Approaching the ultimate superconducting properties of (Ba,K)Fe$_2$As$_2$ by naturally formed low-angle grain boundary networks

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Abstract
The most effective way to enhance the dissipation-free supercurrent in the presence of a magnetic field for type II superconductors is to introduce defects that act as artificial pinning centers (APCs) for vortices. For instance, the in-field critical current density of doped BaFe$_2$As$_2$ (Ba122), one of the most technologically important Fe-based superconductors, has been improved over the last decade by APCs created by ion irradiation. The technique of ion irradiation has been commonly implemented to determine the ultimate superconducting properties. However, this method is rather complicated and expensive. Here, we report a surprisingly high critical current density and strong pinning efficiency close to the crystallographic $c$-axis for a K-doped Ba122 epitaxial thin film without APCs, achieving performance comparable to ion-irradiated K-doped Ba122 single crystals. Microstructural analysis reveals that the film is composed of columnar grains with widths of approximately 30–60 nm. The grains are rotated around the $b$- (or $a$-) axis by 1.5° and around the $c$-axis by $-1°$, resulting in the formation of low-angle grain boundary networks. This study demonstrates that the upper limit of in-field properties reached in ion-irradiated K-doped Ba122 is achievable by grain boundary engineering, which is a simple and industrially scalable manner.

Introduction
Significant progress in the growth of Fe-based superconductor (FBS) thin films has been achieved over the past decade. As a result, high-quality, epitaxial thin films of technologically important FBS [e.g., Fe(Se, Te), doped AeFe$_2$As$_2$ (Ae: alkaline earth elements) and doped LnFeAsO (Ln: lanthanoid elements)] are realized on different kinds of single-crystalline substrates and technical substrates$^{1-5}$ except for (Ba,K)Fe$_2$As$_2$ (K-doped Ba122). The realization of epitaxial K-doped Ba122 has been challenging due to the difficulty in controlling volatile potassium. We have recently succeeded in growing K-doped Ba122 epitaxial thin films on fluoride substrates$^6$, which gives a great opportunity to investigate their electrical transport properties. Our preliminary study shows that grain boundaries (GBs) are present in K-doped Ba122 despite no sign of weak-link behaviors.

GB with a misorientation angle larger than the critical angle $\theta_c \sim 9°$ becomes a detrimental defect to the critical current for most FBS$^{3,7,8}$. On the other hand, GBs with a small misorientation angle less than $\theta_c$ do not impede the supercurrent flow. Rather, dislocation arrays in low-angle GBs (LAGBs) contribute to flux pinning$^{3,7,8}$, leading to improvements in the critical current properties of FBS thin films. Indeed, several studies have shown a proof of principle of this concept by growing P- and Co-doped Ba122 thin films on technical substrates with oxide buffer layers having a different in-plane spread prepared by ion
beam-assisted deposition (IBAD)\(^9,10\). In both compounds, the larger the texture spread of Ba122 within \(\theta_{dr}\), the higher the critical current density \(J_c\), typically a few MA cm\(^{-2}\) at 4 K. Additionally, \(J_c\) for the applied field parallel to the crystallographic c-axis (\(H||c\)) is similar to or even higher than that for \(H||ab\)\(^10,11\). It was later demonstrated that enhanced pinning performance is due to LAGBs acting as flux pinning centers\(^12\).

However, other well-known techniques, such as irradiation with protons and heavy ions, produce either isotropic or anisotropic defects (i.e., artificial pinning centers, APCs) significantly enhanced \(J_c\) above the value obtained by the aforementioned GB engineering. For instance, a SmFeAs(O,F) single crystal with columnar defects produced by heavy-ion irradiation exhibits a high self-field \(J_c\) of 18–20 MA cm\(^{-2}\) at 5 K, which is approximately 9–10 times the \(J_c\) of the pristine sample\(^13\). Similarly, Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) single crystals with point defects created by 3-MeV proton irradiation show a self-field \(J_c\) of 11 MA cm\(^{-2}\) at 2 K, which is ~4.6 times the \(J_c\) of the pristine sample\(^14\). Recently, a Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) single crystal irradiated by 320-MeV Au ions shows a very high self-field \(J_c\) of over 20 MA cm\(^{-2}\) at 2 K\(^15\), corresponding to a 12% depairing current density \(J_{ab}\sim166\) MA cm\(^{-2}\)\(^16\).

