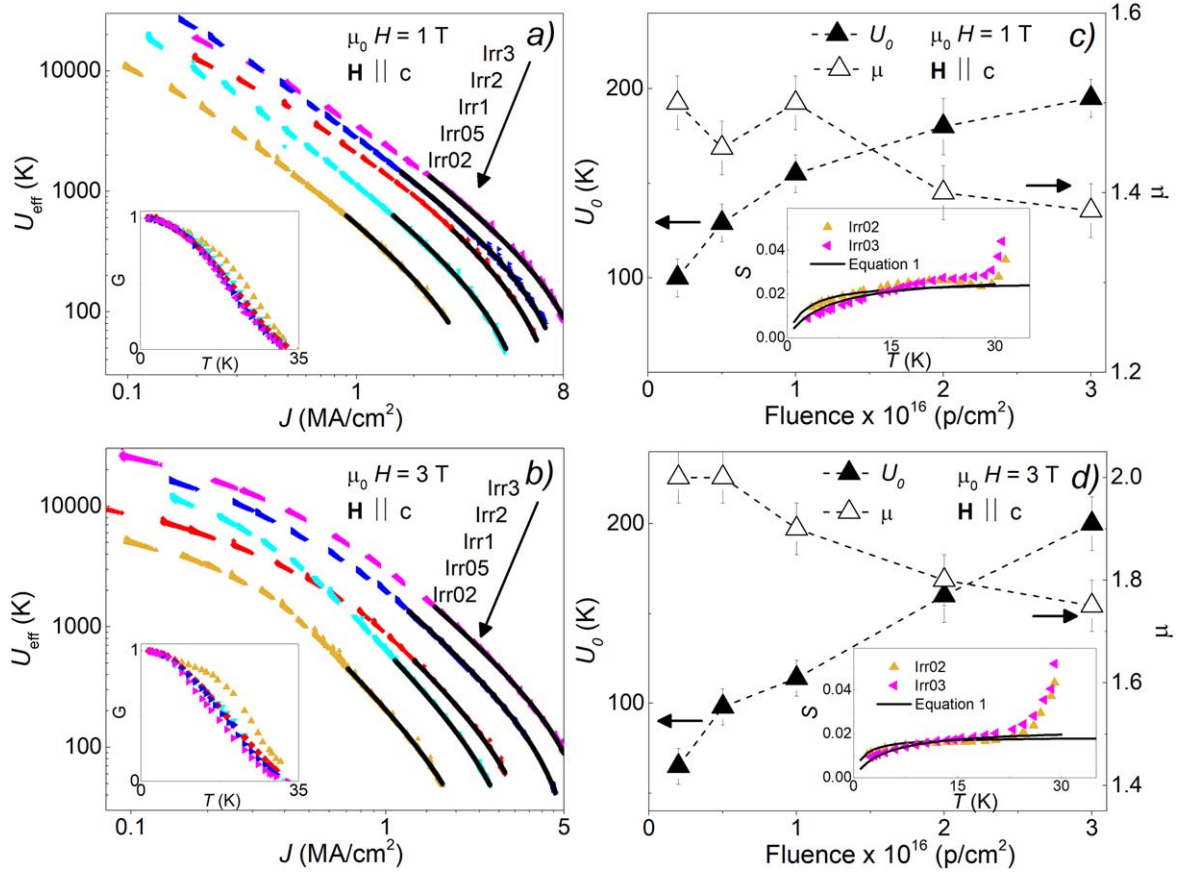
temperatures. On the other hand, for irradiated samples, minimal changes are observed at low temperatures, and slight variations occur at high temperatures. The small shifts in the crossover to faster creep near  $T_c$  indicate that random disorder slightly modifies the temperature in which the crossover from elastic to plastic motion takes place [35].

