Difference in anisotropic vortex pinning in pristine and proton-irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals

W. J. Choi

Daegu Gyeongbuk Institute of Science and Technology

Y. I. Seo

Daegu Gyeongbuk Institute of Science and Technology

K. Park

Daegu Gyeongbuk Institute of Science and Technology

Yong Seung Kwon (✉ yskwon@dgist.ac.kr)

Daegu Gyeongbuk Institute of Science and Technology

Research Article

Keywords: vortex dynamics, proton irradiation, thermally activated flux motion, vortex pinning anisotropy

Posted Date: May 5th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-476287/v1>

License: This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

**Difference in anisotropic vortex pinning in pristine and proton-irradiated
(Ca_{0.85}La_{0.15})₁₀(Pt₃As₈)(Fe₂As₂)₅ single crystals**

W. J. Choi[†], Y. I. Seo[†], K. Park and Yong Seung Kwon^{*}

Department of Emerging Materials Science, DGIST, Daegu 711-873, Republic of Korea

[†] Contributed equally

^{*} Corresponding author: yskwon@dgist.ac.kr

Abstract

To study the difference in the vortex pinning mechanism between in-plane and out-of-plane and the change of these pinning mechanisms due to defects induced by proton irradiation, the in-plane electrical resistivity for pristine and proton irradiated (Ca_{0.85}La_{0.15})₁₀(Pt₃As₈)(Fe₂As₂)₅ single crystals in $B//c$ and $B//ab$ up to $B = 13$ T. Both samples showed a monoclinic crystal structure, which was different from previously known, through crystal structure analysis using the selected area electron diffraction (SAED) method. The protons irradiation incident along the c -axis caused expansion of the lattice constants a and b . The electronic structure changed by the expansion of this lattice constant affected the change of the coherence length ξ_c significantly shorter than the distance between the superconducting Fe₂As₂ layers. The vortex in $B//ab$ was mainly pinned by the newly formed normal component with zero Cooper pair due to the short ξ_c , and ξ_c increased by proton irradiation reduced the pinning energy. On the other hand, the vortex in $B//c$ was mainly pinned by point defects, and the point defects increased by proton irradiation increased the pinning energy.

Keywords: vortex dynamics, proton irradiation, thermally activated flux motion, vortex pinning anisotropy

INTRODUCTION

The limitation to technological advances in high- T_c superconductors essentially comes from the low critical current density J_c . Since J_c is in many cases limited by thermally activated vortex motion, the most important way to increase J_c is to introduce a vortex pinning center in the superconductor during or after sample preparation¹. Therefore, knowing how to controllably change the vortex pinning energy in the process of maximizing J_c is very important for superconductor applications.

It is known that various types of defects that occur naturally during the sample fabrication play a major role of vortex pinning in high- T_c superconductors. These defects include atomic defects (As-vacancies, impurities, etc.) and structural defects (twin boundaries, stacking faults, dislocations and grain boundaries, etc.) in the superconducting Fe_2As_2 layer in iron-based superconductors²⁻⁵. The pinning center can also be introduced by artificially irradiating high energy particles, and the resulting defects can vary in shape from point defects to columnar defects⁶⁻⁸. The biggest advantage of using particle irradiation is that it can generate defects at a precisely controlled concentration, allowing us to explore the relationship between pinning energy and defect concentration and defect type.

The parent $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ (herein called Ca10-3-8) compound is semiconducting, not like a typical iron-based superconductor, and is ordered antiferromagnetically without further reduction in crystal symmetry at $T_N \sim 110$ K. The doping effect in the parent compound leads to superconductivity⁹. The electron doping effect of replacing Ca site with La raises the superconducting temperature to $T_c \sim 35$ K¹⁰, but the electron doping effect replacing Fe site with Pt raised T_c to 13 K¹¹, which is weak. The parent compound has multiple Fermi surfaces like other FeAs superconductors, and the subtle difference between these Fermi surfaces induces superconductivity at $T_c = 8$ K without doping¹².

The Ca10-3-8 family has a stacked crystal structure with $\text{Ca}-(\text{Pt}_3\text{As}_8)-\text{Ca}-(\text{Fe}_2\text{As}_2)$ layers, where the Ca layer serves as a reservoir layer for supplying electric charges to the Fe_2As_2 layer, and the Pt_3As_8 layer serves as an intermediate layer controlling the distance between the Fe_2As_2 layers¹³. This crystal structure is similar to the oxide superconductor $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4+x}$ (BSCCO) with an intermediate layer¹⁴. This BSCCO superconductivity has attracted a lot of attention due to the relationship between the magnitude of the superconducting coupling between the superconducting CuO_2 layers, which is determined by the change in the distance between the superconducting layers due to the insertion of the intermediate layer, and the T_c enhancement¹⁵⁻¹⁷. In this context, the Ca10-3-8 family is one of the good candidates to study T_c enhancement in FeAs-superconductors.

For the study of T_c enhancement as well as the delicate relationship between electronic structure and superconductivity, many experiments such as transport^{13,18}, APRES^{12,19}, pressure effect²⁰, infrared spectroscopy²¹⁻²³, upper critical field^{11,18,24}, penetration depth^{25,26} and thermally activated flux motion (TAFM)²⁷ have been performed. Recently, TAFM experiments²⁷ have already been performed for La-

doped Ca₁₀₋₃₋₈ in the $B//c$ -axis direction up to $B=6$ T. The result of TAFM analysis for this experiment showed the magnetic field dependence of $U_0 \sim B^{-\alpha}$ on the zero-temperature activation energy in the entire measured magnetic field region, which was suggested to be due to the entanglement of vortex lines pinned to the point defect. To understand this more clearly in this paper, we performed TAFM experiments in two directions of $B//c$ -axis and $B//ab$ -plane up to 13 T for $\text{Ca}_{8.5}\text{La}_{1.5}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal. In addition, the TAFM experiment was performed under the same conditions in the $\text{Ca}_{8.5}\text{La}_{1.5}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal irradiated with protons. For the $B//c$ -axis, both pristine and proton irradiated samples showed magnetic field dependence on the zero-temperature activation energy of $U_0 \sim B^{-\alpha}$, similar to the previously reported results²⁷. Interestingly, in proton irradiated samples with more point defects, the magnitude of U_0 increased over the entire magnetic field region compared to that in pristine samples. This clearly indicates that the form $U_0 \sim B^{-\alpha}$ is due to point defects. For $B//ab$ -plane, the magnetic field dependence on the zero-temperature activation energy deviated from the form of $U_0 \sim B^{-\alpha}$, and the magnitude of the activation energy was significantly increased compared to the $B//c$ -axis. This indicates that the pinning of the vortex lines was not caused by point defects. Our analysis found that this pinning energy is related to the short coherence length in the c -axis direction. The coherence length in the c -axis direction increased due to the change of the structural and electronic structures by proton irradiation, causing the activation energy to decrease.

RESULTS AND DISCUSSION

XRD and SAED analysis

The XRD results for pristine and proton-irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals are shown in Fig. 1. In both samples, only $(00l)$ -oriented peaks were observed, which indicates that the direction of the incident X-ray is parallel to the c -axis and that the measured sample is a single phase without a secondary phase. Each peak position was the same within the error in the two samples, indicating that the estimated interplanar distance in the two samples was equal to 10.37 Å. This value is almost consistent with the results reported for pristine samples¹⁸. As shown in the inset of Fig. 1, the FWHM (the full width at half maximum) of the pristine sample and the proton-irradiated sample is evaluated at a very narrow 0.0293° and 0.0353° , respectively. This indicates that the pristine single crystal was aligned with almost no structural defects, and that the proton irradiated single crystal had slightly more structural defects than the pristine sample.

