Magnetic relaxation and collective vortex creep in FeTe$_{0.6}$Se$_{0.4}$ single crystal

YUE SUN$^{1,2}$, TOSHIHIRO TAEN$^2$, YUJI TSUCHIYA$^2$, SUNSENG PYON$^2$, ZHIXIANG SHI$^1$(a) and TSUYOSHI TAMEGAI$^2$(b)

1 Department of Physics, Southeast University - Nanjing 211189, PRC
2 Department of Applied Physics, The University of Tokyo - 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

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Abstract - We study the vortex dynamics in a high-quality FeTe$_{0.6}$Se$_{0.4}$ single crystal by performing magnetization measurements of the screening current density $J_s$ and flux creep rate $S$. The temperature dependence of $S$ shows a plateau in the intermediate-temperature region with a high creep rate $\sim 0.03$, which is interpreted in the framework of the collective creep theory. A crossover from elastic to plastic creep is observed. The glassy exponent and barrier height for the flux creep are directly determined by the extended Maley’s method. $J_s$ with flux creep, obtained from magnetic hysteresis loops, is successfully reproduced based on the collective creep analysis. We also approach the critical current density without flux creep by means of the generalized inversion scheme, which proves that the $\delta l$ and $\delta T_c$ pinning coexist in the FeTe$_{0.6}$Se$_{0.4}$ single crystal.

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The recently discovered iron-based superconductors (IBSs) with superconducting transition temperature $T_c$ above 55 K are another member of the high-temperature superconductors (HTS) after cuprate superconductors [1, 2]. IBSs share some similarities with cuprate superconductors like layered structure, very high upper critical fields and unconventional pairing mechanism [2,3]. The study of IBSs is helpful to solve some puzzles remained in HTS and testify some notions and theories originated from the cuprate superconductors. Among these, vortex dynamics is one of the central issues related to both basic science and technological applications. For cuprate superconductors, due to large anisotropy, short coherence lengths and high operation temperature, the vortex motion and fluctuations are quite strong [4]. This leads to a collective pinning with small characteristic pinning energy, which gives rise to intriguing experimental results, such as a plateau observed in the temperature dependence of the normalized relaxation rate $S \equiv \left| \frac{d \ln M}{d \ln t} \right|$ in contrast to the linear increase with temperature predicted by the Anderson-Kim model in low-temperature superconductors (LTS). To understand these behaviors, some theories have been proposed in the past decades [4]. Among these, the collective creep theory successfully interpreted the plateau region, and the large creep rate [5,6]. The discovery of IBSs provides another opportunity to study the vortex dynamics as well as collective creep theory in HTS, and its intermediate $T_c$ is also meaningful to understand the crossover between LTS and HTS.

Magnetic relaxation measurements in IBSs reported so far have revealed that they also show giant flux creep and collective pinning, which implies that IBSs and cuprate superconductors may share common vortex physics [7–13]. Until now, most detailed studies about the vortex dynamics on IBSs have been performed on the “122” phase since high-quality single crystals are readily available. Especially in Co-doped BaFe$_2$As$_2$, it shows fast flux creep and a transition from collective to plastic creep [7,9,11,12]. In order to know if the large creep rate governed by collective flux creep is an intrinsic property of IBSs, detailed vortex dynamics studies should be done on other IBS samples. Among IBSs, FeTe$_{1-x}$Se$_x$, composed of only Fe(Te,Se) layers is a preferred choice to study vortex dynamics because of its simple crystal structure. On the other hand, its less toxic nature makes FeTe$_{1-x}$Se$_x$ a more suitable candidate for applications among the family of IBSs. However, although large single crystals can be easily obtained, the

