Critical current density and vortex pinning mechanism of $K_xFe_{2-y}Se_{2-z}S_z$ with S doping

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We report critical current density $J_c$ in $K_xFe_{2-y}Se_{2-z}S_z$ crystals. The $J_c$ can be enhanced significantly with optimal S doping ($z = 0.99$). For $K_{0.70(7)}Fe_{1.55(7)}Se_{1.01(2)}S_{0.99(2)}$ the weak fishtail effect is found for $H||c$. The normalized vortex pinning forces follow the scaling law with maximum position at 0.41 of reduced magnetic field. These results demonstrate that the small size normal point defects dominate the vortex pinning mechanism.

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I. INTRODUCTION

Since the discovery of LaFeAsO$_{1-x}$F$_x$ (FeAs-1111 type) with $T_c = 26$ K, extensive studies have been carried out in order to understand the superconducting mechanism, explore new materials and possible technical applications. Among discovered iron-based superconductors, FeAs-1111 materials and AFe$_2$As$_2$ (A = alkaline or alkaline-earth metals, FeAs-122 type) exhibit high upper critical fields ($\mu_0H_c^2$) and good current carrying ability which are important for energy applications. On the other hand, even though FeCh (Ch = S, Se, and Te, FeCh-11 type) materials have nearly isotropic high $\mu_0H_c^2$ and considerable critical current density, their relatively low $T_c$ when compared to FeAs-1111 and FeAs-122 superconductors is a serious disadvantage. Recently, $A_xFe_{2-y}Se_2$ (A = K, Rb, Cs, and Tl, AFeCh-122 type) materials attracted much attention due to rather high $T_{c, onset}$ ($\sim 32$ K), and $\mu_0H_c^2$ ($\sim 56$ T for $H||c$ at 1.6 K). However, preliminary studies indicate that the critical current density in $K_xFe_{2-y}Se_2$ is lower than in other iron based superconductors. Therefore, it is important to explore pathways for the critical current density $J_c$ enhancement in AFeCh-122 compounds.

In present work, we report the enhancement of critical current density and vortex pinning mechanism in $K_xFe_{2-y}Se_{2-z}S_z$ single crystals. Point defect pinning dominates the vortex pinning mechanism whereas critical current density is maximized for $z = 0.99(2)$.

II. EXPERIMENT

Details of crystal growth and structure characterization were reported in previous work. Crystals were polished into rectangular bars and magnetization measurements were performed in a Quantum Design Magnetic Property Measurement System (MPMS-XL5) up to 5 T. The average stoichiometry and homogeneity of samples were determined by examination of multiple points using an energy-dispersive x-ray spectroscopy (EDX) in a JEOL JSM-6500 scanning electron microscope.

III. RESULTS

Fig. 1(a) shows magnetization hysteresis loops (MHLs) of $K_xFe_{2-y}Se_{2-z}S_z$ at 1.8 K for $H||c$ with field up to 5 T. The shapes of MHLs for all of samples are typical of type-II superconductors. However, for different S doping, they exhibit different flux pinning behavior. For low S doping ($z = 0$ and $z = 0.32$), the MHLs are asymmetric. This asymmetry suggests that the bulk pinning is small and that the influence of the surface barrier is important. On the other hand, for higher S dop-
ing (z = 0.99 and z = 1.04), the shapes of MHLs are symmetric indicating that the bulk pinning is dominant. For $K_{0.70(7)}Fe_{1.55(5)}Se_{1.01(2)}S_{0.99(2)}$ crystal, a small fishtail hump appears at 0.8 T, similarly to FeAs-122 single crystals. We determine the critical current density $J_{c}(\mu_0H)$ is given by

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

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 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$^2$. From Fig. 1(b), it can be seen that the $J_{c}^{ab}(\mu_0H)$ shows small increase at high field region for $z = 0.32$ when compared to $z = 0$ sample. On the other hand, it is enhanced about one order of magnitude for $z = 0.99$ in the whole magnetic field range. For higher S content, the $J_{c}^{ab}(0)$ is still much larger than in pure $K_{0.64(4)}Fe_{1.44(4)}Se_{2.00(0)}$, but the $J_{c}^{ab}(\mu_0H)$ at high fields is smaller. It should be noted that the $T_c$ decreases significantly when $z > 0.32$. When compared to $z = 0$ ($T_{c, onset} = 33.0$ K), $z = 0.99$ crystal has $T_{c, onset} = 24.6$ K, whereas $z = 1.04$ has $T_{c, onset} = 18.2$ K. Therefore, sample with $z = 0.99$ exhibits the best performance and we studied its field dependence of magnetization and critical current density in detail. K, Fe, Se and S are uniformly distributed in $z = 0.99$ crystal (Fig. 2), as is the case with all crystals we investigated.

Fig. 3 shows the MHLs of crystal with $z = 0.99$ for both field directions. The fishtail effect is only observed for H||c. It diminishes gradually with increasing temperature. Similar behavior has also been seen in $BaFe_{2−x}Co_xAs_2$ suggesting anisotropic flux pinning. On the other hand, linear $M(\mu_0H)$ background exists for both field directions, being more obvious for H||ab. This is also observed in pure $K_{0.64(4)}Fe_{1.44(4)}Se_{2.00(0)}$. The slope of this background for crystal with $z = 0.99$ is nearly the same as in pure material, suggesting that high temperature magnetism changes little with $z$ for $z \leq 0.99$. The linear $M(\mu_0H)$ background has no effect on the calculation of $\Delta M(\mu_0H)$, and is due to incomplete superconducting volume fraction of the crystals used in this study. We will discuss the effects of electromagnetic granularity in the next section.

