Artificially Engineered Nanostrain in Iron Chalcogenide Superconductor Thin Film for Enhancing Supercurrent

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KEYWORDS

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ABSTRACT

Although nanoscale deformation, such as nanostrain in iron chalcogenide (FeSe\textsubscript{x}Te\textsubscript{1-x}, FST) thin films, has drawn attention due to the enhancement of general superconducting properties such as critical current density ($J_c$) and critical transition temperature, its formation has proven to be a very challenging and complex process thus far. Herein, we successfully fabricated an epitaxial multilayer FST thin film with uniformly distributed nanostrain via the injection of a trace of CeO\textsubscript{2} between each FST layer using sequential pulsed laser deposition. Using transmission electron microscopy and geometrical phase analysis, we verified that a trace of CeO\textsubscript{2} injection forms nanoscale fine defects with nanostrained region, which has a tensile strain ($\varepsilon_{zz} \approx 0.05$) for the c-axis of the FST matrix. The CeO\textsubscript{2} injected FST thin film achieves a remarkable $J_c$ of 3.6 MA/cm\textsuperscript{2} for a self-field and 90% enhanced $J_c$ under 13 T with respect to a pristine FST thin film.
INTRODUCTION

Superconductors are essential materials for high magnetic field applications such as in nuclear fusion energy devices, magnetic resonance imaging (MRI), and superconducting magnetic energy storage systems. In high magnetic field applications, the field penetrates into the superconductor in the form of quantized magnetic fluxes, frequently referred to as vortices, that move under the influence of a Lorentz force when a current is applied if not properly pinned. The moving vortices deteriorates superconducting properties such as critical current density ($J_c$) and it is therefore necessary to minimize vortices motion. To achieve this objective, artificial defects acting as pinning centers can be introduced into the prospective sample.\textsuperscript{1-2} However, controlling the pinning center formation inside at superconductor is challenging and can also result in deterioration of the critical transition temperature ($T_c$). Thus, the formation of pinning centers requires precise optimization in order to minimize the degradation of the crystallinity, to improve $J_c$ while minimizing or avoiding the decrease of $T_c$.

In recent years, iron-based superconductors (FeSCs) have attracted attention for high magnetic field applications because of their high upper critical field ($H_{c2}$) and low magnetic anisotropy ($\gamma$)\textsuperscript{2-3}. Besides, the FeSCs epitaxial thin films have demonstrated enhanced overall superconducting properties compared to correspondent bulk materials\textsuperscript{4-12}. Among several FeSCs iron chalcogenides, (FeSe\textsubscript{x}Te\textsubscript{1-x}, FST) which has a simple PbO type and layered-like structures\textsuperscript{13} is an excellent candidate for use as a practical superconducting material for several reasons. Firstly, the $T_c$ of FST abruptly increases due to an increasing Se ratio with the suppression of phase separation, which is generally observed in Se rich FST bulk, when FST is fabricated as an epitaxial thin film\textsuperscript{14-16}. In addition, the FeSe monolayer can achieve a $T_c$ of 100 K, which is the maximum $T_c$ for FeSCs.\textsuperscript{17} Secondly, FST thin films exhibit a promising $J_c$ over 1 MA/cm$^2$ at self-field regardless of the
substrate including coated conductor substrate $^{6,18-23}$. This indicates that these films can potentially be used as a superconductor tape. However, significant $J_c$ enhancement via an artificial pinning center is a critical requirement in order for FST to be used in high magnetic field applications.

Several approaches have been used to improve $J_c$ in FST to date such as using a buffer layer$^{18}$, oxygen annealing$^{24}$, and ion irradiation$^{19,25-26}$. In particular, low energy proton irradiation (190 KeV) is a very effective method for this purpose because it causes a nanoscale cascade defect accompanied by nanostrain which enhances both $T_c$ and $J_c$ simultaneously in FST thin film$^{19}$. However, proton irradiation is a complicated and ex-situ process which is not suitable for practical application. Therefore, an in-situ and straightforward process for forming artificially controlled nanostrains is necessary to improve $J_c$ in FSTs.