Here, we report a surprisingly high self-field \(J_c\) of 14.4 MA cm\(^{-2}\) at 4 K and a strong pinning efficiency close to the crystallographic c-axis for the K-doped Ba122 epitaxial thin film with LAGB networks. The pinning force density \(F_p\) for \(H||c\) exceeds 200 GN m\(^{-3}\) at 4 K and above 6 T, which is at a level comparable to the K-doped Ba122 single crystal with Pb-ion irradiation\(^17\).

**Materials and methods**

**Thin film growth**

K-doped Ba122 thin films were grown on CaF\(_2\)(001) at 395°C, a slightly lower temperature than in our previous investigation, by custom designed molecular beam epitaxy using solid sources of Fe, As, Ba and In-K alloy\(^6\). Here, we used an In-K alloy rather than pure K because of the good controllability of the K content in the film as well as safety issues. The CaF\(_2\) substrate was fixed on the sample holder using Ag paste to ensure good thermal conduction. Prior to deposition, the substrate was heated to 600°C, kept at this temperature for 15 min for thermal cleaning, and subsequently cooled to 395°C. The compositions of all fluxes except for As were monitored in situ by electron impact emission spectrometry (Ba and Fe) and atomic absorption spectrometry (K). The obtained real-time information was fed back to a personal computer that controls the proportional–integral–differential (PID) of the resistive heaters. The As flux was provided constantly during growth. Compared with our previous films, the growth parameters (i.e., deposition temperature and evaporation rate for each flux) were fully optimized, as evidenced in Supplementary Fig. S1. Unlike our previous investigation, no impurity phases were observed. Additionally, the average full width at half maximum value of the 103 \(\phi\)-scan is 1.1°, which is smaller than our previous film\(^6\).

**Microstructural analysis by transmission electron microscopy**

Cross-sectional samples were prepared by a focused ion beam. Scanning transmission electron microscopy observations were performed by a TEM (JEOL ARM200F) operated at an acceleration voltage of 200 kV. TEM-based scanning precession electron diffraction (PED) analysis was performed by TEM (Thermo Fisher Scientific Tecnai G2 F20 equipped with NanoMEGAS ASTAR system) operated at an acceleration voltage of 200 kV. Details of crystal orientation mapping based on PED are described in ref.\(^18\). In this PED analysis, the convergence semiangle of the incident electron beam was 1 mrad, and the precession angle was 0.55°. The crystal orientation at each measurement point was determined by matching the PED pattern with template patterns pre-generated from the crystal structural data of K-doped Ba122\(^19\) and CaF\(_2\).\(^20\). \(\beta\) and \(y\) are defined as the angles between [001]CaF\(_2\) and [001]K-doped Ba122 and [100] (or [010]) CaF\(_2\) and [100] (or [010]) K-doped Ba122, respectively. Note that the \(\beta\) and \(y\) values in this measurement include an uncertainty of ~0.4°, which was estimated from the standard deviation of crystal orientation determination on the CaF\(_2\) substrate.

**Electrical transport measurements**

A small bridge 38 \(\mu\)m wide and 1 mm long was fabricated by laser cutting. The sample was mounted on a rotator holder in the maximum Lorentz force configuration. The angle \(\theta\) is measured from the crystallographic \(ab\)-plane. Current–Voltage (\(I–V\)) characteristics were measured by a 4-probe method in a commercial physical property measurement system [PPMS) Quantum Design]. The upper critical fields \(H_{c2}\) were defined as 90% of the normal state resistivity. The irreversibility fields \(H_{irr}\) were defined as the intersection between the resistivity traces and the resistivity criterion of \(10^{-5}\) m\(\Omega\)cm. An electric field criterion of 1 \(\mu\)V/cm is used to estimate \(J_c\).

