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# Critical current anisotropy in Fe(Se,Te) films irradiated by 3.5 MeV protons

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**Abstract.** Irradiation effects are often used to improve the effective pinning in superconductors, but sometimes they can reveal detrimental for superconducting properties. Fe(Se,Te) has been proven to be a very robust material against irradiation, in particular proton irradiation, thus configuring as an ideal material to work in harsh environments such as particle accelerators or fusion reactors. Anyway, the study of the pinning activation energy in Fe(Se,Te) thin film irradiated by 3.5 MeV protons suggests that this treatment can modify the anisotropy of the films pinning. Thus here we present the result of further investigation analyzing the effect of proton irradiation on the critical current and the pinning force both for the magnetic field applied parallel and perpendicular to the sample surface. We find that, although a slight effect on the critical current anisotropy is observed, the pinning landscape is not affected by the irradiation process. This confirms that Fe(Se,Te) can be considered for devices working in harsh environments.

## 1. Introduction

Thinking abstractly about the best superconductor for applications, anyone would think to the material with higher critical temperature,  $T_c$ , higher critical current,  $J_c$ , and higher critical field,  $H_{c2}$ , (or irreversibility field,  $H_{irr}$ ) values, which can be produced with low-cost techniques and possibly without dangerous elements. Then, one has to come back to the concrete use and other material properties have to be considered, even at the expense of some of the properties mentioned above. For example, a superconducting material suitable for devices working in harsh environments such as magnets for particle accelerators [1], [2], fusion reactors [3], or in space applications [4], [5], should have superconducting properties which are not affected by high-energy particle irradiation. Another relevant property is the anisotropy: higher is the anisotropy, the more complicate is the design of magnets, while the lower it is, better performance are attained.

Iron Based Superconductors (IBS) can be used to fabricate coated conductors that can work at high magnetic field with fairly high critical currents [6], [7], [8]. 11-IBS, i.e. IBS with chemical composition FeCh (where Ch is a chalcogenide as Se, Te or S), have also the advantage to not



include poisonous elements in their fabrication. In particular the Fe(Se,Te) compound can reach fairly high critical temperatures when grown on the right substrate [9], [10]. Although this material shows vortex dynamics effects resembling those of highly anisotropic high temperature superconductors [11], [12], it exhibits fairly isotropic pinning properties [13] and very low field anisotropy factor values [14].

The effects of different kinds of particle irradiation on different types of IBS have been analyzed so far [15], [16], [17], [18], with opposite effects depending on the material and the irradiation process. In particular, it has been observed that irradiation with low-energy protons can enhance critical current and critical temperature of Fe(Se,Te) thin films grown on a CeO<sub>2</sub> buffer layer by inducing collision cascade defects and modified film strain [19]. On the contrary,  $J_c$  and  $T_c$  of Fe(Se,Te) thin films grown on CaF<sub>2</sub> revealed to be robust against high-energy proton irradiation [20].

In this work, we search for possible effects of high-energy proton irradiation on the critical current and the pinning force anisotropy of Fe(Se,Te) grown on CaF<sub>2</sub>. In particular, samples irradiated with the interposition of an Al foil screen have been considered, since in this case an anisotropic variation on the pinning activation energy,  $U_0$ , behavior as a function of the applied magnetic field has been observed [21]. We anticipate that the considered irradiation process is actually responsible for the mentioned anisotropic variation in the  $U_0(\mu_0 H)$ , but such an irradiation does not change the pinning landscape.

