



Corrigendum: Enhanced critical current in superconducting $\text{FeSe}_{0.5}\text{Te}_{0.5}$ films at all magnetic field orientations by scalable gold ion irradiation (2018 *Supercond. Sci. Technol.* **31** 024002)

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Enhanced critical current in superconducting $\text{FeSe}_{0.5}\text{Te}_{0.5}$ films at all magnetic field orientations by scalable gold ion irradiation

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Abstract

The loss-less electrical current-carrying capability of type II superconductors, measured by the critical current density J_c , can be increased by engineering desirable defects in superconductors to pin the magnetic vortices. Here, we demonstrate that such desirable defects can be created in superconducting $\text{FeSe}_{0.5}\text{Te}_{0.5}$ films by 6 MeV Au-ions irradiations that produce cluster-like defects with sizes of 10–15 nm over the entire film. The pristine $\text{FeSe}_{0.5}\text{Te}_{0.5}$ film exhibits a low anisotropy in the angular dependence of J_c . A clear improvement in the J_c is observed upon Au-ion irradiation for all field orientations at 4.2 K. Furthermore, a nearly 70% increase in J_c is observed at a magnetic field of 9 T applied parallel to the crystallographic c -axis at 10 K with little reduction of the superconducting transition temperature T_c . Our studies show that a dose of 1×10^{12} Au cm⁻² irradiation at a few MeV is sufficient in order to provide a strong isotropic pinning defect landscape in iron-based superconducting films.

Keywords: iron-based superconductor, thin film, irradiation, critical temperature, critical current density, vortex pinning

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

1. Introduction

The critical temperature, T_c , critical current density, J_c , and upper critical field, H_{c2} , of superconducting materials determine the limits of a superconductor in energy applications. The current-carrying capability, measured by J_c , reflects the ability of defects in superconductors to pin magnetic vortices. Defects create local regions with depressed pairing potential, and tend to preclude the Cooper pair formation, and hence drive down the T_c . This is well known in cuprate (Cu–O based) high- T_c superconductors (HTS) as well as iron-based superconductors that have short coherence lengths ξ . Improvements in J_c have been obtained in these materials by

introducing artificially pinning defects, such as precipitate and columnar defects, generally at the cost of T_c degradation [1–10].

Iron-based superconductors hold a great promise for high-field applications due to their relatively high T_c 's and high H_{c2} 's, together with relatively low mass anisotropies γ [11–13]. Recently, we demonstrated a route to raise T_c and J_c concomitantly in iron-based superconductors by using low-energy proton irradiation, in which we created small cascade defects in $\text{FeSe}_{0.5}\text{Te}_{0.5}$ films. The proton irradiation resulted in an enhanced T_c due to nanoscale compressive strain and the proximity effect, while J_c nearly doubled at zero magnetic field at 4.2 K due to an increased vortex pinning by cascade

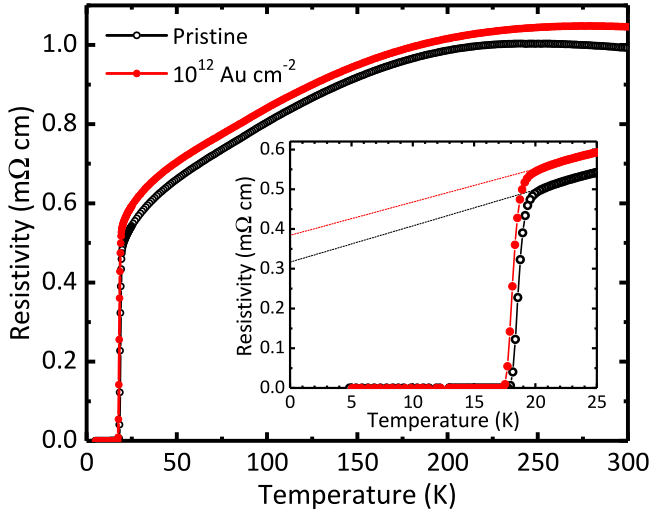


Figure 1. Temperature dependences of electrical resistivity for the $\text{FeSe}_{0.5}\text{Te}_{0.5}$ film before and after 6 MeV Au-ion irradiation with $1 \times 10^{12} \text{ Au cm}^{-2}$ dose. Inset presents magnified temperature region near T_c .

