Transport properties of Li$_x$(NH$_3$)$_y$Fe$_2$(Te$_z$Se$_{1-z}$)$_2$ single crystals in the mixed state

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Abstract

We study the electric transport properties of Li$_x$(NH$_3$)$_y$Fe$_2$(Te$_z$Se$_{1-z}$)$_2$ single crystals with $z = 0$ and 0.6 in the mixed state. Thermally-activated flux-flow, vortex glass and flux-flow Hall effect (FFHE) behaviors are observed. Experimental results show that there are rich vortex phases existing in these systems and the vortex liquid states occupy broad regions of phase diagrams. Further analysis suggests that thermal fluctuation plays an important role in the vortex phase diagrams of Li$_x$(NH$_3$)$_y$Fe$_2$(Te$_z$Se$_{1-z}$)$_2$. Moreover, for Li$_x$(NH$_3$)$_y$Fe$_2$Se$_2$, there is no sign reversal of FFHE in the mixed state and a scaling behavior $|\rho_{xy}(\mu_0 H)| = A \rho_{xx}(\mu_0 H)^{\beta}$ with $\beta \sim 2.0$ is observed.

Keywords: Li$_x$(NH$_3$)$_y$Fe$_2$(Te$_z$Se$_{1-z}$)$_2$, single crystals, electric transport properties, mixed state, thermally-activated flux-flow, vortex glass, flux-flow Hall effect

(Some figures may appear in colour only in the online journal)

1. Introduction

Since the discovery of LaOFeAs [1], iron-based superconductors (IBSCs) have become another family of high-$T_c$ superconductors besides cuprate SCs and have attracted intense attention. On the one hand, because of high superconducting transition temperature $T_c$ and large upper critical fields $\mu_0 H_c^2$, IBSCs are important not only for basic science but also for practical applications. On the other hand, the values of Ginzburg number $G_i$ ranging from $10^{-4}$ to $10^{-2}$ in IBSCs indicate that the effect of thermal fluctuation varies in different members of IBSCs. It leads to rich vortex phase diagrams in these SCs [2].

Among IBSCs, the iron-chalcogenide SC FeSe has a relative low and nearly isotropic $\mu_0 H_c^2$ with low $T_c$ (~8 K) [3, 4] when compared to iron-pnictide SCs [5]. After intercalating alkali metals A in between FeSe layers using a high-temperature synthesis method, the $T_c$ can be dramatically enhanced up to about 31 K for A$_x$Fe$_{2-x}$Se$_2$ [6]. However, the obvious mesoscopic phase separation between the superconducting phase and the inter-grown antiferromagnetic insulating phase in these compounds makes it difficult to study their intrinsic physical properties [7]. On the other hand, superconducting (Li$_{1-x}$Fe$_x$)OFeSe synthesized by a novel hydrothermal method exhibits the features of high $T_c$ (>40 K), rather large $\mu_0 H_c$ (>60 T for H||c) and $G_i$ (~$1.3 \times 10^{-2}$) [8–11]. It results in the existence of various vortex phases in (Li$_{1-x}$Fe$_x$)OFeSe single crystals [11, 12].

Besides A$_x$Fe$_{2-x}$Se$_2$ and (Li$_{1-x}$Fe$_x$)OFeSe, AM-NH$_3$ intercalated FeSe (AM = alkali, alkali-earth, and rare-earth metals) forms another class of iron-chalcogenide SCs with high $T_c$ (>40 K) [13–17]. But the absence of single crystals impedes the study of the transport properties and vortex dynamics of these SCs. Very recently, we have grown Li$_x$(NH$_3$)$_y$Fe$_2$Se$_2$ single crystals successfully and they show rather high $T_c$ and $\mu_0 H_c^2$ with significant anisotropy [18], similar to (Li$_{1-x}$Fe$_x$)OFeSe. In this work, we present a comprehensive study of the electric transport properties of Li$_x$(NH$_3$)$_y$Fe$_2$(Te$_z$Se$_{1-z}$)$_2$ single crystals with $z = 0$ and 0.6 (denoted as LiFeSe-122 and LiFeTeSe-122 for brevity) in the mixed state. The observed thermally-activated flux-flow (TAAF), vortex glass (VG) and flux-flow Hall effect (FFHE) behaviors indicate that there are diverse vortex phases and rather broad vortex liquid region in these systems. Large $G_i$ in
LiFeSe-122 suggests the thermal fluctuation is essential to vortex dynamics. Moreover, the scaling behavior of

\[ H_{xy} \propto \frac{1}{T} \]

implies that the strength of the pinning force is relatively weak in LiFeSe-122 when \( H_c \) and the temperature is close to \( T_c \). On the other hand, with Te doping, the Gi decreases with decreased \( T_c \).