\( \text{(a)} \) E-mail: zxshi@seu.edu.cn
\( \text{(b)} \) E-mail: tamegai@ap.t.u-tokyo.ac.jp
existence of excess Fe affects the sample quality leading to an inhomogeneous distribution of $T_c$ and $J_c$ [14,15]. Thus only a limited number of works have been done on the vortex dynamics in iron chalcogenides, and some reported results are still in controversy because of the difference in sample quality [16–20]. Furthermore, there is no direct evidence for the collective pinning like the plateau in the temperature dependence of the relaxation rate:

Recently, we developed a controllable way of removing the excess Fe by annealing with a controlled amount of O$_2$, and obtained a high-quality single crystal with large and homogeneous $J_c$ [21,22]. In this paper, we report a detailed study of vortex dynamics in a well-annealed FeTe$_{0.6}$Se$_{0.4}$ single crystal. The temperature dependence of $S$ shows a plateau in the intermediate-temperature region with a large vortex creep rate $S \sim 0.03$, which can be interpreted in the framework of the collective creep theory. A crossover from elastic to plastic creep was observed. The screening current density with flux creep, obtained from magnetic hysteresis loops (MHLs), is successfully reproduced based on the collective creep analysis. We also approach the critical current density without flux creep by means of the generalized inversion scheme (GIS), which proves that the $\delta I$ and $\delta T_c$ pinning coexist in FeTe$_{0.6}$Se$_{0.4}$ single crystals.

Single crystals with a nominal composition FeTe$_{0.6}$Se$_{0.4}$ were grown by the self-flux method. The obtained as-grown crystals were further annealed with a fixed amount of O$_2$ to induce bulk and homogeneous superconductivity. Details of the crystal growth and O$_2$-annealing have been reported in our previous publications [21–23]. Magnetization measurements were performed using a commercial SQUID magnetometer (MPMS-XL5, Quantum Design). Magneto-optical (MO) images were obtained by using the local field-dependent Faraday effect in the in-plane magnetized garnet indicator film employing a differential method [24,25]. Screening current density calculated by the Bean model is denoted as $J_s$ in field-sweep measurements or simply $J$ in relaxation measurements.

The temperature dependence of magnetization was measured to check the quality of FeTe$_{0.6}$Se$_{0.4}$ single crystal. As shown in the main panel of fig. 1, the crystal displays a superconducting transition temperature, $T_c \sim 14.5$K with a transition width less than 1 K (obtained from the criteria of 10 and 90% of the magnetization). To further confirm the homogeneity of the crystal, we took MO images on the same piece of single crystal in the remanent state. A typical MO image taken at 8K after cycling the field up to 400 Oe along the c-axis is shown in the inset of fig. 1. The MO image manifests a typical roof-top pattern, indicating a nearly uniform current flow in the crystal. Besides, the typical current discontinuity lines (so-called d-line), which cannot be crossed by vortices, can be observed and marked by the dotted line. The angle $\theta$ of the d-line for our rectangular sample is $\sim 45^\circ$, indicating that the critical current density within the $ab$-plane is isotropic, consistent with the fourfold symmetry of the superconducting plane. The homogeneous current flow within the sample proved by the MO result also guarantees the reliability of using the Bean model to estimate $J_s$ from MHLs.

Figure 2(a) shows the MHLs obtained from the same piece of crystal at temperatures ranging from 2 to 14 K for $H \parallel c$. A second magnetization peak (SMP), also known as the fish-tail effect (FE), can be witnessed, which is a common feature of iron-based superconductors. The SMP can be witnessed more clearly in the field-dependent critical current density in fig. 2(b) obtained by using the Bean model [26]:

$$J_c = \frac{20 \Delta M}{a(1-a/3b)} .$$

where $\Delta M$ is $M_{down} - M_{up}$, $M_{up}$ [emu/cm$^2$] and $M_{down}$ [emu/cm$^2$] are the magnetization when sweeping fields up and down, respectively, $a$ [cm] and $b$ [cm] are sample widths ($a < b$). Here we should point out that the $J_s$ calculated from the MHLs is the critical current density with flux creep, since there is a finite time delay between the measurement and the preparation of the critical state. The self-field $J_s$ reaches a large value about $3 \times 10^5$ A/cm$^2$ at 2 K, and keeps a value $\sim 1 \times 10^5$ A/cm$^2$ even at 50 kOe. Although the value of $J_s$ is lower than that of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystal [27], it is one of the largest values among those reported in Fe(Fe,Se) [21]. The large value of $J_s$ again manifests the high quality of the crystal and ensures that the vortex dynamics study probes the intrinsic property of FeTe$_{0.6}$Se$_{0.4}$ single crystal.