defects and surrounding nanoscale strain [14]. To broadly implement this strategy with other ion species, several questions need to be answered. What are the structural characteristics of the defects in iron-based superconductors created by other irradiation ion species? Higher energy heavy ions are required in order to penetrate the entire sample, likely resulting in large size defects. Large defects are expected to have different morphologies from those produced by proton irradiation. This raises questions as to: (i) whether those defects are effective in pinning vortices in the iron-based systems, and (ii) the T_c dependence on defect morphology and landscape. Iron-based superconductors have lower T_c 's than the cuprate HTS. Hence, too much of T_c reduction will make them less attractive for applications. We performed Au-ion irradiation studies in the iron-based superconductors, aimed at elucidating the relationship between heavy-ion irradiation-induced defect structure and the two critical parameters— T_c and J_c .

Recent reports show that low-energy (1–20 MeV) ion irradiation delivers strong J_c enhancement in high- T_c superconducting wires [15–17]. Low-energy ion beam (\sim several MeV) can produce vacancy-interstitial type of defects, including Frenkel pairs and their clusters along cascades. Energy loss due to ionization on the passage through target materials at such low-energies would be low so that local heating could be minimized. Also comparing light ions with Heavy ions, Heavier ions, such as Ag and Au appear to be more suitable for defect generation. Research on single-crystal iron-based superconductors reveals that ion-irradiated iron-based superconductors retain J_c enhancement up to higher dosage compared with ion-irradiated cuprate HTS, indicative of strong tolerance to artificially introduced defects [18–20].

In this paper we describe the isotropic pinning enhancement achieved in $\text{FeSe}_{0.5}\text{Te}_{0.5}$ (FST) films irradiated with 6 MeV Au-ions with $1 \times 10^{12} \text{ Au cm}^{-2}$. This low dosage is

three orders of magnitude lower than that for proton irradiation ($1 \times 10^{15} \text{ p cm}^{-2}$). Such a drastic reduction in dosage is particularly important for mass production, such as the Reel-to-Reel irradiation process [17], as much less time is needed for irradiating production scale superconducting tapes that reduces cost greatly. We believe that irradiation with several MeV Au-ions is a viable option for creating isotropic and strong pinning centers.

2. Experimental details

We grew 100–130 nm thick FST films with a CeO_2 buffer layer on SrTiO_3 single-crystal substrates by pulsed laser deposition (PLD) method [4, 5, 14]. The FST films were characterized before and after irradiation with the same film to eliminate the influence of film variations. Transport measurements were carried out using the conventional four-probe method in a physical property measurement system (Quantum Design). The electric contacts were not replaced in the process. J_c was determined using $1 \mu\text{V cm}^{-1}$ criterion. T_c^{zero} was determined using $0.01\rho_n$ criteria, where ρ_n means the normal-state resistivity at the transition temperature. The magnetic field was rotated at an angle from the crystallographic c -axis in a maximum Lorentz force configuration, i.e. the current flowing perpendicular to the applied magnetic field. The FST films characterized before the irradiation were glued on a metal plate with silver paste. The FST films were irradiated at fluence $\phi = 1 \times 10^{12} \text{ ion cm}^{-2}$ with 6 MeV Au^{3+} ions in vacuum at room temperature using the 15 MV tandem Van de Graaff accelerator of Brookhaven National Laboratory. The beam was directed to the film surface at normal incidence. The flux was kept around $2 \times 10^{10} \text{ ion cm}^{-2} \text{ s}^{-1}$ to dissipate sample heating, corresponding to a beam current density of $\sim 34 \text{ nA cm}^{-2}$. Microstructures of the films were characterized using high-resolution transmission electron microscopy (HRTEM).

3. Results and discussion

Figure 1 shows the temperature dependence of the electrical resistivity before and after irradiation with Au-ions. The FST films before and after irradiation exhibit metallic behavior below 200 K. We have found that Au-ion irradiation slightly suppresses T_c by about 0.5 K, from 17.9 to 17.4 K, without exhibiting transition broadening. This behavior is almost the same as the one reported in a BaFe_2As_2 film irradiated with $1 \times 10^{16} \text{ p cm}^{-2}$ dose of 3 MeV protons [21]. The simulation using the stopping range of ions in matter software package [22] predicts that the 6 MeV Au-ions to a doses of $1 \times 10^{12} \text{ Au cm}^{-2}$ could produce 6.42×10^{-3} displacements per atom (dpa), which is higher by a factor of ~ 6 for the FST film with $1 \times 10^{15} \text{ p cm}^{-2}$ dose of 190 keV proton [14]. Prior work on irradiation of $\text{YBa}_2\text{Cu}_3\text{O}_y$ (YBCO) thin films has shown that significant T_c degradation begins to occur at about $2 \times 10^{-3} \text{ dpa}$ [23]. Figure 1 also provides further insight into the created defects. The normal-state resistivity shows nearly upwards parallel-shift upon irradiation, indicating that Matthiessen's rule is well verified. The irradiated