2. Experiment

Single crystals of LiFeSe-122 and LiFeTeSe-122 were synthesized via the low-temperature ammonothermal technique. The detailed experimental procedure and characterizations of crystals were described in previous works [18, 19]. The elemental analysis was performed using the inductively coupled plasma atomic emission spectroscopy (ICP-AES) and the atomic ratio of \( \text{Li} : \text{Fe} : \text{Se} = 0.18 : 1 : 0.9 \) for \( z = 0 \) and \( \text{Li} : \text{Fe} : \text{Te} : \text{Se} = 0.16 : 1 : 0.60 : 0.38 \) for \( z = 0.6 \), respectively. Electrical transport measurements were performed using a four-probe configuration with current flowing in the \( ab \) plane of crystals in a Quantum Design PPMS-14. The Hall resistivity was obtained from the difference of the transverse resistivity measured at the positive and negative fields in order to remove the longitudinal resistivity contribution due to voltage probe misalignment, i.e., \( \rho_{xy}(\mu_0 H) = [\rho(\mu_0 H) - \rho(-\mu_0 H)]/2 \).

3. Results and discussions

Figures 1 and 2 present the longitudinal resistivity \( \rho(T, \mu_0 H) \) of LiFeSe-122 and LiFeTeSe-122 single crystals near the superconducting transition region for \( H || ab \) and \( H || c \), respectively. The field-induced broadenings of resistive transitions and resistive tail behaviors are obvious, especially for the field along the \( c \) axis. Similar behaviors have also been observed in (Li, Fe)OHFeSe, SmFeAsO\(_{0.85}\) and cuprates [9, 12, 20, 21]. It can be ascribed to field-induced TAFF. Based on theoretical model describing the TAFF behavior, the resistivity in the TAFF region can be expressed as [22–24]:

\[
\rho = (2\nu_0 LB/J)\exp(-J\nu_0 BVL/T)\sinh(JBVL/T)
\]  

where \( \nu_0 \) is an attempt frequency for a flux bundle hopping, \( L \) is the hopping distance, \( B \) is the magnetic induction, \( J \) is the

Figure 1. (a) and (b) The natural logarithm of \( \rho(T, \mu_0 H) \) of LiFeSe-122 as a function of inverse of temperature at various fields for \( H || ab \) and \( H || c \), respectively. The orange solid and black dashed lines are fitting results from the Arrhenius relation and equation (5).Inset: temperature dependence of in-plane resistivity \( \rho(T) \) for LiFeSe-122 at zero field.

Figure 2. (a) and (b) \( \ln \rho(T, \mu_0 H) \) versus \( 1/T \) for LiFeTeSe-122 at various fields with \( H || ab \) and \( H || c \), respectively. The orange solid and black dashed lines are the fitting results from the Arrhenius relation and equation (5). Inset: temperature dependence of in-plane resistivity \( \rho(T) \) for LiFeTeSe-122 at zero field.
applied current density, \( J_c \) is the critical current density in the absence of flux creep, \( V \) is the bundle volume and \( T \) is the temperature. If \( J \) is small enough and \( J_BVL \ll 1 \), equation (1) can be expressed as

\[
U = J_BVLc \rho_0 \exp(-U/|H|) = J_BVLc \rho_0 \exp(-U/|H|)
\]

where \( U = J_BVL \) is the thermally-activated energy (TAE) or flux-pinning energy, and \( \rho_0 = \gamma_0 LB/J_0 \), which is usually considered to be temperature independent. According to the condensation model, \( U(T, \mu_0 H) = U_0(\mu_0 H)/(1 - \gamma) \), where \( \gamma = T/T_c \) \( (T_c \) is the superconducting transition temperature), \( q = 2 - n/2 \) \((n \) depends on the dimensionality of the vortex system with the range from 0 to 3) and \( U_0(\mu_0 H) \) is the apparent activated energy \([22, 24, 25]\). Generally assuming \( n = 2 \) and the prefactor \( \rho_0 \) is a constant for cuprate SCs \([22]\), we can obtain

\[
\ln \rho = \ln \rho_0(\mu_0 H) = \ln \rho_0(\mu_0 H) - U_0(\mu_0 H)/T
\]

