Direct Current Measurement of Hall Effect in the Mixed State for the Iron-chalcogenide Superconductors

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Abstract. In order to investigate how the composition and the pinning affect the behavior of the Hall effect in the mixed state, we measure the Hall resistivity for iron-chalcogenide superconductor FeSe$_{1-x}$Te$_x$ films near the superconducting transition temperature. We observe a sign change of the Hall resistivity in FeSe$_{0.5}$Te$_{0.5}$ ($x = 0.5$) films, but FeSe$_{0.4}$Te$_{0.2}$ ($x = 0.2$) films do not show the sign reversal. Increasing applying current density, the sign change of the Hall resistivity in the film with $x = 0.5$ disappeared. We evaluated the strength of the pinning in samples from an activation energy and a scaling law, and found that the film with $x = 0.5$ has the stronger pinning than that of the film with $x = 0.2$. All of those results suggest that the pinning has related to the Hall resistivity anomaly in Fe chalcogenides.

1. Introduction
Properties of the type-II superconductors have attracted significant interest for many years. In particular, one of the interesting and puzzling phenomenon is the Hall effect of superconductors in the mixed state. It is known that the sign of the Hall resistivity in the mixed state is different from that in the normal state, for some high $T_c$ superconductors and conventional superconductors, e.g. V and Nb [1-3], and it was recently reported that the Hall resistivity of an iron-based superconductor, Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, also showed the sign reversal below $T_c$ [4]. Moreover, a double sign reversal was observed in some cuprates, such as Tl$_2$Ba$_2$CaCu$_2$O$_{8+}$ [5].

In general, the behavior of the Hall effect in the normal state reflects the electronic structure at Fermi surface. On the other hand, in the mixed state, a vortex motion contributes to the behavior of the Hall effect. However, such anomalous sign changes of the Hall resistivity cannot be explained by basic models of vortex motion. For example, in the Bardeen-Stephen model [6], where the vortex cores are considered to be in the normal state, the sign of Hall resistivity in the mixed state is the same as in the normal state. Likewise, the Nozières-Vinen model, in which vortex cores experience Magnus force, also leads to the same sign of Hall resistivity as in the normal state [7,8].

Since the discovery of the Hall anomalies, several theoretical approaches have been taken to explain this phenomenon, and as the origin of sign reversal, many possibilities were proposed. One is the influence of the superconducting fluctuations. Fukuyama, Ebisawa and Tsuzuki have shown that Hall conductivity appears as a result of the electron-hole asymmetry, in the time dependent Ginzburg-Landau (TDGL) equation [9]. Similarly, Aronov, Hikami and Larkin have shown that the sign of Hall conductivity is determined by a general gauge invariance requirement of the TDGL equation [10]. However, the application of those theories to cuprates gives the
opposite result to the experiments [11]. Wang et al., have shown that a backflow current due to the pinning has an influence on the Hall effect [12]. Kopinin and Vinokur have also shown that a sufficiently strong pinning can result in a sign reversal [13]. On the other hand, experiments in samples with columnar defects showed the contradictory results about whether a pinning modify the Hall conductivity or not [14,15]. In addition, other possibilities were pointed out, such as the vortex core charging [16], the vortex many-body correlation [17]. However, the origin of the Hall anomaly still remains controversial, and consensus regarding this subject is not reached yet.

Another interesting issue on the Hall effect in the mixed state is a scaling behavior, $|\rho_{xy}| \sim \rho_{xx}^\beta$. The scaling law was first discovered for YBCO films [18], and then it has been observed in other superconductors in the mixed state [19]. A phenomenological model considering the effect of pinning on the Hall resistivity gives a scaling exponent $\beta$ of 2 [20]. Wang et al., proposed a theory for the Hall effect in the mixed state including both the flux pinning effect and thermal fluctuations, and they demonstrated that the scaling exponent $\beta$ decrease with increasing pinning strength [12].

