Enhancement of critical current density and mechanism of vortex pinning in H\textsuperscript{+}-irradiated FeSe single crystal

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We report a comprehensive study of the effect of H\textsuperscript{+} irradiation on the critical current density \(J_c\) and vortex pinning in an FeSe single crystal. The value of \(J_c\) for FeSe is enhanced by more than a factor of 2 after 3-MeV H\textsuperscript{+} irradiation, which is explained by the introduction of point pinning centers. Vortex creep rates are found to be strongly suppressed after irradiation. Detailed analyses of the pinning energy based on collective-creep-theory and an extended Maley’s method show that the H\textsuperscript{+} irradiation enhances the value of \(J_c\) before the flux creep and also reduces the size of the flux bundle, which suppresses the field dependence of \(J_c\) owing to vortex motion. © 2015 The Japan Society of Applied Physics

Iron-based superconductors (IBSs) display some fascinating fundamental properties for applications, such as their reasonably high superconducting transition temperature \(T_c\), very high critical field \(H_{c2}\), and relatively small anisotropy.\textsuperscript{1,3} Among the IBSs, iron chalcogenides have stimulated great interest because they are possible candidates for breaking the \(T_c\) record (\(\sim 55\) K) in IBSs. Although the initial \(T_c\) of FeSe is <10 K,\textsuperscript{2,5} it increases to 14 K with appropriate Te substitution\textsuperscript{3,10} and to 37 K under high pressure.\textsuperscript{4,5} Furthermore, the monolayer of FeSe film grown on SrTiO\textsubscript{3} even shows a sign of superconductivity at \(\sim 100\) K.\textsuperscript{10} For applications, high-quality Te-doped FeSe tapes with transport \(J_c > 10^8\) A/cm\textsuperscript{2} under self-field and >10\textsuperscript{7} A/cm\textsuperscript{2} under 30 T at 4.2 K have already been fabricated.\textsuperscript{11,12} Moreover, \(J_c\) was found to be almost isotropic and homogeneously distributed in both single crystals and thin films.\textsuperscript{8,9} In addition, its less toxic nature than iron pnictides is also advantageous for applications.

From the application viewpoint, the value of \(J_c\) is a key factor. \(J_c\) is determined not only by the material’s intrinsic properties, but also by extrinsic conditions, such as defects. Thus, the introduction of artificial pinning centers either by chemical or physical methods is effective in enhancing the value of \(J_c\). The chemical method introduces extended defects, such as Y\textsubscript{2}O\textsubscript{3} nanoparticles in bulk cuprates.\textsuperscript{10} The physical method is usually performed by particle irradiation, including proton irradiation to cause point defects and heavy-ion irradiation to cause columnar defects. The physical method is more advantageous to probe the pinning mechanism because it is easy to control the number and type of pinning centers without affecting the structure of the crystal. For IBSs, both methods have proven to be effective for enhancing \(J_c\).\textsuperscript{11-17} However, until now, attempts have been made mostly in iron pnictides, especially in the “122” phase since high-quality single crystals are available. For FeSe, such a study is still left unexplored because of the difficulty in growing high-quality single crystals. Actually, the study of the irradiation effect in FeSe is not only important to the enhancement of \(J_c\) for applications but also crucial for understanding the pinning mechanism because FeSe possesses some unique characteristics: It has the simplest structure, is composed of only Fe–Se layers, and is also a clean system free from doping-introduced inhomogeneities and charged quasiparticle scattering because of its innate superconductivity.\textsuperscript{22}

Recently, high-quality and sizable single crystals of FeSe have been grown.\textsuperscript{18} In this report, we present a study of the effect of H\textsuperscript{+} irradiation on FeSe single crystal. Introduction of defects into FeSe using 3-MeV H\textsuperscript{+} results in the enhancement of \(J_c\) by a factor of >2, which is explained by the successful introduction of point pinning centers into the crystal. A vortex dynamics study reveals that proton irradiation enhances the value of the critical current density before the flux creep and also reduces the size of the flux bundle, which will further suppress the strong field dependence of \(J_c\) from the vortex motion.

High-quality single crystals of tetragonal \(\beta\)-FeSe were grown by using the vapor transport method as described elsewhere.\textsuperscript{19} Our previous report has shown the high quality of the grown FeSe single crystal, which exhibits \(T_c \sim 9\) K with residual resistivity ratios RRR > 40. Scanning tunneling microscope (STM) observations also demonstrated that the crystal contains extremely small levels of impurities and defects.\textsuperscript{19,20} Those results ensure that our irradiation experiment was performed on a clean crystal with less influence from second phase or inhomogeneities. FeSe crystals were cleaved to thin plates with thickness \(\sim 25\) µm along the \(c\)-axis, which is much smaller than the projected range of 3-MeV H\textsuperscript{+} for FeSe of \(\sim 50\) µm, calculated by the stopping and range of ions in matter-2008.\textsuperscript{21} To avoid any possible sample-dependent influence, all the measurements were performed on one identical piece of crystal, which was divided into two parts: pristine and irradiated samples. The 3-MeV H\textsuperscript{+} irradiation with a total dose of \(5 \times 10^{16}\) cm\textsuperscript{-2} was performed parallel to the \(c\)-axis at the National Institute of Radiological Sciences’ Heavy Ion Medical Accelerator in Chiba. Magnetization measurements were performed by using a commercial SQUID magnetometer. After the irradiation, the value of \(T_c\) is almost unchanged, which is similar to the case of Ba\textsubscript{1−x}K\textsubscript{x}Fe\textsubscript{2}As\textsubscript{2}.\textsuperscript{15}