(3)

and

\[
\ln \rho_0(\mu_0 H) = \ln \rho_0 + U_0(\mu_0 H)/T_c.
\]

(4)

Thus, there is a linear relationship between \( \ln \rho(T, \mu_0 H) \) and \( 1/T \) in TAFK region (Arrhenius relation) and the slope and y-axial intercept correspond to \( U_0(\mu_0 H) \) and \( \ln \rho_0(\mu_0 H) \), respectively. As shown in figures 1(a) and (b), the solid lines represent the results of linear fitting in the TAFK region using the Arrhenius relation. All the linear fittings cross at \( T_{cross} \), approximately, which are about 44.56 K and 44.59 K for \( H_{ab} \) and \( H_{c} \), respectively. Ideally, all the lines at different fields should be crossed into one point, \( T_{cross} \), which should be equal to \( T_c \) \([26]\). Obviously, the values of \( T_{cross} \) are consistent with the \( T_{onset} \) in the \( \rho(T) \) curves for both field directions (44.59 K and 44.56 K). Although the \( \ln \rho(T, \mu_0 H) \) versus \( 1/T \) can be fitted linearly, there are relative large fitting errors for \( H_{c} \) because the Arrhenius relation can only be satisfied in a narrow region. It suggests that the assumptions of linear temperature dependence of \( U(T, \mu_0 H) \) and temperature-independent \( \rho_0 \) may not be valid, leading to \( \ln \rho(T, \mu_0 H) \) versus \( 1/T \) deviating from the Arrhenius relation \([27]\). Using the relation \( U(T, \mu_0 H) = U_0(\mu_0 H)/(1 - \gamma) \), the equation (2) can be rewritten as

\[
\ln \rho = \ln(2\rho_0 U_0) + q \ln(1 - \gamma) - \ln(T) - U_0(1 - \gamma) / T
\]

(5)
Figure 4. \((d \ln \rho/dT)^{-1}\) versus \(T\) at \(\mu_0H = 8\) and 14 T of (a, b) LiFeSe-122 and (c, d) LiFeTeSe-122 for \(H||ab\) and \(H||c\), respectively.

where \(\rho\) and \(U_0\) are temperature independent and the value of \(T_c\) is derived from the Arrhenius relation. All fits (black dashed lines) using equation (5) are also plotted in figures 1(a) and (b). It can be seen that this more general method is in better agreement with experimental data than the Arrhenius relation, especially for \(H||c\).

The same analysis procedure can be applied to LiFeTeSe-122. As shown in figures 2(a) and (b), \(T_{\text{cross}}\) obtained from the linear fitting is about 20.28 K and 20.10 K for \(H||ab\) and \(H||c\), respectively. The values of \(T_{\text{cross}}\) are close to the \(T_{\text{onset}}\) in the \(\rho(T)\) curves (20.78 K and 20.79 K). In contrast to LiFeSe-122, the \(\ln(\rho(T, \mu_0H)\) of LiFeTeSe-122 for both field directions are not linearly proportional to \(1/T\), i.e. the Arrhenius relation cannot capture the bending trends of curves. However, the data can be fitted well using equation (5).

The fitted \(U_0\) and \(q\) at various fields for both samples are shown in figure 3. According to the Anderson-Kim model, \(U_0(\mu_0H)\) is related to the effective pinning energy. \(U_0(\mu_0H)\) for \(H||ab\) are much larger than that for \(H||c\) in both crystals (figures 3(a) and (b)), confirming that the flux pinning is much stronger for \(H||ab\). It is consistent with the layered structure of Li\(_4\)(NH\(_3\))\(_2\)Fe\(_2\)(Te\(_{1-x}\)Se\(_x\)). When compared with \(U_0(\mu_0H) \sim 250\) K at \(\mu_0H = 0.5\) T for FeSe\([28]\), the \(U_0(\mu_0H)\) for \(H||c\) at the same field enhances to about \(10^4\) K in LiFeSe-122, indicating a much stronger vortex-pinning energy with an increased \(T_c\) after intercalating the Li-NH\(_3\) layers in between FeSe layers. In contrast, the value of \(U_0(\mu_0H)\) remains in the same order of magnitude \((10^2 \sim 10^3\) K) after intercalating the Li-NH\(_3\) layers into Fe\(_{1-x}\)Se\(_x\)\([29, 30]\). That is to say, their effective pinning energies are comparable. According to Kramer’s law\([31]\), the field dependence of \(U_0(\mu_0H)\) can be expressed as