We performed crystal structure analysis by selected area electron diffraction (SAED) method using transmission electron microscopy (TEM) to find crystallographic changes between pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals. Figs. 2(a), 2(b) and 2(c), 2(d) show the

experimental results of the SAED analysis for the zone axes [001] and [010] in pristine and proton-irradiated single crystals, respectively. These two patterns for both samples do not show a ring pattern at the center point, indicating that both samples are almost perfect single crystals with no polycrystalline components. Interestingly, as will be discussed below, these two results were not explained by the patterns obtained using the lattice parameters and spatial groups of triclinic structures previously reported²⁸. Instead, this measured SAED pattern is well described as a monoclinic structure with a spatial group of $P2_1/n$, which is similar to the recently published pattern of γ -1048²⁹.

La-doped $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ cannot have a Pt_4As_8 layer as confirmed by EDS analysis using TEM. According to the results of the triclinic structure La-doped $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$, the location of the La atom in the unit cell is located just above and below the Pt vacancies in the $\text{Pt}_3\square\text{As}_8$ layer²⁸. Here, $\square$ means Pt vacancy. La^{3+} , which has a higher charge than Ca^{2+} , is known to prefer this site by lowering its lattice energy when it is located directly above and below the Pt vacancy²⁸. In our compound, the La atom is located almost near the Pt vacancies in the $\text{Pt}_3\square\text{As}_8$ layer, which is expected to have a similar structure to the Pt_4As_8 layer structure, resulting in a change from triclinic to monoclinic crystal structure. In order to obtain more accurate information on the atomic position of the crystal structure, additional experiments with single crystal XRD or high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) are required. In the SAED pattern, diffused stripes appear along the zone axis [010]. These diffused stripes would have been caused by unavoidable sample cross-section damage due to Focused Ion Beam (FIB) in the TEM pretreatment of $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ with a layered structure.

Figs. 2(e) and 2(f) show simulated SAED patterns of pristine and irradiated samples for each zone axis, respectively. The blue color spot represents the simulated SAED pattern for the pristine sample, and the red color one represents the proton irradiated sample. As in the figures, the two spots almost overlap over a large area of the reciprocal lattice, so the two spots are hardly distinguished. However, in Fig. 2(e), an enlarged view for the spot diffracted from the Miller index (660), there is a clear difference between the location of the blue spot for the pristine sample and the location of the red spot for the proton irradiated sample. This small difference arises from the difference in small lattice constants between the two samples. These theoretical simulations agree well with the experimental results indicated by large circles, as shown in the inset in Fig. 2(e). The lattice parameters for the pristine and proton-irradiated samples were obtained by performing simulations on the SAED pattern of each zone axis. The lattice parameters a and b were obtained from the zone axis [001] of each sample, and the lattice parameters a and c were obtained from the zone axis [010]. The lattice parameter a obtained from the zone axis [001] and [010] of each sample was the same. From the results of the simulated pattern, the lattice parameter of the pristine sample was evaluated as $a = 8.727 \pm 0.0122$, $b = 8.776 \pm 0.0096$, $c = 20.800 \pm 0.0083$ Å, $\gamma = 89.05 \pm 0.035^\circ$, and the lattice parameter of the proton irradiated

sample was evaluated as $a = 8.764 \pm 0.0087$, $b = 8.829 \pm 0.0088$, $c = 20.800 \pm 0.0042$ Å, $\gamma = 89.85 \pm 0.026^\circ$. In other words, the lattice parameters a and b for the proton irradiated sample increase by 0.42 and 0.60%, respectively, compared to the pristine sample, but the lattice parameter c does not change. This is a very interesting result. The difference in the constants for the lattice between the two samples was very small, but it was certainly larger than the error for each. The interlayer distance obtained by XRD is obtained by assuming a triclinic structure, but as discussed above, the crystal structure of this compound is a monoclinic structure as a result of SAED analysis. According to the reported γ -1048 compound, the unit cell of the monoclinic structure is a super cell having twice the size of the lattice constant of the c -axis in the triclinic structure²⁹. Therefore, the interlayer distance obtained by XRD is modified to 20.74 Å in a monoclinic structure. This value is similar to the value of the lattice parameter c of the sample obtained from the result of the simulated pattern.

Electrical resistivity in zero magnetic field

Fig. 3 shows the in-plane electrical resistivity near T_c for pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals with zero magnetic field. The abrupt increase due to the superconducting transition was observed in both samples. Compared to the pristine sample, as can be seen in the inset, the normal-state electrical resistivity of the proton irradiated sample is large just above T_c , but becomes the same in the higher temperature region. A similar increase in the limited temperature range just above T_c is observed in La/Pt-underdoped Ca 10-3-8 samples^{18,22}. As the temperature approaches T_c in the normal state, the electrical resistivity bends down as shown in the figure, so we evaluate T_c^{on} and ΔT_c as follows: T_c^{on} is defined as the temperature at which the linearly extrapolated line in the temperature range where the electrical resistivity suddenly increases from zero and the linear extrapolated line performed in the temperature range where the electrical resistivity changes linearly in the high temperature normal state intersect. From this definition, the superconducting transition temperature was evaluated as $T_c^{\text{on}} = 32.7$ K in the pristine sample and $T_c^{\text{on}} = 30.8$ K in the proton irradiated sample. The T_c^{on} of the pristine sample is almost similar to the reported results¹⁸. Meanwhile, the temperature width ΔT_c of T_c was determined from $2 \cdot [0.5 \cdot \rho(T_c^{\text{on}}) - 0.1 \cdot \rho(T_c^{\text{on}})]$. As a result, the width was evaluated as $\Delta T_c = 0.46$ K in the pristine sample and $\Delta T_c = 0.79$ K in the proton irradiated sample. ΔT_c of the pristine sample is very narrow, indicating that the sample has very few defects such as the secondary phase. Compared to the pristine sample, the proton-irradiated sample showed slightly larger ΔT_c . This increase was not caused by the appearance of the secondary phase because no small peak due to the secondary phase was observed in the XRD experiment results. Instead, since the FWHM for the observed peaks in the XRD pattern slightly increased, this increase in ΔT_c is thought to be due to crystallographic distortion. However, since the electrical resistivity of the proton-irradiated sample is

almost the same as that of the pristine sample at high temperature normal state, the crystallographic distortion that occurs in the proton-irradiated sample is very small.

As described above, the T_c of the proton-irradiated sample was lowered by about 2 K compared to the pristine sample. It is known that the decrease in T_c occurs due to the spin flip scattering of the electron pair and the magnetic defect³⁰. Proton irradiation causes distortion of the lattice, but does not excite nonmagnetic ions as magnetic ions. Since lattice defects do not cause spin flip scattering of the electron pair, this causes a small decrease in T_c , not the large decrease in T_c observed here. Another cause of the large decrease in T_c may be the increase in the lattice constants of a and b discussed above. As discussed in the SAED results above, proton irradiation increased the lattice constants a and b by 0.42 and 0.60%, respectively. The increase in the lattice constants a and b is expected to reduce the overlap between the bands, resulting in a decrease in the concentration of doped electrons and to further enhance the localization of the $3d$ -electron orbit. The former will cause an underdoping effect, resulting in a decrease in T_c , and the latter will cause an increase in electrical resistivity in the normal state just above T_c as in the underdoped Ca 10-3-8 samples. Since these two behaviors were observed in the proton irradiated sample, the large decrease in T_c observed in the proton irradiated sample can be understood as an increase in lattice parameters a and b .

Electrical resistivity under magnetic field and the higher critical field $B_{c2}(T)$ curves

Figs. 4(a)-4(d) show the temperature dependence of the in-plane electrical resistivity measured with $B//c$ and $B//ab$ for the pristine and proton-irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals, respectively, on a semi-logarithmic scale. As the magnetic field increases, the electrical resistivity near T_c for both magnetic fields of both samples exhibits a typical broadening, which is known to be due to the thermally activated vortex motion occurring in the vortex liquid. This broadening occurs more strongly in a magnetic field parallel to the c -axis. On the other hand, the first-order phase transition from the vortex liquid to the vortex lattice was not observed up to low temperatures because abruptly changes in the temperature dependence of the electrical resistivity under the magnetic field were not observed up to low temperatures. As shown in the figure, the electrical resistivity fluctuates at a value less than $\rho \sim 4 \cdot 10^{-7} \Omega \cdot \text{cm}$, which is caused because the electrical resistivity of the sample approaches zero and becomes smaller than the minimum value that can be accurately measured.