Figure 1 shows the magnetic field dependence of \(J_c\) for (a) pristine and (b) H\textsuperscript{+}-irradiated FeSe single crystals obtained by using the extended Bean model:\textsuperscript{22}

\[
J_c = 20 \frac{\Delta M}{a(1 - a/3b)},
\]
where $\Delta M$ is $M_{\text{down}} - M_{\text{up}}$, $M_{\text{up}}$ (emu/cm$^3$) and $M_{\text{down}}$ (emu/cm$^3$) are the magnetization when sweeping the field up and down, respectively, and $a$ (cm) and $b$ (cm) are sample widths ($a < b$). It is obvious that H$^+$ irradiation enhances the value of the self-field $J_c$ at 2 K from $3 \times 10^9$ to $8 \times 10^4$ A/cm$^2$.

The value of $J_c$ changes only slightly with increasing field below 1 kOe in the pristine sample, which is followed by a power-law decay $H^{-\alpha}$ in the field range of 4–10 kOe with $\alpha_1 \sim 0.5$. Such a power-law dependence of $J_c$ is also observed in most IBSSs and is attributed to strong pinning by sparse nanometer-sized defects as in the case of YBCO films.\(^{23}\) Such a result is consistent with the STM observation, where randomly distributed defects with an effect range covering a few nanometers are dispersed.\(^{19}\) After that, the decay rate of $J_c$ increases to $\alpha_2 \sim 1.2$. Such behaviors may be explained by the small amount of strong pinning centers with density $<1$ per 2000 Fe atoms, as observed by STM.\(^{19}\) In this case, all the pinning centers will be easily occupied by the flux above some characteristic field. Above 10 kOe, the pinning force $F_p$ will remain constant in spite of the increase in $H$. Thus, the value of $J_c$ will decrease with the rate of $H^{-1}$ since $F_p = \mu_0 H \cdot J_c$. After H$^+$ irradiation, $J_c$ also exhibits field-insensitive behavior at small field and $H^{-1}$ decaying behavior at fields $>10$ kOe, similar to the pristine sample. However, in the field range of 4–10 kOe, $J_c$ decays with the field at a rate of $H^{-0.3}$ rather than the $H^{-0.5}$ behavior observed in the pristine sample. The change of $J_c$ decaying with the field from $H^{-0.3}$ to $H^{-0.5}$ was also observed in H$^+$-irradiated Ba(Fe$_{0.9}$Co$_{0.07}$)$_2$As$_2$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$.\(^{24,25}\)

To see the H$^+$-irradiation effect more clearly and gain more insight into the vortex pinning, the normalized vortex pinning forces $f = F_p/F_p^{\text{max}}$ as a function of the reduced field $h = H/H_{\text{irr}}$ at different temperatures are shown in Figs. 2(a) and 2(b) for pristine and H$^+$-irradiated FeSe, respectively. The pinning force $F_p$ was obtained from the critical current density by using $F_p = \mu_0 H \cdot J_c$, and $F_p^{\text{max}}$ corresponds to the maximum pinning force. $H_{\text{irr}}$ is the irreversibility field, which is obtained from the linear extrapolation of $J_c^{1/2} - \mu_0 H$ curves to the zero value of $J_c$. It is obvious that $f$ for pristine and H$^+$-irradiated FeSe falls into one curve. The peak position of the overlapped curves for the pristine sample is located at a value of $h = 0.14$. After H$^+$ irradiation, the peak position of $f$ was changed to $h = 0.28$, which is close to the values of 0.33 for the core normal point pinning according to the Dew–Hughes model.\(^{26}\) Moreover, the insets of Figs. 2(a) and 2(b) show the curves of $F_p^{\text{max}}$ versus $H_{\text{irr}}$ for pristine and H$^+$-irradiated FeSe. Obviously, the magnitude of $F_p^{\text{max}}$ was enhanced, and the scaling parameter $\alpha$ of $F_p^{\text{max}} \propto H^\alpha$ is $\sim 1.9$ for the irradiated crystal, which is close to the theoretical value of 2 for the core normal pointlike pinning.\(^{26}\) Thus, the peak position change in $f$ indicates that the H$^+$ irradiation successfully introduces point pinning centers into the FeSe single crystal, which enhances the pinning force and critical current density.