To analyze in more detail the influence of random disorder on vortex dynamics, we extract  $U_0$  and the  $\mu$  exponent that determines the  $S$  values at intermediate temperatures using the Maley analysis [41]. According to vortex-glass and collective creep theories [39], the effective activation energy as a function of current density ( $J$ ) is given by

$$U_{\text{eff}} = \frac{U_0}{\mu} \left[ \left( \frac{J_c}{J} \right)^\mu - 1 \right]. \quad (2)$$

The Maley method considers that the current density decays as  $\frac{dJ}{dt} = -\left(\frac{J_c}{\tau}\right) e^{-\frac{U_{\text{eff}}(J)}{T}}$ , and the effective activation energy  $U_{\text{eff}}(J)$  is described by  $U_{\text{eff}} = -T \left[ \ln \left| \frac{dJ}{dt} \right| - C \right]$  (with  $C$ , a constant factor). The analysis requires the consideration of a thermal factor  $G(T)$ , which results in  $U_{\text{eff}}(J, 0) \approx U_{\text{eff}}(J, T)/G(T)$  [42]. Figures 6(a) and (b) show the Maley analysis and the corresponding fits using equation (2) for the different samples with  $\mu_0 H = 1$  T and  $\mu_0 H = 3$  T, respectively. Insets show the respective  $G(T)$  dependences to maintain the ‘piecewise’ continuity. The data for Irr0 are not included due to changes in  $U_0$  with temperature [13]. It is important to note that the analysis considers that the vortex mechanism is not affected by temperature. Therefore, the fits were performed at  $T$  typically smaller than 15 K, which corresponds to the temperature range where the data for  $\mu_0 H = 1$  T and  $\mu_0 H = 3$  T have the power-law behavior (see figures 3(a)–(e)). Figures 6(c) and (d) show the exponent  $\mu$  and the  $U_0$  values for the different irradiation fluencies. The results show  $\mu$  exponents in a range between 1.4 and 2, which in collective creep theory are within the expectations for small bundles. The values are systematically higher for  $\mu_0 H = 3$  T than  $\mu_0 H = 1$  T. Moreover, as irradiation fluence increases,  $\mu$  reduces. A similar behavior is observed in cuprates [43] and has been related to the influence of random point disorder in mixed pinning landscapes [44]. On the other hand, the irradiation produces a significant increment in  $U_0$ . The initial values of a few tens of K (evidenced from the high  $S$  values at low temperatures) in the pristine crystal systematically increase with the irradiation fluence, reaching  $\approx 200$  K for Irr3. Inset figures 6(c) and (d) show the  $S(T)$  dependences for Irr05 and Irr3 and the corresponding data using equation (1) with the obtained  $U_0$  and  $\mu$ , and  $\ln(t/t_0) = 26$  [45]. A good correlation between the data is obtained.

The results show that the addition of random point disorder and nanoclusters enhances  $J_c$  at low and intermediate temperatures. The optimum pinning enhancement is achieved with a dose of  $3 \times 10^{16}$  p cm $^{-2}$ . The changes in  $J_c(H, T)$  show a clear correlation with an increment in  $U_0$ . We found that for the optimum dose,  $J_c$  at 5 K increases by factors  $\approx 5$



**Figure 6.** (a), (b) Maley analysis with  $\mu_0 H = 1$  T and  $\mu_0 H = 3$  T, respectively. Insets show the corresponding  $G(T)$  functions used to maintain the piecewise continuity. (c), (d)  $U_0$  (left y-axis) and  $\mu$  (right y-axis) for  $\mu_0 H = 1$  T and  $\mu_0 H = 3$  T, respectively. Insets shows  $S(T)$  functions for Irr02 and Irr3 using equation (1) with  $\ln(t/t_0) = 26$  and the respective data.