The temperature dependence of the higher critical fields $B_{c2}^{90\%}$ and $B_{c2}^{50\%}$ in $B//c$ and $B//ab$ is determined by the 90 and 50% criteria in the electrical resistivity data of pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals, and the results are shown in Figs. 5(a)-5(d), respectively. The $B_{c2}^{90\%}$ and $B_{c2}^{50\%}$ curves determined in each magnetic field direction for each sample show weak upward curvature in the low magnetic field region. This behavior is often observed in high- T_c

superconductors and is known to be due to a weak link of proximity type between superconducting grains. $B_{c2}(0)$ is generally evaluated using the slope of $B_{c2}^{90\%}$, which is less sensitive to flux motion, but for our samples, the determination of $B_{c2}(0)$ from $B_{c2}^{90\%}$ is expected to cause a large error because a large rounding shape in the electrical resistivity is observed even at $B = 0$ in the temperature range just above T_c . In order to reduce the error due to this rounding effect and the weak link between the superconducting grains, $B_{c2}(0)$ was determined from $B_{c2}^{50\%}$ above $B = 2$ T. The orbital limited higher critical field $B_{c2}(0)$ was determined by applying the Werthamer-Helfand-Hohenberg (WHH) expression, $B_{c2}(0) = -0.693T_c(dB_{c2}/dT)T_c$, to $B_{c2}^{50\%}$ above $B = 2$ T: $B_{c2,c}(0) = 77.0$ T, $B_{c2,ab}(0) = 631.1$ T for pristine samples and $B_{c2,c}(0) = 74.6$ T, $B_{c2,ab}(0) = 197.6$ T for the proton irradiated sample. The magnitude of $B_{c2}(0)$ for pristine samples is 30-50% larger than that reported¹⁸. The in-plane Ginzburg-Landau coherence length $\xi_{ab}(0)$ is evaluated from the equation $\xi_{ab}(0) = \left(\frac{\phi_0}{2\pi B_{c2,c}(0)}\right)^{1/2}$, and the out-of-plane Ginzburg-Landau coherence length $\xi_c(0)$ is evaluated from the determined value of $\xi_{ab}(0)$ and the equation $\xi_c(0) = \frac{\phi_0}{2\pi B_{c2,ab}(0)\xi_{ab}(0)}$, where ϕ_0 is the magnetic flux quantum, 2.07×10^{-15} Wb: $\xi_{ab}(0) = 20.7$, $\xi_c(0) = 2.5$ Å for pristine samples, and $\xi_{ab}(0) = 21.0$, $\xi_c(0) = 7.9$ Å for proton irradiated samples. The magnitude of anisotropy evaluated from these coherence lengths was $\gamma \sim 8.3$ in the pristine sample and $\gamma \sim 2.7$ significantly decreased in the proton irradiated sample. The decrease in anisotropy in the proton irradiated sample is due to a three-fold increase in $\xi_c(0)$. In a single crystal, proton irradiation causes a decrease in the mean free path due to scattering caused by the distortion of the lattice. The coherence length simply decreases as well due to the decrease in the mean free path, but as discussed above, the out-of-plane coherence length $\xi_c(0)$ in the proton irradiated sample, on the contrary, increased significantly and thus this increase is not understood by the change of the mean free path.

When La is optimally doped to the parent Ca 10-3-8, lattice constants a and b are almost invariant and c increases by 0.45%. On the other hand, when Pt is optimally doped, the lattice constants a , b , and c increase by almost 0.25%. This increase in these samples was caused by the doping of La/Pt with a large ion radius, which would cause a change in the distance between Fe-As ions and the angle between As-Fe-As ions in the conducting Fe_2As_2 layer. The distance and angle in the Fe_2As_2 layer will change the magnitude of the overlap between the Fe-3d wavefunctions and the hybridization between the Fe-3d wavefunctions and the As-4p wavefunctions. In the proton-irradiated sample, the lattice constant c remains unchanged, but a and b increase by 0.42-0.6%. Inferred from the increase of the lattice constant in Pt-doped Ca 10-3-8, this increase is the same as the increase when the parent Ca 10-3-8 is doped with a much larger amount of Pt than in the optimal doping, which suggests that the electron band structure and anisotropy, etc., may change significantly. Note that the anisotropy changes more than three times between the Pt underdoped Ca 10-3-8 and the optimally doped one. Unlike Ca 10-3-8 doped with Pt/La, proton-irradiated samples show an increase in the distance between ions without doping with large ions.

This clearly reduces the magnitude of the overlap between the Fe-3d wavefunctions in the Fe₂As₂ layer. An increase in low-temperature electrical resistivity caused by the reduced overlap was observed in the proton-irradiated sample. This can be inferred from the fact that the Pt-underdoped Ca 10-3-8 system, which shows a strong localization of the Fe-3d wavefunction and an increase in the low-temperature electrical resistivity, exhibits larger anisotropy than the Pt-optimal doping system, which shows less localization. However, compared to pristine samples, proton-irradiated samples showed three times less anisotropy. According to the band theory³¹, band hybridization induces the three-dimensionality of the carrier by reducing the localization of the band. Based on this, this small anisotropy in the proton irradiation sample is due to a large change in the magnitude of hybridization between the Fe-3d wavefunction and the As-4p wavefunction. In pristine and proton-irradiated samples, the lattice constant c was constant, but the angle γ in the unit cell was changed by $\sim 1^\circ$ as discussed previously. This change in angle γ can change the distance between Fe-As and the angle between As-Fe-As, resulting in a change in the hybridization magnitude between the Fe-3d wave function and the As-4p wave function. Therefore, the decrease in the anisotropy in the proton-irradiated sample is considered to be caused by the change in the electronic structure determined by the hybridization between the Fe-3d wavefunction and the As-4p wavefunction and the overlap between the Fe-3d wavefunction having the opposite effect. Accurate band calculation is required to make this clear.

Thermally activated flux motion in the vortex liquid state

The temperature dependence of the in-plane electrical resistivity measured under the magnetic field for the pristine and proton irradiated (Ca_{0.85}La_{0.15})₁₀(Pt₃As₈)(Fe₂As₂)₅ single crystals shown in Figs. 4(a)-4(d) is understood as thermally activated flux motion (TAFM) in the vortex liquid state as described above. According to TAFM theory, the electrical resistivity is given by

$$\rho(T) = (2\rho_c U(B, T)/T) \exp(-U(B, T)/T). \quad (1)$$

In this equation, the prefactor $2\rho_c U(B, T)/T$ is precisely temperature dependent, but since it has been assumed to be temperature constant in many high- T_c superconductor analyzes, the prefactor is assumed to be a temperature constant in this study. In order to physically understand the flux motion of this activation type, it is first necessary to determine whether or not the activation energy changes linearly with temperature. The linearly temperature dependent activation energy is given by $U(T, B) = U_0(B)(1 - t)$, where $t = T/T_c$. This means that $U_a(T) = -\partial \ln \rho / \partial (1/T)$ is constant over a limited temperature range that satisfies the linear activation energy. As shown in Figs. 6a-1)-6a-4), there is no temperature range showing a constant $U_a(T)$, but rather $U_a(T)$ shows a nonlinear change. This means that the activation energy linearly dependent on the temperature is not suitable for our two samples. Therefore, in our two samples, we intend to analyze using a nonlinear temperature-dependent activation

energy $U(T, B) = U_0(B)(1 - t)^q$, which was previously performed on high- T_c superconductors by Zhang et al.^{32,33}. Using the nonlinear activation energy, the logarithm of Eq. (1) is given by

$$\begin{aligned} D(T) &= \ln \rho(T) \\ &= \ln(2\rho_c U_0(B)) + q \ln(1 - t) - \ln T - U_0(B)(1 - t)^q / T, \end{aligned} \quad (2)$$

and then the derivative of Eq. (2) by $1/T$ is expresses as

$$\begin{aligned} U_a(T) &= -\partial \ln \rho / \partial (1/T) \\ &= [U_0(B)(1 - t)^q - T] [1 + q t / (1 - t)]. \end{aligned} \quad (3)$$