Sequential pulsed laser deposition (S-PLD) is a an appropriate technique for artificially forming nanoscale pinning centers during the growth of epitaxial FST thin films as an in-situ process. To date, S-PLD has been widely used to fabricate superlattice structures, secondary phases, and uniformly dispersed defects in superconductor thin films through artificial insertion or doping of the desired material while fabricating an epitaxial thin film$^{5,27-30}$. The ideal case is to form minimal nanoscale defects using S-PLD for inducing nanostrain in FST thin films thereby improving $J_c$ to minimize crystallinity degradation. In particular, minimal nanoscale fine defects and nanostrain can prevent Cooper pair breaking by a short coherence length (~2 nm) of FST$^{31}$. However, because the injected material can also form sub-microscale defects and layers, selection of the appropriate material to be injected is very important.

Among a diverse list of compounds or atoms as an insertion material, cerium oxide (CeO$_2$) is the most promising candidate. So far, CeO$_2$ has been used as a buffer layer$^{18,32}$, capping layer$^{32}$, and interlayer$^{33}$, to improve crystalline quality and to enhance $J_c$ in FST thin films due to its
excellent chemical stability with these films. In addition, the appropriate difference of the lattice constants between FST and CeO$_2$ can minimize the deterioration of the crystallinity with the application of fine strain. However, inserting oxide materials periodically in FeSCs has proven to be significantly challenging because the range of optimized condition for inserting oxide materials may be very narrow, so many factors should be taken into consideration$^{5,34-35}$.

Herein, we report that nanostrain with nanoscale pinning centers was successfully formed in an epitaxial FST thin film through the injection of precisely controlled trace amounts of CeO$_2$ using S-PLD. We confirm the crystallinity and structure for CeO$_2$ injected FST (Ce-FST) using X-ray diffraction (XRD) measurement, and the nanostrain was verified using an atomic resolved scanning transmission electron microscopy (STEM) with geometrical phase analysis (GPA). The Ce-FST thin film with nanostrain shows significantly enhanced $J_c$ compared to that of pristine FST (P-FST) thin film.

EXPERIMENTAL DETAILS

Sample preparation

The Ce-FST and the P-FST thin films were grown on a (001)-oriented CaF$_2$ substrate by PLD using a KrF (248 nm) excimer laser (Coherent, COMPEX PRO 205F) in a vacuum of $2 \times 10^{-5}$ Pa at 400 °C. We used a FeSe$_{0.45}$Te$_{0.55}$ target, which was made by an induction melting method. Both FST thin films were grown using a laser energy density of 3 J/cm$^2$, a repetition rate of 3 Hz, and target to substrate distance of 4 cm. The thickness of each FST layer was 20 nm (445 laser pulses) in the Ce-FST thin film, and the total thickness of both FST thin films was 100 nm (2225 layer pulses). CeO$_2$ was deposited between 20 nm FST layers in a Ce-FST thin film. Only two laser pulses of CeO$_2$ were deposited at each FST layer, corresponding to less than a half unit cell under
our growth conditions (laser energy density of 1.5 J/cm² and a repetition rate of 1 Hz). The target changing time between the FST and CeO₂ targets was 10 seconds which is the drive time when the laser is turned off and then on again.

