Strong and nonmonotonic temperature dependence of Hall coefficient in superconducting $K_x\text{Fe}_{2-y}\text{Se}_2$ single crystals

Xiaxin Ding, Yiming Pan, Huan Yang and Hai-Hu Wen

Center for Superconducting Physics and Materials, National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing 210093, China

In-plane resistivity, magneto-resistance and Hall effect measurements have been conducted on quenched $K_x\text{Fe}_{2-y}\text{Se}_2$ single crystals in order to analysis the normal-state transport properties. It is found that the Kohler’s rule is well obeyed below about 80 K, but clearly violated above 80 K. Measurements of the Hall coefficient reveal a strong but non-monotonic temperature dependence with a maximum at about 80 K, in contrast to any other FeAs-based superconductors. With the two-band model analysis on the Hall coefficient, we conclude that a gap may open below 65 K. The data above 65 K are interpreted as a temperature induced crossover from a metallic state at a low temperature to an orbital-selective Mott phase at a high temperature. This is consistent with the recent data of angle resolved photoemission spectroscopy. These results call for a refined theoretical understanding, especially when the hole pockets are absent or become trivial in $K_x\text{Fe}_{2-y}\text{Se}_2$ superconductors.

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I. INTRODUCTION

In iron-based superconductors (FeSCs), it is very important to understand the electron correlation in different bands and the orbital selective Mott transitions. As revealed by angle resolved photoemission spectroscopy (ARPES) and band structure calculations, the superconductivity and normal state of FeSCs are governed by their electronic structure involving the Fe 3d orbitals crossing the Fermi energy. Due to different structures and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrication, the iron chalcogenide superconductors show up with differences and probably different methods for fabrica

In this paper, we study the normal-state transport properties of quenched $K_x\text{Fe}_{2-y}\text{Se}_2$ superconducting single crystals with $T_c = 32$ K through the in-plane resistivity, transverse magnetoresistance (MR) and Hall effect measurements. We find that the Kohler’s rule is obeyed in the low temperature region. The Hall coefficient has a strong but non-monotonic temperature dependence below 150 K. This is in contrast with the FeAs-based systems in which the Hall coefficient shows a monotonic temperature dependence. These abnormal temperature-dependent behaviors cannot be de-
scribed by one single band model, suggesting the multi-band nature in K$_x$Fe$_{2-y}$Se$_2$. Using a two-band model (mainly $d_{xz/yz}$ and $d_{xy}$) analysis, we conclude that a gap may open below 65 K, while the non-monotonic temperature dependence of the Hall coefficient could be understood as a consequence of an orbital-selective Mott transition. These results would trigger further theoretical and experimental studies of the orbital selective correlation effect in FeSCs.

II. MEASUREMENTS OF MAGNETORESISTANCE AND HALL EFFECT

The K$_x$Fe$_{2-y}$Se$_2$ single crystals used for the transport measurements were fabricated by a self-flux method with a starting synthesizing composition of K:Fe:Se=0.8:2:2. The crystals are rapidly quenched in liquid nitrogen after heating to 350 °C and staying for several hours. Details of the preparation were described elsewhere. The quenching process can greatly improve the connection of the tiny superconducting networks (paths), and thus the global appearance of superconductivity is much better than the slowly cooled samples of K$_x$Fe$_{2-y}$Se$_2$. X-ray diffraction patterns (XRD) taken on the quenched crystals show only (00l) peaks with some small accompanying peaks. We think that the main diffraction (00l) peaks of the XRD pattern are coming from the major part of the sample, i.e., the K$_2$Fe$_4$Se$_5$ matrix, while the small accompanying peaks are coming from the minority superconducting networks. Because there is clear evidence of phase separation in the sample, it is meaningless to claim an uniform composition through out the sample. The microanalysis using energy-dispersive-spectrum (EDS) on the samples reveals that the background has a composition close to K:Fe:Se=2:4:5. For the present quenched sample, since the minority superconducting phase (path) has very small size which is smaller than the size of the electron beam in the EDS analysis, we could not use the EDS technique directly to get valid values of compositions for the three elements. Transport measurements were carried out with the six-lead method in a Quantum Design instrument physical property measurement system (PPMS). The electric contacts were made using silver paste in a glove box filled with nitrogen atmosphere. We have worked on two samples from different batches and the results are similar to each other.

In the K$_x$Fe$_{2-y}$Se$_2$ system, one concern is the phase-separation property, the superconducting and insulating phases could both contribute in transport measurements. As shown in Fig. 1, a broad hump with the peak at $T_H \sim 240$ K is observed in the normal-state, being similar to those reported previously. This anomaly, being
sensitive to the preparation process, could be caused by the connection in series between the metallic and the insulating phases as a result of the phase-separation picture. Hence, we mainly focus on the normal-state properties below $T_H$ which are dominated by contributions from the metallic phase (superconducting paths below $T_c$). The small residual resistivity with sharp superconducting transition ($T_c = 32$ K) indicates good connectivity of the superconducting paths.