## 2. Experimental details

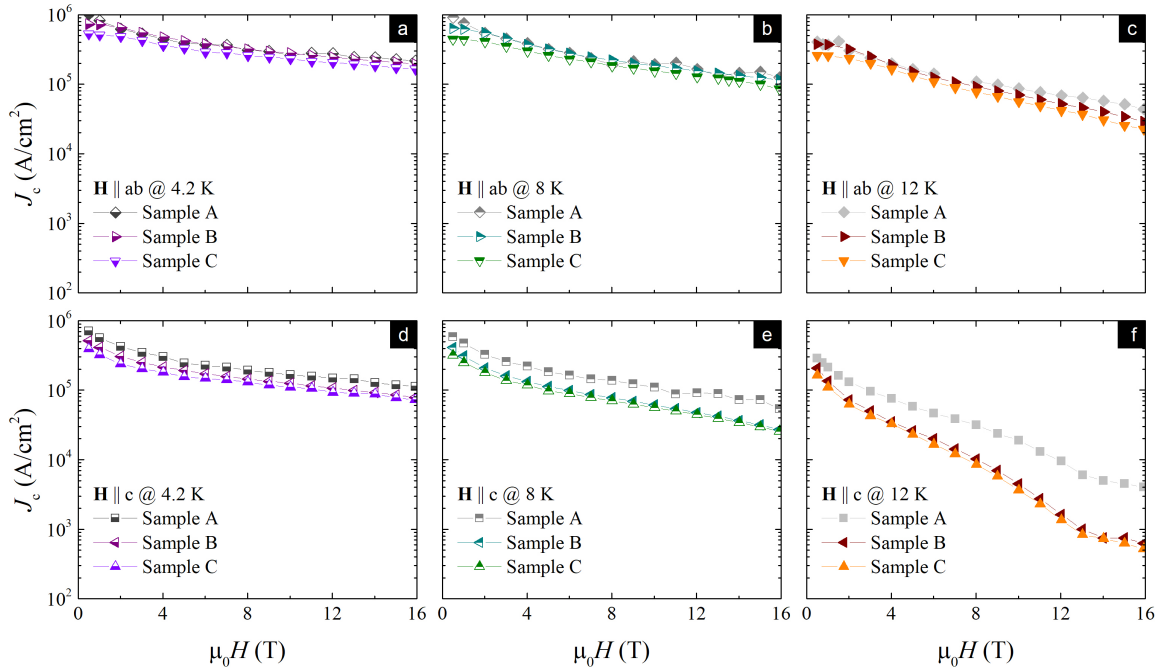
For the purpose of this study, Fe(Se,Te) thin films have been grown on [001] CaF<sub>2</sub> substrates. The deposition has been performed by a Ultra-High Vacuum Pulsed Laser technique using a Nd:YAG laser at 1024 nm. The starting target stoichiometry is FeSe<sub>0.5</sub>Te<sub>0.5</sub>. Film thickness results to be 100 nm. On each film, nine Hall-bars have been patterned through standard optical lithography and Ar ion-milling etching. The bars are 20  $\mu\text{m}$  wide and 50  $\mu\text{m}$  long.

In this paper, three different type of samples have been considered. The first type, named Sample A, is a pristine Hall-bar. Sample B type is a Hall-bar irradiated with 3.5 MeV protons with a fluence of  $2.68 \cdot 10^{16} \text{cm}^{-2}$ . For Sample C the fluence is  $5.35 \cdot 10^{16} \text{cm}^{-2}$ . In both cases, protons are decelerated through an 80  $\mu\text{m}$  thick Al foil, which reduce the average proton energy to 1.4 MeV, leading to the implantation of the protons in a region of the substrate near to the interface with the Fe(Se,Te) film [20] (see Table 1 for a summary on the used proton fluence and samples critical temperature values.).

**Table 1.** In the table the 3.5 MeV proton fluence and the resulting critical temperature are reported for each of the analyzed samples.