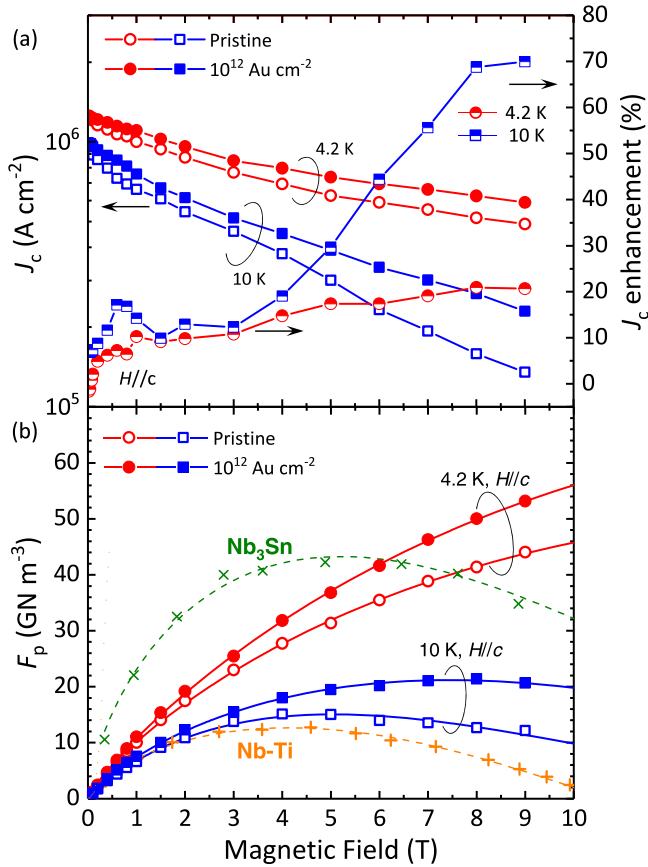


Figure 2. (a) Magnetic field dependence of J_c at 4.2 and 10 K of the FeSe_{0.5}Te_{0.5} (FST) film in the pristine state and following irradiation with 6 MeV Au-ion to a dose 1×10^{12} Au cm⁻² in $H//c$ direction. Magnetic field dependence of J_c enhancement for the FST film at 4.2 and 10 K is shown on the right y-axis. (b) Corresponding magnetic field dependence of vortex pinning force F_p for the FST films, compared with the data of Nb-Ti^{27,28} and Nb₃Sn^{29,30} wires.

FST film does not show low- T upturns of resistivity which is associated with a Kondo-like spin-flip scattering. This indicates that the irradiation with 6 MeV Au-ions produces nonmagnetic defects. Similar behavior has been reported in Ba(Fe_{1-x}Co_x)₂As₂ [24] and Ba_{1-x}K_xFe₂As₂ [25] irradiated with protons, which is in clear contrast to the behavior in α -particle irradiated NdFeAs (OF) [26]. Even though we found no low- T upturns of resistivity, the parallel-up-shift in the normal-state resistivity can lead to the increase of residual resistivity ρ_0 by $\sim 21\%$ ($\Delta\rho_0 = \rho_0^{\text{irr}} - \rho_0^{\text{unirr}}$). The ρ_0 value was estimated as a zero-temperature intercept of the linear extrapolation of $\rho(T)$ from above T_c to $T = 0$, as shown in the inset of figure 1. The increase of ρ_0 would correspond to the density of defects produced by the Au-ion irradiation.