Characterization

To characterize the crystal structure, we performed a θ-2θ scan at the beamline 3A in the Pohang Accelerator Laboratory with a six circle XRD (λ = 1.148 Å). The azimuthal phi scan and rocking curve were measured using a four-circle XRD (PANalytical, X’Pert pro, λ = 1.5406 Å). STEM images and EDS were obtained in a Cs-corrected FEI Titan Themis G2 at an accelerating voltage of 300 KV with a beam current of 70 pA, a convergence semi-angle of 15 mrad, and a collection semi-angle snap in the range of 80-379 mrad. The resistivity-temperature measurement was performed using a physical property measurement system (PPMS, Quantum Design). \( T_c^{\text{onset}} \) and \( T_c^{\text{zero}} \) were determined using the 0.9 \( \rho_n \) criteria and 0.01 \( \rho_n \) criteria, respectively, where \( \rho_n \) is \( \rho(23 \text{ K}) \). Magnetization \( J_c \) was measured using a vibrating sample magnetometer (VSM, Oxford) by applying a magnetic field perpendicular to the film. It was estimated using a Bean model for thin film: \( J_c = 15 \Delta M/Vr \), where \( V \) is the thin film volume in cubic centimeter, \( r \) is the equivalent radius of the sample size (\( \pi r^2 = a \times b \); \( a \) and \( b \) are width and length of the sample), and \( \Delta M \) is the width of magnetic moment from the M-H loop.
RESULTS AND DISCUSSIONS

We first fabricated the Ce-FST thin films to form nanostrain inside FST (Figure 1). To date, nanostrain has been generated via the introduction of various defect formations. For example, the insertion of a desired material with a slightly different lattice constant can induce strain through the formation of a secondary phase \(^{36-37}\), and the doping of certain elements which generate lattice change with nanoscale strain\(^{38}\). Additionally, a formation of cascade\(^{19}\) or point defect\(^{39}\) by ion irradiation also induces deformation of a lattice with a nanoscale defect. However, because large-scale and excessive amounts of defects can degrade the entire superconducting matrix in FST thin films\(^{25,40}\), it is critical to form nanoscale defects to induce nanostrain at a minimum extent which does not deteriorate the superconducting state. Thus, we only inserted infinitesimal CeO\(_2\) (two laser pulses, below 0.5 unit cell) such that the formation of nucleation clusters or layers at the 20 nm interval between the FST layers was minimized.

To identify the out-of-plane crystalline quality of the Ce-FST thin film, we performed a \(\theta-2\theta\) scan using a Synchrotron-based XRD. Figure 2a shows an out-of-plane \(\theta-2\theta\) XRD pattern of the Ce-FST thin film on CaF\(_2\) (001). The \(\theta-2\theta\) scan clearly shows only (00\(l\)) peaks for FST thin film without any other phase peaks despite the periodically injected CeO\(_2\). Figure 2b represents an enlarged section of Figure 2a close to the (001) reflection of the Ce-FST thin film. As shown in Figure 2b, the Ce-FST thin film shows satellite peaks which have been generally observed in superlattice thin film\(^5\). However, it is difficult to form an intact CeO\(_2\) layer because a trace of CeO\(_2\) was injected at the interface between the 20 nm FST layers. Thus, the structure of the Ce-FST is similar to a superlattice but substantially different. We speculated that the satellite peaks are due to very small changes such as the nanostrain at the interface between the FST layers, as a result of the dispersion of an inappreciable amount of CeO\(_2\). When we calculated the thickness of each FST
layer using the satellite peak position as shown in Figure 2b, the FST layer thickness was determined to be 19.7 nm, which is consistent with the expected thickness of one FST layer (20 nm). Additionally, we measured the rocking curve for the (001) reflection of both P-FST and Ce-FST thin films using four circles XRD to compare the out-of-plane crystalline quality and the mosaicity (Figure 2c). The calculated full width at half maximum (FWHM) of the (001) reflections was 0.67° for the Ce-FST and 0.55° for the P-FST, respectively. The difference in the FWHM between P-FST and Ce-FST thin films is minimal, and the FWHM of Ce-FST is similar to other reported FeSexTe1-x thin films\(^6,11,14\). This indicates that the Ce-FST thin film was well-epitaxially grown along the \(c\)-axis despite the insertion of the oxide materials into the FST matrix.

To confirm in-plane texture and epitaxial quality, we performed an azimuthal phi scan using four circles XRD. Figure 2d indicates the azimuthal \(\phi\) scan of the (113) peak from the CaF\(_2\) substrate and the (112) peak from the Ce-FST thin film. The \(\phi\) scan of the Ce-FST thin film shows clear four peaks which have 90° intervals without extra broader intermediate-angle peaks. These peaks are at 45° with respect to the CaF\(_2\) (113) peaks because the [100] FST is parallel to the [110] CaF\(_2\). These results indicate that Ce-FST has the characteristic of a genuine epitaxial film with excellent in-plane texture.