\[
U_0(\mu_0H) = a(\mu_0H)^\gamma(1 - \mu_0H/\mu_0H_m)^\delta
\]

where \(\mu_0H_m\) is an irreversible field and \(\gamma\), \(\delta\) are scaling parameters. In LiFeSe-122, when assuming \(\delta = 2\), the field is parallel to \(ab\) plane, the value of \(\gamma\) is 0.57(3) and 0.67(5) for \(H||ab\) and \(H||c\), respectively (figure 3(a)). On the other hand, the value of \(\gamma\) is 0.17(5) for \(H||ab\) and 0.38(5) for \(H||c\) in LiFeTeSe-122 (figure 3(b)). The weak field dependence of \(U_0\) implies that single-vortex pinning should be dominant in LiFeSe-122. In contrast, the faster decreases of \(U_0(\mu_0H)\) in LiFeSe-122 imply that the collective flux pinning may dominate\([32]\). The better fits using equation (5) than the Arrhenius relation can be partially ascribed to considering the temperature dependence of prefactor \(\rho_0\). When the assumption of \(U \gg T\) with \(q = 1\) is satisfied, the Arrhenius relation is valid and it can well describe the TAFB behavior, as in LiFeSe-122 for \(H||ab\). But because the obtained \(U_0\) of LiFeSe-122 for \(H||c\) and LiFeTeSe-122 for both field directions are much smaller than that of LiFeSe-122 for \(H||ab\), the assumption of temperature-independent \(\rho_0\), i.e. \(U \gg T\), might be improper and the Arrhenius relation will become invalid.
As shown in figures 3(c) and (d), the $q$ for both crystals are almost independent of the field strength for both directions. When $H||c$, the values of $q$ for both samples are about 2, but it changes from 1 in LiFeSe-122 to 2 in LiFeTeSe-122 when $H||ab$. The $q = 1$ for $H||ab$ in LiFeSe-122 is consistent with the good linear behavior of $\ln \rho(T, \mu_0 H)$ versus $1/T$ shown in figure 1(a). In contrast, the value of $q = 2$ should be another reason leading to the deviation of experimental curves from the Arrhenius relation.

According to the VG model [33, 34], the linear resistivity disappears as a power law

$$\rho = \rho_0 \left( \frac{T}{T_g} \right)^{-1} \quad (7)$$

close to the glass transition temperature, $T_g$. In equation (7), $\rho_0$ is identified as a characteristic resistivity and should in some way be related to the normal state resistivity and $s$ is the glass critical exponent. $T_g$ can be extracted by applying the relation ($d \ln \rho / dT \sim 1 \propto (T - T_g)$ to the resistive tail, and $T^*$ is defined as the temperature deviating from the straight line, as shown in figure 4.

Based on the values of $T_g$ and $T^*$ extracted from VG model and $\mu_0 H_g$ defined from the 90% $\rho_c$, we plot the vortex phase diagrams of LiFeSe-122 and LiFeTeSe-122 (figure 5). The different regions indicate different flux-pinning mechanisms. The region below the $\mu_0 H_g$ line is a vortex glass phase and the region above $\mu_0 H_g$ is the normal state, while the region between $\mu_0 H_g$ and $\mu_0 H_c$ is the vortex liquid phase. Besides, $\mu_0 H^*$ represents the divider of unpinned and strongly pinned regions of vortex liquid. Obviously, the vortex liquid region is broad in these crystals, especially in LiFeSe-122 with $H||c$, suggesting a weak vortex-pinning ability even at relative low field in this system when $T$ approaches $T_c$.

