Transport properties and asymmetric scattering in $Ba_{1-x}K_xFe_2As_2$ single crystals

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Resistivity, Hall effect, and magnetoresistance have been investigated systematically on single crystals of $Ba_{1-x}K_xFe_2As_2$ ranging from undoped to optimally doped regions. A systematic evolution of these properties has been observed. It is found that the resistivity in the normal state of $Ba_{1-x}K_xFe_2As_2$ is insensitive to the doped potassium concentration, which is very different from the electron-doped counterpart $Ba(Fe_{1-x}Co_x)_2As_2$, where the resistivity at 300 K reduces to the half value of the undoped one when the system is optimally doped. In stark contrast, the Hall coefficient R_H changes suddenly from a negative value in the undoped sample to a positive one with slight K doping, and it keeps lowering with further doping. We interpret this dichotomy as being due to the asymmetric scattering rate in the hole and the electron pockets with much higher mobility of the latter. The magnetoresistivity shows also a nonmonotonic doping dependence indicating an anomalous feature at about 80 to 100 K, even in the optimally doped sample, which is associated with a possible pseudogap effect. In the low-temperature region, it seems that the resistivity has similar values when superconductivity sets in, regardless of the different T_c values, which indicates that the influence of impurity scattering on superconductivity is weak. A linear feature of resistivity ρ_{ab} versus T was observed just above T_c for the optimally doped sample, suggesting a quantum criticality.

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I. INTRODUCTION

The discovery of iron-based superconductors has triggered great interest in the field of condensed matter physics. A lot of theoretical and experimental works suggest multiple Fermi surfaces (or gaps) and unconventional pairing mechanism.^{2–8} There is a common issue between the iron pnictides and the cuprates: in both systems the superconductivity is in the vicinity of the antiferromagnetic (AF) order, leading to a very similar phase diagram. However, the feature of the undoped (parent) phase of the two systems is actually quite different. The cuprate may be categorized as the so-called Mott insulator, while the iron pnictide is a poor metal. The phase diagram has thus been a focus of intense research in order to achieve a comprehensive understanding of the relationship between the AF and the superconducting (SC) states, and intimately the superconducting mechanism. 9-13 For the electron-doped 122 family, such as $Ba(Fe_{1-x}Co_x)_2As_2$, a lot of works, both in experiment and theory, have been done to illustrate the systemic evolution of the transport properties and electronic structure. These works all indicate the importance of the multiband effect. 10,14-18 It was presumably believed that in the underdoped regime, the AF and the SC phase compete for the density of states on the Fermi surfaces. In the overdoped regime, superconductivity suffers from the weakening of the spin fluctuations, 15 probably due to the loss of the Fermi surface nesting. These two effects lead to an asymmetric superconducting dome, which is somewhat different from the case in the cuprate superconductors. By analyzing the transport¹⁰ and optical data 16,19,20 in Ba(Fe_{1-x}Co_x)₂As₂, it has been claimed that the mobility of the electron band is much higher than that in the hole band. It is interesting to know in the hole-doped samples, would the mobility disparity survive, disappear, or change sign? In this paper, we report systematic studies on resistivity, Hall effect, and magnetoresistivity on selected hole-doped $Ba_{1-x}K_xFe_2As_2$ single crystals. The detailed investigations and analysis suggest the asymmetric quasiparticle scattering in the hole and electron bands, with still a much larger mobility in the electron band. Meanwhile, we present evidence that the electron doping induced by substituting the Fe sites with Co results in impurity scattering, but without breaking too much Cooper pairs. These Fe-sites doping may generate the impurities, which can only scatter the electrons with small momentum transfer, with the pairing induced by the interpocket scattering intact.

II. EXPERIMENTAL

The $Ba_{1-x}K_xFe_2As_2$ single crystals were grown by the self-flux method using FeAs as the flux the detailed procedures of synthesizing the samples are similar to the previous reports.^{21–23} The crystal structure and chemical composition were checked by x-ray diffraction and energy dispersive x-ray microanalysis. For the transport measurements, all the samples were cut into rectangular shape and the standard six electric probes were made by silver paste. The electronic transport measurements were carried out in a physical properties measurement system (PPMS, Quantum Design) with the temperature down to 2 K and magnetic field up to 9 T. The superconducting transition temperatures of the samples were determined by using the criterion of 50% of the normal-state resistivity ρ_n . The in-plane longitudinal and the Hall resistance were measured by either sweeping the magnetic field at a fixed temperature or sweeping the temperature at a fixed magnetic field. Both sets of data coincide with each other.

