Anisotropy of upper critical fields and thermally-activated flux flow of quenched K$_x$Fe$_{2-y}$Se$_2$ single crystals

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(Dated: October 25, 2011)

We report the anisotropy of the upper critical fields $\mu_0 H_{c2}(T)$ and thermally-activated flux flow (TAFF) behavior of quenched K$_x$Fe$_{2-y}$Se$_2$. Even though the post-annealing and quenching process enhances the superconducting volume fraction, it has a minor effect on the upper critical fields for $H|c$ and $H|ab$. Analysis of the angular-dependence of resistivity $\rho_{ab}(\theta, H)$ indicates that it follows the scaling law based on the anisotropic Ginzburg-Landau (GL) theory and the anisotropy $\Gamma(T)$ increases with decreasing temperature with $\Gamma(T) \sim 3.6$ at 27 K. The resistivity of quenched sample exhibits an Arrhenius TAFF behavior for both field directions. Field dependence of thermally activated energy $U_0(H)$ implies that the collective flux creep is dominant in high fields and point defects are the main pinning source in this regime.

PACS numbers: 74.70.Xa, 74.25.Op, 74.25.Wx, 74.25.F-

I. INTRODUCTION

Since the discovery of iron-based superconductors, these materials have attracted great interests because of their complexity and possible unconventional superconductivity. They contain Fe with local moment, exhibit band effects and possible s$\pm$ pairing. Among iron-based superconductors, recently discovered A$_x$Fe$_{2-y}$Ch$_2$ (A = K, Rb, Cs, Ti, and Ch = S, Se, Te, AFeCh-122 type) superconductors have some unique characteristics. These include the proximity to an antiferromagnetic (AFM) semiconducting state distinct from SDW, possible coexistence with long/short range AFM order, considerable Fe deficiencies in Fe plane and the absence of hole pockets which are necessary for s$\pm$ pairing. Structurally, AFeCh-122 materials are rather complex, i.e., they exhibit phase separation with superconducting and insulating regions. Insulating and superconducting state in K$_x$Fe$_{2-y}$Se$_2$ crystals can be even tuned reversibly by post-annealing and quenching process which is very rare in known superconductors. It implies that superconducting and insulating regions are intimately connected and could transform into each other. Therefore, it is of interest to investigate the influence of post-annealing and quenching on the superconducting properties.

In this work, we report the anisotropy of the upper critical fields $\mu_0 H_{c2}(T)$ and thermally-activated flux flow (TAFF) of post-annealed and quenched K$_x$Fe$_{2-y}$Se$_2$ crystals. Our results show that the anisotropy of $\mu_0 H_{c2}(T)$ increases with decreasing temperature, whereas the collective flux creep with point defects pinning source is important at high magnetic fields.

II. EXPERIMENT

Crystal growth method and structure characterization of K$_x$Fe$_{2-y}$Se$_2$ were reported elsewhere. The as-grown crystals were sealed into Pyrex tube under vacuum ($\sim 10^{-1}$ Pa). The ampoule was annealed at 400 °C for 1h and quenched in the air. Crystals were cleaved and cut into rectangular bars and the in-plane resistivity $\rho_{ab}(T)$ was measured using a four-probe configuration in a Quantum Design PPMS-9. The sample dimensions were measured with a Nikon SMZ-800 optical microscope with 10-μm resolution. Magnetization measurements were performed in a Quantum Design Magnetic Property Measurement System (MPMS-XL5).

III. RESULTS AND DISCUSSION

After quenching, the resistivity $\rho_{ab}(T)$ of K$_x$Fe$_{2-y}$Se$_2$ crystals decreases significantly (Fig. 1(a)) and the crossover temperature of metal-semiconductor transition shifts from about 134 K to 186 K. This is consistent with the results in the literature. On the other hand, quenching has a minor effect on superconducting transition tem-
nearly linear for both field directions and the initial slope of the resistivity curves deviate from the linear extrapolation of the normal state behavior.

Temperature $T_c$ which is about $\sim 31$ K for both samples as shown in the inset of Fig. 1(a) and (b). Although the $T_c$s are similar for the as-grown and quenched samples, the quenched crystal exhibits a very sharp magnetic transition at zero-field-cooling curve and saturates at about 10 K whereas the diamagnetic signal increases gradually for the as-grown crystal (Fig. 1(b)). The calculated volume fraction at 1.8 K from dc magnetic susceptibility is increased after quenching.\textsuperscript{12} Transport and magnetic results indicate that the post-annealing and quenching process significantly enhances the metallicity and superconducting volume fraction of K$_2$Fe$_{2-y}$Se$_2$ crystals.

