# Giant increase in critical current density of $K_xFe_{2-y}Se_2$ single crystals

Hechang Lei(雷和畅) and C. Petrovic
*Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, New York 11973, USA*

(Received 6 September 2011; published 28 December 2011)

The critical current density $J_c^{ab}$ of $K_xFe_{2-y}Se_2$ single crystals can be enhanced by more than one order of magnitude, up to $\sim 2.1\times 10^4$ A/cm$^2$ by the post annealing and quenching technique. A scaling analysis reveals the universal behavior of the normalized pinning force as a function of the reduced field for all temperatures, indicating the presence of a single vortex pinning mechanism. The main pinning sources are three-dimensional (3D) point-like normal cores. The dominant vortex interaction with pinning centers is via spatial variations in critical temperature $T_c$ ("$\delta T_c$ pinning").

DOI: 10.1103/PhysRevB.84.212502 PACS number(s): 74.25.Sv, 74.25.Wx, 74.25.Ha, 74.70.Xa

## I. INTRODUCTION

Recently discovered iron-based superconductors[1] induce great interest in the scientific community because of rather high $T_c$, proximity to the spin-density wave state, and multiband nature of electronic transport.[2–4] However, these materials also encourage potential technical applications due to high upper critical fields $\mu_0H_{c2}$ and critical current densities $J_c$.[4–7]

In the family of iron-based superconductors, FeCh (Ch = S, Se, and Te, FeCh-11 type) materials have the simplest crystal structure, nearly isotropic high $\mu_0H_{c2}$ and rather high $J_c$,[8,9] but their relatively low $T_c$ impedes prospects for applications. Superconducting $T_c$ was raised up to about 32 K in $A_xFe_{2-y}Se_2$ (A = K, Rb, Cs, and Tl, FeCh-122 type) iron selenide superconductors with rather high $\mu_0H_{c2}$ ($\sim$56 T for $H \parallel c$ at 1.6 K).[10,11] Preliminary results indicate that the $J_c$ of $K_xFe_{2-y}Se_2$ is much lower when compared to iron arsenides or binary FeCh-11 type iron selenides.[6,7,9,12–15] Post annealing and quenching treatment can induce metallic and superconducting state in as-grown and insulating $K_xFe_{2-y}Se_2$ crystals,[16] yet current carrying characteristics of such materials are not known.

In this work we report on the significant enhancement of critical current density in $K_xFe_{2-y}Se_2$ single crystals obtained via the post-annealing and quenching process. We also give detailed insight into the vortex pinning mechanism. Main pinning sources are the 3D normal cores, whereas dominant vortex interaction with pinning centers is via spatial variations in $T_c$.

## II. EXPERIMENT

Details of crystal growth and structure characterization were reported elsewhere.[12] The as-grown crystals were sealed into a Pyrex tube under vacuum ($\sim 10^{-1}$ Pa). The samples were annealed at 400 °C for 1 h and quenched in the air as reported previously.[16] Crystals were claved and cut into rectangular bars. Magnetization measurements were performed in a Quantum Design magnetic property measurement system (MPMS-XL5).

## III. RESULTS AND DISCUSSION

Calculated volume fractions from ac susceptibility at 1.8 K are rather similar, 75% for as-grown and 88% for quenched crystal. However, the quenched crystal shows a very steep transition at 31 K and saturates at about 10 K, whereas for the as-grown sample the diamagnetic signal increases gradually with slightly lower $T_c$ [Fig. 1(a)]. The single sharp peak of $4\pi\chi''$ in quenched crystals [Figs. 1(a) and 1(b)] indicates a more homogeneous superconducting state. The calculated volume fraction from dc susceptibility [Fig. 1(b)] significantly increased after quenching, consistent with previous results.[16] Hence, the post-annealing and quenching process significantly advances superconducting volume fraction in quenched $K_xFe_{2-y}Se_2$. The small volume fraction estimated from the FC curve suggests possible strong magnetic flux pinning effects.