III. RESULTS AND DISCUSSION

A. Temperature and doping dependence of resistivity

Figure 1(a) shows the temperature dependence of resistivity for the $Ba_{1-x}K_xFe_2As_2$ single crystals with doping levels ranging from undoped (parent) phase to optimally doped compounds. It is clear that the potassium doping makes the system evolve from an AF state (with a resistivity anomaly) to a superconducting one. The sharp drop of resistivity at about 138 K due to the AF/structural transition can be observed in the parent phase. With doping holes, the resistivity anomaly is suppressed and shifts to lower temperatures, which is consistent with the peak in the derivative of the resistivity $d\rho/dT$ shown in Fig. 1(b). When the doping level x reaches 0.25, the resistivity anomaly disappears and a little pit can be observed in $d\rho/dT$, and the superconducting transition temperature is 29 K. With further doping, the T_c reaches a maximum value

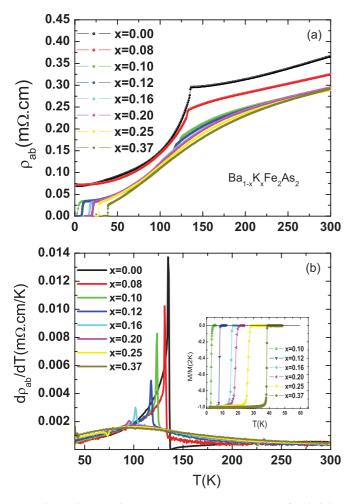


FIG. 1. (Color online) (a) Temperature dependence of resistivity of $\mathrm{Ba}_{1-x}\mathrm{K}_x\mathrm{Fe}_2\mathrm{As}_2$ ($x=0\sim0.37$) single crystals. The AF/structural transition is shifted to lower temperatures and becomes invisible with further doping potassium. The resistivity anomaly in the normal state can not be explicitly resolved when the doping level is 0.25 with $T_c=29$ K and beyond. (b) Temperature dependence of derivative of the resistivity $d\rho_{ab}/dT$. The peak in $d\rho_{ab}/dT$ associated with the AF/structural transition is suppressed with doping and disappears at x=0.25. Inset: zero field cooling magnetization of six superconductive samples.

at x = 0.37 with $T_c = 39$ K. For the electron-doped Fe-based 122 family, the magnetic and structural transitions are slightly separated²¹ leading to two close peaks on the $d\rho/dT$ versus T curves. While for hole-doped BaFe₂As₂, previous studies show that the AF and structure transitions occur at the same temperature in underdoped region, ^{12,13,24} in our measurements, we observed only one single sharp peak in $d\rho/dT$ versus T curves for underdoped Ba_{1-x}K_xFe₂As₂, which is consistent with previous results.

Here, we would like to emphasize several contrasting phenomena by comparing the resistivity curves in electron-doped Ba(Fe_{1-x}Co_x)₂As₂ (Co-122)¹⁰ and hole-doped $Ba_{1-x}K_xFe_2As_2$ (K-122) samples (see Fig. 1). Firstly, in the Co-122 samples, the resistivity at 300 K reduces to half value of undoped sample when the system is optimally doped, ¹⁰ this is actually not the case in K-doped Ba122. One can see from Fig. 1(a) that the resistivity at 300 K drops only about 20-30% when the doping is getting to the maximum value. Interestingly, the resistivity values when the superconductivity sets in are quite close to each other although T_c changes from 10 K (x = 0.10) to 29 K (x = 0.25). This may suggest that the resistivity is mainly dominated by the scattering of the electron band, which is weakly influenced by the hole doping. The interest stimulated by this observation is two fold: the threshold for the occurrence of superconductivity seems to be related to the residual resistivity at T_c while the T_c value is determined by the hole concentration, perhaps by how strong the suppression of the antiferromagnetic phase is. It is not clear at present, what leads to this strange behavior, but clearly it indicates a weak influence of the impurity scattering on the superconductivity. Secondly, the resistivity exhibits an uprising step at the AF/structural transition in the electron-doped Co-122, while in K-122, this transition exhibits always as a drop of resistivity at the antiferromagnetic transition temperature (T_{AF}) and it is smeared up gradually with more doping. Thirdly, the ratio between the room-temperature resistivity and the residual resistivity just above T_c (namely, RRR), is about 2.4 in Co-122,¹⁰ indicating a strong impurity scattering. But it seems that such a strong scattering does not suppress the superconductivity completely. Based on the picture of pairing through interpocket scattering, ^{2,3} the nonmagnetic impurities may be detrimental to the superconductivity if they induce the interpocket scattering. In this sense, the impurities here may only induce the scattering with small momentum transfer, for example, the intrapocket scattering. In the optimally holedoped samples, the RRR can get up to 14, indicating a weak impurity scattering. At high temperatures, the ρ_{ab} -T curve shows a bending down feature for the hole-doped samples. In a conventional metal with a single band, the bending down of resistivity was interpreted as approaching the Ioffe-Regel limit, which corresponds to the case when the mean free path induced by the phonon scattering is comparable to the atomic lattice constant. This seems not to be the case in the electron-doped samples Co-122, although they have the same atomic structure and similar phonon spectrum as the hole-doped samples, but the bending-down feature of resistivity has not been observed up to 300 K.