Figure 3(b) shows the temperature dependence of the magnetic relaxation rate $S \equiv |d \ln M/d \ln t|$ at an applied field ranging from 5 to 30 kOe, where $M$ is the magnetization and $t$ is the time from the moment when the critical state is prepared. In these measurements, the magnetic
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Fig. 2: (Colour on-line) (a) Magnetic hysteresis loops of FeTe\(_{0.6}\)Se\(_{0.4}\) at different temperatures ranging from 2 to 14 K for \(H \parallel c\). (b) Magnetic field dependence of critical current densities for \(H \parallel c\). The field was swept more than 5 kOe higher before decreasing to the target field. Obviously, there is a plateau in the intermediate-temperature range with a high vortex creep rate \(S \approx 0.03\). The plateau and large vortex creep rate were also observed in YBa\(_2\)Cu\(_3\)O\(_{7−δ}\) [6], and iron-based “122” [7,11,12,28] and “1111” samples [13], which can be interpreted by the collective creep theory [6]. On the other hand, the \(S-T\) curve undergoes an upturn at low temperatures especially at small fields like 5 and 10 kOe.

In the context of the collective creep theory, the increase of \(S\) corresponds to the smaller value of \(μ\), suggesting that, at least, a part of the vortex system approaches the single-vortex regime with \(μ = 1/7\). As the temperature is lowered, \(J_c\) increases leading to a wider distribution of the local field in the sample. When the applied field is not considerably larger than the self-field, the local magnetic induction in the region close to the edge of the sample becomes much smaller than the applied field, making this region close to the single-vortex regime with smaller \(μ\). On the other hand, when the applied field becomes much larger than the self-field, no regions in the sample can experience a low enough field for the single-vortex regime. Hence, the increase of \(S\) becomes not obvious at higher fields like 20 or 30 kOe. Actually it can be seen more clearly in the field dependence of \(J_c\)’s under different fields which show crossovers between \(\sim 3\) K and \(\sim 7\) K. In this temperature range, \(J_c\) at high field is larger than that at low fields, which comes from the SMP. Actually, this temperature range is exactly the same range where \(S\) manifests a hump-like behavior. A similar effect of SMP on the magnetic relaxation rate was also reported in Co-doped BaFe\(_2\)As\(_2\) [7,11] and Na-doped CaFe\(_2\)As\(_2\) [28].

Fig. 3: (Colour on-line) (a) Temperature dependences of log \(J_s\) at applied fields ranging from 5 to 30 kOe. The inset shows the field dependence of the magnetic relaxation rate at 2 and 5 K. (b) Temperature dependence of \(S\) at applied fields ranging from 5 to 30 kOe. The inset is the enlarged part of \(S\) at 20 and 30 kOe from 2 to 9 K.
Based on the discussion above, we chose the S-T data at 10 kOe, which is little affected by SMI and by the self-field effect compared to the case of 5 kOe to further probe the vortex dynamics of the FeTe$_{0.6}$Se$_{0.4}$ single crystal. According to the collective creep theory [6], the magnetic relaxation rate $S$ can be described as

$$S = \frac{T}{U_0 + \mu T \ln(t/t_{cG})},$$

(2)

where $U_0$ is the temperature-dependent flux activation energy in the absence of flux creep, $t_{cG}$ is the effective hopping attempt time, and $\mu > 0$ is a glassy exponent for elastic creep. The value of $\mu$ contains information about the size of the vortex bundle in the collective creep theory. In a three-dimensional system, it is predicted as $\mu = 1/7$, (1) $5/2$, $7/9$ for single-vortex, (intermediate) small-bundle, and large-bundle regimes, respectively [4,31].