Sample	Fluence ( $10^{16} \text{cm}^{-2}$ )	Critical Temperature (K)
A	0	17.7
B	2.68	17.3
C	5.35	17.1

Current-voltage measurements,  $V(I)$ , have been carried out in a cryogen-free cryostat by Cryogenic Limited, equipped with a superconducting 16 T magnet and a variable temperature insert (VTI) ranging from 1.6 to 300 K where samples are cooled by He gas flow. Samples are mounted on a mechanical rotating platform able to rotate along two perpendicular axis. For the considered measurements, samples have been rotated with respect to the fixed applied field



**Figure 1.** In panels a, b and c the the critical current curve as a function of the applied magnetic field are reported for each of the three Samples at 4.2, 8 and 12 K respectively. The field direction in this case is parallel to the superconducting film ab-plane. In panels d, e and f  $J_c(\mu_0 H)$ 's are reported for the field direction parallel to the superconducting film c-axis.

direction,  $\mathbf{H}$ , keeping  $\mathbf{H}$  always perpendicular to the current flow direction. The rotation angle  $\Theta$  is the angle formed by  $\mathbf{H}$  and the thin film surface, such as  $\Theta = 90^\circ$  corresponds to the configuration with the c-axis parallel to  $\mathbf{H}$  (we will refer to this configuration as  $\mathbf{H} \parallel \mathbf{c}$ ), while  $\Theta = 0^\circ$  corresponds to the ab-planes parallel to  $\mathbf{H}$  ( $\mathbf{H} \parallel \mathbf{ab}$  configuration in the following).

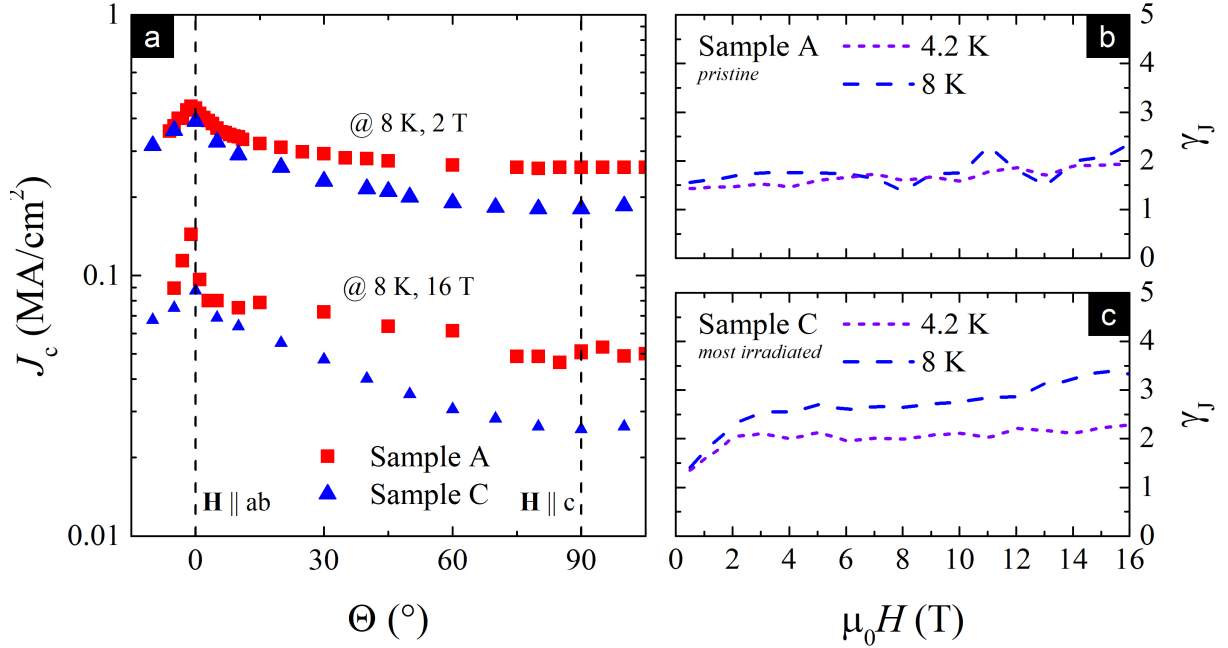
Voltage values have been measured by a delta-mode 4-probe technique using a Keithley Nanovoltmeter model 2182. Current bias is in shape of current pulses whose duration is set to 100 ms, with inter-pulse separation time of 2 s, generated by a Keithley Current Source model 6221. The criterion used for critical current estimation is set at  $10 \mu\text{V cm}^{-1}$ .

