Effects of Splayed Columnar Defects on Critical Current Density in CaKFe$_4$As$_4$

A Takahashi$^1$, S Pyon$^1$, Y Kobayashi$^1$, T Kambara$^2$, A Yoshida$^3$, S Okayasu$^3$, A Ichinose$^4$ and T Tamegai$^1$

$^1$Department of Applied Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
$^2$Nishina Center, RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan
$^3$Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
$^4$Central Research Institute of Electric Power Industry, Nagasaka, Yokosuka, Kanagawa 240-0196, Japan

E-mail: ayumu-takahashi141@g.ecc.u-tokyo.ac.jp

Abstract. Introduction of columnar defects to superconductors through particle irradiation enhances their critical current density ($J_c$). Further enhancement of $J_c$ by dispersing the direction of columnar defects has been confirmed. Moreover, in such systems with splayed columnar defects, an anomalous peak effect in $J_c$ at a certain magnetic field determined by the irradiation dose accompanied by the in-plane anisotropy of $J_c$ between those parallel and perpendicular to the splay direction were observed. We introduce splayed columnar defects to CaKFe$_4$As$_4$ single crystals, which was recently found as a new type of iron-based superconductors (1144-type IBS), by irradiating 2.6 GeV U and 320 MeV Au ions and measure their $J_c$ properties.

1. Introduction

Introduction of columnar defects to superconductors through swift-particle irradiation enhances their critical current density ($J_c$) [1-4]. Further enhancement of $J_c$ by dispersing the direction of columnar defects has been confirmed in cuprates YBa$_2$Cu$_3$O$_{7-\delta}$ [5] and iron-based superconductors (IBSs) Ba$_{1.8}$K$_4$Fe$_2$As$_2$ [6] single crystals. Moreover, in such systems with splayed columnar defects, an anomalous peak effect in $J_c$ at a certain magnetic field determined by the irradiation dose accompanied by the in-plane anisotropy of $J_c$ between those parallel and perpendicular to the splay direction were observed [6, 7].

IBSs have been investigated as promising materials for practical applications because of their large $J_c$ at high magnetic fields and temperatures. In previous studies, remarkable effects have been demonstrated in IBSs by irradiating heavy ions and protons into Co or K doped BaFe$_2$As$_2$ (Ba-122) single crystals [3, 8]. Recently, another promising IBS CaKFe$_4$As$_4$ (1144-type IBS) was found [9]. Its crystal structure is similar to 122-type IBSs. CaKFe$_4$As$_4$ has a tetragonal structure (P4/mmm), where Ca and K layers stack alternatively along the $c$-axis [9, 10]. CaKFe$_4$As$_4$ shows similar superconducting properties, such as critical temperature ($T_c$) or upper critical field ($H_{c2}$), to those of optimally K-doped Ba-122-type IBS Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ [11-13]. However, alternate stacking of Ca and K along the $c$-axis may lead to different physical properties.
Here, we introduce splayed columnar defects to CaKFe$_4$As$_4$ single crystals by irradiating 2.6 GeV U and 320 MeV Au ions, and compare their $J_c$ properties with those in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$.

2. Experimental Methods

CaKFe$_4$As$_4$ single crystals ($T_c \sim 36$ K) were synthesized by FeAs self-flux method. Ca granules (99.5%), K ingots (99.5%), and FeAs powder were used as starting materials. FeAs was prepared by sealing stoichiometric amounts of As grains (7N) and Fe powder (99.9%) in an evacuated quartz tube and reacting them at 700 °C for 40 h after heating at 500 °C for 10 h. A mixture with a ratio of Ca : K : FeAs = 1 : 1 : 10 was placed in an alumina crucible in an argon-filled glove box. The alumina crucible was sealed in a niobium tube using arc welding. The niobium tube was sealed in an evacuated quartz tube. The whole assembly was heated for 5 h at 1180 °C after a preliminary heating at 650 °C for 5 h, and cooled to 1050 °C at a rate of 26 °C/h, followed by cooling to 930 °C at a rate of 1.5 °C/h for the crystal growth. The crystals were shaped into rectangular parallelepipeds with lengths and widths of approximately 500 μm and with thicknesses less than half of the projected range of irradiated ions (63 μm for 2.6 GeV U ions and 17 μm for 320 MeV Au ions).