The above mentioned behavior of the resistivity can be qualitatively understood by the two-band scenario with asymmetric scattering rate in the hole and the electron pockets. According to the simple two-band model, the conductivity can be written as $\sigma = \sum_i \sigma_i$, where $\sigma_i = n_i e^2 \tau_i / m_i$ is the conductivity, n_i is the charge carrier density, τ_i is the relaxation time, and m_i is the mass of the *i*th band (i = e or h for the electron and hole bands, respectively). Therefore the resistivity can be described as

$$\rho = \frac{m_e m_h}{e^2 (n_e \tau_e m_h + n_h \tau_h m_e)}.$$
 (1)

It is known that the parent phase has identical areas of electron and hole Fermi pockets in the nonmagnetic phase, therefore we can assume identical charge carrier densities n_e and n_h for the two bands $(n_e \approx n_h \approx n_0)$. Considering $m_h > m_e$, as revealed by ARPES²⁵ and specific heat data, ²⁶ and assuming that $\tau_e > \tau_h$, the conductivity is thus dominated by the electron band. With electron doping, the term $n_e \tau_e m_h$ is getting much larger than $n_h \tau_h m_e$, which reduces the resistivity further. At the optimal doping at about x = 0.08, n_e has increased a lot, perhaps doubled. This reduces the resistivity to almost its half value. In the case of K-122, the situation is different. If we still adopt the relation of $n_e \tau_e m_h \gg n_h \tau_h m_e$, doping holes will decrease n_e but increase n_h , in this case, the resistivity should increase instead of decrease. Actually, hole doping will decrease n_e and increase n_h , but more important it will lower down the m_h and $1/\tau_h$. In this case, the resistivity is determined by a balance between the quantities of $n_e \tau_e m_h$ and $n_h \tau_h m_e$, and shows a weak doping dependence. A quantitative understanding would require a detailed doping dependence of n_i , τ_i , and m_i (i = e, h). This is beyond what we can get from a simple resistivity measurement and analysis. Some calculations indicate that the conductivity for electrons grows strongly upon electron doping, while the hole conductivity varies weakly compared to that of the electrons.²⁷ Thus the resistivity of hole-doped $Ba_{1-x}K_xFe_2As_2$ at 300 K changes less than that for electron-doped Ba($Fe_{1-x}Co_x$)₂As₂. The multiband effect in which one band is strongly coupled and relatively clean (electron band), while the other band is weakly coupled and characterized by much stronger impurity scattering (hole band) will cause anomalous T dependence of the in-plane resistivity: a convex curve with the tendency to saturate at high temperatures.²⁸

B. Trace of possible quantum critical points at the optimal doping

In Fig. 2(a), we present the temperature dependence of the resistivity $\rho_{ab}(T)$ normalized by the room-temperature resistivity $\rho_{ab}(300 \text{ K})$. From a glance at the data, we can immediately see that the ρ -T curve in the low-temperature region changes from a nonlinear to a linear behavior toward optimal doping. In order to know precisely the evolution of the resistivity with doping, we fit the data in the low-temperature region by the equation

$$\rho_{ab} = \rho_0 + A \times T^n \tag{2}$$

with three fitting parameters ρ_0 , A, and n for each curve. Due to the saturation in the resistivity at high temperatures and the anomalies of resistivity induced by the AF/structure transition, we fit the data from just above T_c to 80 K for all doped samples. We also did the fitting for the undoped (parent) phase with