### 3. Results

In order to analyze the effect of the proton irradiation on the anisotropy of our samples pinning properties, several current-voltage measurements have been carried out. For all three samples, we acquired  $V(I)$  curves as a function of the applied magnetic field at three different temperature values, namely 4.2, 8 and 12 K, for both the orientation  $\mathbf{H} \parallel \mathbf{c}$  and  $\mathbf{H} \parallel \mathbf{ab}$ .

In Figure 1, the critical current curves as a function of the applied magnetic field as evaluated from  $V(I)$  measurements are reported. It is evident that the proton irradiation has an almost null effect on the critical current in the case  $\mathbf{H} \parallel \mathbf{ab}$ , while for  $\mathbf{H} \parallel \mathbf{c}$  a decrease in the  $J_c$  values can be observed as temperature increases and at high applied critical field. Figure 1 also show that an higher proton fluence does not reduce further the  $J_c$  values, so that in the following we will focus on comparing the results relate to the pristine sample A with those related to the most irradiated sample C.

As a consequence of the variation in the critical current values after the irradiation, also



**Figure 2.** (a) The critical current behavior as a function of the angle between magnetic field direction and superconducting film c-axis. (b) Sample A anisotropy factor dependence on the applied magnetic field at two different temperature values (short-dashed line is for 4.2 K, long-dashed line is for 8 K). (c) Sample C  $\gamma_J(\mu_0 H)$  at the same two temperature values.

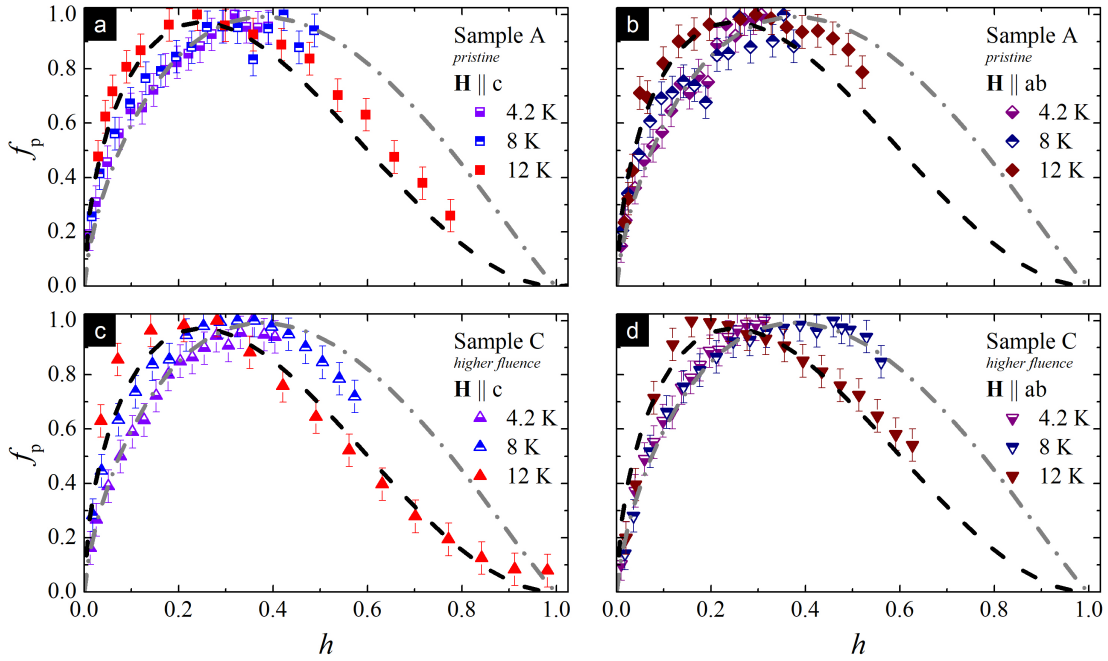
the anisotropy of the critical current results to be modified. Anyway, we can observe that the irradiation process does not modify the general behavior of the critical current as a function of the angle between the sample and the applied magnetic field. Indeed, looking at Figure 2a, we note that the  $J_c(\Theta)$  of sample A presents a peak for  $\Theta = 0^\circ$ , i.e.  $\mathbf{H} \parallel ab$ , as expected [22], [23]. The  $J_c(\Theta)$  of sample C shows the same behavior, even if a suppression of  $J_c(90^\circ)$  can be inferred.