**Magnetic measurements**

Magnetization measurements were performed on the rectangular-shaped sample using a superconducting quantum interference device magnetometer [SQUID VSM, (MPMS3) Quantum Design]. The temperature dependence of susceptibility was measured with a magnetic field of 1 mT applied parallel to the \(ab\)-plane. Magnetic \(J_c\) was determined using the Bean model from the field dependence of magnetization curves.
Results
Microstructure
As revealed by structural characterization using X-ray diffraction, K-doped Ba122 was phase-pure and epitaxially grown on CaF2(001) (Supplementary Fig. S1). To evaluate the nanostructure of the grain boundaries, a cross section was observed by scanning transmission electron microscopy (STEM, Fig. 1a) and analyzed by TEM-based scanning PED. The incident direction of the electron beam is approximately parallel to the [110] direction of the CaF2(001) substrate. An annular dark-field (ADF) image in Fig. 1a shows columnar grains growing in the direction, which is more clearly seen in a virtual dark-field image of the 008 reflection (Fig. 1b) of K-doped Ba122. The width of columnar grains is 30–60 nm. The epitaxial relationship is revealed as (001)[110]K-doped Ba122 || (001)[100]CaF2 by the PED patterns (Fig. 1c, d), which is consistent with the structural characterization by X-ray diffraction (Supplementary Fig. S1). Crystal rotations of K-doped Ba122 around the b-axis (equivalent to the a-axis) and the c-axis were calculated from the crystal orientation data separately and are plotted as two-dimensional maps in Fig. 1e, f. For clarity, the crystal rotation angles β (around the b- and a-axis) and γ (around the c-axis) with respect to CaF2 are shown in Fig. 1g, h. As clearly seen in the line profiles (Fig. 1i), the average grain rotation around the b- (or a-) axis is \( \Delta \beta_{\text{average}} = 15^\circ \) and around the c-axis is \( \Delta \gamma_{\text{average}} = -1^\circ \) with respect to the ideal values (i.e., \( \beta_{\text{ideal}} = 0^\circ \) and \( \gamma_{\text{ideal}} = 45^\circ \)), resulting in the formation of LAGB networks. As seen in Supplementary Fig. S2, the [001] of K-doped Ba122 was tilted toward [010] in our coordinate system. The distribution of β over the 2880 points shows that a large fraction is located between 0° and 3.5° with a peak of 1.5° (Supplementary Fig. S2). For completeness, the distribution of γ is also shown in Supplementary Fig. S3. This fact reflects the angular dependence of \( J_c \) measurements, which will be discussed later.

Resistivity measurements
\( T_{c,90} \), defined as 90% of the normal state resistivity, of our K-doped Ba-122 thin film is 35.2 K (Supplementary Fig. S4). The zero-resistivity temperature \( T_{c,0} \) is 33 K, corresponding to the onset temperature of the diamagnetic signal measured by the temperature dependence of susceptibility. Therefore, the transition width, defined as \( T_{c,90} - T_{c,0} \), is 2.2 K.

To determine the upper critical field \( H_{c,2} \) and the irreversibility field \( H_{\text{irr}} \), the temperature dependence of resistivity was measured in the field up to 16 T (Fig. 2a, b). As the applied magnetic field increases, a clear shift of \( T_c \) to lower temperatures together with a broadening of the superconducting transition is observed for both main crystallographic orientations. The broadening of the transition is more obvious for \( H||c \) than \( H||ab \); however, such broadening is not as significant compared with