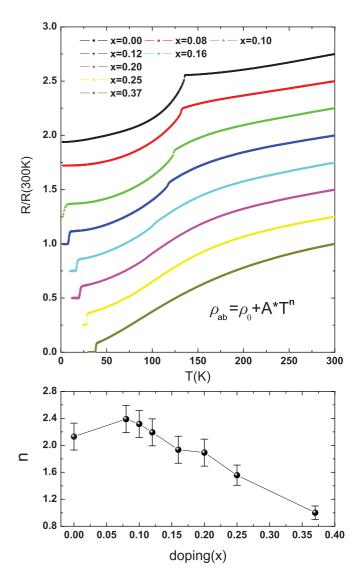


FIG. 2. (Color online) (a) Temperature dependence of the inplane resistivity normalized by its value at 300 K of $Ba_{1-x}K_xFe_2As_2$. The data for each doping are offset vertically by 0.25 for clarity. The corresponding bars near zero-temperature mark the zero resistance for each doping level. (b) The fitting parameter n for the undoped and seven doped samples (see text).

the data from 2 to 100 K. The resulted fitting parameters are presented in Table I and the exponent n is shown in Fig. 2(b). The evolution from a power law with exponent $n = 2 \pm 0.2$ for the undoped sample to a linear temperature dependence at optimal doping can be easily observed in Fig. 2(b), which may indicate the crossover from a Fermi liquid to non-Fermi liquid when the quantum critical point is approached. It was previously pointed out that the exponent n in metals near an AF quantum critical point (QCP) may be sensitive to disorder.²⁹ Although in K-122, the impurity scattering is quite weak, this can be corroborated by the negligible ρ_0 value at the optimal doping. At the optimal doping, the T-linear resistivity in the low-temperature region may suggest a quantum critical point. Similar behavior has been observed in $Sr_{1-x}K_xFe_2As_2$, ³⁰ $BaFe_2As_{2-x}P_x$, ³¹ $Ba(Fe_{1-x}Co_x)_2As_2$, ¹⁵ etc. It has been pointed out that the quantum fluctuation

TABLE I. Fitting parameters.

x (charges/Fe)	T_c (K)	$\rho_0 \; (\mathrm{m}\Omega \mathrm{cm})$	$A~(\times 10^{-5}~\text{m}\Omega~\text{cm/K}^2)$	n
0.00	0	0.07178	0.3662	2.13
0.10	5.5	0.03387	0.1767	2.32
0.12	10	0.03304	0.2898	2.20
0.15	18	0.02686	1.168	1.94
0.20	22	0.02682	1.4735	1.892
0.25	31	0.01651	7.24	1.56
0.37	39	-0.00146	104.68	1.01

becomes very strong when the Neel temperature of the AF order becomes zero. It is this strong quantum fluctuation that heavily couples to the itinerant electrons and modifies the transport property. Although it was argued that this linear feature may be reconstructed with a residual term and a T^2 term in the optimally doped Ba(Fe_{1-x}Co_x)₂As₂ system,²⁰ the systematic evolution shown in our present study can rule out this possibility.

For the cuprate superconductors, the antiferromagnetic order of the magnetic moments of the Cu²⁺ is completely suppressed before superconductivity sets in. They do not coexist at any point of the $T_c(p)$ (p is a doped hole number) phase diagram (an exception was suggested in the electron doped cuprate systems). In contrast, the coexistence of AF order and superconductivity can be observed in the underdoped region of the dome of K-122. 13,15,24,32,33 Figure 3 shows the phase diagram of $Ba_{1-x}K_xFe_2As_2$ and $Ba(Fe_{1-x}Co_x)_2As_2$. Although there are some reports claiming that magnetically ordered phases and SC state are probably mesoscopically/microscopically separated, 36-38 most of the studies on K- or Co-doped samples are in favor of the coexistence of magnetic order and superconductivity, and have consistently ruled out the presence of phase separation. 15,33,34,39 The very small residual specific heat coefficient $\gamma_0 (=C/T|_{T\to 0})$ in the optimally doped Ba_{1-x}K_xFe₂As₂ also strongly suggests the

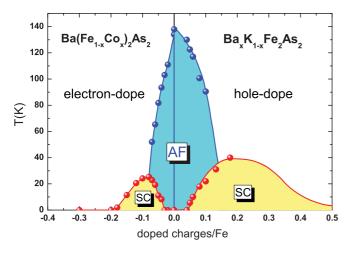


FIG. 3. (Color online) The phase diagram of $Ba_{1-x}K_xFe_2As_2$ and $Ba(Fe_{1-x}Co_x)_2As_2$. No splitting between the structural and AFM transitions has been observed in the hole-doped samples. The curve of T_c vs doping in a wide hole-doping region (solid line) was taken from Ref. 12. The data in the electron-doped regions were adopted from Ref. 10.

absence of macroscopic phase separation, since otherwise one should be able to see a large residual term of specific heat. Therefore we argue that the QCP occurs near the optimally doped samples where the AF order vanishes at about zero K. In the specific heat measurements, we also found that the mass enhancement m^*/m goes up quickly when the optimal doping point is approached. So Confirmation of the existence of a quantum critical point and the coexistence of magnetic order and superconductivity needs, certainly, extra investigations using other local probes.