With increasing magnetic fields, the $T_c$ shifts to lower temperature for both H$||c$ and H$||ab$ as shown in Fig. 2 (a) and (b). The shift is more pronounced when the field is parallel to the c axis of quenched K$_2$Fe$_{2-y}$Se$_2$ crystal than the field is in the ab plane, suggesting that the $\mu_0 H_{c2}(T)$ is anisotropic. The $\mu_0 H_{c2}(T)$ curves are nearly linear for both field directions and the initial slope $d\mu_0 H_{c2}/dT$ for H$||ab$ is much larger than for H$||c$ (Fig. 2 (c) and Table 1). The slopes are similar to reported values\textsuperscript{14–16} indicating that post-annealing and quenching does not have major effect on the intrinsic superconducting properties of K$_2$Fe$_{2-y}$Se$_2$ crystals. Using the Werthamer-Helfand-Hohenberg (WHH) formula $\mu_0 H_{c2}(0) = -0.693 T_c (d \mu_0 H_{c2}/dT)|_{T_c}$\textsuperscript{27} and the slope determined from 10% $\rho_n(T, H)$ with $T_c$ = 32.27 K, the $\mu_0 H_{c2}(0)$ is estimated to be 33.3(5) T and 100(1) T for H$||c$ and H$||ab$, respectively. The Pauli limiting field is $\mu_0 H_{p}(0) = 1.86 T_c (1 + \lambda_{e-\text{ph}})\frac{1}{2}$ where $\lambda_{e-\text{ph}}$ is electron-phonon coupling parameter.\textsuperscript{28} Using the typical value for weak-coupling BCS superconductors ($\lambda_{e-\text{ph}} = 0.5$)\textsuperscript{19} we obtain $\mu_0 H_{p}(0) = 73.5$ T for quenched K$_2$Fe$_{2-y}$Se$_2$ crystal. This value is larger than calculated $\mu_0 H_{c2,c}(0)$ but smaller than that for H$||ab$. This could imply that the electron-phonon coupling is strong, similar to PbMo$_6$S$_8$\textsuperscript{20} or that the real value for H$||ab$ may be influenced by gradual setting in of the spin-paramagnetic effect in the high field limit. On the other hand, experiments in high field indicate that the $\mu_0 H_{c2,c}(0)$ is larger than the calculated value from WHH formula, suggesting that the multiband effects might need to be considered as well.\textsuperscript{28} The superconducting coherence length $\xi_{zero}(0)$ estimated using the Ginzburg-Landau formula $\mu_0 H_{c2}(0) = \Phi_0/2\pi \xi^2(0)$, where $\Phi_0 = 2.07 \times 10^{-15}$ Wb is the flux quantum, is listed in Table 1. Furthermore, the anisotropy of $\Gamma(0) = H_{c2,ab}/H_{c2,c}(0)$ is about 3, consistent with the previous reports.\textsuperscript{14,21}

Because of the uncertainty in the upper critical field values using different criterion, there is an uncertainty in estimated anisotropy ratio $\Gamma(T)$. The measurements of angular-dependent resistivity $\rho_{ab}(\theta, H)$ can to some extent diminish this uncertainty. According to the anisotropic Ginzburg-Landau (GL) model, the effective upper critical field $\mu_0 H_{c2}^{GL}(\theta)$ can be represented as:\textsuperscript{22}

$$\mu_0 H_{c2}^{GL}(\theta) = \mu_0 H_{c2,ab}/(\sin^2 \theta + \Gamma^2 \cos^2 \theta)^{1/2}$$

where $\Gamma = H_{c2,ab}/H_{c2,c} = (m_c/m_{ab})^{1/2} = \xi_{ab}/\xi_c$. Since the resistivity transition only depends on the effective field $H/H_{c2}^{GL}(\theta)$, the resistivity can be scaled using reduced $H/H_{c2}^{GL}(\theta)$ and all curves measured in different magnetic fields but at the same temperature should collapse in a single curve when choosing a proper $\Gamma(T)$ value.\textsuperscript{23} Fig. 3 (a) presents the angular-dependent resistivity of quenched K$_2$Fe$_{2-y}$Se$_2$ crystal at 30 K in various fields. All curves exhibit similar cup-like shape and the minimum values of resistivity is at $\theta = 90^\circ$.\textsuperscript{24}