In order to determine the nanoscale strain caused by the infinitesimal CeO\(_2\) injection at the interface of each FST layers, we analyzed atomically resolved-STEM images for the Ce-FST thin film (Figure 3). Figure 3a shows a cross-sectional atomically resolved-STEM image of the Ce-FST thin film. Fine strained lines are observed at the interface of each FST layer with 20 nm intervals without large-scale defects. To better understand the strain field in Ce-FST thin films, we performed GPA based on the atomically resolved-STEM image of Figure 3a. GPA is generally used to generate show strain distribution and to determine deformation of the lattice constant in
crystalline structure. Figure 3b shows an extracted strain map of out-of-plane strain ($\varepsilon_{zz}$) for the same region in Figure 3a. GPA map undoubtedly shows the strained region with 20 nm intervals, and the type of strain is a tensile strain ($\varepsilon_{zz} \approx 0.05$) along the c-axis. The nanostrain position is in good agreement with the expected site where we intentionally inserted CeO$_2$. For a more accurate comparison, we performed STEM analysis of the P-FST thin film. The P-FST thin film shows a relatively clear phase as shown in Figure 3c, and there is no particular strain field in the out-of-plane GPA strain map from the atomically resolved-STEM image of the P-FST (Figure 3d). Based on these results, we can confirm that the injected infinitesimal CeO$_2$ indeed forms nanostrain in the Ce-FST thin film, but it is challenging to determine why the injected CeO$_2$ forms nanostrain in the FST matrix due to the wide range of possibilities.

In general, nanostrain is induced at various types of defect perimeters. Interestingly, nanoscale lattice distortion points, which indicate nanoscale fine defects such as dislocation core, were prominently observed at the nanostrain region in the GPA image for the Ce-FST thin film (dashed circle in Figure 3b). In contrast, there are few nanoscale lattice distortion points in P-FST thin film (dashed circle in Figure 3d). When we examine the enlarged image of the nanostrain region where lattice distortion points were not observed in the GPA map for Ce-FST thin film as shown in Figure 3b, nanoscale defects are observed, and we confirm that the spaces between the FST tetrahedral layers are actually filled by interstitial atoms in the more strained region (See supporting information, Figure S1).

There are several possibilities with regard to the formation of nanoscale defects due to the introduction of a trace amount of CeO$_2$. Firstly, the ionized species with high kinetic energy in the plume can cause re-sputtering and fine defects on the surface in the initial stage before these species form the cluster or layer. In the PLD system, the laser ablation of the target forms a plume
in which ionized target species exist. In effect, complex Ce compounds such as ionized Ce, ionized CeOx, Ce atom, and CeOx particles are actually injected into the Ce-FST matrix due to the high laser energy and high vacuum conditions. Interestingly, when the CeO2 layer was deposited into or onto the FST thin film, a nanoscale damaged layer (or transition layer) was formed between the CeO2 and FST interface, although CeO2 has excellent chemical stability with FST.32-33, 43 In other words, since a damaged FST layer is formed at the initial stage of CeO2 deposition, a small amount of CeO2 insertion, which typically does not form an intact CeO2 layer, can generate a fine defect inside the FST layer without a CeO2 layer.

Moreover, the residual Ce atoms or its compounds inside the FST layers can form nanoscale defects such as substitutional and interstitial defects. Given that the CeO2 phase has not been observed in atomically resolved-STEM images and XRD analysis of Ce-FST thin films, the injected CeO2 can exist as a Ce atom. However, the similar atomic weights of Ce and Te makes it difficult to distinguish between the two atoms in the common Z-contrast (Z = atomic number) STEM image of Ce-FST, and it is also challenging to identify Ce atoms because the strained area is commonly blurred in the image due to defects44-45. In addition, when composition analysis was performed using energy dispersive spectroscopy (EDS) with STEM to confirm the presence of residual Ce and O atoms at the interface of each FST layers, Ce and O were not observed at the nanostrain region corresponding to the injection of the infinitesimal CeO2 (Figure S2). Conversely, these results indicate that the nanostrain was successfully formed without crystallization degradation via inappreciable Ce injection.