For the pristine and proton irradiated samples, $U_a(T)$ calculated from the data in the directions of $B//c$ and $B//ab$ at $B = 13$ T agrees well with the red line fitted with Eq. (3) over a limited temperature range as shown in Figs. 6a-1)-6a-4), respectively. The best fit for different magnetic fields is shown by red lines in Figs. 7(a)-7(d). From the best fittings in both magnetic field directions, $T_c = 32.3$ K and $q = 1.52$ were achieved for the pristine sample, and $T_c = 30.1$ K and $q = 1.52$ for the proton irradiated sample. Also, the zero-temperature activation energy $U_0(B)$ in each magnetic field evaluated by these fittings is shown in Fig. 8. The best fitting using Eq. (2) with the values of T_c , q , and U_0 obtained from the $U_a(T)$ fitting at $B = 13$ T performed just above is shown as a red line in Figs. 6b-1)-6b-4). ρ_c was determined from this fitting. In both fittings, the temperature range consistent with the experimental results is almost the same. The fitting results using Eq. (2) in different magnetic fields are drawn with red lines in Figs. 9(a)-9(d). The $\rho_c(B)$ determined in this fitting is shown in the inset in Fig. 8, and $\rho_c(B)$ increases rapidly in the low magnetic field region and then almost saturates above $B = 2$ T.

As shown in Figs. 6a-1)-6a-4), experimentally calculated $U_a(T)$ in both magnetic field directions for pristine and irradiated samples deviates from the red line analyzed with TAFM below T^* and fluctuates significantly in the lower temperature region. This fluctuation is due to a measurement error that occurs because the electrical resistance of the sample is very close to zero and exceeds our measurement limit. This fluctuation indicates that these two samples possess a second-order phase transition from the vortex liquid phase to the vortex glass phase. From this point of view, the gradual deviation of $U_a(T)$ obtained by the experiment to the larger side compared to the TAFM model below T^* is due to the development of vortex glass gradually appearing as the temperature decreases in the vortex liquid state. In this temperature region, the activation energy increases effectively due to the development of vortex glass, and this region is called a critical region. According to the vortex glass theory, which quantitatively explains the temperature dependence of the electrical resistivity in this critical region, the electrical resistivity satisfies $\rho(T) \sim |T - T_g|^{\nu(z+2-d)}$ and disappears completely

at T_g , where d is the dimensionality of the sample ($d = 3$ in both samples), ν is a static index for the vortex-glass correlation length, and z is a dynamic index for the correlation time. The derivative of the logarithmic electrical resistivity with temperature in this region shows a linear temperature dependence, that is,

$$R(T) = \left(\frac{d \ln \rho(T)}{dT} \right)^{-1} = \frac{1}{s} (T - T_g), \quad (4)$$

where $s = \nu(z+2-d)$ is the reciprocal of the slope of Eq. (4). For the pristine and proton irradiated samples, $R(T)$ calculated from the experimental data in the directions of $B//c$ and $B//ab$ at $B = 13$ T is shown in Figs. 6c-1)-6c-4), respectively. As shown in the figure, a linear change of $R(T)$ was observed in the temperature range below T^* , indicating that it was well explained by the vortex glass theory. $R(T)$ in different magnetic fields is shown in Figs. 10(a)-10(d), and linear changes in each magnetic field were observed in the low temperature range. The best fit obtained using Eq. (4) is also shown as green lines in Figs. 10(a)-10(d). These fittings agree well with the experimental results up to T^* . $R(T)$ calculated from the TAFM model discussed above is drawn with red lines in Figs. 6c-1)-6c-4). It showed good agreement with the experiment in the limited temperature range above T^* . Therefore, T^* becomes the temperature that divides the TAFM temperature region and the critical temperature region. From the fitting using Eq. (4) [Figs. 10(a)-10(d)] the vortex glass temperature T_g and critical exponent s were determined. The magnetic field dependence of T_g , which is termed $B_g(T)$, and s are shown in Figs. 5(a)-5(d) and 12(a)-12(d), respectively.

According to Liu et al., the thermal activation energy in the critical region is given by $U_{\text{eff}} = k_B T (T_c - T)/(T_c - T_g)$, and the temperature dependence of the electrical resistivity by this activation energy is given by $\rho(T) = \rho_n [\exp(U_{\text{eff}}/k_B T)]^s$, which is consistent with the Eq. (4) obtained by the vortex-glass theory^{34,35}. When the electrical resistivity $\rho(T)$ is plotted on the new temperature scale $(T - T_g)/(T_c - T_g)$ in the critical region, according to Eq. (4), $\rho(T)$ is scaled to one in all magnetic fields if s is constant with respect to the magnetic field change. Figs. 12(a)-12(d) show the electrical resistivity in each magnetic field direction for each sample on a temperature scale of $(T - T_g)/(T_c - T_g)$, respectively. Here, for T_c and, values obtained from the fittings discussed above were used. For the magnetic field direction of $B//ab$ in the pristine sample and the magnetic field direction of $B//c$ in the proton irradiated sample, $\rho(T)$ scales well to $(T - T_g)/(T_c - T_g)$, indicating that s has little dependence on the magnetic field. The s value obtained from the slope of the graph in the critical region is plotted as a red circle in Figs. 11(a)-11(d). As shown in the figures, it shows good agreement with the s value determined by the vortex glass model. $U_d(T)$ and $D(T)$ calculated from $\rho(T) = \rho_n [\exp(U_{\text{eff}}/k_B T)]^s$ along with the values obtained for s , T_c , and T_g in each magnetic field

are shown by green lines in Figs. 7(a)-7(d) and Figs. 9(a)-9(d), respectively. These lines agree well with the experimental results in the temperature range below T^* .

Next, let's discuss the magnetic field dependence of the zero-temperature activation energy $U_0(B)$. As shown in Fig. 8, $U_0(B)$ for the pristine and proton-irradiated samples in the magnetic field direction of $B//c$ decreases linearly with increasing magnetic field on a log-log scale in the entire measured magnetic field region. That is, it decreases in the form of $U_0(B) \sim B^{-\alpha}$. In both samples, the exponent is almost the same as $\alpha = 1.21 - 1.22$. These determined $U_0(B)$ and α in $B//c$ are almost similar to those obtained by TAFM analysis up to $B = 6$ T in pristine $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal. Compared with iron-based superconductors showing similar T_c , the magnitude of $U_0(B)$ in our sample is significantly larger, which means that strong pinning occurred in our sample^{27,36-38}. Since the coherence length in the direction perpendicular to the c -axis is about 20 Å, which is about 2.5 times larger than the lattice constants a and b , as discussed above, the normal component region with zero Cooper pair does not exist in the ab -plane due to the large coherence length. Therefore, the normal components that pin the vortex in the pristine sample are atomic point defects such as impurity atoms and vacancies present in the crystal as already discussed in the XRD result. In the proton-irradiated sample, the normal components pinning the vortex are new point defects created by proton irradiation in addition to such atomic defects. The vortex pinning due to random point defects is according to the plastic creep theory. According to this theory, the vortex lines weakly pinned to point defects in the vortex liquid becomes entangled, so that the zero activation energy is dependent on $U_0(B) \sim B^{-\alpha}$ with $\alpha = 0.5$. Due to the relative motion that occurs between the entangled vortex lines, the vortex lines will be cut off or recombined and eventually change to a new state of vortex motion. This new motion changes the magnetic field dependence of $U_0(B)$ and the exponent α changes around 0.5. As an example, untwinned YBCO showed $\alpha \sim 0.7$ by vortex entanglement in weakly pinned vortex liquid. Meanwhile, it is known that the magnetic field dependence of $U_0(B)$ in strongly pinned vortex liquid becomes stronger and has a larger exponent α . In $B//c$, both samples show the dependence of $U_0(B) \sim B^{-\alpha}$, and the exponent α has a larger value than the observed value in YBCO. Therefore, the magnetic field dependence of the activation energy observed in both samples at $B//c$ was caused by the movement of vortex lines strongly pinned by point defects. The magnitude of the activation energy in the proton-irradiated sample increased by about 15% compared to that in the pristine sample. Since the magnitude of the coherence length in the ab plane is almost the same in both samples, the difference in activation energy is due to pinning of point defects caused by proton irradiation.