Magnetic hysteresis loops (MHL) of the quenched sample are much bigger and more symmetric [Fig. 1(c)]. The pinning force is enhanced significantly and the bulk pinning is dominant when compared to the as-grown sample. The MHL of the as-grown crystal is small and asymmetric, suggesting that the surface barrier may be important.[17,18] Moreover, there is no fishtail effect up to 5 T, which has been observed in S-doped $K_xFe_{2-y}Se_{2-x}S_x$ single crystal with $S = 0.99$ at low field and in FeAs-122 single crystals at high field.[7,14,19–21]

The in-plane critical current density $J_c^{ab}(\mu_0H)$ for a rectangularly shaped crystal with dimension $c < a < b$ when $H \parallel c$ is[22,23]

$$J_c^{ab}(\mu_0H) = \frac{20\Delta M(\mu_0H)}{a(1-a/3b)}, \tag{1}$$

where $a$ and $b$ ($a < b$) are the in-plane sample size in cm, $\Delta M(\mu_0H)$ is the difference between the magnetization values for the increasing and decreasing field at a particular applied field value (measured in emu/cm$^3$), and $J_c^{ab}(\mu_0H)$ is the critical current density in A/cm$^2$. As shown in Fig. 1(d), the calculated $J_c^{ab}(0)$ for the quenched sample from Fig. 1(c) is enhanced about 50 times when compared to the as-grown sample. This cannot be simply ascribed to the improvement of the superconducting volume fraction, because the volume fraction of the quenched crystal is only about 4 times larger than the volume fraction of the as-grown crystal. Critical current values in the quenched crystal are higher than that in $K_xFe_{2-y}Se_2$ crystals grown using the one-step technique and are the highest known $J_c^{ab}$ among FeCh-122 type materials.[15] The quenched sample also exhibits better performance at high field. The $J_c^{ab}$ for the quenched sample is still larger than $10^4$ A/cm$^2$ at 4.8 T, whereas for the as-grown sample,

 

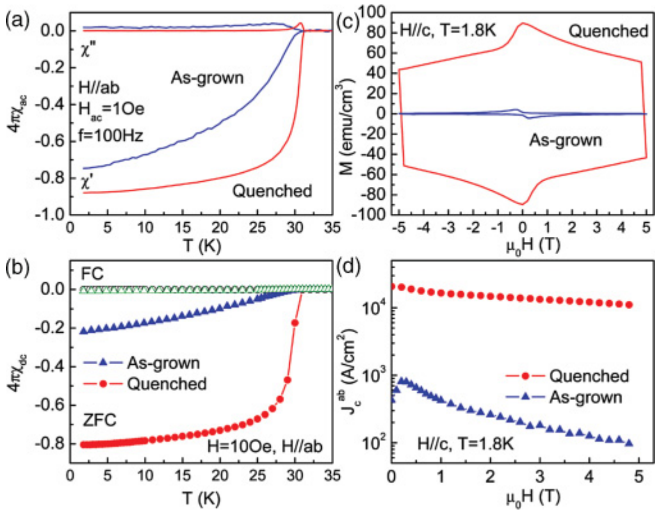


FIG. 1. (Color online) Temperature dependence of the (a) ac and (b) dc magnetic susceptibility of as-grown and quenched $K_xFe_{2-y}Se_2$ crystals taken in $\mu_0 H = 0.1$ (ac) and 1 mT (dc) field, respectively. (c) Magnetization hysteresis loops of as-grown and quenched samples at 1.8 K for $H \parallel c$. (d) Superconducting critical current densities $J_c^{ab}(\mu_0 H)$ of as-grown and quenched samples.

it has decreased about one order of magnitude. The $J_c^{ab}$ (4.8 T, 1.8 K) is also larger than for $K_xFe_{2-y}Se_{2-z}S_z$ with $z = 0.99$.[14]

