Proton Irradiation Effects on the Superconducting Properties of Fe(Se,Te) Thin Films

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***Abstract*—Ion irradiation of superconductors allows both the establishment of their radiation hardness and the modification and optimization of their properties useful for applications. In this work, we investigate the effects of proton irradiation with different energies on Fe(Se,Te) thin films grown on CaF_2 and on buffered YSZ substrates. These systems allowed us to perform preliminary studies for the development of Fe(Se,Te) coated conductors. Critical temperature and critical current were measured for different levels of displacement damage, and with ion implantation happening at various depths inside the substrate, as evaluated by simulations with Monte Carlo codes. All measurements evidenced that these Fe(Se,Te) films are robust against proton induced damage, and that an increase of critical current can be achieved introducing pointlike defects in the superconductor. However, we find that damage induced in the substrate also plays a crucial role in modifying superconducting film properties through a variation of the strain exerted on the film. This strain degrades superconducting parameters and should be minimized.**

***Index Terms*—Iron-based superconductors, proton irradiation, strain, critical temperature, critical current, critical fields, radiation hardness.**

I. Introduction

IRRADIATION is a powerful tool to introduce a controlled distribution of defects in a superconductor [1]–[3], introducing carriers scattering useful for fundamental studies [4]–[8], tuning the pinning mechanism [9], [10] and improving parameters useful for applications such as the in-field critical current density [11]. This approach was proven useful even for the optimization of coated conductors on an industrial scale [12], [13]. In addition, irradiation allows defining damage thresholds above which a significant degradation of the superconductor properties occurs [14]. This is crucial for applications in locations where high radiation levels are expected [15], [16], such as in the aerospace field and in nuclear fusion reactors [17]. When working with superconducting materials deposited on a different material, however, the situation is further complicated by the interplay between the substrate (that is itself damaged by the irradiation) and the film [18]–[20]. This interplay can take place through a change of the strain level exerted by the damaged substrate on the film and can induce modifications on the superconducting properties [21]. Here, we analyze the response of Fe(Se,Te) thin films deposited on single crystal substrates (buffered and not) to proton irradiation with different energies. The irradiation conditions are chosen to produce comparable, moderate damage levels in the superconducting film while varying the implantation depth of protons in the substrate (i.e. the distance from the interface with the film). In this way, the effect of the strain induced by the damaged substrate on the superconductor can be tuned. We find that the critical current (J_c) and critical temperature (T_c) modifications with increasing irradiation damage are strongly dependent on the implantation depth. This is particularly relevant in view of the possible application of Fe(Se,Te) as coated conductors in radiation hard environments. In this paper, we organize and expand the set of irradiation conditions and transport experiments performed on Fe(Se,Te) thin films and attempt to correlate the implantation depth of protons in the substrate with the observed changes of T_c and J_c.

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II. Experimental Details

A. Samples

Thin films were deposited on single crystalline substrates starting from a stoichiometric $FeSe_{0.5}Te_{0.5}$ target in an Ultra-High Vacuum Pulsed Laser deposition system equipped with a Nd:YAG laser at 1024 nm [22]. The resulting film thickness was about 100 nm. The substrates employed were either oriented [001] CaF_2 single crystals [23] or Y_2O_3-stabilized ZrO_2 (YSZ) buffered with Zr-doped CeO_2 [24]. For transport measurements, Hall-bars shaped patterns were obtained from the films through standard optical photolithography and Ar ion-milling etching prior to the irradiation process. The bars width was 20 μm, while their length was 50 μm. The same process was employed to obtain rectangular shapes for magneto-optical measurements. All films were analyzed with x-ray diffraction using a four-circle diffractometer to confirm their phase purity and their optimal epitaxial growth [25].

B. Irradiation and Simulations

Details of the irradiation experiment were guided by simulations of the damage induced both in the film and in the substrate, performed with the SRIM code [26]. As visible in Fig. 1, in all cases the protons crossed the films and implanted into the substrate with implantation depths varying from 4.5 to 86 μm. In the superconductor, protons produce randomly distributed point defects, and small cascades forming clusters of defects with typical dimension of the order of few nm, as investigated both in IBSs [27], [28] and cuprates [29]. Following [30], the modified Kinchin Pease approach assuming a displacement energy of 25 eV for all atoms of the target [31] was employed to estimate the damage in the film in terms of displacements per atom (dpa) starting from the SRIM output. The simulations showed that the damage is very homogeneous across 100 nm thick films (<5% relative variation in the worst case). The irradiation experiments were performed in high vacuum and at room temperature at the CN and AN2000 facilities of the INFN-LNL laboratories.