Both pristine sample and proton-irradiated sample in $B//ab$ showed significantly larger zero-temperature activation energy $U_0(B)$ in the entire measured magnetic field region compared to those in $B//c$. In particular, the increase in $U_0(B)$ for the pristine sample is much larger. The coherence length ξ_c in both samples discussed above is significantly smaller than the distance between the superconducting

Fe₂As₂ layers ($d = 10.4 \text{ \AA}$), and in particular, is smaller in the pristine sample. This short ξ_c creates a normal region with zero Cooper pairs along the c -axis in both samples. The normal region forms a long plate shape parallel to the ab -plane. Therefore, vortex pinning in $B//ab$ is caused by a normal component region formed by a short coherence length ξ_c along with impurity atoms, vacancies, and point defects caused by proton irradiation, as in $B//c$. According to the theory of normal component pinning caused by short coherence length³⁹, the pinning of vortex lines by the long normal component is stronger than that by atomic defects or point defects created by proton irradiation. The vortex pinning of the normal component created by the short coherence length becomes stronger the shorter the coherence length is. Since the coherence length ξ_c in both samples is sufficiently small compared to the distance between the superconducting Fe₂As₂ layers, the vortex pinning by this normal component is expected to be sufficiently large. Therefore, since pinning by point defects can be neglected due to the strong vortex pinning of the normal component, the linear magnetic field dependence of $U_0(B)$ due to entanglement between vortex lines occurring in point defects will not appear in our both samples. In fact, as shown in Fig. 8, $U_0(B)$ in $B//ab$ is bent downward in the high magnetic field region. According to this pinning mechanism in $B//ab$, we can infer that the coherence length increases in the high magnetic field from the bending of $U_0(B)$ in the high magnetic field. A detailed study of this is needed. In both samples, $U_0(B)$ in $B//ab$ increased significantly than $U_0(B)$ in $B//c$ axis, but this increase was less significant in proton irradiated samples compared to pristine samples. This is due to the increased coherence length ξ_c in the proton irradiated sample. Note that increasing the coherence length decreases the ratio of the normal component of the plate shape.

As discussed above, $U_0(T)$ in the vortex liquid depends on the temperature power law, i.e. $U(T, B) = U_0(B)(1 - t)^q$. The exponent q was determined to be 1.52 for both magnetic field directions in the pristine and proton-irradiated samples. Note that $q = 1.5$ is observed in 3-dimensional high- T_c superconductor and $q = 2$ is observed in 2-dimensional high- T_c superconductor. The pristine sample showed anisotropy $\gamma \sim 8.3$ and the proton irradiated sample showed $\gamma \sim 2.7$, but both samples formed 3D vortex without forming 2D-disk type vortex in vortex liquid state.

The magnetic field dependence of s representing the vortex glass dimensions determined in the $B//c$ and $B//ab$ directions for pristine and proton-irradiated samples is shown in Figs. 11(a)-11(d), respectively. In both magnetic field directions for both samples, s is greater than 2.7 in the entire measured magnetic field region, indicating that a three-dimensional vortex glass is formed. Each s shows a value for three-dimensional vortex glass, but s for $B//c$ in pristine samples and $B//ab$ in proton-irradiated samples shows a distinct magnetic field dependence. On the other hand, s for $B//ab$ in pristine sample and $B//c$ in proton-irradiated sample have almost constant values for the change of magnetic field. In particular, s in the $B//c$ direction for the pristine sample decreases as the magnetic field increases, and approaches 2.7 that appears in 2D glass above $B = 4 \text{ T}$. To understand this complex magnetic field

dependence, a supplementary study on static index ν for vortex-glass correlation length and dynamic index z for correlation time is needed.

In pristine and proton-irradiated samples, $B(T)$ phase diagrams for $B//c$ and $B//ab$ directions are shown in Figs. 5(a)-5(d), respectively. Each $B(T)$ phase diagram is largely divided into four different regions. The vortex glass-to-vortex liquid transition takes place in $B_g(T)$ and is divided into vortex glass and vortex liquid regions. This vortex glass line is well represented by $B_g(T) = B_0(1 - T/T_c)^m$ where B_0 is the zero-temperature vortex glass field and m is the exponent. The best fitting from this equation is shown as green lines in Figs. 5(a)-5(d). The parameters determined in the pristine sample are $B_0 = 45.2$, $m = 1.7$ for $B//c$ and $B_0 = 279.52$, $m = 1.6$ for $B//ab$. On the other hand, the parameters determined in the proton-irradiated sample are $B_0 = 40.7$, $m = 1.6$ for $B//c$, and $B_0 = 234.4$, $m = 1.7$ for $B//ab$. The evaluated m is similar to the values determined for cuprates and iron-based superconductors. The vortex liquid region is divided by $B^*(T)$ into a critical region in contact with T_g and a thermally activated flux flow region. The thermally activated flux flow region and the unpinned vortex liquid region were separated by a wine-colored dashed line. This separation line is obtained from the higher temperature that deviates between the results using the TAFM model and the experimental data.

CONCLUSION

We grew optimally doped $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals and measured in-plane electrical resistivity with $B//c$ and $B//ab$ to study the difference between vortex pinning in pristine and proton-irradiated samples. As a result of analyzing the crystal structure of the two samples by the selected area electron diffraction (SAED) method using TEM, the diffraction patterns of both samples were not explained by the known triclinic structure but were explained by the monoclinic crystal structure. In addition, in the lattice structure, the lattice constants of a and b in the proton-irradiated sample increased compared to that of the pristine sample, but the lattice constant of c did not change. The increase in the lattice constant caused a change in their electronic structure due to the change in the overlap effect between Fe-3d wavefunctions and the hybridization effect of Fe-3d wavefunction and As-4p. This change showed a significant difference in superconducting transition temperature and anisotropy between the two samples. The two samples are divided into a vortex glass region showing zero electrical resistivity and a vortex liquid region showing electrical resistivity values. This vortex liquid region is divided into a critical region, a pinned vortex liquid region, and an unpinned vortex liquid region. The critical region has a large activation energy due to the development of vortex glass, and its activation energy is expressed as $U_{\text{eff}} = k_B T (T_c - T)/(T_c - T_g)$. Meanwhile, the pinned vortex liquid region is described by the thermally activated flux motion (TAFM) with an activation

energy $U(T, B) = U_0(B)(1 - t)^q$ that is nonlinearly dependent on temperature. In the unpinned vortex liquid region, there is no restriction on the flux motion due to the activation energy that is small compared to temperature. The magnetic field dependence of the zero-temperature activation energy in the $B//c$ direction determined by TAFM motion in the pinned vortex liquid region shows the form of $U_0(B) \sim B^{-\alpha}$ with $\alpha = 1.21 - 1.22$ due to the entanglement of flux lines strongly pinned to point defects in both pristine and proton-irradiated samples. On the other hand, $U_0(B)$ for both samples in the direction of $B//ab$ increases significantly over the entire measured magnetic field region compared to that in $B//c$, and decreases non-linearly with respect to magnetic field changes at high magnetic fields. This is due to the vortex pinned from the long normal component formed along the c -axis direction by the short coherent length ξ_c .

METHODS

Single-crystals growth

$(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals were grown by a Bridgman method. In order to accurately make a compound of the desired molar ratio with arsenic having a gaseous phase and Ca, La, Pt, and Fe having a liquid phase at high temperature, the precursors of CaAs, LaAs and FeAs was synthesized at 550, 800 and 1050 °C in evacuated quartz ampoules, respectively. The powdered precursors and Pt element were mixed in a mortar and then placed in a BN crucible. In order to prevent the evaporation of each element, especially arsenic, into a vapor state at high temperature, the filled BN crucible was put into a Mo-crucible and covered with a lid, and the contact part between the crucible and the lid was welded in a high purity Ar-gas atmosphere using an arc welding machine. The welded Mo-crucible was placed in an electric furnace with a tungsten mesh heater with ± 0.1 °C temperature stability, and a single crystal was grown at 1250 °C.