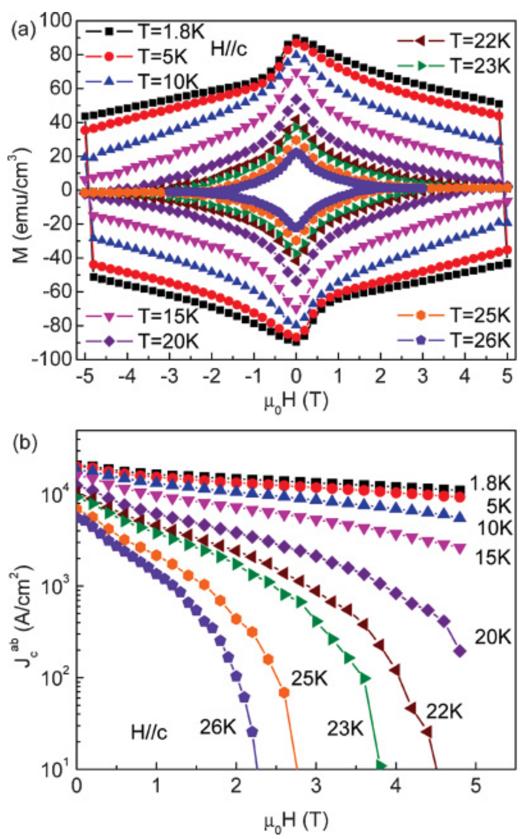


FIG. 2. (Color online) (a) MHLs of quenched $K_xFe_{2-y}Se_2$ crystal for $H \parallel c$. (b) Magnetic field and temperature dependencies of superconducting critical current densities $J_c^{ab}(\mu_0 H)$ for quenched $K_xFe_{2-y}Se_2$ crystal determined from MHLs.

The temperature-dependent symmetric curves for all MHLs imply that the bulk pinning dominates in the crystal at all temperatures. The hysteresis area decreases with the temperature suggesting gradual decrease of $J_c^{ab}$ as the temperature is increased [Fig. 2(b)]. The current carrying performance of the quenched crystals is superior at all temperatures and fields when compared to crystals prepared using the one-step technique.[15]

In order to explain the mechanism of flux pinning in the quenched sample, we studied the temperature and field dependencies of the vortex pinning force $F_p = \mu_0 H J_c$. Based on the Dew-Huges model,[24] if there is a dominant pinning mechanism then the normalized vortex pinning forces $f_p = F_p/F_p^{\max}$ from different measurement temperatures should overlap and a scaling law of the form $f_p \propto h^p(1-h)^q$ will be observed. Here $h$ is the reduced field $h = H/H_{\rm irr}$ and $F_p^{\max}$ corresponds to the maximum pinning force. The irreversibility field $\mu_0 H_{\rm irr}$ is the magnetic field where $J_c^{ab}(T,\mu_0 H)$ extrapolates to zero. The indices $p$ and $q$ provide the information about the pinning mechanism. As shown in Fig. 3(a), the normalized curves