In order to clarify the behavior of the Hall effect in the mixed state, we measured the Hall resistivity for the iron-chalcogenide superconductor FeSe$_{1-x}$Te$_x$ films near the transition temperature, and investigate how the composition and the pinning strength affect the Hall effect in the mixed state. We observed the sign reversal of Hall resistivity for FeSe$_{0.5}$Te$_{0.5}$ ($x = 0.5$) films, while FeSe$_{0.8}$Te$_{0.2}$ ($x = 0.2$) films do not show the sign anomaly. We also observed the relation of $|\rho_{xy}| \sim \rho_{xx}^\beta$, which indicate the validity of the scaling in those samples. Evaluation of the activation energy and the scaling exponent suggest that the Hall anomaly has deeply related to the vortex motion and pinning in the iron-chalcogenide superconductor.

2. Experiment

In this study, we measured FeSe$_{1-x}$Te$_x$ films with $x = 0.2$ and 0.5, which were grown by the PLD method with a KrF laser[21-23]. FeSe$_{1-x}$Te$_x$ polycrystalline pellets were used as targets and commercially available CaF$_2$ (100) substrates were used. Measurements were carried out with a Physical Property Measurement System of Quantum Design. The longitudinal resistance and Hall resistance were measured by the standard six-probe method using silver paste for contact with the magnetic field up to 9 T applied perpendicular to Fe-chalcogenide planes. Unless otherwise noted, the measurements were carried by applying a dc current density of 100 A/cm$^2$.

3. Results and discussions

Figure 1 shows the temperature dependence of the longitudinal resistivity, $\rho_{xx}$, and Hall resistivity, $\rho_{xy}$, for a film with $x = 0.2$ and a film with $x = 0.5$. For the film with $x = 0.5$ near the superconducting transition temperature, the Hall resistivity approaches to zero from positive as temperature decreases and does not show a sign change. On the other hand, for the film with $x = 0.5$, we observed that the sign of the Hall resistivity changes from positive to negative as temperature decreases at $B = 0.5$ T. Figure 2 represents a current-voltage characteristics for the film with $x = 0.5$. The gradient of I-V curve is approximately unity in the region where the sign changes are observed, which means that the longitudinal resistivity is ohmic and Hall resistivity anomaly is observed in the thermally assisted flux-flow (TAFF) region.

The magnetic field dependence of the Hall conductivity are expressed as $\sigma_{xy} = \sigma_{xy}^n + \sigma_{xy}^{SC}$, where $\sigma_{xy}^n$ and $\sigma_{xy}^{SC}$ are the Hall conductivity due to the normal quasiparticles inside and around the vortex and the Hall conductivity due to the motion of vortices. $\sigma_{xy}^n$ is proportional to $B$ and $\sigma_{xy}^{SC}$, according to TDGL theory, is proportional to $1/B$ [24]. Therefore, in low magnetic fields, the superconducting term becomes dominant and the Hall conductivity is approximately proportional to $1/B$. However, the Hall conductivity drops faster than $1/B$ for the film with $x = 0.5$ at lower temperatures as the magnetic field increases, which are shown in Figure 3.
Figure 1. (a),(b) Temperature dependences of the longitudinal resistivity and the Hall resistivity for film with $x = 0.2$. (c),(d) Temperature dependences of the longitudinal resistivity and the Hall resistivity for film with $x = 0.5$.

Figure 2. DC current-voltage characteristics of film with $x = 0.5$. 
Figure 3. Magnetic field dependences of the absolute values of Hall conductivity for films with $x = 0.2$ and $x = 0.5$ respectively. Solid lines are proportional to $1/B$.