The flux activation energy $U$ as a function of the current density $J$ can be defined as [32]

$$U(J) = \frac{U_0}{\mu}[(J_0/J)^\mu - 1].$$

(3)

Combining this with $U = T \ln(t/t_{cG})$ extracted from the Arrhenius relation, we can deduce the so-called interpolation formula

$$J(T,t) = \frac{J_0}{[1 + (\mu T/U_0) \ln(t/t_{cG})]^{1/\mu}},$$

(4)

where $J_0$ is the temperature-dependent critical current density in the absence of flux creep. From eqs. (3) and (4), the effective pinning energy $U^* = T/S$ can be derived as

$$U^* = U_0 + \mu T \ln(t/t_{cG}) = U_0(J_0/J)^\mu.$$  

(5)

Thus, the value of $\mu$ can be easily obtained from the slope in the double logarithmic plot of $U^*$ vs. $1/J$, as shown in fig. 4(a). The evaluated value of $\mu$ is $1.34$ close to that reported in YBa$_2$Cu$_3$O$_{7-\delta}$ [33], and IBSs [10,12,20]. The value of $\mu$ resides between the prediction of single-vortex (1/7) and small-bundle (5/2) regimes, indicating that contributions of those two kinds of pinnings coexist. Usually, at low-field and low-temperature region, the flux creep is dominated by the motion of individual flux lines without interaction. When temperature and field are increased, the interaction between flux lines becomes non-negligible, and they will creep collectively in the form of small (or intermediate) bundles. However, it is very difficult to determine the boundary of these two regimes [4]. In our case, $S$ increases at low temperature and small field as shown in fig. 3, which means the single-vortex motion becomes more dominant. Contrary to the above prediction of $\mu > 0$, a negative slope $p = -0.48$ is obtained at small $J$, which is very close to the value of $-0.5$ predicted by the plastic creep theory [34]. Thus the temperature dependence of $S$ shows a crossover between elastic and plastic creep regimes.

![Fig. 4](p4)

Fig. 4: (Colour on-line) (a) Inverse current density dependence of the effective pinning energy $U^*$ at 10 kOe in FeTe$_{0.6}$Se$_{0.4}$ single crystal. (b) Current density dependence of the flux activation energy $U$ constructed by the extended Maley’s method. The solid line indicates power-law fitting in the large-$J$ region.

In the following, we analyze the $U(J)$ relation by the extended Maley’s method [35], which considers the temperature dependence of $U$ and $J$ into the original Maley’s method [36]. This method allows to scale $U$ in a wide range of $J$. The temperature-dependent $U_0$ and $J_0$ are assumed as

$$U_0(T) = U_{00}[1 - (T/T_c)^2]^n,$$

(6)

$$J_0(T) = J_{00}[1 - (T/T_c)^2]^n.$$

(7)

Here, the exponent $n$ is set to 3/2 as in the case of YBa$_2$Cu$_3$O$_{7-\delta}$ [33,35] and Co-doped BaFe$_2$As$_2$ [12]. $U = -T \ln\{dM(t)/dt\} + CT$, and $C = \ln(B\omega/2\pi r)$ is assumed as a constant, where $B$ is the magnetic induction, $\omega$ is the attempt frequency for vortex hopping, $\alpha$ is the hopping distance, and $r$ is the sample radius. By selecting $C = 18$, all the curves can be scaled together as shown in fig. 4(b). The solid line indicates power-law fitting by eq. (3) to the large-$J$ region where the slope in fig. 4(a) is positive. Deviation of the data from the fitting line in the small-$J$ region is reasonable since vortex creep is plastic there. The fitting gives the glassy exponent $\mu = 1.27$, activation energy $U_{00} = 22$ K, and $J_{00} = 2.5 \times 10^5$ A/cm$^2$. The value of the glassy exponent obtained from the extended Maley’s method is very close to that evaluated in
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![Graph](image)

Fig. 5: (Colour on-line) Temperature dependence of $J_c$ determined from the MHLs, model function of length $ξ$ in this scheme we have to assume the empirical temperature dependence of $S$ is fitted by eq. (2) with a single free parameter $μ\ln(t/t_{c,0}) = 35$ shown as the dashed line in fig. 3(b).