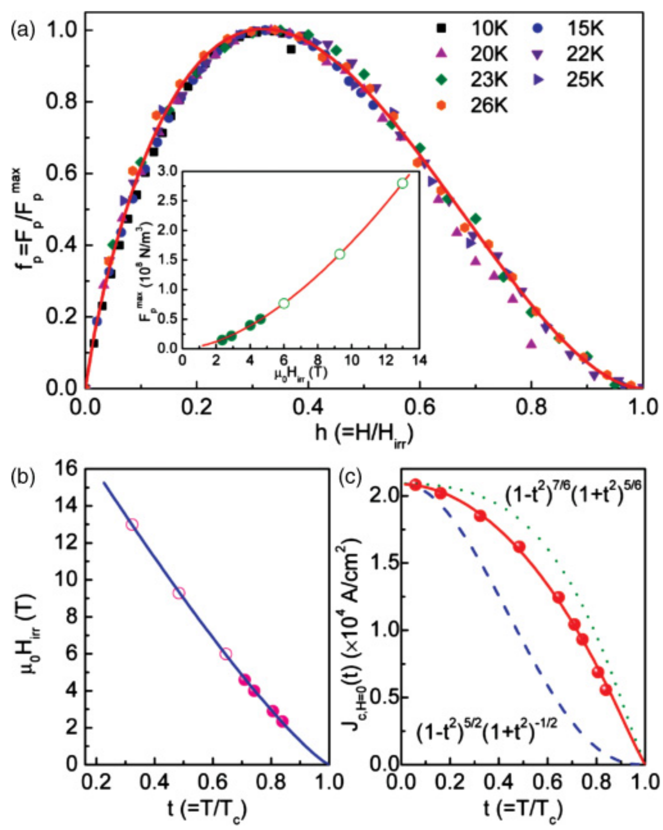


FIG. 3. (Color online) (a) Reduced field dependence of normalized flux pinning force $f_p(h)$ at various temperatures. Solid line is the fitting curve using $f_p = Ah^p(1-h)^q$. Inset shows $F_p^{\max}$ as a function of $\mu_0 H_{\rm irr}$. The fitting result using $F_p^{\max} = A(\mu_0 H_{\rm irr})^\alpha$ is shown as solid lines. (b) Reduced temperature dependence of $\mu_0 H_{\rm irr}(t)$ with the solid line standing for the fitting result obtained by using the $(1-t)^\beta$ law. (c) Reduced temperature dependence of the $J_c(t)$ at zero field. The dotted, dashed, and solid lines show the $J_{c,H=0}^{\delta T_c}(t)$, $J_{c,H=0}^{\delta l}(t)$, and the fitting result using $J_{c,H=0}(t) = x J_{c,H=0}^{\delta T_c}(t) + (1-x) J_{c,H=0}^{\delta l}(t)$, respectively (see text). The measured and estimated $\mu_0 H_{\rm irr}$ are shown as closed and open circles in the inset of (a) and (b).

of $f_p(h,T)$ for $T \geqslant 22$ K present a temperature independent scaling law. Using the scaling function $h^p(1-h)^q$ we estimate $p = 0.86(1)$ and $q = 1.83(2)$, respectively. The value of $h_{\max}^{\text{fit}}$ $[= p/(p+q)] \approx 0.32$ is consistent with the peak positions ($h_{\max}^{\text{exp}} \approx 0.33$) of the experimental curves at different temperatures. Those values are close to the expected values for core normal point-like pinning ($p = 1$, $q = 2$, and $h_{\max}^{\text{fit}} = 0.33$).[24] Moreover, for $T \leqslant 20$ K, the $H_{\text{irr}}$ can be estimated by the $F_p^{\max}$ location at $h_{\max} = 0.33$. Partial $f_p(h,T)$ curves measured between 10 and 20 K also exhibit the same scaling law, suggesting that the core normal point-like pinning mechanism is dominant above 10 K. These point-like pinning centers could come from the random distribution of Fe vacancies after quenching, similar to FeAs-122 type materials.[7,19,20] On the other hand, the $F_p^{\max}$ obeys the $F_p^{\max} \propto (\mu_0 H_{\text{irr}})^\alpha$ scaling with $\alpha = 1.67(1)$ [inset of Fig. 3(a)], close to the theoretical value ($\alpha = 2$) for the core normal point-like pinning.[24] Moreover, as shown in Fig. 3(b), the temperature dependence of $\mu_0 H_{\text{irr}}$ can be fitted by using $\mu_0 H_{\text{irr}}(T) = \mu_0 H_{\text{irr}}(0)(1-t)^\beta$, where $t = T/T_c$ and we obtained $\beta = 1.21(1)$, close to the characteristic value of 3D giant flux creep ($\beta = 1.5$).[25] A similar index has been observed in overdoped $Ba(Fe_{1-x}Co_x)_2As_2$.[26]