2.6 GeV U ions were irradiated at RIKEN Ring Cyclotron in R1 Beam Factory operated by RIKEN Nishina Center and CNS, The University of Tokyo, and 320 MeV Au ions were irradiated at the tandem accelerator in JAER. The incident directions of ions were changed by tilting the crystals about their $ab$-plane, and the angle of CDs ($\theta_{CD}$) is denoted by the angle between their $c$-axis and the ion beam. The irradiation dose is evaluated by the dose-equivalent magnetic field called “matching field”, at which all defects are occupied by single vortices;

$$B_0 = n \Phi_0.$$  \hspace{1cm} (1)

Here, $n$ is the areal density of defects and $\Phi_0$ is a flux quantum.

Magnetization of the crystal was measured by a superconducting quantum interference device (SQUID) magnetometer (MPMS-5XL, Quantum Design). Only the data shown in Figure 3(b) was measured up to 9 T by Physical Properties Measurement System (PPMS) with vibrating sample magnetometer (VSM). The single crystal was placed in a quartz sample holder and fixed with Apiezon N grease. Average in-plane $J_c$ was calculated using the extended Bean model from the results of the magnetization measurements after subtracting the reversible component approximated by linear field dependence.

In order to observe the in-plane anisotropy of $J_c$, magneto-optical (MO) imaging was also performed. The single crystal was mounted on a copper sample holder and held together with Apiezon N grease to fix and to increase thermal conductivity to the sample holder. On top of the sample holder, a ferrimagnetic garnet indicator film with a reflective mirror was placed in direct contact with the crystal. The whole assembly was placed on a cold finger and shrouded by a radiation shield, then cooled by a He-flow cryostat. The light intensity versus flux density reference was obtained after locating the sample above the $T_c$ between -500 Oe and 500 Oe using a copper solenoid. The reference were fitted with a second order polynomial, in which the coefficients were used to calculate the flux density. Remanent state images were obtained after applying a field of 1 kOe for 1 second.

In the usual MO imaging method using a ferrimagnetic garnet indicator, an in-plane field is applied to diminish the effect of in-plane magnetic domain formed in the indicator. However, an in-plane field can induce an in-plane anisotropy of $J_c$. In order to observe the pure in-plane anisotropy of the samples, MO imaging was performed without applying in-plane field, which leads to MO images with noises due to in-plane magnetic domains.

3. Results and Discussion

Figures 1(a) and (b) show the splay angle dependence of $J_c$ at various temperatures under self-field for CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$, respectively, that are irradiated by 2.6 GeV U ions with $B_0 = 4$ T + 4 T. As shown in Fig. 1(a), $J_c$ under self-field for CaKFe$_4$As$_4$ irradiated by 2.6 GeV U ions with $B_0 = 4$ T + 4 T has the maximum value when $\theta_{CD}$ is around ±15°. On the other hand, as shown in Fig. 1(b), $J_c$
under self-field for Ba$_{0.6}$K$_0$Fe$_2$As$_2$ irradiated by 2.6 GeV U ions with $B_\Phi = 4$ T + 4 T has the local minimum value when $\theta_{CD}$ is around ±15°.

Figures 2(a) and (b) show the magnetic field dependence of $J_c$ at various temperatures for CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_0$Fe$_2$As$_2$, respectively, that are irradiated by 2.6 GeV U ions with $B_\Phi = 4$ T + 4 T and $\theta_{CD} = ±20°$. The anomalous peak effect at $-1/3B_\Phi$ as observed in Ba$_{0.6}$K$_0$Fe$_2$As$_2$ (Fig. 2(b)) does not show up in CaKFe$_4$As$_4$ (Fig. 2(a)) at the same irradiation condition.

![Figure 1](image1.png)

**Figure 1.** Splay angle dependence of $J_c$ at various temperatures under self-field for (a) CaKFe$_4$As$_4$ and (b) Ba$_{0.6}$K$_0$Fe$_2$As$_2$ irradiated by 2.6 GeV U ions with $B_\Phi = 4$ T + 4 T. Dashed lines at $T = 2$ K indicate the estimated $J_c$ values since the exact values could not be measured due to the flux jump.

![Figure 2](image2.png)

**Figure 2.** Magnetic field dependence of $J_c$ at various temperatures for (a) CaKFe$_4$As$_4$ and (b) Ba$_{0.6}$K$_0$Fe$_2$As$_2$ irradiated by 2.6 GeV U ions with $B_\Phi = 4$ T + 4 T and $\theta_{CD} = ±20°$.