Using the parameters obtained above, we can calculate $J_c$ after flux creep from eq. (4), which is shown as the solid line in fig. 5. Obviously $J_c$ is reasonably reproduced except for the deviation at 2 K, which comes from the self-field effect as already discussed above. The good match between experimental results and the calculation means that the present analysis based on the collective creep theory is appropriate. To get more insight into the pinning mechanisms in FeTe$_{0.6}$Se$_{0.4}$, we also calculated $J_c$ without flux creep using GIS [37,38]. Although in this scheme we have to assume the empirical temperature dependence of penetration depth $λ$ and the coherence length $ξ$ as $\propto (1 - t^2)^{-1/2}$ and $\propto (1 + t^2)^{1/2}(1 - t^2)^{-1/2}$, respectively, we can directly reconstruct the true critical current density $J_c$, without creep from $J_s$, and can discuss the pinning mechanism. To compare with the theoretical prediction, we simply choose parameters for the three-dimensional single-vortex pinning, and assume $\ln(t/t_{c,0}) = 28$, consistent with the analysis in fig. 3(b). $J_c$ is reconstructed as presented in fig. 5. Shown together with $J_c$ are the theoretical predictions for $δl$ pinning $J_c(t)/J_c(0) = (1 - t^2)^{5/2}(1 + t^2)^{-1/2}$ and for $δT_c$ pinning $J_c(t)/J_c(0) = (1 - t^2)^{7/6}(1 + t^2)^{5/6}$ [39]. Obviously, $J_c$ resides between the predictions of $δl$ and $δT_c$ pinning. Thus, the $δl$ pinning associated with charge-carrier mean free path fluctuations, and the $δT_c$ pinning associated with spatial fluctuations of the transition temperature, coexist in FeTe$_{0.6}$Se$_{0.4}$ single crystal. Such a result is similar to those reported in Co-doped [11,12] and K-doped [40] BaFe$_2$As$_2$. However, it should be noted that a dominance of $δl$ pinning in FeTe$_{0.7}$Se$_{0.3}$ [18] and FeTe$_{0.4}$Se$_{0.5}$ is reported [19]. These conclusions may come from the underestimated critical current density because of the sample quality (lower $T_c$ [18] and smaller $J_s$ [19]) and the insufficient lowest measuring temperature of $\sim 0.4 T_c$. More importantly, a direct comparison of the theoretical curve with the experimental $J_c$ after flux creep is also inappropriate.

In summary, we have studied the vortex dynamics in FeTe$_{0.6}$Se$_{0.4}$ single crystal by magnetization measurements and its time relaxation. Sharp superconducting transition width, large critical current density and homogeneous current distribution revealed by MO imaging manifest the very high quality of the crystal. The temperature dependence of $S$ shows a plateau in the intermediate-temperature region with a high vortex creep rate $S \sim 0.03$, which is further interpreted by the collective creep theory. A crossover from elastic to plastic creep regime was also observed. With an extended Maley’s method, the glassy exponent and pinning energy were directly determined as $μ = 1.27$ and $U_{00} = 22$ K at $H = 10$ kOe. With these parameters, we successfully reproduce the screening current density obtained from magnetic hysteresis loops, which is actually after flux creep. GIS was applied to analyze the critical current density without flux creep, which proves that the $δl$ and $δT_c$ pinning coexist in FeTe$_{0.6}$Se$_{0.4}$ single crystals.

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