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**Fig. 1** Microstructural analyses by TEM. a Cross-sectional view obtained by ADF-STEM. b Virtual dark-field image of the 008 reflection of K-doped Ba122. c Typical PED patterns extracted from the K-doped Ba122 thin film (red cross in b) and (d), the CaF2 substrate (green cross in b). e β rotation map and (f), γ rotation map obtained from the K-doped Ba122 thin film. The z-axis shows the distance from the interface between K-doped Ba122 and CaF2, the same direction as shown in a. The ideal angles, 0° and 45°, are defined as light-green and red color in (e) and (f), respectively. g Schematic illustrations of the crystal rotation angles β (around the [100]-axis) and (h) and γ (around the [001]-axis) with respect to CaF2 as the reference. i Line profiles of β rotation and γ rotation extracted along the black broken lines in (e) and (f), respectively. The lines of β = 0° and γ = 45° are marked for comparison.


\[ \text{LnFeAsO}^{21} \text{ due to the weak thermal fluctuation. It is also worth mentioning that the foot structure in the vicinity of zero resistance arising from the presence of high-angle GBs, previously observed in ref. 22, is not present here. Such a foot structure is also due to poor connectivity. The temperature dependence of the upper critical field } H_{c2} \text{ and the irreversibility field } H_{irr} \text{ are summarized in Fig. 2c. The slopes of } H_{c2} \text{ in the field range } 0 \leq \mu_0 H \leq 2 \text{T are } -20.1 \text{TK}^{-1} \text{ and } -11.5 \text{TK}^{-1} \text{ for } H || ab \text{ and } || c, \text{ respectively. These values are much higher than those of a single crystal}^{23}. \text{Another feature is that the slope of the } H_{irr}-\text{line for } H || c \text{ changes at approximately 2T} (\text{inset of Fig. 2c}), \text{which is reminiscent of } \text{REBa}_2\text{Cu}_3\text{O}_7 (\text{RE: rare earth elements, REBCO}) \text{ thin films with } c\text{-axis correlated defects}^{24}. \text{To identify the matching field, } \mu_0 H_{irr} \text{ is plotted as a function } 1-T/T_{irr}, \text{where } T_{irr} \text{ is the irreversibility temperature (Fig. 2d). The slope of the } \mu_0 H_{irr}-\text{line changes from } 1.77 \text{ to } 1.02 \text{ at } 2 \text{T.}

\text{Pinning potential} \text{ To obtain the activation energy } U_0 \text{ for vortex motion at given fields, linear fits of the Arrhenius plots for the resistivity curves are conducted (Fig. 3a, b). Based on the thermally activated flux-flow model}^{25}, \text{the slope of the linear fits corresponds to } -U_0. \text{In fact, on the assumption of the linear temperature dependence, } U(T,H) = U_0(H)(1-T/T_c), \text{the following two formulae, } \ln \rho(T,H) = \ln \rho_0(H) - U_0(H)/T \text{ and } \ln \rho_0(H) = \ln \rho_{0f} + U_0(H)/T_c, \text{are obtained with } \rho_{0f} \text{ being the pre-factor. As seen in Fig. 3c, the activation energy } U_0 \text{ for both } H || c \text{ and } || ab \text{ shows the same power law relation } H^{-\alpha} \text{ in low fields up to } 2 \text{T: the exponent } \alpha \text{ is } -0.05 \text{ to } -0.07, \text{which indicates that single vortex pinning prevails. In this regime, } U_0 \text{ for both directions is } 12,000 \text{ to } 13,000 \text{K, whereas the respective values of the } \text{Ba}_{0.72}\text{K}_{0.28}\text{Fe}_2\text{As}_2 \text{ single crystal with } T_c = 32 \text{K (i.e., underdoped sample) for } H || ab \text{ and } || c \text{ at } 1 \text{T.}