In order to get more information of the normal-state properties, further investigations are provided by the transverse MR and Hall effect measurements on the same sample. In general, for most normal metals, the MR exhibits a $H^2$-dependence in the weak-field limit, and the MR normally affords a useful method to investigate the nature of electronic scattering. In Fig. 2(a), we show the field dependence of the transverse MR, $\Delta \rho_{xx}/\rho_0$, below 80 K, solid lines are fits to $\Delta \rho_{xx}/\rho_0 = aH^2$, $\rho_0$ is the resistivity under zero magnetic field. It is clear that the MR increases as $H^2$ in the sweeping magnetic field up to 5 T, signaling metallic behavior for $T \leq 80$ K, while the slight deviation at 35 K can be attributed to the superconducting fluctuations at a finite magnetic field. Besides, there is a fundamental difference in the MR between $T \leq 80$ K and $T \geq 85$ K. In the latter, as shown in Fig. 2(b), the MR seems not follow the $H^2$-dependence, which indicates a transition of the electronic characteristics from one to another. The inset of Fig. 2(a) shows the temperature dependence of MR at 5 T , a clear minimum occurs at about 80 K and the MR below 80 K shows a nice fit to the $T^{-2}$-dependence. It is interesting to note that, the crossover at around 80 K is consistent with the measurement of the finite-frequency dielectric function by means of terahertz spectroscopy in a Rb-based sister compound.

The Kohler’s rule, which assumes a simple scaling function of $\Delta \rho/\rho_0 = F(H/\rho_0)$, should be satisfied for a single band metal with an isotropic Fermi surface, with $\rho_0$ the resistivity at a fixed temperature and zero field. For a multiband system, this rule is also applicable as long as the number of charge carriers from each band is independent on temperature and the scattering rates of different bands have the similar temperature dependence. From the first glance at the data below 60 K, as shown in Fig. 3(a), it seems that the Kohler’s rule is slightly violated. Actually this slight ”deviation” of Kohler’s rule may not be true. The reason is that the resistivity at zero field, namely $\rho_0(T)=\rho_0(T = 0) + A/\tau$ does not really reflect directly the scattering rate $1/\tau$ when the residual resistivity is sizable. In addition, as argued above, there maybe a partial gap opening below 65 K, which may give an influence of the Kohler’s scaling rule. Instead of using the original scaling function $\Delta \rho/\rho_0 = F(H/\rho_0)$, we use here a more accurate form of the Kohler’s scaling rule $\Delta \rho/\rho_0 = F(H/\tau)$ where $\tau$ is the relaxation rate. Since the system exhibits metallic properties in the low temperature region, we could assume the relaxation rate as $\tau \propto T^{-2}$. Fig. 3(b) shows the refined Kohler’s plot of $\Delta \rho/\rho_0$ vs $(H/\tau)^2 \propto (HT^{-2})^2$. One can see that the Kohler’s rule is well obeyed below 80 K if we assume a general scattering rate $1/T^2$. In contrast, the Kohler’s rule is drastically violated above 85 K, as shown with the enlarged view in the inset of Fig. 3(a).

We now switch our attention to the temperature dependence of Hall coefficient. In the present $K_xFe_2-ySe_2$ system, Hall effect measurements may provide the message concerning the temperature dependence of the charge carrier density and mobilities of electrons in different bands of the superconducting phase. Since the insulating phase $K_xFe_2Se_5$ has the nearest band 300 meV below the Fermi energy, they should not contribute in the Hall effect measurements below $T_H$. In Fig. 4(a), we show the Hall resistivity $\rho_{xy}$ versus magnetic field up to 5 T, a linear relation between $\rho_{xy}$ and magnetic field $H$ has been found in wide temperature region (35 K to 150 K). From the $\rho_{xy}(H)$ data, the Hall coefficient $R_H$ is determined through $R_H = \rho_{xy}/H$ and shown in Fig. 4(b). The negative $R_H$ over the whole temperature region up to 150 K reveals that the conduction is dominated by electron-like charge carriers. However, the most remarkable feature in Fig. 4(b) is that the $R_H(T)$ shows a strong but non-monotonic temperature dependence. This is in sharp contrast with the FeAs-based 122 samples in which the Hall coefficient is monotonically dependent on temperature. By having a closer scrutiny to the temperature dependence of Hall coefficient, two characteristic temperatures could be defined: $T_{gap} = 65$ K and $T_{mott} = 85$ K. Below 65 K, $R_H$ decreases rapidly with a suppression towards lower temperatures. This is quite similar to that in the FeAs-based superconductors. Between 65 K and 85 K, $R_H$ is almost temperature independent, while it decreases upon raising temperature from 85 K to 150 K. This anomalous behavior has never been reported in previous studies in $K_xFe_2-ySe_2$ and suggests that something beyond the multi-band physics is very important here in determining the electric conduction.

