Two-Fold Reduction of J_c Anisotropy in FeSe_{0.5}Te_{0.5} Films Using Low-Energy Proton Irradiation

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Abstract—High in-field performance and low anisotropy of critical current density J_c are important for superconducting wires/tapes in rotating machine and magnet applications. Here, we report measurements of angular dependence of critical current density J_c in proton-irradiated FeSe_{0.5} Te_{0.5} (FST) films at magnetic field up to 35 T. We observed two-fold reduction in the J_c anisotropy of FST films at 4.2 K and 15 Tesla magnetic field by irradiation with low-energy (190 keV) proton. In light of recent demonstration of roll-to-roll irradiation process successfully incorporated in the standard 2G HTS long length production wire, low-energy proton irradiation can be a practical solution to enhance the performance of iron-based superconducting wires and tapes, as low-energy ion sources are inexpensive to operate and are readily available commercially.

Index Terms—Critical current density, flux pinning, iron-based superconductors, iron chalcogenide thin films, irradiation.

I. INTRODUCTION

S UPERCONDUCTING wire applications include electricity transmission cables that operate at relatively low fields of a few tenth of a Tesla or less, rotating machines that operate in moderate fields up to 5 T, superconducting magnetic energy storage (SMES) devices or high-field magnets expected to operate at 5 T to several tens of Tesla [1], [2]. Besides high in-field critical current density J_c performance of superconducting wires, low J_c anisotropy are particularly desirable for open bore magnet and rotator/stator in superconducting rotary machine applications, such as generators for wind turbines, hydro power, and marine propulsion.

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Iron-based superconductors have potentials for high-field applications due to their relatively high superconducting transition temperature T_c , high upper critical fields H_{c2} , and relatively low J_c anisotropy [3]–[6]. The high in-field J_c performance could be obtained by the introduction of nano-sized precipitates and defects, which can pin the vortices. Up to now, great advances in the in-field $J_{\rm c}$ performance have been achieved in iron-based superconductors [3], [7]–[9]. The desirable pinning structures could be afforded by ion irradiation, which enable the creation of various defects, such as points, clusters and tracks, by opting appropriate ion species and energy. In iron-chalcogenide superconductors, some different results on the irradiation effect have been reported depending on ion species, irradiation energies, fluences and target phases [9]-[12]. Sylva et al. reported that proton implantation depths were important for superconducting properties in Fe(Se,Te) films: When protons implanted far from the substrate-film interface, J_c enhancement could be observed [13]. On the other hand, when the implantation layer was close to the interface, both T_c and J_c were degraded. Of particular interest reported on Fe(Se,Te) compounds is that an increase of $T_{\rm c}$ was found after electron [11] and neutron [12] irradiation. Recently, we demonstrated a route to simultaneously raise $T_{\rm c}$ and J_c in superconducting FeSe_{0.5} Te_{0.5} (FST) thin films by using low-energy proton irradiation [14]. T_c is enhanced due to the nanoscale compressive strain induced by cascade defects created by the low-energy proton irradiation in FST films. $J_{\rm c}$ is nearly doubled at 4.2 K from self-field to 35 T perpendicular to the film surfaces through strong vortex-pinning by the cascade defects and surrounding nanoscale strain. Effect of low energy proton irradiation on the J_c anisotropy in FST films has not been reported. Here, we present a transport study of angular dependence of J_c in 190 keV proton irradiated FST films at magnetic field up to 35 T. Two-fold reduction was observed in the J_c anisotropy of these films at 4.2 K and 15 T magnetic field.

II. EXPERIMENTAL DETAILS

The FST films (~130 nm thick) are grown on SrTiO₃ singlecrystal substrates with CeO₂ buffer layer by pulsed laser deposition (PLD) method [6], [14]–[16]. The FST films were covered by Al foil with 1.5 μ m thickness and irradiated with 190 keV protons at a dose of 1 × 10¹⁵ p·cm⁻² [14]. The flux was kept around 6 × 10¹² p·cm⁻²·s⁻¹ to avoid sample heating during the irradiation. The proton beam was directed to the film surface at normal incident.

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Fig. 1. (a) Angular field dependence of the critical current density J_c at 4.2 K for the 190 keV proton-irradiated FeSe_{0.5}Te_{0.5} films at 15, 25, and 30 T. (b) Angular field dependence of the normalized critical current density $J_c(\theta)/J_c(90^\circ)$ at 4.2 K for the same films at 15, 25, and 30 T.

Low-field (≤ 9 T) transport measurements were performed by the standard four-probe method in a physical property measurement system (Quantum Design), whereas the high-field measurements were done in 35 T direct current magnet at National High Magnetic Field Laboratory in Tallahassee. Transport J_c was determined using 1 μ V·cm⁻¹ criterion. Irreversibility field H_{irr} and H_{c2} was determined using $0.01\rho_n$ and $0.9\rho_n$ criteria, respectively, where ρ_n is the normal state resistivity at the transition temperature. The applied magnetic field was rotated at an angle from the crystallographic *c*-axis of the crystalline film in a maximum Lorentz force configuration, corresponding to the current flowing perpendicular to the applied magnetic field.