Figure 2(a) presents the magnetic field dependence of transport critical current density J_c with $H//c$ for the sample before and after irradiation with 6 MeV Au-ions to a dose of 1×10^{12} Au cm⁻² at 4.2 and 10 K. The self-field J_c values are almost the same for FST films before and after irradiation at both 4.2 and 10 K. However, as the magnetic field increases, the enhancing effect of the irradiation is clearly visible, especially at 10 K. Figure 2(a) also shows the J_c

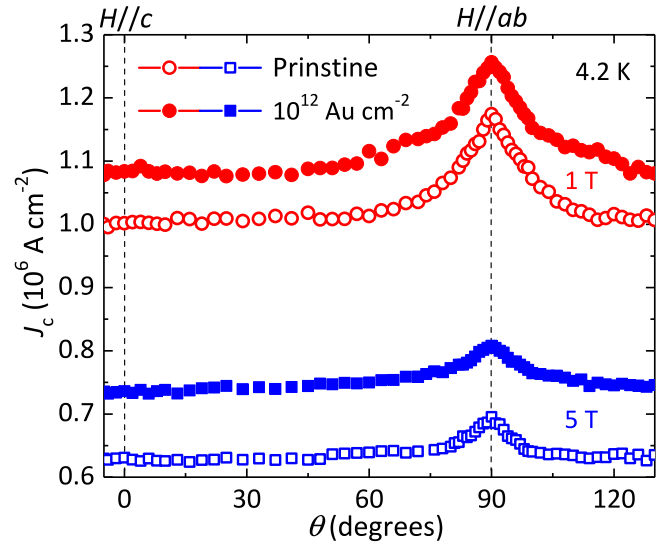


Figure 3. Angular field dependence of the critical current density at 4.2 K for the FeSe_{0.5}Te_{0.5} films before and after Au-ion irradiation at 1 and 5 T.

enhancement, $(J_c^{\text{after}} - J_c^{\text{before}})/J_c^{\text{before}}$, for the irradiated sample relative to the sample prior to the Au-ion irradiation for 4.2 and 10 K. At 4.2 K, the J_c enhancement shows gradual increase with increasing magnetic field ($\sim 20\%$ increase at 9 T). At 10 K, the Au-ion irradiation yields up to 70% J_c enhancement at 9 T. These behaviors are considerably different from those reported by Tamegai *et al* [18], who demonstrated J_c increase at low fields, but reduced J_c at high fields after 200 MeV Au-ion irradiation of FeSe_{0.39}Te_{0.61} single crystals. The above authors have claimed that it is difficult to appreciate the effect of heavy-ion irradiation into FeSe_xTe_{1-x} crystals quantitatively, since the superconducting properties of the crystals are sensitive to the annealing process which could eliminate the influence of excess Fe at the interstitial sites of the Te/Se layer in FeSe_xTe_{1-x} crystals. In our films grown via the PLD method, however, no excess Fe has been observed in HRTEM and scanning TEM with high-angle annular dark field detector characterization [14], resulting in more quantitative comparison of the effects of Au-ion irradiation.

In order to assess the effects of Au-ion irradiation into FST films on pinning properties, we plot the pinning force F_p ($=J_c \times \mu_0 H$), where μ_0 is the permeability in vacuum, with $H//c$ for the sample before and after the irradiation at 4.2 and 10 K, together with the literature data for thermo-mechanically processed Nb-Ti [27, 28] and small grain Nb₃Sn [29, 30] wires at 4.2 K in figure 2(b). Solid lines and dashed lines present the fitting curve using $F_p = A(H/H_{\text{irr}})^p(1 - H/H_{\text{irr}})^q$ with four free parameters (p, q, A, H_{irr}) [31]. The F_p values of the FST films at both 4.2 K and 10 K are higher than those of Nb-Ti wires at 4.2 K. At 10 K, the pristine film exhibits F_p maximum (F_p^{max}) at 4 T, whereas the irradiated film has a maximum at 8 T. Moreover, the (F_p^{max}) increases from ~ 15 GN m⁻³ in the pristine film to ~ 21 GN m⁻³ in the irradiated film. The irradiation with 6 MeV Au-ions yields both a broader F_p peak width and