![FIG. 2. Temperature dependence of $\rho_{ab}(T)$ of quenched K$_2$Fe$_{2-y}$Se$_2$ crystal in various fields up to 9 T for (a) H$||c$ and (b) H$||ab$. (c) Temperature dependence of $\mu_0 H_{c2}(T)$ for quenched K$_2$Fe$_{2-y}$Se$_2$ crystal determined from the resistivity drops to 90%, 50%, and 10% of the normal-state resistivity $\rho_n(T, H)$. The $\mu_0(T, H)$ were determined from the first points where the resistivity curves deviate from the linear extrapolation of the normal-state behavior.](image)

### Table I. Upper critical fields and coherence lengths of quenched K$_2$Fe$_{2-y}$Se$_2$ crystals.

| $H$ | Onset | Middle | Zero |
|-----|-------|--------|------|
| $H||c$ | 32.27(3) | 2.14(7) | 1.67(5) | 1.49(2) | 33.3(5) | 3.15(2) |
| $H||ab$ | 32.26(3) | 8.3(3) | 6.0(2) | 4.46(6) | 100(1) | 1.05(2) |
FIG. 3. Angular dependence of in-plane resistivity \( \rho_{ab}(\theta, H) \) of quenched K\(_x\)Fe\(_{2-y}\)Se\(_2\) crystal at 30 K in various fields. (b) Scaling behavior of the resistivity versus \( \mu_0 H_s = \mu_0 H (\cos^2 \theta + \Gamma^2 \sin^2 \theta)^{1/2} \) at different magnetic fields and temperatures. Inset: the temperature dependence of determined \( \Gamma(T) \) from GL theory and \( H_{c2,ab}/H_{c2,c} \) with 10% \( \rho_n \) criterion.

where \( \theta \) is the angle between the direction of external filed and the \( c \) axis of quenched K\(_x\)Fe\(_{2-y}\)Se\(_2\) crystal. This shape indicates that the \( \mu_0 H_{c2,ab} \) is larger than the \( \mu_0 H_{c2,c} \), consistent with previous results obtained when fields are fixed along ab plane or c axis. Using scaling field \( \mu_0 H_s = \mu_0 H (\sin^2 \theta + \Gamma^2 \cos^2 \theta)^{1/2} \) as abscissa axis and by adjusting \( \Gamma(T) \), the angular-dependent resistivity measured at same temperature in various fields show excellent scaling behavior as shown in Fig. 3(b). Because there is only one adjustable parameter \( \Gamma(T) \) for scaling at each temperature, the obtained value of \( \Gamma(T) \) is more reliable than that determined from \( H_{c2,ab}(T)/H_{c2,c}(T) \), which may be influenced by a choice of criterion.

As shown in the inset of Fig. 3(b), the \( \Gamma(T) \) increases with decreasing temperature and \( \Gamma \) is about 3.6 at 26 K, which is consistent with the data on unquenched crystal.\(^{16,24}\) The \( \Gamma(T) \) determined from \( H_{c2,ab}(T)/H_{c2,c}(T) \) with 10% \( \rho_n \) criterion also show same trend, but the temperature region is much higher and narrower, which is limited by the rather high \( \mu_0 H_{c2,ab}(T) \). Temperature dependence of \( \Gamma(T) \) implies that the multiband effect may have an influence on anisotropy of upper critical fields. The results obtained at higher field show that the \( \Gamma(T) \) decreases below 26 K, similar to Rb\(_x\)Fe\(_{2-y}\)Se\(_2\).\(^{16,24}\) This suggests that the spin-paramagnetic effect could also influence \( \Gamma(T) \). The \( \Gamma(T) \) of quenched K\(_x\)Fe\(_{2-y}\)Se\(_2\) crystal is much larger than of Fe(Se,Te) and Fe(Te,S).\(^{25,26}\)

FIG. 4. \( \log \rho(T, H) \) vs. \( 1/T \) in various field for (a) \( H||c \) and (b) \( H||ab \). The corresponding solid lines are fitting results from the Arrhenius relation. (c) \( \ln \rho_c(H) \) vs. \( U_c(H) \) derived from Arrhenius relation for both field directions. The solid lines are linear fitting results. (d) Field dependence of \( U_c(H) \). The solid lines are power-law fitting using \( U_c(H) \sim H^{-\alpha} \).