We interpret that the difference in the splay angle dependence and the suppression of the anomalous peak effect in CaKFe$_4$As$_4$ are due to the presence of planar defects parallel to the $ab$-plane, which is unique to this material [14]. The optimal splay angle for $J_c$ is determined by the competition between the beneficial effect of suppressing vortex motion by the variable inter-defect distance and forcing vortices to entangle, and the adverse effect of vortex-field misalignment [5, 6]. Planar defects in CaKFe$_4$As$_4$ can affect this competition and lead to the different splay angle dependence of $J_c$ from that without such planar defects. Moreover, figure 3 shows the magnetic field dependence of $J_c$ at various temperatures for CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_0$Fe$_2$As$_2$ irradiated by 2.6 GeV U ions with $B_\Phi = 8$ T and $\theta_{CD} =$
0°, in other words, parallel to the c-axis. Similar to the case of systems with splayed columnar defects in Fig. 2, non-monotonic behavior of $J_c$ due to the self-field effect, where columnar defects become ineffective for curved vortices when the external magnetic field is below the self-field [3, 15, 16], observed at low fields in $\text{Ba}_0.6\text{K}_{0.4}\text{Fe}_2\text{As}_2$ (Fig. 3(b)) at the same irradiation condition disappears in $\text{CaKFe}_4\text{As}_4$ (Fig. 3(a)). The weakening of $J_c$ suppression at low fields by the self-field effect in $\text{CaKFe}_4\text{As}_4$ can be due to the pinning effects of planar defects for curved vortices. In systems with splayed columnar defects, the coexistence of columnar defects and planar defects may also contribute to the suppression of the anomalous peak effect.

![Figure 3](image1.png)

**Figure 3.** Magnetic field dependence of $J_c$ at various temperatures for (a) $\text{CaKFe}_4\text{As}_4$ and (b) $\text{Ba}_0.6\text{K}_{0.4}\text{Fe}_2\text{As}_2$ irradiated by 2.6 GeV U ions with $B_\phi = 8$ T and $\theta_{CD} = 0°$ (parallel to the c-axis).

![Figure 4](image2.png)

**Figure 4.** (a) The MO image of the remanent state of $\text{CaKFe}_4\text{As}_4$ irradiated by 2.6 GeV U ions with $B_\phi = 4$ T + 4 T and $\theta_{CD} = 15°$ after applying a field of 1 kOe along the c-axis at $T = 33$ K. The white arrow indicates the splay direction. (b) Line profiles along the yellow line in (a). (c) Line profiles along the red line in (a).

Figure 4(a) shows the remanent state MO image of $\text{CaKFe}_4\text{As}_4$ irradiated by 2.6 GeV U ions with $B_\phi = 4$ T + 4 T and $\theta_{CD} = 15°$ after sweeping up to 1 kOe and back to zero field at $T = 33$ K. Since the crystal has a macroscopic defect in the right half, the in-plane anisotropy should be discussed focusing on the left half of the sample. A critical state with a double Y-shaped current discontinuity line is formed. For the isotropic case, the angle between the discontinuity line and the sample edge should be 45°. The in-plane anisotropy of $J_c$ can be observed as a skewed double Y-shaped discontinuity line. For quantitative analysis of the in-plane anisotropy of $J_c$, line profiles were taken along two axes as shown by the yellow (parallel to the splay direction) and red (perpendicular to the splay direction) lines in Fig. 4(a), as shown in Fig. 4(b) and (c). The in-plane anisotropy of $J_c$, namely the ratio of $J_c$, parallel to the splay direction ($J_{c||}$) and that perpendicular to the splay direction ($J_{c\perp}$), can be estimated by the ratio of...
Moreover, these results imply that the two distances of the flux density peak from the sample edge. For CaKFe$_4$As$_4$ irradiated by 2.6 GeV U ions with $B_0 = 4 \ T + 4 \ T$ and $\theta_{CD} = 15^\circ$, the in-plane anisotropy is estimated as $J_{//}/J_{\perp} \sim 1.19$ at $T = 33 \ K$. For Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ irradiated by 2.6 GeV U ions with $B_0 = 4 \ T + 4 \ T$ and $\theta_{CD} = 15^\circ$, much larger in-plane anisotropy, $J_{//}/J_{\perp} \sim 4.17$, was reported [7]. Planar defects parallel to the $ab$-plane mentioned above can suppress the preferential enhancement of $J_c$ in a certain direction due to the spilled columnar defects and suppress the in-plane anisotropy. Moreover, these results imply that $J_{//}$, preferentially enhanced in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$, may play a significant role in the anomalous peak effect observed only in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. This is the subject of future studies.

**Figure 5.** Splay angle dependence of $J_c$ at various temperatures under self-field for (a) CaKFe$_4$As$_4$ and (b) Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ irradiated by 320 MeV Au ions with $B_0 = 2 \ T + 2 \ T$.