Despite a similar behavior as a function of the angle, Figure 2a suggests that the ratio between the critical current value for  $\mathbf{H} \parallel ab$  and the one for  $\mathbf{H} \parallel c$ , i.e. the critical current anisotropy factor  $\gamma_J$ , is different. This is clearly seen looking at the  $\gamma_J(\mu_0 H)$  curves. Indeed, in Figure 2b it is shown that for the pristine sample A the  $\gamma_J$  value is almost constant in field and independent of temperature in a range  $T \leq 10$  K interesting from an application point of view. In Figure 2c, it can be recognized an increasing trend with the increasing field for the irradiated samples, trend which is more pronounced as temperature increases.

In general, we can note that there is an overall weak anisotropy of this superconductor probed by the quite low values of the critical currents anisotropy factor even for the highest irradiation dose and by field anisotropy factor values  $\gamma_H < 4$  [21], [24].

#### 4. Discussion

The suppression in  $J_c$  for  $\mathbf{H} \parallel c$  has been associated with the variation of the strain in the superconducting films due to the position of the defects created in the substrate by the irradiation process [20]. In our samples, indeed, the use of an Al foil reduces the proton implantation depth, thus defects in the substrate are created close to the interface with the superconducting film. As a consequence, the strain in the Fe(Se,Te) is changed in such a way that is detrimental to its



**Figure 3.** The normalized pinning curve as function of the magnetic field at three different temperature values for: (a) Sample A in field applied along the film  $c$ -axis, (b) Sample A in field applied along the film  $ab$ -plane, (c) Sample C in field applied along the film  $c$ -axis, and Sample C in field applied along the film  $ab$ -plane. In all four panels, the dashed line is related to the curve resulting from Dew-Hughes fitting procedure with two contributions, while the dot-dashed line is related to the curve resulting from a standard Dew-Hughes fitting procedure.

superconducting properties, as it is well known that this material superconducting properties are strongly influenced by the crystalline structure [9], [10]. However, in the case of the critical current it could not be excluded that the changes can be determined also by a change in the pinning landscape, as reported in [19]. Nevertheless, we have to remark that in our case only the critical current in the  $\mathbf{H} \parallel c$  case is modified, contrary to the results reported in [19] where both the  $J_c^{\parallel c}$  and  $J_c^{\parallel ab}$  are affected.

A way to determine whether the considered irradiation changes the nature of the pinning centers or not is looking at the pinning forces as function of the applied magnetic field  $F_p(\mu_0 H)$ . In particular, in the framework of the Dew-Hughes model [25], the normalized pinning force  $f_p = F_p/F_{p,max}$  ( $F_{p,max}$  being the maximum value of the  $F_p(\mu_0 H)$  curve) can be expressed in terms of the normalized magnetic field  $h = H/H_{irr}$  as:

$$f_p = C \cdot h^p (1 - h)^q. \quad (1)$$

The values of the exponent  $p$  and  $q$  and of the constant  $C$  are strictly determined by the nature and the dimensionality of the pinning centers.