\text{Fig. 2 In-field resistivity measurements and the magnetic phase diagram for the K-doped Ba122 thin film. a Resistivity curves for } H \text{ parallel to the crystallographic } c\text{-axis and (b) } H || ab\text{-plane. The field increment was 2 T from 2 to 16 T. Below 2 T, measurements were conducted at 0, 0.5, 1, and 2 T. c Both } H_{c2} \text{ and } H_{irr} \text{ are plotted as a function of temperature. The solid symbols represent } H_{c2} \text{ and the open symbols show } H_{irr}. \text{For } H || c, \text{the slope of } H_{irr} \text{ changes by approximately 2T, as indicated by the arrow. d Logarithmic presentation of } H_{irr} \text{ vs. } 1-T/T_{irr}, \text{where } T_{irr} \text{ is the irreversibility temperature at the self-field. The slope changes at 2T, corresponding to the matching field.}
are 8500 K and 5000 K\(^{26}\). Above 2 T, for \(H\parallel ab\), \(\alpha \approx 0.5\) is consistent with a plastic pinning regime\(^{27}\). On the other hand, for \(H\parallel c\), \(\alpha = 0.68\), which is located between 0.5 and 1, where the exponent \(\alpha = 1\) is the theoretical prediction for collective pinning\(^{28}\). It is interesting to note that for the high field regime (i.e., 13–16 T) \(U_0\) of our film is comparable to that of single crystals\(^{26}\).

The relationship between \(\ln[\rho_0]\) and \(U_0\) for both orientations is shown in Fig. 3d, where the slope of the linear fits corresponds to \(1/T_c\). The respective \(T_c\) for \(H\parallel c\) and \(||ab\) are 35.4 K and 35.5 K, which is close to \(T_c,90\). This perfect scaling justifies the initial assumption of \(U(T, H) = U_0(H)(1−T/T_c)\) in a wide range of temperatures.

**Field dependence of \(J_c\) obtained from the transport and magnetization measurements**

Figure 4a shows the in-field \(J_c\) properties for the K-doped Ba122 thin film measured by the \(I−V\) (or current density \(J\)—electric field \(E\)) characteristics at various temperatures. \(E−J\) curves for \(H\parallel c\) are shown in Supplementary Fig. S5. At 30 K for both \(H\parallel c\) and \(||ab\), \(J_c\) gradually decreases with increasing fields. However, \(J_c\) below 25 K is almost insensitive to applied magnetic fields, and a high \(J_c\) above \(2 \times 10^5\) A cm\(^{-2}\) is maintained over the entire investigated field range. The most striking feature is that \(J_c\) for \(H\parallel c\) exceeds that for \(H\parallel ab\) with decreasing temperature, opposite to the expected intrinsic behavior related to the anisotropy of \(H_c^2\). Similar features with inverse anisotropy caused by strong \(c\)-axis correlated defects were previously observed, for instance, in Cdoped Ba122\(^{25}\) and REBCO\(^{24,30,31}\). These results infer that strong \(c\)-axis pinning is active at \(T \leq 25\) K. It is worth mentioning that the \(J_c\) peak for \(H\parallel c\) is prominent at high temperatures for REBCO, but it is strongly suppressed with decreasing temperature\(^{32}\), which is different from FBS.

![Fig. 3 Arrhenius plots of the resistivity curves shown in Fig. 2a, b and the resultant pinning potential \(U_0\) and prefactor \(\rho_0f\) for the K-doped Ba122 thin film. a For \(H\parallel c\) and (b) \(||ab\). c \(U_0\) dependence of \(\ln[\rho_0 (m\Omega\text{cm})]\) for \(H\parallel c\) and \(||ab\).](image-url)
To prevent overheating of the contact leads/pads and possible sample damage, the $E$–$J$ characterization was limited at low fields and temperatures. Hence, for completeness, the field dependence of magnetization to extract $J_c$ was measured on a rectangular sample cut from the same film used for transport measurements over a wider temperature range (Supplementary Fig. S6). $J_c$ calculated from the Bean model is shown in Fig. 4b. Except for 28 K, $J_c$ has a weak field dependence, which is consistent with the transport $J_c$. At 4 K, self-field $J_c$ reaches 14.4 MA cm$^{-2}$, corresponding to ~9% of the depairing current density $J_d$. The temperature dependence of $J_c$ measured by electrical transport measurements well follows the magnetization $J_c$ (Fig. 4c), although the electric field criterion $E_c$ of the former is higher than that of the latter. The data at 30 K slightly deviating from the trend are likely due to the fluctuations close to $T_c$.