Experimental conditions of proton irradiation

Proton irradiation along the c -axis of the single crystal was performed at 300 K using an MC-50 cyclotron installed at KIRAM (Korea institute of radiological & medical sciences). The proton energy is 3 MeV and the total dose of the incident proton is $5 \times 10^{15} \text{ cm}^{-2}$. To prevent the temperature-rise of the sample during proton irradiation, we fixed the specimen to a water copper block and used a weak proton dose rate of $\sim 2 \times 10^{11} \text{ cm}^{-2} \cdot \text{s}^{-1}$.

Electrical transport measurements under magnetic fields

Using the grown single crystal, the in-plane electrical resistivity in $B//c$ and $B//ab$ using grown single crystals were measured by the four-probe method at a current density of about 20 A/cm² using the Oxford superconducting system. The gold wires were attached to the sample electrodes using silver epoxy (EPO-TEK H20E). Gold was deposited in advance on the sample surface for the probes to reduce the contact resistance below 1 Ω . The electrical resistivity was measured from 2 to 85 K with $B = 0.5, 1, 2, 3, 5, 7, 9, 11$ and 13 T.

X-ray diffraction

The X-ray diffraction experiments were performed using a PANalytical X-ray powder diffractometer with Cu K α radiation (40 kV, 30 mA and $\lambda = 1.5406$ Å) at step size of 0.026° (2 θ) and scan rate of 0.78°/min.

Measurements and analysis of SAED patterns

Selected area electron diffraction (SAED) patterns were measured at 300 kV accelerating voltage using a high-resolution transmission electron microscope (HR-TEM, Hitachi HF-3300). As a pretreatment process for HR-TEM, the thickness of all samples was processed to less than 100 nm using a dual-beam focused ion beam (dual-beam FIB, Hitachi NB 5000) under an acceleration voltage of 40 kV. To analyze the SAED pattern, the ReciPro program was used. All spots of the pattern were fitted with the ReciPro program to obtain d -spacing of the sample and the angle between the spots⁴⁰. From these fitting results, we found a pattern that closely matches the experimental SAED pattern based on the crystal structures and space groups of Ca 10-3-8²⁸ and Ca 10-4-8²⁹, which are known to date. The lattice constants of all samples were obtained by refinement using UNITCELL software⁴¹. The simulated SAED pattern was obtained using dynamical theory and simulated by adjusting both the Bloch waves and thickness parameters.

DATA AVAILABILITY

The x-ray diffraction data, selected area electron diffraction (SAED) data and magnetoresistivity data used in this work are available upon request from the corresponding author.

References

1. Huse, D. A., Fisher, M. P. A. & Fisher, D. S. Are superconductors really superconducting? *Nature* **358**, 553–559 (1992).

2. Dai, P. Antiferromagnetic order and spin dynamics in iron-based superconductors. *Rev. Mod. Phys.* **87**, 855–896 (2015).
3. Kalisky, B. *et al.* Behavior of vortices near twin boundaries in underdoped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$. *Phys. Rev. B* **83**, 064511 (2011).
4. Hänisch, J. *et al.* High field superconducting properties of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ thin films. *Sci. Rep.* **5**, 17363 (2015).
5. Iida, K., Hänisch, J. & Yamamoto, A. Grain boundary characteristics of Fe-based superconductors. *Supercond. Sci. Technol.* **33**, 043001 (2020).
6. Kwok, W. K. *et al.* Vortices in high-performance high-temperature superconductors. *Rep. Prog. Phys.* **79**, 116501 (2016).
7. Mizukami, Y. *et al.* Disorder-induced topological change of the superconducting gap structure in iron pnictides. *Nat. Commun.* **5**, 5657 (2014).
8. Eisterer, M. Radiation effects on iron-based superconductors. *Supercond. Sci. Technol.* **31**, 013001 (2018).
9. Stürzer, T., Stürzer, C. & Johrendt, D. Iron arsenide superconductors $(\text{CaFeAs})_{10}\text{MnAs}_8$ with metallic interlayers ($\text{M} = \text{Pt, Pd}$; $n = 3, 4$). *Phys. Status Solidi B* **254**, 1600417 (2017).
10. Stürzer, T., Derondeau, G., Bertschler, E. M. & Johrendt, D. Superconductivity by rare earth doping in the 1038-type compounds $(\text{Ca}_{1-x}\text{RE}_x)_{10}(\text{FeAs})_{10}(\text{Pt}_3\text{As}_8)$ with $\text{RE} = \text{Y, La-Nd, Sm-Lu}$. *Solid State Commun.* **201**, 36–39 (2015).
11. Xiang, Z. J. *et al.* Transport properties and electronic phase diagram of single-crystalline $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_{1-x}\text{Pt}_x)_2\text{As}_2$. *Phys. Rev. B* **85**, 224527 (2012).
12. Neupane, M. *et al.* Fermi-surface topology and low-lying electronic structure of the iron-based superconductor $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$. *Phys. Rev. B* **85**, 094510 (2012).
13. Ni, N., Allred, J. M., Chan, B. C. & Cava, R. J. High T_c electron doped $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ and $\text{Ca}_{10}(\text{Pt}_4\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ superconductors with skutterudite intermediary layers. *Proc. Natl. Acad. Sci. U. S. A.* **108**, E1019 (2011).
14. Damascelli, A., Hussain, Z. & Shen, Z. X. Angle-resolved photoemission studies of the cuprate superconductors. *Rev. Mod. Phys.* **75**, 473 (2003).
15. Sterne, P. A. & Wang, C. S. Higher T_c through metallic inter-layer coupling in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$. *J. Phys. C Solid State Phys.* **21**, L949 (1988).

16. Parkin, S. S. P. *et al.* Bulk superconductivity at 125 K in $\text{Ti}_2\text{Ca}_2\text{Ba}_2\text{Cu}_3\text{O}_x$. *Phys. Rev. Lett.* **60**, 2539 (1988).
17. Gao, L. *et al.* Study of superconductivity in the Hg-Ba-Ca-Cu-O system. *Phys. C* **213**, 261 (1993).
18. Ni, N. *et al.* Transport and thermodynamic properties of $(\text{Ca}_{1-x}\text{La}_x)_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ superconductors. *Phys. Rev. B* **87**, 060507(R) (2013).
19. Thirupathaiah, S. *et al.* Why T_c of $(\text{CaFeAs})_{10}\text{Pt}_{3.58}\text{As}_8$ is twice as high as $(\text{CaFe}_{0.95}\text{Pt}_{0.05}\text{As})_{10}\text{Pt}_3\text{As}_8$. *Phys. Rev. B* **88**, 140505(R) (2013).
20. Gao, P. *et al.* Pressure-Induced Superconductivity and Its Scaling with Doping-Induced Superconductivity in the Iron Pnictide with Skutterudite Intermediary Layers. *Adv. Mater.* **26**, 2346–2351 (2014).
21. Seo, Y. I., Choi, W. J., Kimura, S. I., Bang, Y. & Kwon, Y. S. Optical properties of optimally doped single-crystal $\text{Ca}_{8.5}\text{La}_{1.5}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$. *Phys. Rev. B* **95**, 094510 (2017).
22. Seo, Y. I., Choi, W. J., Ahmad, D., Kimura, S. I. & Kwon, Y. S. Temperature dependence of the superconducting energy gaps in $\text{Ca}_{9.35}\text{La}_{0.65}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal. *Sci. Rep.* **8**, 8648 (2018).
23. Yang, R. *et al.* Unravelling the mechanism of the semiconducting-like behavior and its relation to superconductivity in $(\text{CaFe}_{1-x}\text{Pt}_x\text{As})_{10}\text{Pt}_3\text{As}_8$. *Phys. Rev. B* **99**, (2019).
24. Watson, M. D. *et al.* Field-induced magnetic transitions in $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)[(\text{Fe}_{1-x}\text{Pt}_x)_2\text{As}_2]_5$ compounds. *Phys. Rev. B* **89**, 205136 (2014).
25. Kim, J. *et al.* Large magnetic penetration depth and thermal fluctuations in a superconducting $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)[(\text{Fe}_{1-x}\text{Pt}_x)_2\text{As}_2]_5$ ($x=0.097$) single crystal. *Phys. Rev. B* **85**, 180504(R) (2012).
26. Cho, K. *et al.* Doping-dependent superconducting gap anisotropy in the two-dimensional pnictide $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)[(\text{Fe}_{1-x}\text{Pt}_x)_2\text{As}_2]_5$. *Phys. Rev. B* **85**, 020504(R) (2012).
27. Choi, W. J., Seo, Y. I., Ahmad, D. & Kwon, Y. S. Thermally activated flux motion in optimally electron-doped $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ and $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)[(\text{Fe}_{0.92}\text{Pt}_{0.08})_2\text{As}_2]_5$ single crystals. *Results Phys.* **19**, 103430 (2020).
28. Stürzer, T., Derondeau, G. & Johrendt, D. Role of different negatively charged layers in $\text{Ca}_{10}(\text{FeAs})_{10}(\text{Pt}_4\text{As}_8)$ and superconductivity at 30 K in electron-doped $(\text{Ca}_{0.8}\text{La}_{0.2})_{10}(\text{FeAs})_{10}(\text{Pt}_3\text{As}_8)$. *Phys. Rev. B* **86**, 060516(R) (2012).