The  $f_p(h)$  curves for samples A and C, evaluated by the acquired  $J_c(\mu_0 H)$  for both  $\mathbf{H} \parallel c$  and  $\mathbf{H} \parallel ab$  orientation, are reported in Figure 3. The normalizing field value  $H_{irr}$  as been estimated by a linear extrapolation on the  $J_c^{1/2}(\mu_0 H)$  curves [26]. It is quite evident that the irradiation process does not modify the  $f_p(h)$ , thus the pinning landscape.

In particular, we found that the data for  $\mathbf{H} \parallel c$  (3a and c) at 12 K can be fitted well by the modified two-contributions  $f_p(h)$  curve  $w \cdot f_{p,1}(h) + (1-w) \cdot f_{p,2}(h)$  (black dashed line in Figure 3) where  $w$  is the only fitting parameter and each  $f_{p,x}$  is expressed by Equation (1) with the values  $C_1 = 3.49, p_1 = 0.5, q_1 = 2$  and  $C_2 = 6.75, p_2 = 1, q_2 = 2$ . The resulting  $w$  value is  $0.68 \pm 0.05$ , which is in perfect agreement with previous results enlightening the presence of both 2D  $\delta l$  and 1D  $\delta l$  pinning centers [14], [27].

At lower temperature, the maximum of the  $f_p(h)$  curve is shifted at higher  $h$  values and the data are fitted by a single contribution  $f_p(h)$  curve where  $C = 3.4 \pm 0.3, p = 0.71 \pm 0.04, q = 1.17 \pm 0.10$  (grey dot-dashed line in the mentioned Figures). These values are close to those that the Dew-Hughes model associates to  $\Delta T_c$  pinning (i.e.  $C = 4, p = 1, q = 1$ ) [25]. This additional contribution, which plays the same role in both  $\mathbf{H} \parallel ab$  and  $\mathbf{H} \parallel c$  orientations and which dominates at lower temperature, should be ascribed to the intrinsic nature of this material.

We remark that in the case  $\mathbf{H} \parallel ab$  (see Figure 3b and d) no difference with the  $\mathbf{H} \parallel c$  case can be observed, thus probing that the material pinning properties are not modified by the irradiation process. Thus, we can confirm that observed reduction in  $J_c$  with the proton irradiation is due to the suppression of the superconducting properties induced by the change in the material strain.

Finally, a previous analysis by the authors on the pinning activation energy as a function of the applied magnetic field,  $U_0(\mu_0 H)$ , of the same samples here investigated pointed out that the  $U_0(\mu_0 H)$ 's related to  $\mathbf{H} \parallel ab$  shows a change after the irradiation. In particular, it has been observed a progressive shift to higher values of the threshold field between two different pinning regimes as the proton fluence increases [21]. The results here shown enlighten that there are no changes in the pinning landscape of the considered Fe(Se,Te) thin films by proton implantation near the film-substrate interface. Thus, the origin of the  $U_0(\mu_0 H)$  behavior can be ascribed to the changes in the strain of the layered crystallographic structure induced by the irradiation process [20].

## 5. Conclusions

Summarizing, we have investigated the effect of high-energy proton irradiation on the pinning properties anisotropy of Fe(Se,Te) thin films grown on CaF<sub>2</sub>. The pinning properties of pristine samples and irradiated samples have been compared both for magnetic field applied parallel and perpendicular to the material c-axis.

We considered samples which have been irradiated with the interposition of an Al foil between the proton source and the film, a procedure which leads to the implantation of protons in a region of the CaF<sub>2</sub> substrate near to the interface with the superconducting film. Such an irradiation has been proven to affect slightly the critical temperature and the critical current values. Moreover, the analysis of the pinning forces in the framework of the Dew-Hughes model revealed no changes in the pinning landscape.

Since an anisotropic effect in the behavior of the pinning activation energy has been observed previously, our findings let us suggest that this effect is related to the change induced by the considered irradiation in the material strain, which changes the balance between vortex-vortex and vortex-pinning interactions.

Finally, the results here reported confirm one more time that the iron based superconductor Fe(Se,Te) is suitable for the use in device which have to work in high magnetic field and harsh environments.

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