C. Temperature and doping dependence of Hall coefficient

The Hall coefficient R_H from the undoped BaFe₂As₂ to the optimally doped K-122 is presented in Fig. 4(a), and the systematic evolution can be observed. The Hall coefficient R_H changes suddenly from a negative value in the undoped sample to a positive one with slight K doping, and it keeps lowering

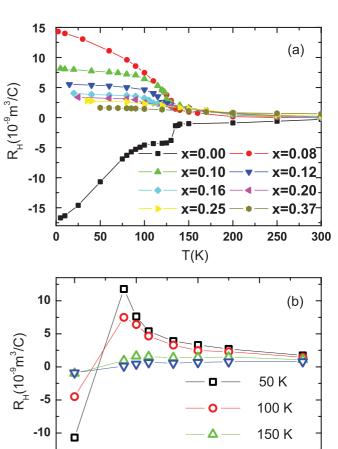


FIG. 4. (Color online) (a) Temperature dependence of Hall coefficient of $\text{Ba}_{1-x}K_x\text{Fe}_2\text{As}_2$ ($x=0\sim0.37$). The Hall coefficient R_H changes suddenly from a negative value in the undoped sample to a positive one with slight K doping. The AF/structure transition can be found in the underdoping dome (0.08 $\leq x \leq$ 0.25) as an onset of the rising of the Hall coefficient R_H . For the optimally doped sample, R_H variation with temperature is less pronounced. (b) The doping dependence of Hall coefficient at different temperatures.

0.2

Doping(x)

0.1

200 K

0.4

0.3

-15

0.0

with further doping in the low-temperature region. For each doping level, the sudden increase of the Hall coefficient corresponds to the AF/structure transition, which is consistent with the resistivity anomaly for underdoped K-122 ($x=0.1\sim0.25$). Above the AF/structure transition temperature the Hall coefficient varies weakly. The general formula for the Hall coefficient in the Boltzmann approximation reads

$$R_H = \sum_i \frac{\sigma_i^2}{en_i} / \left(\sum_i \sigma_i\right)^2. \tag{3}$$

For fully compensated semimetals within the two-band model, Eq. (3) reduces to

$$R_H = \frac{n_h \mu_h^2 - n_e \mu_e^2}{e(n_e \mu_e + n_h \mu_h)^2}.$$
 (4)

By definition, undoped samples are compensated, that is, $n_h = n_e = n_0$. Then Eq. (4) can be written as

$$R_{H} = n_{0}^{-1} \frac{\mu_{h} - \mu_{e}}{\mu_{h} + \mu_{e}} = n_{0}^{-1} \frac{\sigma_{h} - \sigma_{e}}{\sigma_{h} + \sigma_{e}},$$
 (5)

where $\mu_i = \sigma_i/n_i = \tau_i/m_i$ (i = e,h) is the mobility. If $\mu_e \gg \mu_h$, then $R_H \approx 1/en_e$, the transport is dominated by the electron band. However, with the potassium doping, the hole pocket increases in size instantly and the electron pocket contracts. At the hole doping with an $x \sim n_0$, the Hall coefficient changes sign. With further doping, the hole-doped systems have a presence of γ pocket near (π,π) . In addition to $(\pi,0)$ scattering between α and β sheets, new phase space for scattering opens up.²⁷ The asymmetric scattering rate in the hole and the electron pockets play an important role on R_H . Figure 4(b) shows the doping dependence of Hall coefficient at 50, 100, 150, and 200 K. In the low-temperature region, R_H changes sign and reaches a large value with little potassium doping. With further doping, R_H decreases gradually. In the high-temperature region the R_H varies very little, which is consistent with the recent calculation.²⁷ In principle, one can get the n_0 value from the temperature dependence of R_H where R_H becomes zero. Although for samples below x = 0.08 the superconducting transition looks very broad, the samples are apparently quite inhomogeneous, while the undoped (parent) sample is still quite pure. In this case, we can only estimate the n_0 value by connecting the data at zero doping to x = 0.08, and determine the n_0 at the point where the R_H changes sign. As shown in Fig. 4(b), this happens at about x = 0.04, therefore we get $n_0 = 0.02$. This value seems too low as compared with the local density approximation (LDA) calculations, which may suggest that the mobility of the hole band will get immediately enhanced after doping a very small amount of holes. Another reason for this contradiction would be that Eq. (4) does not take the effect of how the AF is suppressed by the hole doping into account. In the electron-doped 122 system, the transport property is dominated by the electrons. In the compensated case, the results can be explained by the remarkably different mobilities of holes and electrons. ¹⁰ For hole-doped K-122, the asymmetric scattering rate in the hole and the electron pockets still holds, but the relative ratio between τ_e/m_e and τ_h/m_h may change a little bit, namely, τ_h/m_h will get enhanced. Therefore, according to Eq. (4), the Hall coefficient will change sign because $n_h \mu_h^2$ is getting larger