Given the presence of 3D core pinning in quenched $K_xFe_{2-y}Se_2$ single crystals, it is important to distinguish between the case of $\delta T_c$ and $\delta l$ pinnings. For type-II superconductors, vortices interact with pinning centers either via the spatial variations in the $T_c$ ("$\delta T_c$ pinning") or by scattering of charge carriers with reduced mean free path $l$ near defects ("$\delta l$ pinning").[27] These two pinning types have different temperature dependence and therefore result in a different relationship between $J_c(t)$ and $t = T/T_c$ in the single vortex-pinning regime (low-field and zero-field regions). For $\delta T_c$ pinning $J_{c,H=0}^{\delta T_c}(t) = J_{c,H=0}(0)(1-t^2)^{7/6}(1+t^2)^{5/6}$, while for $\delta l$ pinning $J_{c,H=0}^{\delta l}(t) = J_{c,H=0}(0)(1-t^2)^{5/2}(1+t^2)^{-1/2}$.[28] As shown in Fig. 3(c), the $J_{c,H=0}(t)$ is between the two curves corresponding to $\delta T_c$ and $\delta l$ pinnings, respectively, but much closer and similar in shape to the $\delta T_c$-pinning curve. Using $J_{c,H=0}(t) = x J_{c,H=0}^{\delta T_c}(t) + (1-x) J_{c,H=0}^{\delta l}(t)$, the experimental data can be fitted very well with $x = 0.74(2)$, suggesting that both $\delta T_c$ and $\delta l$ pinnings play roles in the quenched $K_xFe_{2-y}Se_2$ single crystals, but the former mechanism is dominant. It also implies that the main pinning centers lead to the distribution of $T_c$ in their vicinity or even might be nonsuperconducting like $Y_2O_3$ and Y-Cu-O precipitates in $YBa_2Cu_3O_{7-x}$ thin films.[29]

Even though the $J_c^{ab}$ of quenched $K_xFe_{2-y}Se_2$ single crystals is still one or two order(s) smaller than that of other iron pnictide superconductors,[7,19–21] the post-annealing and quenching technique is an effective way to increase the $J_c^{ab}$ of $K_xFe_{2-y}Se_2$.

## IV. CONCLUSION

In summary, we report a giant increase in the $J_c^{ab}$ of $K_xFe_{2-y}Se_2$ single crystals by the post-annealing and quenching technique. We demonstrate that quenched $K_xFe_{2-y}Se_2$ crystals carry the highest observed $J_c^{ab}$ among FeCh-122 type materials and exhibit good performance at high field. Detailed analysis of the vortex pinning mechanism points out to the presence of a 3D point-like normal core pinning in quenched samples. Moreover, the analysis of temperature dependence of $J_c^{ab}$ at zero field indicates that the $\delta T_c$ pinning is dominant at measured temperature range.


## ACKNOWLEDGMENTS

Work at Brookhaven is supported by the US DOE under Contract No. DE-AC02-98CH10886 and in part by the Center for Emergent Superconductivity, an Energy Frontier Research Center funded by the US DOE, Office for Basic Energy Science.



[1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. **130**, 3296 (2008).

[2] Z.-A. Ren, G.-C. Che, X.-L. Dong, J. Yang, W. Lu, W. Yi, X.-L. Shen, Z.-C. Li, L.-L. Sun, F. Zhou, and Z.-X. Zhao, Europhys. Lett. **83**, 17002 (2008).

[3] C. de la Cruz, Q. Huang, J. W. Lynn, J. Li, W. Ratcliff II, J. L. Zarestky, H. A. Mook, G. F. Chen, J. L. Luo, N. L. Wang, and P. Dai, Nature (London) **453**, 899 (2008).

[4] F. Hunte, J. Jaroszynski, A. Gurevich, D. C. Larbalestier, R. Jin, A. S. Sefat, M. A. McGuire, B. C. Sales, D. K. Christen, and D. Mandrus, Nature (London) **453**, 903 (2008).

[5] H. Q. Yuan, J. Singleton, F. F. Balakirev, S. A. Baily, G. F. Chen, J. L. Luo, and N. L. Wang, Nature (London) **457**, 565 (2009).

[6] J. Karpinski, N. D. Zhigadlo, S. Katrych, Z. Bukowski, P. Moll, S. Weyeneth, H. Keller, R. Puzniak, M. Tortello, D. Daghero, R. Gonnelli, I. Maggio-Aprile, Y. Fasano, O. Fischer, K. Rogacki, and B. Batlogg, Physica C **469**, 370 (2009).