The $J_{c}^{ab}(\mu_0H)$ is calculated using eq. (1) for H||c and shown in Fig. 4(a). The evaluation of critical current density becomes more complex for H||ab, since there are two different contributions. One is vortex motion across the planes, $J_{c}^{ab}(\mu_0H)$, and the other is vortex motion parallel to the planes, $J_{c}^{ab}(\mu_0H)$. Usually, $J_{c}^{ab}(\mu_0H) \neq J_{c}^{ab}(\mu_0H)$. Assuming $a, b \gg c/3$, we obtain $J_{c}^{ab}(\mu_0H) \approx 20\Delta M(\mu_0H)/c$. The calculated $J_{c}^{ab}(\mu_0H)$ is shown in Fig. 3(b).

The $J_{c}^{ab}(0)$ and $J_{c}^{ab}(0)$ are 7.4 and $8.4 \times 10^5$ A/cm$^2$ at 1.8 K and 2 K, respectively. Even though they are still smaller than in other iron pnictide superconductors (where critical current densities are usually above $10^5$ A/cm$^2$ at 5 K), $S$ doping significantly
enhances the critical current density when compared to pure K$_{0.64(4)}$Fe$_{1.44(4)}$Se$_{2.00(0)}$. It suggests that S doping introduces effective pinning center and therefore enhances the $J_c$ for both field directions. On the other hand, the ratio of $J_c^c(\mu_0H)/J_c^{ab}(\mu_0H)$ is approximately 1 and is smaller than in BaFe$_{1-x}$Co$_x$As$_2$. The field dependence of $J_c^c(\mu_0H)$ is somewhat weaker than $J_c^{ab}(\mu_0H)$. This could be related to the layered structure of K$_4$Fe$_{2-y}$Se$_{2-z}$.

IV. DISCUSSION

In order to gain more insight into the vortex pinning mechanism in K$_{0.70(7)}$Fe$_{1.55(7)}$Se$_{1.01(2)}$S$_{0.99(2)}$, we plot the normalized vortex pinning force $f_p = F_p/F_p^{\text{max}}$ as a function of the reduced field $h = H/H_{\text{irr}}$ at various temperatures for $H\parallel c$ (Fig. 5). The pinning force $F_p$ was obtained from the critical current density using $F_p = \mu_0 H J_c$, and $F_p^{\text{max}}$ corresponds to the maximum pinning force. The irreversibility field $\mu_0 H_{\text{irr}}$ is the magnetic field at which $J_c^{\text{ab}}(T, \mu_0 H)$ is zero. It can be clearly seen that the $f_p$ vs $h$ curves exhibit scaling behavior, independent of temperature, suggesting dominance of single vortex pinning mechanism. Scaling law $f_p \propto h^{\beta}(1-h)^{\gamma}$ explains well our data. The obtained parameters are $p = 1.10(1)$ and $q = 1.64(2)$. The value of $h_{\text{max}}^{ \text{fit}} = (p)/(p+q) \approx 0.40$ is consistent with the peak positions ($h_{\text{max}}^{\text{exp}} \approx 0.41$) of the experimental $f_p$ vs $h$ curves at various temperatures. According to the Dew-Hughes model for pinning mechanism, the $h_{\text{max}} = 0.33$ with $p = 1$ and $q = 2$ corresponds to small size normal point defects pinning. Our results indicate that small normal point defects pinning dominates vortex pinning mechanism. These defects could be related to distribution of S ions on a submicron scale, similarly to FeAs-122 system. Moreover, the $F_p^{\text{max}}$ can be fitted using $F_p^{\text{max}} = A(\mu_0 H_{\text{irr}})^\gamma$ and we obtain $\alpha = 1.94(3)$. This is consistent with the theoretical value ($\alpha = 2$).

Since our crystals are rather homogeneous (Fig. 2), the shape of M(H) for $z = 0$ and $z = 0.32$ suggests some electromagnetic granularity similar to SmFeAsO$_{0.85}$F$_{0.15}$. Since we used the full sample dimensions in $J_c^{\text{ab}}$ and $J_c$ calculation, the values we obtained represent the lower limit of bulk superconducting crystals. Indeed, very recently Gao et al. reported the $J_c$ of K$_4$Fe$_{2-y}$Se$_{2-z}$ can be enhanced significantly (about $1.7 \times 10^4$ A/cm$^2$ at 5 K) using one-step technique. It implies that with S doping, the $J_c$ of AFeCh-122 might increase further if preparation process can be optimized.

V. CONCLUSION

In summary, we show an order of magnitude increases in $J_c$ by S doping in K$_x$Fe$_{2-y}$Se$_2$. The optimum S content in K$_x$Fe$_{2-y}$Se$_{2-z}$ single crystals is $z = 0.99$. For the optimally doped sample, the weak fishtail effect is observed when $H\parallel c$. The analysis of vortex pinning force indicates that the dominant pinning sources are small size normal point defects which could originate from distribution of doped S. The results demonstrate that by further optimizing the vortex pinning force, higher values of $J_c$ could be achieved, raising the prospects for technical applications of AFeCh-122 compounds.
VI. ACKNOWLEDGEMENTS

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