29. Wang, Z. *et al.* Atomically imaged crystal structure and normal-state properties of superconducting $\text{Ca}_{10}\text{Pt}_4\text{As}_8((\text{Fe}_{1-x}\text{Pt}_x)_2\text{As}_2)_5$. *Phys. Rev. B* **100**, 94103 (2019).
30. Bulaevskil, L. N., Guselnov, A. A. & Rusinov, A. I. Superconductivity in crystals without symmetry centers. *Sov. Phys. JETP* **44**, 1243–1251 (1977).
31. Majid, A., Batool, A., Khan, S. U. D. & Ahmad, A. First-principles study of f-orbital-dependent band topology of topological rare earth hexaborides. *Int. J. Quantum Chem.* **121**, e26452 (2021).
32. Zhang, Y. Z., Wen, H. H. & Wang, Z. Thermally activated energies of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{Y}_{0.8}\text{Ca}_{0.2}\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films. *Phys. Rev. B* **74**, 144521 (2006).
33. Zhang, Y. Z., Ren, Z. A. & Zhao, Z. X. Thermally activated energy and critical magnetic fields of $\text{SmFeAsO}_{0.9}\text{F}_{0.1}$. *Supercond. Sci. Technol.* **22**, 065012 (2009).
34. Fisher, M. P. A. Vortex-glass superconductivity: A possible new phase in bulk High-T_c oxides. *Phys. Rev. Lett.* **62**, 1415 (1989).
35. Liu, S. L., Wu, G. J., Xu, X. B., Wu, J. & Shao, H. M. Scaling of the vortex-liquid resistivity in high temperature superconductors. *Supercond. Sci. Technol.* **18**, 1332 (2005).
36. Chong, S. V., Williams, G. V. M., Sambale, S. & Kadowaki, K. Doping dependence of the upper critical field, superconducting current density and thermally activated flux flow activation energy in polycrystalline $\text{CeFeAsO}_{1-x}\text{Fx}$ superconductors. *Phys. C* **507**, 35 (2014).
37. Wang, X. L. *et al.* Very strong intrinsic flux pinning and vortex avalanches in $(\text{Ba},\text{K})\text{Fe}_2\text{As}_2$ superconducting single crystals. *Phys. Rev. B* **82**, 024525 (2010).
38. Xing, X. Z. *et al.* Thermally activated flux flow, vortex-glass phase transition and the mixed-state Hall effect in 112-type iron pnictide superconductors. *Sci. China-Phys. Mech. Astron.* **61**, 127406 (2018).
39. Tachiki, M. & Takahashi, S. Strong vortex pinning intrinsic in high-T_c oxide superconductors. *Solid State Commun.* **70**, 291–295 (1989).
40. Seto, Y. Crystallography Software ReciPro. <http://www2.kobe-u.ac.jp/~seto/> (2008).
41. Holland, T. J. B. & Redfern, S. A. T. UNITCELL: a nonlinear least-squares program for cell-parameter refinement and implementing regression and deletion diagnostics. *J. Appl. Crystallogr.* **30**, 84 (1997).

ACKNOWLEDGEMENTS (not compulsory)

Y.S.K was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (NRF-2019R1F1A1040989).

AUTHOR CONTRIBUTIONS

Y.S.K. designed the research. W.J.C. and Y.I.S. performed single crystal growth, proton irradiation, X-ray diffraction, electron diffraction, and magnetoresistivity experiments. K.P. helped in data interpretation and analysis. Y.S.K, W.J.C. and Y.I.S. wrote this manuscript.

COMPETING INTERESTS

The authors declare no competing interests.

Figure captions

Fig. 1. XRD patterns for pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals. The inset showed an extended (002) peak for the determination of full width at half maximum (FWHM).

Fig. 2. (a) and (b) are experimental electron diffraction patterns for the [001] and [010] directions in the pristine $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal, respectively. (c) and (d) are experimental electron diffraction patterns for the [001] and [010] directions in the proton-irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal, respectively. (e) and (f) are simulated electron diffraction patterns in [001] and [010] directions, respectively, where the blue is for the pristine sample and the red circle is for the proton irradiated sample. The inset of (e) is the diffraction spot at Miller index (660): large circles are experimentally measured and small dots are theoretically calculated points.

Fig. 3. Temperature dependence of electrical resistivity around superconducting transition temperature for pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals. The inset represents the temperature dependence of the electrical resistance for the two samples over a wider temperature range. The blue dashed lines represent extrapolated lines to determine T_c^{on} .

Fig. 4. Temperature dependence of the in-plane electrical resistivity measured with $B//c$ (a) and $B//ab$ (b) in a pristine $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal. Temperature dependence of the in-plane electrical resistivity measured with $B//c$ (c) and $B//ab$ (d) in a proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal.

Fig. 5. Temperature dependence of the higher critical fields $B_{c2}^{90\%}$ and $B_{c2}^{50\%}$ determined by the 90 and 50% criteria in the electrical resistivity data of $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals in $B//c$ and $B//ab$: (a) for $B//c$ and (b) for $B//ab$ in the pristine sample, and (c) for $B//c$ and (d) for $B//ab$ in the proton irradiated sample. The figure also shows the vortex glass magnetic field B_g and the magnetic field B^* that separates CR and TA in the vortex liquid. Here, VG, CR, TA and UV denote the vortex glass, critical, thermally activated flux motion and unpinning vortex regions, respectively. The wine-colored dashed line represents the separation between the thermally activated flux flow region and the unpinning vortex liquid region.

Fig. 6. Derivatives from in-plane electrical resistivity measured on pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals at $B = 13$ T. a-1)-a-4): derivatives $U_a = (d \ln \rho(T))/ (d T^{-1})$ in $B//c$ and $B//ab$ for each sample, respectively. The red line represents the best fit using Eq. (3) and the green line is the best fit using expression $\rho(T) = \rho_n [\exp(U_{\text{eff}}/k_B T)]^S$ with $U_{\text{eff}} = k_B T (T_c - T)/(T_c - T_g)$. b-1)-b-4): derivatives $D = \ln \rho(T)$ in $B//c$ and $B//ab$ for each sample, respectively. The red line represents the best fit using Eq. (2) and the green line is the best fit using expression $\rho(T) = \rho_n [\exp(U_{\text{eff}}/k_B T)]^S$ with $U_{\text{eff}} = k_B T (T_c - T)/(T_c - T_g)$. c-1)-c-4):

derivatives $R = (d \ln \rho(T) / d T)^{-1}$ in $B//c$ and $B//ab$ for each sample, respectively. The red line is derived using the parameters obtained from the fitting for the TAFM model indicated by the red line in the graphs of a-1)-a-4) and the green line represents the best fit using Eq. (4).