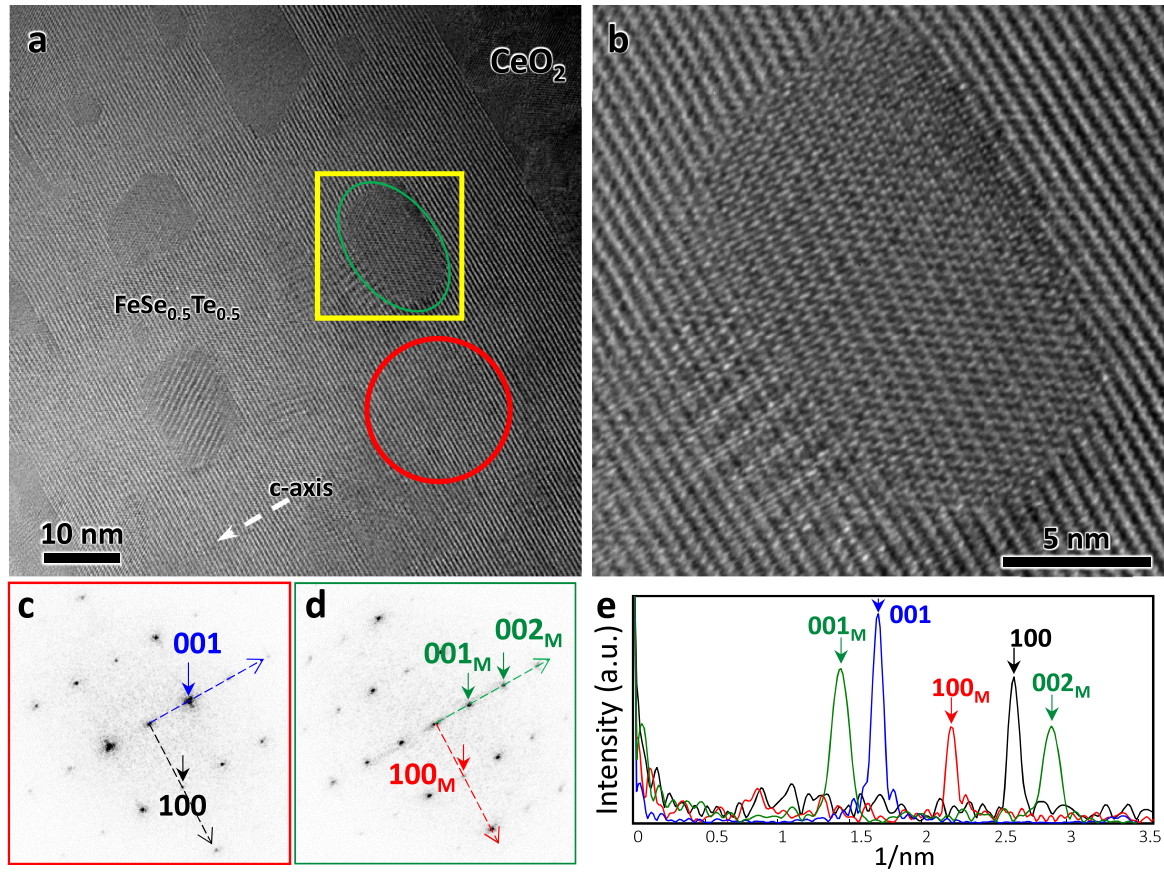


Figure 4. (a) High resolution TEM image of Au-ion irradiated $\text{FeSe}_{0.5}\text{Te}_{0.5}$ (FST) film. (b) Magnified image from the area marked by yellow square in (a), showing structure has been modified by Au-ion irradiation (denoted as M). (c), (d) Diffraction patterns from (c) the red circle (unmodified area) and (d) the green ellipse (modified area) area in (a). (e) Intensity profiles from the scan line along [100] and [001] in the diffraction patterns. The modified structure has the similar lattice to the FST, but larger lattice parameters with $a \sim 0.45$ nm and $c \sim 0.69$ nm.

enhancement of (F_p^{\max}). A similar trend was observed in a FST film irradiated with 1×10^{15} p cm $^{-2}$ dose of 190 keV proton beam [14].

The angular dependence J_c gives us further insight into the pinning effectiveness of the irradiated FST films. We show $J_c(\theta)$ for the FST films before and after irradiation under 1 and 5 T at 4.2 K in figure 3. The pristine film has an isotropic J_c angular dependence up to 5 T. J_c exhibits a broad maximum $H//ab$ and no prominent J_c peak at $H//c$. Very small J_c -anisotropies, $\gamma_{J_c}(J_c^{H//ab}/J_c^{H//c})$, of 1.17 and 1.10 are observed at 1 T, and 5 T, respectively. These values are much smaller than the corresponding values of $\text{FeSe}_{1-x}\text{Te}_x$ films grown on Fe-buffered MgO substrates ($\gamma_{J_c} = 2.6$ [32]). Upon irradiation with 6 MeV Au-ions, the J_c increases for all orientations, retaining small γ_{J_c} 's of 1.15 and 1.10 at 1 T and 5 T, respectively, indicating that the vortex pinning centers are isotropic and randomly distributed.