\[
\ln \rho(T, H) = \ln \rho_0(H) - \frac{U_c(H)}{T}
\]

where \( \ln \rho_0(H) = \ln \rho_{0f} + U_c(H)/T_c \) is temperature-independent and \( U_c(H) \) is apparent activated energy. Therefore, the relationship between \( \ln \rho(T, H) \) and \( 1/T \) should exhibit linear behavior in TAFF region. As shown in Fig. 4 (a) and (b), the experimental data can be fitted using the Arrhenius relation (solid lines) very well for \( H||c \) and \( H||ab \). The results are shown in the common logarithmic scale in the figures but calculated in the natural one. The good linear behavior of \( \ln \rho(T, H) \) vs. \( 1/T \) indicates that the temperature dependence of thermally activated energy (TAE) \( U(T, H) \) is approximately linear, i.e., \( U(T, H) = U_c(H)(1 - H/T_c) \).\(^{27,28}\) Furthermore, as shown in Fig. 4(c), \( \ln \rho_0(H) - U_c(H) \) exhibit linear behavior for both field directions which is expected from eq. (1). From fitting using \( \ln \rho_0(H) = \ln \rho_{0f} + U_c(H)/T_c \), we obtained \( \rho_{0f} = 12.4(1) \) and \( 5.3(1) \Omega \cdot \text{cm} \) and \( T_c = 32.3(2) \) and 31.8(3) K for \( H||c \) and \( H||ab \), respectively. On the other hand, the log \( \rho(T, H) \) lines for different fields extrapolate to one temperature \( T_{cross} \), which should equal
to $T_c$. The $T_{cross}$ are about 32.2 K for both H||c and 31.7 K for H||ab, consistent with the values of $T_c$ within the error bars. The field dependence of $U_0(H)$ is similar for both field directions at high fields and can be fitted using a power law ($U_0(H) \sim H^{-\alpha}$) which is a characteristic of collective flux creep (Fig. 4(d)). When $\mu_0H > 3$ T, the obtained $\alpha = 0.84(2)$ and $0.78(2)$ for H||c and H||ab, respectively. Because $\alpha = 0.5$ and 1 correspond to a planar-defect pinning and a point-defect pinning, respectively, the fitted $\alpha$ are close to 1, suggesting that the vortex are mainly pinned by the collective point defects in the high field region. On the other hand, the experimental $U_0(H)$ deviates from the extrapolated values in low field. The weaker field dependence of $U_0(H)$ in low fields implies the crossover in vortex pinning mechanism and possibly entry in the single-vortex pinning region. It should be noted that the obtained $U_0(H)$ are much larger than in Fe(Te,S) and comparable to a polycrystalline SmFeAsO$_{0.9}$F$_{0.1}$ but still much smaller than in cuprates.

Previous studies show that there is possible coexistence of superconducting and insulating regions in K$_x$Fe$_{2-\gamma}$Se$_2$ crystals. Post-annealing and quenching may enlarge the superconducting region and/or reduce insulating region, therefore decreasing the resistivity and improving the superconducting volume fraction of sample. On the other hand, post-annealing and quenching has negligible effect on $T_c$, $\mu_0H_{c2}(T)$ and its anisotropy but it has significant influence on the pinning force and critical current density of quenched K$_x$Fe$_{2-\gamma}$Se$_2$ crystal.

IV. CONCLUSION

In summary, we present the superconducting properties of quenched K$_x$Fe$_{2-\gamma}$Se$_2$ crystals. The results show that the post-annealing and quenching process improves the superconducting volume fraction and reduces scattering in K$_x$Fe$_{2-\gamma}$Se$_2$. The hump in resistivity is shifted to higher temperature. The $T_c$ and $\mu_0H_{c2}(T)$ of quenched K$_x$Fe$_{2-\gamma}$Se$_2$ crystals determined by WHH formula and anisotropic GL theory is similar to unquenched samples, hence he quenching has minor effect on the establishment superconducting state. The resistivity of quenched sample shows a clear Arrhenius TAFF behavior. At high field collective flux creep with point defects pinning center is the dominant mechanism for both field directions whereas a possible crossover to single vortex pinning region sets in at low field. The obtained $U_0(H)$ are much larger than in Fe(Te,S) and comparable to a polycrystalline SmFeAsO$_{0.9}$F$_{0.1}$.

V. ACKNOWLEDGEMENTS

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

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