Furthermore, we examined whether or not the formation of the nanostrain is affected by pausing time (10 s) during target exchange between the FST and CeO2 targets, because Se and Te are sensitive and volatile in FST thin films14. The paused FST thin film was fabricated following the
same fabrication process for the Ce-FST thin film except for CeO₂ injection; the CeO₂ plume was screened during the laser ablation of the CeO₂ target. When we performed the θ-2θ scan for paused FST thin film using Synchrotron-based XRD, the results indicated well-oriented (001) peaks without satellite peak, indicating that the pausing time had a negligible effect on the formation of the nanostrain in the FST matrix (Figure S3).

To compare superconducting properties between the Ce-FST and P-FST thin films, we measured the temperature dependence resistivity to obtain the transition temperature ($T_c$). Figure 4a and 4b show the resistivity as a function of temperature up to 9T with H∥c for both films. The measured $T_c^{\text{onset}}$ and $T_c^{\text{zero}}$ are 21.3 K and 19.8 K, respectively, for the P-FST thin film and 20.4 K and 18.9 K, respectively, for the Ce-FST thin film. The Ce-FST thin films have slightly lower $T_c$ than that of the P-FST thin film. The reason for the lower $T_c$ in the Ce-FST thin film is not only the slight degradation of crystalline quality by the nanoscale defects, but also the induced tensile nanostrain where the tensile strain causes the anion height and Ch-Fe-Ch (Ch: chalcogen) bond angle to deviate from ideal values\textsuperscript{46-48}. However, the degradation of $T_c$ by nanostrain is rather negligible because the Ce-FST thin film shows higher or similar $T_c$ compared to other reports of FeSe\textsubscript{x}Te\textsubscript{1-x} films\textsuperscript{19, 46, 49-50}.

The interesting observation with regard to the $R-T$ measurement is that the suppression of $T_c$ depending on the magnetic field ($\Delta T_{c,\text{field}}^{\text{zero}} = T_{c,\text{9T}}^{\text{zero}} - T_{c,\text{0T}}^{\text{zero}}$) for the Ce-FST thin film ($\Delta T_{c,\text{field}}^{\text{zero}} = 2.6$ K) is lower than that of the P-FST thin film ($\Delta T_{c,\text{field}}^{\text{zero}} = 3.2$ K). This indicates that the Ce-FST thin film has a lower magnetic field dependence than the P-FST thin film. We estimated $H_{\text{irr}}$ and $H_{c2}$ of both FST thin films using the 0.01 $\rho_n$ criteria and the 0.9 $\rho_n$ criteria with $\rho_n = \rho(23$ K) as a function of the normalized temperature ($t = T / T_c^{\text{onset}}$), to characterize the magnetic field dependence of temperature (Figure 5). $H_{c2}$ and $H_{\text{irr}}$ are certainly improved after the application of
nanoscale lattice distortion with nanostrain through CeO₂ injection. In particular, the improved $H_{irr}$ is indicative of the beneficial effect of the periodic nanostrained region and the nanoscale defects as pinning centers for high magnetic fields.