III. DISCUSSION

According to the band structure calculations and ARPES studies, two sets of electronic orbitals near the Fermi level, namely $d_{xz}$ and $d_{yz}$ play an important role. Since the $d_{xz}$ and $d_{yz}$ are normally degenerate, we thus use a two band model to handle the issue. Based on the Boltzmann transport theory in the weak field limit, the equation of the Hall coefficient for two-band model could be simplified as

$$R_H = \frac{\sigma_1 R_{H1} + \sigma_2 R_{H2}}{(\sigma_1 + \sigma_2)^2} = \frac{n_1 \mu_1^2 + n_2 \mu_2^2}{-e(n_1 \mu_1 + n_2 \mu_2)}$$

(1)

where $\sigma_1 = e n_1 \mu_1, \sigma_2 = e n_2 \mu_2$ and $R_{H1} = -1/e n_1, R_{H2} = -1/e n_2$ are single band conductivity and Hall coefficients for the two orbitals, respectively. Based
The temperature dependence of the Hall coefficient below 65 K can be characterized by the fact that the recent THz spectroscopy experiments on Rb$_{1-x}$Fe$_2-y$Se$_2$ report a gap-like suppression of optical conductivity near the Fermi surface for the $d_{xy}$ orbital. Moreover, investigations using pump-probe spectroscopy and THz spectroscopy also evidenced the Mott-transition related behavior in the normal state. This scenario could give a reasonable explanation to our data here. Since the $d_{xz/yz}$ orbital remains metallic, we still express the mobility of the two orbitals as $\mu_i = \alpha_1 T^{-2}$ and the carrier density as a constant $n_i$. Meanwhile, we consider that the $d_{xy}$ band goes into the Mott phase with raising temperature. As it is well known, the Mott insulating behavior...
ior has been experimentally identified and theoretically explained in terms of the band narrowing effect associated with the electron-electron correlation. Therefore, the mobility of $d_{xy}$ orbital could be interpolated with the formula $n_2 = \alpha_2 T^{-\beta}/(1 + \gamma T)$, where $1/(1 + \gamma T)$ is the modification term associated with the Mott transition. To approach a solution, we may set the carrier density of the $d_{xy}$ orbital as a constant $n_2'$ for simplicity. Thus, the expression of the Hall coefficient with the orbital-selective Mott phase is written as

$$R_H = \frac{n_1^0 (\alpha T^{-2})^2 + n_2^0 (T^{-\beta})^2 - c (n_1^0 a T^{-2} + n_2^0 (1 + \gamma T))^2}{\alpha T^{-2} + n_2^0 (1 + \gamma T)}$$

(2)

where $\alpha = \alpha_1/\alpha_2$ is the relative ratio of the mobility coefficient of the two orbitals. In Fig. 4(b), the blue solid line above 85 K shows the theoretical fitting result. Consequently, we acquire the parameters as $\Delta = 7.7$ meV, $n_1^0 = 9 \times 10^{26}$ m$^{-3}$, $n_2^0 = 9 \times 10^{26}$ m$^{-3}$, $n_2^0 = 1.1 \times 10^{26}$ m$^{-3}$, $\alpha = 500$, $\beta = 0.2$ and $\gamma = 0.001$ K$^{-1}$. The plateau of the Hall coefficient $R_H$ between 65 K and 85 K may be viewed as the crossover of the two different regions. We note that the appearance of a gap with the value of $\Delta = 7.7$ meV below 65 K could be compared to the observation of high-temperature superconductivity at 65 K in single-layer FeSe films. We also can’t rule out the possibility of a pseudogap opening or other explanations for this gap-like suppression of $R_H$ below 65 K. The decrease of $R_H$ for increasing temperature starting from 85 K can get a strong support from the scenario of the orbital-selective Mott transition in this system. The explanation based on the orbital selective Mott transition should call for further theoretical and experimental efforts.

IV. CONCLUSION

In summary, magnetoresistance and Hall coefficient $R_H$ have been measured in superconducting K$_x$Fe$_{2-y}$Se$_2$ single crystals. The Kohler’s rule is well obeyed below 80 K by assuming a general scattering rate $1/\tau \propto T^2$. We have observed a strong and non-monotonic temperature dependence of the Hall coefficient in the normal state. Using a two-band model analysis and combining with the published data of the time domain optical conductivity measurements, we conclude that a gap may open below 65 K, while the data above 85 K could be understood as a consequence of an orbital-selective Mott transition of the $d_{xy}$ band.

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* lhwen@nju.edu.cn
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