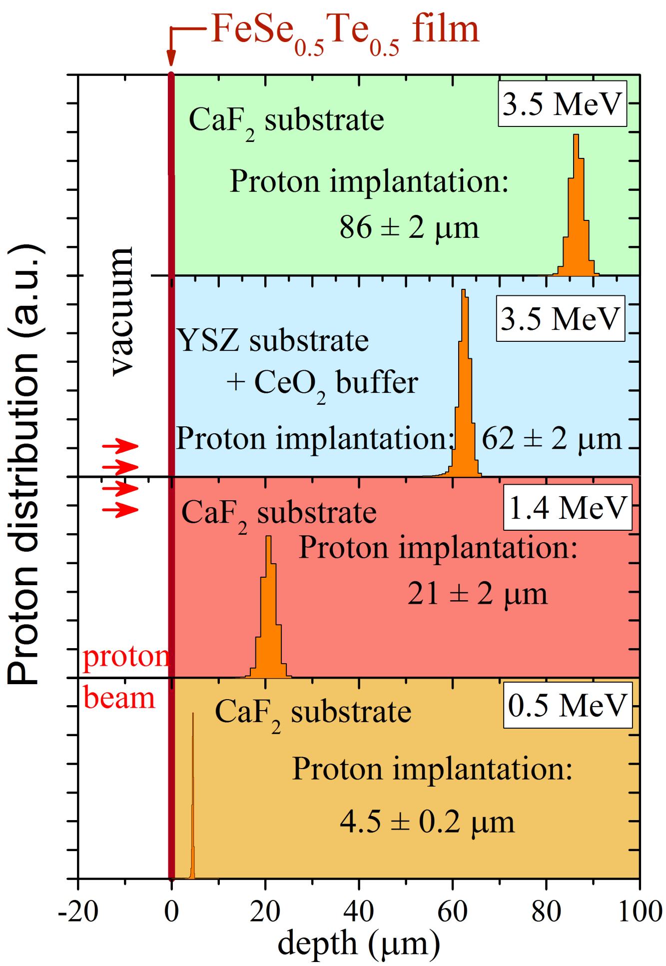

Fig. 1. Summary of the four proton irradiation conditions showing the implantation peak in the substrate of the Fe(Se,Te) thin films.

C. Measurements

1) X-ray Analysis: Large unpatterned samples were investigated by x-ray diffraction to study the strain induced by the substrate after irradiation. The values of the c cell parameter for the different samples were evaluated by means of Rietveld refinement with the programme Fullprof from the $00l$ diffraction patterns. The CaF_2 substrate was used as reference to refine the instrumental resolution function and the zero-shift parameters. A Thompson–Cox–Hastings pseudo-Voigt function convoluted with an axial divergence asymmetry function was used to model diffraction lines. The obtained values were fixed, and the parameters pertaining to the $FeSe_{0.5}Te_{0.5}$ thin films (the lattice parameter c and the Lorentzian strain parameter) were refined by imposing a full c-axis texturing. Lattice microstrain along $[00l]$ was evaluated by the refined strain parameters, and the broadening of the diffraction lines was analysed by means of the Williamson–Hall plot method [32].

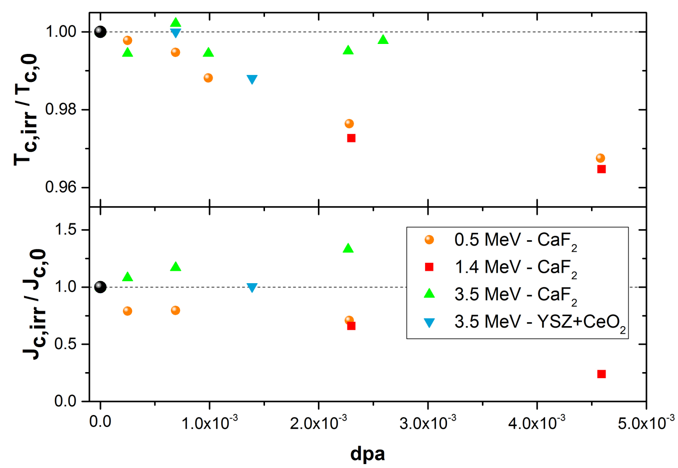

Fig. 2. Critical temperature (defined with the 90% criterion) and critical current modifications (at an applied magnetic field of 8 T) due to irradiation as a function of dpa for different energy and substrate combinations resulting in different implantation depths.