III. RESULTS AND DISCUSSION

The angular dependence of J_c gives us important insight into the pinning effectiveness of the superconducting films. We show $J_c(\theta)$ for the FST films irradiated with $1 \times 10^{15} \text{ p}\cdot\text{cm}^{-2}$ dose of 190 keV proton beam under 15, 25, and 30 T at 4.2 K in Fig. 1(a). The irradiated film is capable of carrying high J_c exceeding $10^5 \text{ A}\cdot\text{cm}^{-2}$ for all orientations even at 30 T. Fig. 1(b) shows the angular dependence of normalized J_c ($J_c(\theta)/J_c(90^\circ)$). The normalized J_c exhibits a broad maximum at H//ab and no prominent



Fig. 2. Angular field dependence of (a) the critical current density J_c . (b) The normalized critical current density $J_c(\theta)/J_c(90^\circ)$ for the pristine and the proton-irradiated FeSe_{0.5} Te_{0.5} films at a dose of $1 \times 10^{15} \text{ p} \cdot \text{cm}^{-2}$ at 4.2 K and 15 T.

 J_c peak at H//c. Small J_c anisotropies, $\gamma_{Jc} (J_c^{H//ab}/J_c^{H//c})$, of 1.9, 2.8, and 2.9 were observed at 15, 25, and 30 T, respectively. We found that the γ_{Jc} increases with increasing magnetic field.

In order to compare the γ_{Jc} in the irradiated film with the one in the pristine film under high magnetic field, the angular dependence of J_c and normalized J_c ($J_c(\theta)/J_c(90^\circ)$) at 4.2 K and 15 T for both the pristine and the proton-irradiated FST films are shown in Figs. 2(a) and 2(b), respectively. Two-fold reduction in the J_c anisotropy of FST films at 4.2 K and 15 T was achieved by 190 keV proton irradiation with a dose of $1 \times 10^{15} \text{ p} \cdot \text{cm}^{-2}$. Through the TEM characterization, we found that the cascade defects and nanoscale strain modulations were produced in FST films irradiated with low-energy proton irradiation, which could provide the reduction of J_c anisotropy [14]. These defects and the extension of strain field that provides pinning potential to vortex are found to be much stronger in the *ab*-plane, that inhibits the in-plane motion of vortices and thus leads to strong enhancement of J_c^c for magnetic field applied along the *c*-axis. In contrast, the strain field are found much weaker across the plane leading to less enhancement of J_c^{ab} for magnetic field applied along the *ab*-plane. This may explain the reduction of J_c anisotropy, defined as J_c^{ab}/J_c^c , in low-energy proton irradiated films. We also found that the γ_{Jc} in the pristine FST film increase by a factor of \sim 3.5 in increasing magnetic field from 5 T to 15 T



Fig. 3. Temperature dependence of (a) the upper critical field H_{c2} , compare with FeSe_{0.4} Te_{0.6} single crystal [17] and (b) the irreversibility field H_{irr} for the pristine and the irradiated FeSe_{0.5} Te_{0.5} films with 190 keV protons at a dose of $1 \times 10^{15} \text{ p} \cdot \text{cm}^{-2}$.

 $(\gamma_{Jc} = 1.10 \text{ for the pristine FST film at 5 T [16]})$, whereas the γ_{Jc} in the proton-irradiated FST film at 15 T increases only by a factor of ~1.7 compared with the γ_{Jc} (= 1.10) in the pristine FST film at 5 T. This is indicative of the trend toward less anisotropic contribution to vortex pinning, as low-energy proton irradiation produces cascade defects and the surrounding strain field [14]. In comparison, MeV range gold ion irradiation that produces nearly isotropic cluster defects appears to be even more effective in the reduction of γ_{Jc} which was previously reported to result in $\gamma_{Jc} = 1.10$ at 5 T [16].

Figs. 3(a) and 3(b) present temperature dependence of H_{c2} and $H_{\rm irr}$ for the pristine and the irradiated FST films, respectively, together with the data for FeSe_{0.4}Te_{0.6} single crystal [17] for comparison. Our data on films were taken from the temperature and magnetic field dependence of the measured resistivity. H_{c2} curves exhibit downward curvature in all case and very steep slopes near T_c , which is consistent with previous reports [18], [19]. Similar to the behavior in FeSe_{0.4}Te_{0.6} single crystals [17], large H_{c2} anisotropies, $\gamma_{Hc2} (J_c^{H//ab}/J_c^{H//c})$, of \sim 3.5 was observed near T_c in the pristine FST film, and γ_{Hc2} decreases with decreasing temperature ($\gamma_{Hc2} = 1.2$ at 15 K). This indicates that the H_{c2} becomes more isotropic as temperature decreases in our irradiated films. Upon proton irradiation, an increase of H_{c2} for H//c was observed. In the case of H_{irr} , $H_{\rm irr}$ in all temperatures goes up slower than $H_{\rm c2}$ with decreasing temperature.

IV. CONCLUSION

We present measurements of angular dependence of J_c in pristine and proton-irradiated FST films at magnetic field up to 30 T. Upon a low-energy (190 keV) proton irradiation with a dose of 1×10^{15} p·cm⁻², two-fold reduction in the J_c anisotropy γ_{Jc} of FST films was achieved under 15 T at 4.2 K. It is found that the γ_{Jc} . increases from 1.9 at 15 T to 2.9 at 30 T. These results indicate that proton irradiation is effective in providing less anisotropic pinning centers in iron-based superconducting films.

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