Fig. 7. (a) to (d) represent the temperature dependence of $U_a = (d \ln \rho(T)) / (d T^{-1})$ for various magnetic fields with $B//c$ and $B//ab$ in pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals, respectively. The red line represents the best fit using Eq. (3) and the green line represents the best fit using expression $\rho(T) = \rho_n [\exp(U_{\text{eff}} / k_B T)]^s$ with $U_{\text{eff}} = k_B T (T_c - T) / (T_c - T_g)$.

Fig. 8. Magnetic field dependence of thermal activation energy $U_0(B)$ in magnetic flux motion for $B//c$ and $B//ab$ in pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals. The black and blue lines represent the linear fitting result of $U_0(B)$ obtained from pristine samples, and the red and green lines are visually drawn to see the trend of $U_0(B)$. The inset shows the magnetic field dependence of the parameter $\rho_c(B)$ included in Eq. (2) for the pristine and proton irradiated single crystals in $B//c$ and $B//ab$.

Fig. 9. (a) to (d) represent $D = \ln \rho(T)$ vs. $1/T$ plots for various magnetic fields with $B//c$ and $B//ab$ in pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals, respectively. The red line represents the best fit using Eq. (2) and the green line is the best fit using expression $\rho(T) = \rho_n [\exp(U_{\text{eff}} / k_B T)]^s$ with $U_{\text{eff}} = k_B T (T_c - T) / (T_c - T_g)$.

Fig. 10. The temperature dependence of $R = (d \ln \rho(T) / d T)^{-1}$ at different magnetic fields in $B//c$ and $B//ab$ for (a)-(b) pristine and (c)-(d) proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals. The green line represents the best fitting using Eq. (4).

Fig. 11. (a) to (d) show the magnetic field dependence of determined fitting parameter s for pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals in $B//c$ and $B//ab$. The solid square in black represents s obtained by fitting the experimental data using Eq. (4) for the vortex glass model in different magnetic fields, and the red solid circle represents s determined from the dependence of the new temperature scale $(T - T_g) / (T_c - T_g)$ of the electrical resistivity in the critical region.

Fig. 12. (a) to (d) show the dependence of the electrical resistivity on the new temperature scale $(T - T_g) / (T_c - T_g)$ for the pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals in $B//c$ and $B//ab$, respectively. The red line represents the linear best fitting in the critical region.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 10

Fig. 11

Fig. 12

Figures

Figure 1

XRD patterns for pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals. The inset showed an extended (002) peak for the determination of full width at half maximum (FWHM).

Figure 2

(a) and (b) are experimental electron diffraction patterns for the [001] and [010] directions in the pristine $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal, respectively. (c) and (d) are experimental electron diffraction patterns for the [001] and [010] directions in the proton-irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal, respectively. (e) and (f) are simulated electron diffraction patterns in [001] and [010] directions, respectively, where the blue is for the pristine sample and the red circle is for the proton irradiated sample. The inset of (e) is the diffraction spot at Miller index (660): large circles are experimentally measured and small dots are theoretically calculated points.

Figure 3

Temperature dependence of electrical resistivity around superconducting transition temperature for pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals. The inset represents the temperature dependence of the electrical resistance for the two samples over a wider temperature range. The blue dashed lines represent extrapolated lines to determine T_{con} .

Figure 4

Temperature dependence of the in-plane electrical resistivity measured with $B \parallel c$ (a) and $B \parallel ab$ (b) in a pristine $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal. Temperature dependence of the in-plane electrical resistivity measured with $B \parallel c$ (c) and $B \parallel ab$ (d) in a proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystal.

Figure 5

Temperature dependence of the higher critical fields $B_{c2}^{90\%}$ and $B_{c2}^{50\%}$ determined by the 90 and 50% criteria in the electrical resistivity data of $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals in $B // c$ and $B // ab$: (a) for $B // c$ and (b) for $B // ab$ in the pristine sample, and (c) for $B // c$ and (d) for $B // ab$ in the proton irradiated sample. The figure also shows the vortex glass magnetic field B_g and the magnetic field B^* that separates CR and TA in the vortex liquid. Here, VG, CR, TA and UV denote the vortex glass, critical, thermally activated flux motion and unpinned vortex regions, respectively. The wine-colored dashed line represents the separation between the thermally activated flux flow region and the unpinned vortex liquid region.

Figure 6

Derivatives from in-plane electrical resistivity measured on pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals at $B = 13 \text{ T}$. a-1)-a-4): derivatives $U_a = (d \ln \rho(T) / d T^{-1})$ in $B // c$ and $B // ab$ for each sample, respectively. The red line represents the best fit using Eq. (3) and the green line is the best fit using expression $\rho(T) = \rho_n [\exp(U_{\text{eff}} / (k_B T))]^s$ with $U_{\text{eff}} = k_B T ((T_c - T) / (T_c - T_g))$. b-1)-b-4): derivatives $D = \ln \square \rho(T) \square$ in $B // c$ and $B // ab$ for each sample, respectively. The red line represents the best fit using Eq. (2) and the green line is the best fit using expression $\rho(T) = \rho_n [\exp(U_{\text{eff}} / (k_B T))]^s$ with $U_{\text{eff}} = k_B T ((T_c - T) / (T_c - T_g))$. c-1)-c-4): derivatives $R = (d \ln \square \rho(T) / d T \square)^{-1}$ in $B // c$ and $B // ab$ for each sample, respectively. The red line is derived using the parameters obtained from the fitting for the TAFM model indicated by the red line in the graphs of a-1)-a-4) and the green line represents the best fit using Eq. (4).

Figure 7

(a) to (d) represent the temperature dependence of $U_a = -(d \ln \rho(T) / d T^{-1})$ for various magnetic fields with B//c and B//ab in pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals, respectively. The red line represents the best fit using Eq. (3) and the green line represents the best fit using expression $\rho(T) = \rho_n [\exp(U_{\text{eff}} / (k_B T))]^s$ with $U_{\text{eff}} = k_B T ((T_c - T) / ((T_c - T_g)))$.

Figure 8

Magnetic field dependence of thermal activation energy $U_0(B)$ in magnetic flux motion for B//c and B//ab in pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals. The black and blue lines represent the linear fitting result of $U_0(B)$ obtained from pristine samples, and the red and green lines are visually drawn to see the trend of $U_0(B)$. The inset shows the magnetic field dependence of the parameter $\rho_c(B)$ included in Eq. (2) for the pristine and proton irradiated single crystals in B//c and B//ab.

Figure 9

(a) to (d) represent $D = \ln \rho(T)$ vs. $1/T$ plots for various magnetic fields with $B // c$ and $B // ab$ in pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals, respectively. The red line represents the best fit using Eq. (2) and the green line is the best fit using expression $\rho(T) = \rho_n [\exp(U_{\text{eff}}/(k_B T))]$ with $U_{\text{eff}} = k_B T_c (T - T_c)/(T - T_g)$.

Figure 10

The temperature dependence of $R = (d \ln \rho(T) / dT)^{-1}$ at different magnetic fields in B//c and B//ab for (a)-(b) pristine and (c)-(d) proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals. The green line represents the best fitting using Eq. (4).

Figure 11

(a) to (d) show the magnetic field dependence of determined fitting parameter s for pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals in B//c and B//ab. The solid square in black represents s obtained by fitting the experimental data using Eq. (4) for the vortex glass model in different magnetic fields, and the red solid circle represents s determined from the dependence of the new temperature scale $((T - T_g) / (T_c - T_g))$ of the electrical resistivity in the critical region.

Figure 12

(a) to (d) show the dependence of the electrical resistivity on the new temperature scale $((T - T_g) / (T_c - T_g))$ for the pristine and proton irradiated $(\text{Ca}_{0.85}\text{La}_{0.15})_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ single crystals in B//c and B//ab, respectively. The red line represents the linear best fitting in the critical region.