To verify the effect of the nanostrain and nanoscale defects as pinning centers on supercurrents in the FST thin film, the magnetization $J_c$ of both the Ce-FST and the P-FST thin films were measured (Figure 6). Figure 6a and 6b show the magnetic field dependence $J_c$ of the Ce-FST and the P-FST thin films at various temperatures (4.2 K, 7 K, 10 K and 12 K) up to 13T($H∥c$). The $J_c$ of the Ce-FST thin film had a value of 3.2 MA/cm² in a self-field and 0.44 MA/cm² under 13 T, at 4.2 K. The self-field $J_c$ of the Ce-FST thin film is the highest value of an iron-chalcogenide superconductors to our best knowledge²⁴. The $J_c$ of the P-FST thin film had a value of 2.3 MA/cm² in a self-field and 0.23 MA/cm² in 13 T at 4.2 K. The $J_c$ of the P-FST thin film is also similar and higher when compared to other reported values¹⁹,²⁴,³³. To verify these magnetization $J_c$ value derived using the Bean model, the transport $J_c$ of the Ce-FST thin film was also measured. The Ce-FST shows a transport $J_c$ of 3.5 MA/cm² in a self-field and of 0.44 MA/cm² under 13 T at 6 K, which is reasonably similar to the magnetization $J_c$ in the Ce-FST thin film. Additionally, $J_c$ enhancement was calculated to confirm the effect of the nanoscale pinning centers in detail. The $J_c$ enhancement of Ce-FST compared to P-FST increased from 40% to 120% up to 5T and gradually decreased for a high magnetic field (Figure 6c). These results clearly show that the Ce-FST maintains high $J_c$ under a high magnetic field as well as a low magnetic field.

Additionally, we estimated the pinning force ($F_p$) to characterize the effect of nanostrain with nanoscale defects. Figure 6d shows the magnetic field dependence of the vortex pinning force ($F_p = J \times B$) of both the Ce-FST and the P-FST thin films up to 13 T ($H∥c$) at 4.2 K. The Ce-FST and P-FST thin films show maximum pinning force ($F_{p,max}$) of 57.4 GN/m³ at 12.5 T and 30.5 GN/m³
at 13.5 T and 4.2 K, respectively. In particular, the Ce-FST thin film shows ~90% higher $F_{p,\text{max}}$ than for the P-FST thin film at 13 T ($H//c$). Consequently, the Ce-FST shows much higher $J_c$ and $F_p$ than the P-FST under all measured magnetic field and temperature settings. These results suggest that nanoscale defect and nanostrain are effective pinning centers in iron chalcogenide thin films.

CONCLUSION

Herein, we have developed a new method of inducing nanostrain at the desired position inside FST thin films via the injection on an infinitesimal CeO$_2$ using S-PLD without additional post-processing. GPA with STEM structural analysis demonstrated that the injected CeO$_2$ forms nanoscale defects that apply out-of-plane tensile strain in the FST layer. The nanostrain and nanoscale defects significantly improve the self-field $J_c$ of the FST thin film from 2.3 MA cm$^{-2}$ up to 3.2 MA cm$^{-2}$ at 4.2 K, while minimizing the degradation of the $T_c$ of the FST thin film. In particular, the Ce-FST thin film with nanostrain exhibited 90% improved $J_c$ under 13 T compared to the pristine sample. This study demonstrates that the formation of the nanostrain using S-PLD is significantly effective in attempting to achieve the ultimate goal of high magnetic field applications of iron chalcogenide. We also believe that this technique will be of great utility in inducing artificial nanoscale strains in other epitaxial thin films.

ASSOCIATED CONTENT

Supporting Information.

Atomic resolved STEM image of Ce-FST, EDS mapping of Ce-FST, and XRD diffraction pattern of paused FST.
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Author Contributions

S.S. and H.N made the FeSe$_x$Te$_{1-x}$ film specimens and performed the XRD characterization with the assistance of J.L. S.G.J. performed electrical resistivity-temperature measurements under the guidance of T.P. J.J. performed electromagnetic characterizations and C.T performed the transport $J_c$ measurement under the guidance of E.E.H. M.L. performed the TEM measurement and GPA analysis under the guidance of P.G. S.L. designed the experiment and supervised the work. S.S., H.N., and S.L. co-wrote and commented on the manuscript. # These authors contributed equally.

Notes

The authors declare no competing financial interest

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FIGURE CAPTIONS

**Figure 1.** Schematic of the Ce-FST thin film. FST and CeO₂ were alternately deposited. The infinitesimal CeO₂ is injected at the interface of each FST layer. The thickness of each FST layer is 20 nm, and the total thickness was 100 nm.