The field dependence of $F_p$ calculated from Fig. 4a is summarized in Fig. 4d. Because of the presence of strong $c$-axis pinning at $T \leq 25$ K, the maximum $F_p$ is always recorded for $H||c$ within our experimental condition (i.e., up to 16 T).

### Angle dependence of $J_c$ obtained from the transport measurement

To obtain a better understanding of the pinning efficiency, measurements of the angular dependences of $J_c$ were conducted at various temperatures and field strengths (Fig. 5). For all fields, the $J_c$ peaks around $H||c$ ($\theta = 90^\circ$) are weak at 30 K; however, they become intense at $T \leq 25$ K. The peak position of $J_c$ around $H||c$ is ~4° away from the $c$-axis, indicating that “the correlated defects” are slightly tilted. This is because the columnar grains of K-doped Ba122, which creates LAGBs along the grains, grew unidirectionally at an incline of a few degrees with respect to the substrate normal. To clearly see the effect of correlated defects on $J_c$, $J_c$ anisotropy defined as $J_c/J_{c^{ab}}$, where $J_{c}^{ab}$ is $J_c$ at $\theta = 180^\circ$, is plotted at the fixed magnetic field (Fig. 5e–g). The black dashed lines are positioned at 94° to clearly see the $J_c$ peaks. At 4 T and 30 K, $J_c/J_{c}^{ab}$ is approximately 0.5 for $H||c$ (Fig. 5e), increasing to ~1.6 at low temperatures. This is a clear indication that the strong pinning around $H||c$ is activated between 30 and 25 K. As increasing applied magnetic fields, a full evolution of
the angular dependence of $J_c/J_{c}^{ab}$ can be observed from a roughly regular behavior with a maximum at 180° for $H||ab$ (e.g., 16 T and 25 K) to an almost isotropic behavior (e.g., 10 T and 25 K as well as 16 T and 20 K) and finally to a behavior strongly affected by $c$-axis correlated pinning at the lowest temperatures.

**Discussions**

Through microstructural analyses and electrical transport measurements, a "$c$-axis correlated defect" in our K-doped Ba122 thin film is identified as a low-angle grain boundary (LAGB). On the assumption that the mean distance $d$ of correlated pinning is identical to that of the...
In our setup, vapor together with the deposition without rotating substrates. Ba122 (917 °C and 988 °C, respectively)34. Hence, the temperature was low compared with the incongruent fuse relatively slowly on the substrate, since the substrate oblique angle. Additionally, adatoms are expected to diffuse relatively slowly on the substrate, while the substrate temperature was low compared with the incongruent melting and decomposition temperatures of K-doped Ba122 (917 °C and 988 °C, respectively)34. Hence, the shadowing effect35, which limits the formation of new nuclei during the deposition behind initially formed nuclei, is pronounced, resulting in inclined columnar growth.

A pinning force density $F_p$ of 114 GN m$^{-3}$ is recorded even at 15 K and 14–16 T (obtained from the transport measurement) and exceeds 200 GN m$^{-3}$ at 4 K and a field above 6 T (the data at 4 K are obtained from the magnetization measurements in Fig. 4b). In Fig. 6, the field dependence of $F_p$ for our K-doped Ba122 thin film is plotted. For comparison, we also plotted the following data of pinning-enhanced Ba122 single crystal and thin films with different dopants: K-doped Ba122 single crystal with Pb-ion irradiation measured at 5 K17, Co-doped Ba122 thin film with large amounts of stacking faults measured at 4.2 K35, Co-doped Ba122 thin film with 3 mol% BaZrO3 (BZO) measured at 4.2 K36, and P-doped Ba122 thin film with 3 mol% BZO measured at 4 K and 15 K38 are also plotted.