**Figure 6.** Magnetic field dependence of $J_c$ at various temperatures for (a) CaKFe$_4$As$_4$ and (b) Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ irradiated by 320 MeV Au ions with $B_0 = 2 \ T + 2 \ T$ and $\theta_{CD} = \pm 20^\circ$.

Figures 5(a) and (b) show the splay angle dependence of $J_c$ at various temperatures under self-field for CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$, respectively, that are irradiated by 320 MeV Au ions with $B_0 = 2 \ T + 2 \ T$. $J_c$ under self-field for both CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ irradiated by 320 MeV Au ions with $B_0 = 2 \ T + 2 \ T$ shows weak splay angle dependence compared with the case of irradiating 2.6 GeV U ions (Fig. 1).
Figures 6(a) and (b) show the magnetic field dependence of \( J \) at various temperatures for CaKFe\(_4\)As\(_4\) and Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 320 MeV Au ions with \( B_\phi = 2T + 2T \) and \( \theta_{CD} = ±20° \). As shown in Fig. 6(a), the anomalous peak effect at \( -1/3 \Phi_0 \) is suppressed in CaKFe\(_4\)As\(_4\) irradiated by 320 MeV Au ions with \( B_\phi = 2T + 2T \) and \( \theta_{CD} = ±20° \), similar to Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 2.6 GeV U ions with \( B_\phi = 4T + 4T \) and \( \theta_{CD} = ±20° \) (Fig. 2 (a)). On the other hand, as shown in Fig. 6(b), the anomalous peak effect at \( -1/3 \Phi_0 \) becomes much weaker in Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 320 MeV Au ions with \( B_\phi = 2T + 2T \) and \( \theta_{CD} = ±20° \) than Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 2.6 GeV U ions with \( B_\phi = 4T + 4T \) and \( \theta_{CD} = ±20° \) (Fig. 2 (b)).

One of the possible origins for the differences between the cases of irradiating 2.6 GeV U ions and that of irradiating 320 MeV Au ions is the continuity of the introduced columnar defects. Figures 7(a)-(d) show scanning transmission electron microscope (STEM) images of cross-sections of CaKFe\(_4\)As\(_4\) irradiated by 2.6 GeV U ions, Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 2.6 GeV U ions, CaKFe\(_4\)As\(_4\) irradiated by 320 MeV Au ions, and Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 220 MeV Au ions, respectively. Columnar defects in CaKFe\(_4\)As\(_4\) and Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 220 or 320 MeV Au ions are discontinuous, while those in CaKFe\(_4\)As\(_4\) and Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 2.6 GeV U ions are more continuous. This difference of the continuity of the introduced columnar defects leads to the difference of pinning efficiency and can affect the splay angle dependence and the anomalous peak effect of \( J \).

![Figure 7](image)

**Figure 7.** STEM images of cross-sections of (a) CaKFe\(_4\)As\(_4\) irradiated by 2.6 GeV U ions with \( B_\phi = 16T \) and \( \theta_{CD} = 0° \) (parallel to the \( c \)-axis), (b) Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 2.6 GeV U ions with \( B_\phi = 4T + 4T \) and \( \theta_{CD} = ±20° \), (c) CaKFe\(_4\)As\(_4\) irradiated by 320 MeV Au ions with \( B_\phi = 16T \) and \( \theta_{CD} = 0° \) (parallel to the \( c \)-axis), and (d) Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) irradiated by 220 MeV Au ions with \( B_\phi = 4T + 4T \) and \( \theta_{CD} = ±10° \). Planar defects parallel to the \( ab \)-plane can be observed only in CaKFe\(_4\)As\(_4\) ((a) and (c)).

### 4. Summary

We introduce splayed columnar defects to 1144-type IBS CaKFe\(_4\)As\(_4\) single crystals by irradiating 2.6 GeV U and 320 MeV Au ions, and compare their \( J \) properties with those in 122-type IBS Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) single crystals. In the case of 2.6 GeV U irradiation, the splay angle dependence of \( J \) for CaKFe\(_4\)As\(_4\) is different from that for Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\), and the anomalous peak effect observed in Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) is strongly suppressed in CaKFe\(_4\)As\(_4\). The in-plane anisotropy of \( J \) in CaKFe\(_4\)As\(_4\) is much weaker than that in Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\). These differences can be due to the presence of planar defects parallel to the \( ab \)-plane, which is unique to CaKFe\(_4\)As\(_4\). In the case of 320 MeV Au irradiation, the splay angle dependence and the anomalous peak effect are much weaker, compared with the case of 2.6 GeV U irradiation. These differences can be interpreted as the difference in the pinning efficiency due to the difference in the continuity of the introduced columnar defects.

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