Figure 4. (a),(b) Arrhenius plots for films with $x = 0.2$ and $x = 0.5$. Solid lines are the fitted lines to obtain activation energies. (c) Calculated magnetic field dependence of the activation energy for $x = 0.2$ and $x = 0.5$, respectively.
To investigate the influence of the pinning on the Hall effect in the mixed state, we show Arrhenius plots for the films with $x = 0.2$ and $x = 0.5$ (Figure 4). The longitudinal resistivity in the TAFF region is expressed as $ho_{xx} \sim \exp(-U/k_BT)$, where $U$ is the activation energy [20]. From the gradients of Arrhenius plots in low temperature region, we find that the values of $U$ are 1200 K for $x = 0.2$ at $B = 1.0$ T, and 4500 K for $x = 0.5$ at $B = 1.0$ T, respectively. Together with the already-published Hall data in other iron-based superconductors [25-28], we summarized the estimated $U$ and the absence/presence of the sign reversal of the Hall effect in Table 1. This suggests that the sign changes occur when the activation energy is relatively high. The result of our measurements for FeSe$_{1-x}$Te$_x$ films shows the same tendency and consistent with the previous studies in other iron-based superconductors.

To further investigate the influence of the pinning, we measured the Hall resistivity for film with $x = 0.5$ at different dc current densities from 10 A/cm$^2$ to 5000 A/cm$^2$. Figure 5 shows the temperature dependence of the longitudinal resistivity and the Hall resistivity at $B = 0.5$ T and $B = 1.0$ T. At $B = 0.5$ T, the sign reversal disappears as the current density becomes stronger. This result suggests that the strong current density decreases the pinning potential, leading to the disappearance of the sign change.

![Figure 5](image-url)  
**Figure 5.** (a),(b) Temperature dependences of the longitudinal resistivity and the Hall resistivity at $B = 0.5$ T for a film with $x = 0.5$ by applying different current densities. (c),(d) Temperature dependences of the longitudinal resistivity and the Hall resistivity at $B = 1.0$ T for a film with $x = 0.5$ by applying different current densities.
Table 1. The relation between the activation energy and the sign reversal for various iron-based superconductors.

|                  | $U(B = 1 \, \text{T})$ (K) | Sign reversal | Ref. |
|------------------|-----------------------------|---------------|------|
| Fe$_{1+y}$(Te$_{1+x}$S)$_z$ | 200 | × | [25] |
| Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ ($x = 0.08$) | 4700 | × | [26] |
| Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ ($x = 0.10$) | 6000 | ○ | [26] |
| BaFe$_2$As$_2$ | 2300 | × | [27] |
| NaFe$_{1-x}$Co$_x$$_2$As$_2$ ($x = 0.0220$) | 1000 | × | [28] |
| NaFe$_{1-x}$Co$_x$$_2$As$_2$ ($x = 0.0205$) | 2000 | × | [28] |
| FeSe$_{1-x}$Te$_x$ ($x = 0.2$) | 1200 | × |         |
| FeSe$_{1-x}$Te$_x$ ($x = 0.5$) | 4500 | ○ |         |

Figure 6. The relation between the longitudinal resistivity and the Hall resistivity for $x = 0.2$ and 0.5. Solid lines are the fitting lines to obtain scaling exponents $\beta$.

To evaluate the strength of pinning in other indicators, we plot the relation between the longitudinal resistivity and the Hall resistivity for FeSe$_{1-x}$Te$_x$ films, which are shown in Figure 6. We measured those values at fixed magnetic fields and sweeping temperatures. We observed $|\rho_{xy}| \sim \rho_{xx}^\beta$, which indicate the validity of the scaling in those samples, and we obtained, at $B = 0.5$ T, the scaling exponents $\beta$ of 2.2 for film with $x = 0.2$, and of 0.7 for film with $x = 0.5$, respectively. According to Wang’s theory [12], a larger $\beta$ value implies weaker pinning. Therefore, those values are consistent with the activation energy analysis shown in Table I.
4. Conclusion

We measured the Hall resistivity of FeSe$_{1-x}$Te$_x$ films near $T_c$ and observed that the sign of the Hall resistivity changes for films with $x = 0.5$, while the sign of the Hall resistivity of films $x = 0.2$ remains the same as that in the normal state. Increasing applying current density, the sign change of the Hall resistivity vanished. Evaluation of the activation energy and the scaling exponent clarify that the film with $x = 0.5$ has the relatively stronger pinning than that of the film with $x = 0.2$. All of those results suggest that the Hall anomaly has deeply related to the vortex motion and the pinning in the iron-chalcogenide superconductor.

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