than $n_e \mu_e^2$. This is especially necessary to interpret the drop of resistance at high temperatures when a small number of holes are doped into the system, as shown in Fig. 1.

D. Magnetoresistance

The temperature dependence of magnetoresistivity for nine samples measured at a magnetic field of 9 T is presented in Fig. 5. The data show also a nonmonotonic doping dependence and a sudden increase below AF/structure transition, which is associated very well with the anomaly found in resistivity and Hall effect. In the undoped sample, a large magnetoresistivity with a magnitude of about 35% (at about 9 T) has been found in the low-temperature region. With increasing doping, the magnetoresistivity decreases instantly. It remains unclear yet what causes this large magnetoresistance within the AF phase. There are two main explanations: (1) the magnetic field will break down the antiferromagnetic order to some extent and lead to stronger spin fluctuations and thus larger scattering to itinerant electrons, and (2) a magnetic field will induce a stronger localization leading to an enhanced resistivity.

In addition to this strong magnetoresistance in the antiferromagnetic state, an anomalous feature at about 80 to 100 K can be observed even in the optimally doped sample in which the AF state does not exist at all. One can see this in the inset of Fig. 5. The magnetoresistance rises up gradually when the temperature is lowered down, but it drops suddenly at about 100 K and reaches almost zero (for the optimally doped sample), then it rises up again in the lower-temperature region and smoothly connects to the magnetoresistance induced by the vortex motion in the mixed state. This anomaly at about 100 K may be associated with a possible pseudogap feature due to some unknown reasons. This is consistent with the recent observation in the c-axis resistive measurements where a maximum of ρ_c is observed. 18 A high-temperature pseudogap was also claimed very recently from the optical conductivity measurements.⁴⁰ Further experimental and theoretical investigations are needed to clarify this point.

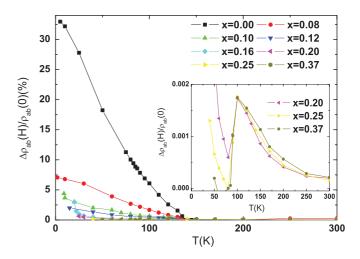


FIG. 5. (Color online) Temperature dependence of magnetoresistivity for $Ba_{1-x}K_xFe_2As_2$ ($x=0\sim0.37$) measured at 9 T. Inset: magnetoresistivity of the samples ($x=0.2,\ 0.25,\$ and 0.37). An anomaly at around 80–100 K can be easily observed.

IV. CONCLUSIONS

In summary, we investigated resistivity, Hall effect, and magnetoresistance systematically on single crystals of $Ba_{1-x}K_xFe_2As_2$ ranging from undoped to optimally doped samples. The resistivity in the normal state of $Ba_{1-x}K_xFe_2As_2$ is insensitive to the potassium doping, in sharp contrast with the electron-doped $Ba(Fe_{1-x}Co_x)_2As_2$ samples. The Hall coefficient R_H changes suddenly from a negative value in the undoped (parent) sample to a positive one with slight K doping, and it keeps decreasing with further doping. This contrasting behavior is interpreted as being due to the asymmetric scattering between the electron and hole bands, with a much larger mobility in the former. An anomalous

feature of magnetoresistivity has been observed at about 80 to 100 K and may be associated with a possible pseudogap feature. A linear feature of resistivity ρ versus T was observed just above T_c for the optimally doped sample, which suggests a quantum criticality.

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