2) Transport Measurements: The electrical transport properties of the micro-bridges were measured in a Physical Properties Measuring System (PPMS) by Quantum Design that goes up to 9 T, and in a Cryogenic Free Measurement System (CFMS) by Cryogenic Ltd up to 16 T. Resistivity measurements were performed by the standard four-probe current-biased measurement technique. Critical temperature was defined by the 90% criterion of the normal state resistivity. Critical current values at different temperatures and magnetic fields were extracted from voltage versus current characteristics by sweeping the current from zero with exponentially increasing steps, with the aim to avoid heating problems. The same sample was always measured in the pristine and irradiated state (also in case of multiple doses) by keeping some of the Hall-bars pristine and exposing the others for different times to the ion beam. This procedure is particularly important to avoid sample-to-sample variability issues: in order to study the effects of radiation damage what is relevant are the relative shifts of superconducting properties with respect to their pristine state rather than their absolute values that can vary from sample to sample (typically $T_c \sim 17.7$ K, $J_c(5K, 0T) \sim 8-14\times 10^5$ A/cm^2 and $J_c(5K, 8T) \sim 2-4\times 10^5$ A/cm^2).

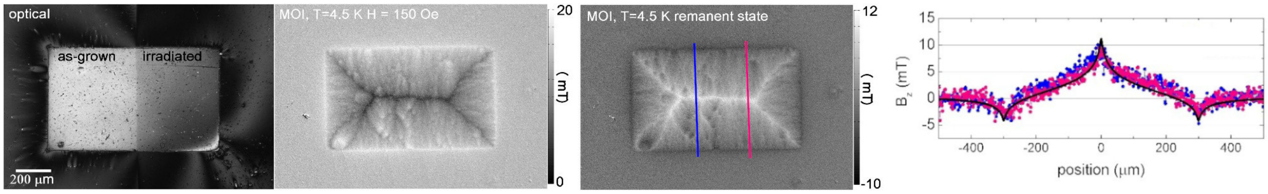

Fig. 3. Optical image of the sample (left), the irradiated part (with 3.5 MeV protons at dpa=1.38×10^{-3}) is clearly identified by the different optical contrast, and MOI quantitative measurements of the magnetic pattern in applied magnetic field and in the remanent state (after a maximum applied field of $\mu_0 H$ = 20 mT) at T = 4.5 K (central panels). The magnetic field profiles in the central region of the remanent state, taken across the irradiated (magenta symbols) and the as-grown (blue symbols) parts of the sample are shown in the right panel, together with the expected curve from the Bean model (black line).

3) Magneto-Optical Analysis: Samples deposited on YSZ were also characterized by means of the Magneto-Optical imaging with an Indicator film (MOI) technique [33], [34] that makes use of the Faraday effect to directly visualize their magnetic field distribution. A nonlinear calibration combined with an iterative algorithm [35], [36] allows obtaining the quantitative measurement of the magnetic field and the reconstruction of the current density distribution (J) [37]. In this case, the rectangular patterns were partially irradiated and partially protected with a suitable screen to keep them pristine. The analysis of such samples ensured that a direct comparison could be carried out without differences in terms of thermal contact or distance to the indicator film, and that the observed modifications can be attributed solely to the introduction of defects by irradiation.

III. Results and Discussion

Fig. 2 shows the modifications of T_c and J_c as a function of dpa for all the energy-substrate combinations, obtained from transport measurements. Despite the scattering of the data, the four datasets clearly show different behaviors, with a better performance when protons are implanted far from the interface with the Fe(Se,Te) film.

Interestingly, the sample deposited on YSZ buffered with Zr-CeO_2 shows no modification of the critical current despite some worsening of the critical temperature (still very limited, $\Delta T_c \sim 0.3$ K for dpa=1.38×10^{-3}). The same result as from the transport measurements was obtained on a twin sample (exposed to the same irradiation conditions) observed with the MOI technique. Fig. 3 shows the optical image of the sample and the low temperature magnetic pattern both with an applied magnetic field of 20 mT, and in the remanent state. The irradiated half of the crystal is clearly visible from the optical image, but no distortion of the magnetic pattern is observed, whereas a modification would be expected if the critical current had been changed by the irradiation. This observation, directly visible from the MOI image, is quantitatively presented in the last panel of Fig. 3 through the magnetic field profiles in the pristine and irradiated central regions of the remanent state: the profiles overlap with each other and with the theoretical curve predicted by the Bean-model. Therefore MOI confirms what was observed by the transport measurements and suggests that the use of a buffer layer might contribute to relax the strain induced by irradiation damage.