**Figure 2.** (a) Out-of-plane θ-2θ XRD pattern of the Ce-FST thin film on CaF₂. (b) The enlarged FST (001) reflection on Figure 2a. (c) Rocking curve of (001) reflection of the Ce-FST and the P-FST thin films. Δω indicates the FWHM of the (001) reflection. (d) Azimuthal ϕ scan of the Ce-FST (113) and CaF₂ (112).

**Figure 3.** (a) Cross-sectional HAADF-STEM image of the Ce-FST thin film. (b) Map of out-of-plane strain for the 100 nm Ce-FST thin film determined by GPA of STEM images from the same area on Figure 3a. Yellow arrows indicate nanostrain region. (c) Cross-sectional STEM image of the P-FST thin film. (d) Map of our-of-plane strain for the 100 nm P-FST thin film determined from GPA of STEM images from the same area on Figure 3c. Dashed circles represent lattice distortion points.

**Figure 4.** Temperature dependence of the resistivity of (a) the Ce-FST thin film and (b) the P-FST thin film.

**Figure 5.** Upper critical field ($H_{c2}(T)$) and irreversibility field ($H_{irr}(T)$) as a function of normalized temperature ($t=T/T_c$) for the Ce-FST and P-FST thin films. Each point shows the resistivity drops to 0.9 of normal resistivity ($\rho_n$) and 0.01 of $\rho_n$. $\rho_n$ is the resistivity at the normal state ($\rho(23\,K)$) of Ce-FST and P-FST thin film, respectively.
Figure 6. Magnetization $J_c$ as a function of the magnetic field ($H/c$) of (a) the Ce-FST and (b) the P-FST thin films which were derived from half magnetization loops at different temperatures (4.2 K, 7 K, 10 K and 12 K). Transport $J_c$ as a function of magnetic field ($H/c$) of Ce-FST thin films up to 14 T at 6 K is represented with magnetization $J_c$ as star open mark in Figure 6b. (c) The percentage of improved $J_c$ of Ce-FST which is calculated based on P-FST as a function of magnetic field. (d) Vortex pinning force ($F_p$) for the Ce-FST and the P-FST thin films as a function of magnetic field ($H/c$).
Figure 1
Figure 2
Figure 3
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Figure 5
Figure 6

(a) P-FST: $J_c$ vs. $H//c$, showing $J_c$ in units of $A/cm^2$ as a function of magnetic field $H$ for temperatures 4.2 K, 7 K, 10 K, and 12 K.

(b) Ce-FST: $J_c$ vs. $H//c$, similar to (a) but with additional data points at 12 K and transport data at 6 K.

(c) $J_c$ enhancement vs. $H//c$ at 4.2 K for P-FST.

(d) $F_c$ vs. $H//c$ for Ce-FST and P-FST at 4.2 K.
Supplementary information

S1. STEM image of nanostrain region in Ce-FST thin film.

Figure S1. The STEM image of nanostrain region in Ce-FST thin film. White scale bar represents 2 nm. Inset image indicates map of out-of-plane strain from same area on STEM image. Yellow dashed shows strained region with defects. Red circle shows interstitial defect which occupy the empty site between tetrahedral FST layers.
S2. EDS mapping for Ce-FST thin film.

**Figure S2.** (a) The large-scale STEM image of the Ce-FST thin film. White arrows represent nanostrained region. EDS elemental map of (b) Ce, (c) O, (d) Fe, (e) Se, and (f) Te. Se signal is observed in Pt area due to peak overlap between Pt and Se. In the nanostrained region, there is no remarkably Ce and O signal.
S3. Magnification of FST (001) reflection for paused FST

Although we confirm the enlarged (001) reflection of the paused FST thin film, there is no satellite peaks, indicating that the paused FST thin film has no superlattice-type structure.

Figure S3. (a) $\theta$-2$\theta$ scan for paused FST thin films using Synchrotron-based XRD. (b) The enlarged FST (001) reflection of paused FST thin film.