[7] H. Yang, H. Luo, Z. Wang, and H. H. Wen, Appl. Phys. Lett. **93**, 142506 (2008).

[8] H. C. Lei, R. W. Hu, E. S. Choi, J. B. Warren, and C. Petrovic, Phys. Rev. B **81**, 094518 (2010).

[9] C. S. Yadav and P. L. Paulose, Solid State Commun. **151**, 216 (2011).

[10] J. Guo, S. Jin, G. Wang, S. Wang, K. Zhu, T. Zhou, M. He, and X. Chen, Phys. Rev. B **82**, 180520(R) (2010).

[11] E. D. Mun, M. M. Altarawneh, C. H. Mielke, V. S. Zapf, R. Hu, S. L. Bud'ko, and P. C. Canfield, Phys. Rev. B **83**, 100514(R) (2011).

[12] H. C. Lei and C. Petrovic, Phys. Rev. B **83**, 184504 (2011).

[13] R. Hu, K. Cho, H. Kim, H. Hodovanets, W. E. Straszheim, M. A. Tanatar, R. Prozorov, S. L. Bud'ko, and P. C. Canfield, Supercond. Sci. Technol. **24**, 065006 (2011).

[14] H. C. Lei and C. Petrovic, Phys. Rev. B **84**, 052507 (2011).

[15] Z. S. Gao, Y. P. Qi, L. Wang, C. Yao, D. L. Wang, X. P. Zhang, and Y. W. Ma, e-print arXiv:1103.2904 (to be published).

[16] F. Han, B. Shen, Z.-Y. Wang, and H.-H. Wen, e-print arXiv:1103.1347 (to be published).

[17] M. Pissas, E. Moraitakis, D. Stamopoulos, G. Papavassiliou, V. Psycharis, and S. Koutandos, J Supercond. Nov. Magn. **14**, 615 (2001).

[18] L. Zhang, Q. Qiao, X. B. Xu, Y. L. Jiao, L. Xiao, S. Y. Ding, and X. L. Wang, Physica C **445-448**, 236 (2006).

[19] D. L. Sun, Y. Liu, and C. T. Lin, Phys. Rev. B **80**, 144515 (2009).

[20] A. Yamamoto, J. Jaroszynski, C. Tarantini, L. Balicas, J. Jiang, A. Gurevich, D. C. Larbalestier, R. Jin, A. S. Sefat, M. A. McGuire, B. C. Sales, D. K. Christen, and D. Mandrus, Appl. Phys. Lett. **94**, 062511 (2008).
[21] R. Prozorov, N. Ni, M. A. Tanatar, V. G. Kogan, R. T. Gordon, C. Martin, E. C. Blomberg, P. Prommapan, J. Q. Yan, S. L. Bud'ko, and P. C. Canfield, Phys. Rev. B **78**, 224506 (2008).
[22] C. P. Bean, Phys. Rev. Lett. **8**, 250 (1962).
[23] E. M. Gyorgy, R. B. van Dover, K. A. Jackson, L. F. Schneemeyer, and J. V. Waszczak, Appl. Phys. Lett. **55**, 283 (1989).
[24] D. Dew-Hughes, Philos. Mag. **30**, 293 (1974).
[25] Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).
[26] B. Shen, P. Cheng, Z. S. Wang, L. Fang, C. Ren, L. Shan, and H.-H. Wen, Phys. Rev. B **81**, 014503 (2010).
[27] G. Blatter, M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Rev. Mod. Phys. **66**, 1125 (1994).
[28] R. Griessen, H.-h. Wen, A. J. J. van Dalen, B. Dam, J. Rector, H. G. Schnack, S. Libbrecht, E. Osquiguil, and Y. Bruynseraede, Phys. Rev. Lett. **72**, 1910 (1994).
[29] A. O. Ijaduola, J. R. Thompson, R. Feenstra, D. K. Christen, A. A. Gapud, and X. Song, Phys. Rev. B **73**, 134502 (2006).