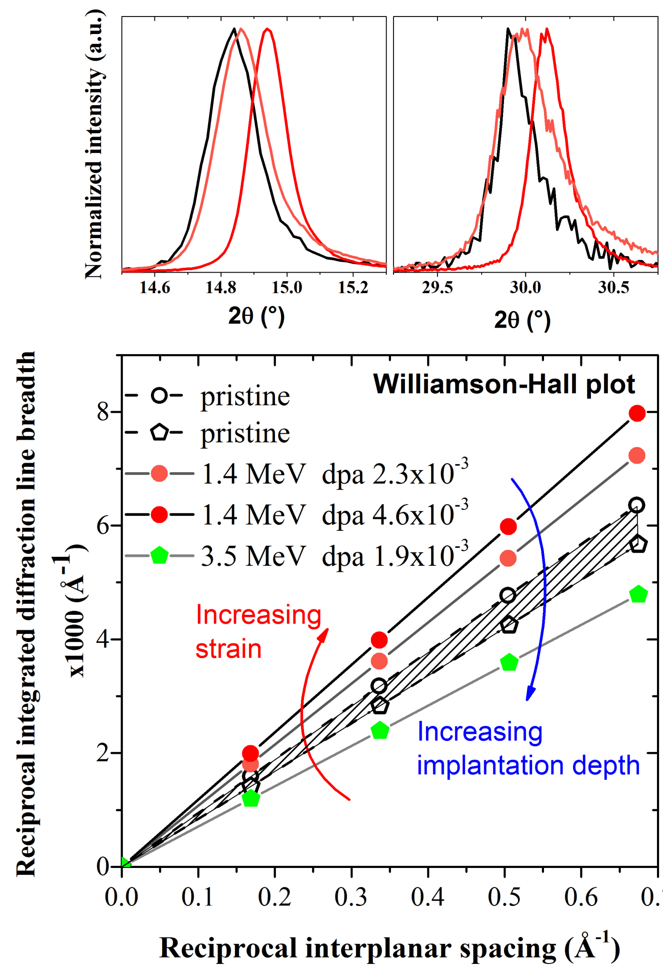

Fig. 4. Williamson–Hall plots (bottom panel) obtained by X-ray diffraction patterns of Fe(Se,Te) samples on CaF_2 before and after irradiation with 1.4 and 3.5 MeV protons. By way of example, modifications of the diffraction peaks after two irradiations doses with 1.4 MeV protons are shown in the top panels for the (001) and (002) reflections (left and right panels, respectively).

The x-ray analysis of the strain induced in the film (Fig. 4) shows that the irradiation process can increase (for shallow implantation) or decrease (deep implantation) the strain in the film, resulting in a worse or better response to the irradiation process of the superconducting material, respectively. It should be noted that the strain is not constant along the thickness of the film due to relaxation. However, since all films have the same thickness and we only consider variations relative to the pristine state, this should not affect the discussion. This is shown in Fig. 5 through the analysis of the critical temperature and critical current modification rate with dpa extracted by fitting the data in Fig. 2 with linear functions with zero intercept.

The results reported here are particularly interesting in light of other results already present in literature. In particular, it has been shown that Fe(Se,Te) thin films have a low anisotropy [38], [39] (a quantity that can be quite critical for applications [40]) that is only slightly affected by irradiation [41], [42]. This, together with the weak field dependence of the critical current, make Fe(Se,Te) coated conductors strong candidates for applications as high-field magnets for radiation harsh environments [43]. In this regard, it is worth comparing the excellent performance of the Fe(Se,Te) samples with that of YBCO films irradiated with 3.5 MeV protons. At a fluence of 1×10^{16}cm^{-2}, corresponding to a dpa value of 2.84×10^{-4}, the critical current of YBCO is reduced to 80% of its pristine value [44]: whereas for the same kind of irradiation Fe(Se,Te) maintains the same value or even increases it slightly.

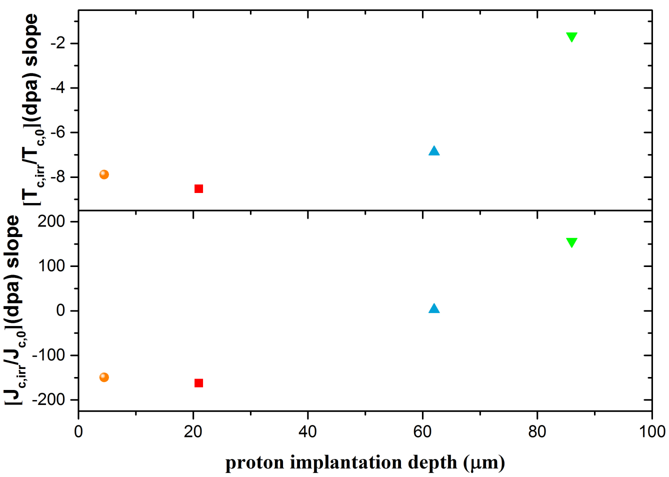

Fig. 5. Critical temperature and critical current modification rate with dpa as a function of proton implantation depth in the substrate.

IV. Conclusions

In summary, we reported on proton irradiation experiments on Fe(Se,Te) thin films deposited on single crystal substrates, finding that the interplay between substrate and film plays a primary role in the response to ion irradiation. The more the damaged substrate induces a strain in the film the more the superconducting properties are affected. The use of a buffer layer, and possibly also of metallic substrates, might help to accommodate the strain and therefore to enhance the radiation hardness of this, already well performing, material. Further systematic studies stemming from these preliminary results are ongoing.

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