and  $\approx 14$  at self-field and  $\mu_0 H = 3$  T, respectively. The obtained values are close to those observed in proton irradiated  $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$  with  $T_c \approx 38.6$  K where a maximum  $J_c$  value of  $\approx 11$  MA  $\text{cm}^{-2}$  at 2 K for a fluence of  $5.8 \times 10^{16}$  p  $\text{cm}^{-2}$  [23]. Furthermore, the enhancement in  $J_c$  is higher than that observed in proton irradiated Co-doped  $\text{BaFe}_2\text{As}_2$  of  $\sim 2.6$  [21, 22]. The differences may be related to the  $\xi$  values, which make the pinning produced by small defects more effective [9, 46–48]. The  $J_c^{sf} \approx 9$  MA  $\text{cm}^{-2}$  for Irr3 corresponds to  $(J_c/J_0) \approx (9/170) \approx 5\%$ . Considering the SVR for random point disorder, the  $U_0$  value can be estimated

$$\text{by } U_c \approx \frac{H_c^2 \xi^3}{\gamma} \left( \frac{J_c}{J_0} \right)^{\frac{1}{2}} \approx T_c \left( \frac{J_c \left( 1 - \left( \frac{T}{T_c} \right) \right)}{J_0 G_i} \right)^{\frac{1}{2}} \quad (\text{with } G_i \text{ the Ginzburg}$$

number) [39]. An estimate for 5 K using  $\xi(0) = 1.4$  nm [9],  $\gamma \approx 1.2$  and  $\lambda \approx 200$  nm [8],  $G_i \approx 5.5 \times 10^{-4}$  and  $J_c^{sf} \approx 9$  MA  $\text{cm}^{-2}$ , yields  $U_0 \approx 300$  K. This value is in a good agreement with the data obtained at high magnetic fields. It is important to note the peak in  $J_c^{H||c}(T)$  usually observed in pristine crystals disappears even for irradiation fluence of  $2 \times 10^{15}$  p  $\text{cm}^{-2}$ . This fact agrees with fundamentally weak pinning for  $\mathbf{H}||c$  at low temperatures in the pristine  $\text{CaKFe}_4\text{As}_4$  single crystals. The substantial enhancement in  $J_c$  produced by random point disorder for  $\mathbf{H}||c$  and the strong pinning usually

observed for  $\mathbf{H}||ab$  make of the  $\text{CaKFe}_4\text{As}_4$  system a potential material for applications based on current carrying capacity at high magnetic fields [4]. Moreover, like in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ , the in-field dependences of  $J_c$  for  $\mathbf{H}||c$  might be significantly improved introducing correlated disorder [25, 26].

#### 4. Conclusions

To summarize, we have studied the effect of proton irradiation for fluencies up to  $4 \times 10^{16}$  p  $\text{cm}^{-2}$  in  $\text{CaKFe}_4\text{As}_4$  single crystals. The  $T_c$  systematically reduces from 34.8 K to 32.4 K as irradiation fluence increases. The addition of extrinsic random point disorder enhances the  $J_c$  values at low and intermediate temperatures in the entire range of magnetic fields. The optimum pinning enhancement is achieved with a proton fluence of  $3 \times 10^{16}$  p  $\text{cm}^{-2}$ , increasing  $J_c$  at 5 K by factors  $\approx 5$  and  $\approx 14$  at self-field and  $\mu_0 H = 3$  T, respectively. Moreover, the peak in  $J_c(T)$  present in pristine crystals is completely masked, even for lower irradiation fluencies. For irradiated samples, the collective creep theory, with a unique  $\mu$  and  $U_0$  for every  $H$ , describe the flux creep relaxation rates. We found that the enhancement in  $J_c$  at low and intermediate temperatures matches with a regular increase in the collective

pinning energy. The highest  $J_c$  values are reached for a fluence of  $3 \times 10^{16}$  p cm<sup>-2</sup> and correspond to glassy exponents  $\mu$  within expectations for small bundles and  $U_0 \approx 200$  K. The substantial enhancement in  $J_c$  produced by random point disorder for  $\mathbf{H}||c$  and the strong pinning usually observed for  $\mathbf{H}||ab$  make CaKFe<sub>4</sub>As<sub>4</sub> a promising material for applications based on current carrying capacity at high magnetic fields.

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