To get a more comprehensive and quantitative understanding of the H$^+$-irradiation effect on the vortex dynamics of FeSe single crystal, we carefully traced the decay of

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**Fig. 1.** Magnetic field dependence of critical current densities with $H \parallel c$ for (a) pristine and (b) H$^+$-irradiated FeSe. The lowest field data were captured at a value very close to zero (usually $<1$ Oe) to estimate the self-field $J_c$. Thus, the lines below 1 kOe are prepared by using the $J_c$ values at $H = 0$. The dashed lines show the power-law decay of $H^{-\alpha}$.

**Fig. 2.** Normalized flux pinning force $f = F_p/F_p^{\text{max}}$ as a function of the reduced field $h = H/H_{\text{irr}}$ at different temperatures for (a) pristine and (b) H$^+$-irradiated FeSe. Insets show $F_p^{\text{max}}$ as functions of $H_{\text{irr}}$. 

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magnetization with time, \(M(t)\), originating from the flux creep for more than one hour, where \(t\) is the time from the moment when the critical state is prepared. The normalized magnetic relaxation rate \(S\) is defined by \(S \equiv |d \ln M/d \ln t|\). In these measurements, the magnetic field was swept more than 5 kOe higher than the target field before starting measurements. Figure 3(a) shows the temperature dependence of the normalized magnetic relaxation rate \(S\) at 500 Oe (larger than the self-fields of ~100 and ~300 Oe for the pristine and H\(^+\)-irradiated crystals, respectively). \(S\) for both crystals shows an obvious temperature-insensitive plateau in the intermediate temperature region with a relatively large vortex creep rate. The plateau and large vortex creep rate were also observed in \(YBa_2Cu_3O_7\) \(\delta\) and other IBSs, \(^{28,29}\) which can be interpreted by using collective creep theory. \(^{27}\) By contrast, after H\(^+\) irradiation, the magnitude of \(S\) is suppressed to half of the value of the pristine sample, and the plateau behavior becomes more obvious. Similar suppression of \(S\) can also be seen in its field dependence, as shown in Fig. 3(b), which shows typical results at 2 K.

Figures 4(a) and 4(b) show the effective pinning energies \(U^* = T/S\) as a function of inverse current density \(1/J\) for pristine and H\(^+\)-irradiated FeSe, respectively. According to collective creep theory, the slope \(\mu\) for the \(U^* - 1/J\) relation on log–log plot contains information about the size of the vortex bundle. In a three-dimensional system, it is predicted as \(\mu = 1/7, (1) 5/2, 7/9\) for single-vortex, (intermediate) small-bundle, and large-bundle regimes, respectively. \(^{10,31}\) The evaluated value of \(\mu\) for the pristine crystal is ~0.71, as expected for collective creep by large bundles. Contrary to the above prediction of \(\mu > 0\), a negative slope with a value \(~ -0.81\) is obtained at small \(J\). The negative slope is often denoted as \(p\) in plastic creep theory, which is thought to lead to faster escape of vortices from the superconductors. \(^{32}\) The crossover is persistent after H\(^+\) irradiation. However, the value of \(\mu\) increases to 1.0, which indicates that vortex creep in the irradiated crystal is in the form of an intermediate bundle.

To get more quantitative insight into the variation in vortex pinning, we analyze the \(U - J\) relation by using the extended Maley’s method. \(^{33}\) We find that all the curves can be well scaled together, as shown in the insets of Figs. 4(a) and 4(b) for pristine and H\(^+\)-irradiated FeSe, respectively. The solid lines indicate the power-law fitting by \(^{33}\)

\[
U(J) = \frac{U_0}{\mu} [(J_{\text{c0}}/J)^{\mu} - 1] 
\]  

to the large-\(J\) region, where \(U_0\) and \(J_{\text{c0}}\) are the flux activation energy and critical current density in the absence of flux creep, respectively. The deviation of the data from the fitting line in the small-\(J\) region is reasonable since vortex creep is plastic there. The fitting gives \(\mu = 0.72, U_0 = 91.1\) K, and \(J_{\text{c0}} = 4.0 \times 10^4\ \text{A/cm}^2\) for the pristine crystal and \(\mu = 1.09, U_0 = 96.2\) K, and \(J_{\text{c0}} = 8.1 \times 10^4\ \text{A/cm}^2\) for the irradiated one. The values of \(\mu\) obtained from the extended Maley’s method are very close to those evaluated in the main panel of Fig. 4 for both crystals, which confirms the correctness of the present analyses. The observed changes in \(J_{\text{c0}}\) and \(\mu\) show that the H\(^+\) irradiation enhances the critical current density without flux creep and also reduces the size of the flux bundle to suppress the reduction of current density from vortex motion.

In conclusion, we report that 3-MeV H\(^+\) irradiation can enhance the critical current density \(J_c\) of FeSe single crystal by a factor of >2 by introducing extra point pinnings. Magnetic relaxation measurements show that the vortex creep rate is strongly suppressed after the irradiation. Detailed analyses of the critical-current-dependent pinning energy based on collective creep theory and the extended Maley’s method demonstrate that the H\(^+\) irradiation enhances the value of \(J_c\) before the flux creep starts and also reduces the size of the flux bundle from large to intermediate. The reduction of the size of the flux bundle will further suppress the field dependence of \(J_c\) owing to vortex motion.
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