Because the resistive transition curve $\rho(T, \mu_0 H)$ not only shifts but also broadens when the field increases, it suggests that thermal fluctuation is essential to vortex dynamics in these materials. The strength of thermal fluctuations can be quantified by

$$Gi = (\gamma^2 / 2) \left[ (\mu_0 k_B T_g) / (4 \pi B_c^2 (0) \xi_{ab}^3 (0)) \right]^2 \quad [35]$$

where $\gamma$ is the electronic mass anisotropy, $B_c(0) = \phi_0 / (2 \sqrt{2} \pi \lambda_{ab} (0) \xi_{ab} (0))$ is the thermodynamic critical field, $\phi_0$ is the flux quantum, $\xi_{ab} (0)$ and $\lambda_{ab} (0)$ is the coherence length and penetration depth at 0 K for $H||ab$. With $\gamma = \xi_{ab} (0) / \xi_c$, we obtain

$$Gi = \left[ (2 \pi \mu_0 k_B T_g \lambda_{ab}^2 (0)) / (\phi_0 \xi_c (0)) \right]^2 / 2$$

where $\xi_c (0)$ is the coherence length at 0 K for $H||c$. Assuming $\lambda_{ab} (0) = 200 \text{ nm}$ is the same as (Li, Fe)OFeSe [11], the value of $Gi$ is $1.4 \times 10^{-2}$ and $2.8 \times 10^{-4}$ for LiFeSe-122 and LiFeTeSe-122 with $\xi_c (0) = 0.27$ and 0.9 nm derived from $\mu_0 H_g (0)$. The value of $Gi$ for LiFeSe-122 is comparable to (Li, Fe)OFeSe (1.3 $\times 10^{-2}$) and YBCO (10$^{-2}$), but much larger than that of LiFeTeSe-122 [11, 35]. It is consistent with the more obvious broadening of $\rho(T, \mu_0 H)$
in the temperature range of 17 K–50 K when \(H|c\). With increasing field, superconductivity is suppressed gradually and the transition of \(\rho(\mu_0H)\) shifts to lower magnetic fields when temperature increases. \(\rho_x(\mu_0H)\) shows a weak positive magnetoresistance at the normal state \((T = 50 \text{ K})\), consistent with previous results [18]. As shown in figure 6(b), at the mixed state, the Hall resistivity \(\rho_y(\mu_0H)\) at low field is zero and becomes negative with increased absolute values at the high fields. The high-field values of \(\rho_y(\mu_0H)\) gradually reach that in the normal state when temperature is slightly higher than \(T_c\). The sign of the Hall resistivity is negative, indicating that the electron type carriers dominate in the mixed state as well as in the normal state, consistent with the electron doping in LiFeSe-122. Moreover, there is no sign reversal of \(\rho_y(\mu_0H)\) in the mixed state, which is often observed in cuprates [36, 37].

Furthermore, there is a scaling behavior of \(|\rho_x(\mu_0H)| = A\rho_x(\mu_0H)^{\beta}\) in LiFeSe-122 (figure 7) and the fitted values of \(\beta\) are close to 2 in the whole measuring temperature range. Different values of \(\beta\) have been observed in IBSCs and cuprate SCs, such as Fe(Te, S) \((\beta = 0.9–1.0)\) [26], Ba(Fe0.9Co0.1)As2 \((\beta = 2.0(2))\) [38], and Bi2Sr2CaCu2O8+\(\delta\) \((\beta = 2.0 \pm 0.1)\) [39]. A number of theories have been proposed to explain the value of \(\beta\). For example, when considering the effect of pinning on the Hall resistivity, Vinokur et al proposed a phenomenological model where \(\beta = 2.0\) in the TAFF region [40]. Additionally, a unified theory for the Hall effect including both the pinning effect and thermal fluctuations was developed by Wang, Dong, and Ting [41, 42]. They explained scaling behavior by taking into account the backflow current effect on flux motion due to pinning and pointed out \(\beta\) changing from 2 to 1.5 for increasing pinning force. The exponent \(\beta \sim 2.0\) and the absence of sign change of \(\rho_y(\mu_0H)\) for LiFeSe-122 single crystals imply that the strength of pinning force is relatively weak when \(H|c\) and \(T \rightarrow T_c\), consistent with the low TAE in the same field direction shown above.

4. Conclusion

In summary, field-induced resistive broadenings of superconducting transitions and resistive tail behaviors have been observed in LiFeSe-122 single crystals. Detailed analysis shows that these results are associated with TAFF and VG behaviors. Moreover, thermal fluctuation plays an important role in the vortex dynamics of LiFeSe-122, resulting in broad vortex liquid regions in the vortex phase diagrams. The FFHE with \(\beta \sim 2\) indicates the relatively weak vortex-pinning force in LiFeSe-122 when the magnetic field is along the \(c\) axis and \(T\) approaches \(T_c\).

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