The tilted growth of K-doped Ba122 is presumably due to the geometrical configuration of the deposition sources together with the deposition without rotating substrates. In our setup, vapor flux arrives at the substrate with an oblique angle. Additionally, adatoms are expected to diffuse relatively slowly on the substrate, while the substrate temperature was low compared with the incongruent melting and decomposition temperatures of K-doped Ba122 (917 °C and 988 °C, respectively)34. Hence, the shadowing effect35, which limits the formation of new nuclei during the deposition behind initially formed nuclei, is pronounced, resulting in inclined columnar growth.

width of K-doped Ba122 grains (i.e., 30–60 nm), the matching field $B_d$ of $\phi_0/d^2$ is approximately 2 T at which a kink of $H_{irr}$ is observed (Fig. 2c, d, $\phi_0$ being the flux quantum). As shown in Fig. 5, this pinning is strongly temperature dependent, which is presumably due to the crossover between the in-plane coherence length of K-doped Ba122 and the defect size. The correlated GB pinning and networks improve not only self-field $J_c$ but also in-field $J_c$ for $H$ close to the $c$-axis. Consequently, the anisotropy of $J_c$ is inverted with respect to $H_c$. A similar observation was reported in ref. 33, where the GBs between columnar grains in MgB2 thin films grown by e-beam evaporation worked as pinning centers.

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The large improvement of the superconducting properties of our K-doped Ba122 thin film without APCs is due to the high density of correlated pinning centers created by LAGB networks. Unlike Co- and P-doped Ba122 thin films, the growth temperature of K-doped Ba122 thin film is quite low (~400 °C). This low-temperature synthesis may lead to a small grain size and hence an increase in the density of LAGBs. It is worth mentioning that the dislocation density increases with increasing grain boundary angle. Hence, further improvement of in-field $J_c$ is possible by enlarging the texture spread within the critical angle $\theta_c$. The grain boundary engineering presented in this study highlights a possible novel approach to improve the superconducting properties, which is a simple and industrially scalable manner.

Conclusion

Herein, we investigated the nanoscale microstructure of a K-doped Ba122 epitaxial thin film grown on CaF2 by molecular beam epitaxy. The nanoscale crystal orientation mapping shows that the film is composed of columnar grains with widths of approximately 30–60 nm. The average grain rotation around the $b$- (or $a$-) axis is 1.5° around the $c$-axis is 1° with respect to the ideal values, resulting in the formation of low-angle grain boundary networks. LAGB networks are used to realize superior superconducting properties of K-doped Ba122: the pinning force density $F_p$ for $H||c$ exceeds 200 GN m$^{-3}$ at 4 K and above 6 T, which is comparable to the best performing K-doped Ba122 by ion irradiation.

Acknowledgements

The authors thank Wai-Kwong Kwok (Argonne National Laboratory) for data17, Yanwei Ma (Chinese Academy of Science) for data16, Jongmin Lee and Sanghan Lee (Gwangju Institute of Science and Technology) for data17, and Masashi Miura (Seikei University) for data38. This work was supported by JST CREST Grant Number JPMJCR18J4. A portion of the work was performed at the National High Magnetic Field Laboratory, which was supported by National Science Foundation Cooperative Agreement No. DMR-1644779 and the State
of Florida. It was also supported by the US Department of Energy Office of High Energy Physics under grant number DE-SC0018750. This work was also partly supported by the Advanced Characterization Platform of the Nanotechnology Platform Japan sponsored by the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan.

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Author contributions
K.I. and A.Y. designed the study. K.I. and C.T. wrote manuscript together with D.Q., H.S., S.H., M.N., and A.Y. Thin film preparation, structural characterization by XRD, and micro bridge fabrication were carried out by D.Q., M.N., K.I., T.H., and C.T. Microstructural characterization by TEM was performed by C.W., Z.G., and H.G., H.S., and S.H., and C.T. conducted in-field electrical transport measurements.

Conflict of interest
The authors declare no competing interests.

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Supplementary information
The online version contains supplementary material available at https://doi.org/10.1038/s41427-021-00337-5.

Received: 8 May 2021 Revised: 20 August 2021 Accepted: 3 September 2021.

Published online: 22 October 2021

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