In order to understand the origin of these pinning properties, we performed extensive HRTEM analysis. Figure 4(a) displays a representative HRTEM image of the FST film irradiated with 6 MeV Au-ions to doses of 1×10^{12} Au cm $^{-2}$. The above micrograph clearly shows evidence for the formation of different microstructures from that in the pristine film and in the 190 keV proton-irradiated FST film [14]. The layered structure of FST is clearly visible in the irradiated FST film. Atomic-scale analysis with HRTEM reveals the presence

of cluster-like defects modified by the Au-ion irradiation over the entire film. The higher magnification image (figure 4(b)) from the area marked with a yellow square shows that the Au-ion irradiation produces 10–15 nm-sized defect clusters. Figures 4(c) and (d) show the diffraction pattern from the red circle (unmodified area) and the green ellipse (modified area), respectively, while figure 4(e) shows the intensity line profile along the [001] and [100] crystallographic axes in the diffraction patterns. The full width at half the maximum (FWHM) are about 0.15 and 0.11 nm for 001 M/002 M and 100 M obtained from figure 4(e), respectively. This FWHM difference is consistent with the difference in the particle size between [001] and [100] direction, e.g. the size is 9.7 nm along [001] and 15.7 nm along [100] direction, respectively, measured from the image in figure 4(b). The shape of the most defects is anisotropic with the size along [100] direction larger than that along [001] direction. The modified structure has a similar lattice to the FST. However, larger lattice parameters, $a \sim 0.45$ nm and $c \sim 0.69$ nm, are obtained from these diffraction patterns. These defects are different from columnar defects with a diameter of about 3 nm and clusters of point-like defects in $\text{FeSe}_{0.45}\text{Te}_{0.55}$ single crystals irradiated with 249 MeV Au-ions with a beam current density of 0.36 nA cm $^{-2}$ [33], and point-like defects smaller than 6 nm in diameter produced by 3 MeV Au-ion irradiation in YBCO films [15]. These defects of various

shapes and sizes produced by Au-ion irradiation may arise from the difference between the ion energy and the beam current. The irradiation with 6 MeV Au-ions of FST films may be responsible for the formation of cluster-like defects with sizes of 10–15 nm, much larger than coherence length ξ for FeSe_{0.5}Te_{0.5} single crystals [34, 35], giving rise to strong isotropic vortex pinning, as shown in figure 3. This indicates that material defects with a size much larger than the coherence length ξ could be effective pinning centers. A similar result has been reported in GdBa₂Cu₃O_y thin films with gold nanorods with average diameter of 34 nm [36]. Kwok *et al* shows that a mixed-pinning landscape composed of defect inclusions with different sizes may be beneficial for enhancement of vortex pinning [37]. We have proposed a route for creating the cascade defect and surrounding nanoscale strain by low-energy proton irradiation in the FST films [14]. Thus, a pinning landscape by way of combined proton and Au-ion irradiation would enable the creation of tailored defects for even higher J_c 's under applied magnetic fields.

4. Conclusions

In summary, Au-ions with 6 MeV to a dose of 1×10^{12} Au cm⁻² have been irradiated into FST films. The irradiation of the FST film is found to result in the formation of structures, which serve as strong vortex pinning sites, as evidenced in the nearly 70% increase of J_c at 9 T// c and 10 K with little subsequent reduction in the critical temperature, T_c . Nonmagnetic defects produced via 6 MeV Au-ion irradiation yields the increase of J_c for all directions with respect to the J_c for the pristine film along all crystallographic directions. This increase in the critical current density is indicative of the isotropic defect contribution to pinning. The cross-sectional HRTEM image and the diffractogram point out that the irradiation produces a moderate density of isotropic cluster-like defects with sizes on the order of 10–15 nm. These results indicate that MeV Au-ions irradiation at low dosage opens up a new avenue for engineering strong isotropic pinning centers in iron-based superconducting films at a much reduced cost.

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