A crossover in the phase diagram of NaFe$_{1-x}$Co$_x$As determined by electronic transport measurements

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Abstract. We report electronic transport measurements on single crystals of the NaFe$_{1-x}$Co$_x$As system. We found that the cotangent of the Hall angle, $\cot \theta_H$, follows $T^4$ for the parent compound with filamentary superconductivity and $T^2$ for the heavily overdoped non-superconducting sample. However, it exhibits approximately $T^3$-dependence in all the samples with bulk superconductivity, suggesting that this behavior is associated with bulk superconductivity in ferropnictides. A deviation develops below a characteristic temperature $T^*$ well above the structural and superconducting transitions, accompanied by a departure from power-law temperature dependence in resistivity. The doping dependence of $T^*$ resembles the crossover line of the pseudogap phase in cuprates.

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1. Introduction

The ferropnictide and cuprate high-temperature superconductors share some key similarities—an antiferromagnetism in parent compounds, the quasi-two-dimensional nature of superconducting CuO$_2$ and FeAs layers [1–4], and the emerging superconductivity realized by suppressing the antiferromagnetic ground states [5, 6]. A natural question is whether the two families have the same mechanism for superconductivity. In cuprate compounds, the nature of the pseudogap phase, which describes a state with partial gaps opening in the regions around antinodal points of the Fermi surface, is thought to be a key issue in understanding the high-temperature superconductivity. Recent evidence from resistivity [7], the Nernst effect [8], inelastic neutron scattering [9–11] and scanning tunneling spectroscopy [12, 13] suggested that the pseudogap phase in cuprates could be an electronic state with the four-fold rotational symmetry of underlying lattice breaking, that is, nematic order. Compared with the phase diagram of cuprates, the current interest in ferropnictides lies in the peculiar normal-state properties of these materials to detect whether there exists a pseudogap-like state. The energy gap developed above $T_c$, which is called a pseudogap, is very important for us to understand the mechanism of high-$T_c$ in cuprates.

In ferropnictides, electronic nematicity has been detected by in-plane resistivity anisotropy [14], angle-resolved photoemission measurements [15], scanning tunneling spectroscopy [16] and inelastic neutron scattering [17]. In these studies, the structural distortions that break the crystal’s $C_4$ rotational symmetry may apply external driving forces to induce electronic nematicity. Very recently, magnetic torque measurements by Kasahara et al [18] revealed a nematic transition at high temperatures above the structural and superconducting transitions in BaFe$_2$(As$_{1-x}$P$_x$)$_2$, with a characteristic temperature similar to the pseudogap crossover in cuprate superconductors. Without the potential external driving forces from the lattice, this transition can be considered to be induced by the electronic system only.

However, there lacks evidence directly related to the electronic properties of the high-temperature nematic transition reported by Kasahara et al. For an evident phase transition, such as the superconducting, antiferromagnetic and structural transitions in ferropnictides, specific heat data show anomalies at corresponding temperatures. Furthermore, in some doped Ba-122 compounds, the opening of a pseudogap was suggested from the maximum in the interplane resistivity [19–21], which happens at a temperature well above structural and superconducting transitions and extends throughout the superconducting doping region. It becomes non-trivial to locate the crossover line and determine the phase diagram. In this paper, we study the resistivity and Hall coefficient measurements on high-quality single crystals of NaFe$_{1-x}$Co$_x$As. In our previous work, we mapped out the phase diagram and studied the calorimetric properties.
of NaFe$_{1-x}$Co$_x$As [22]. However in this paper, we observed a crossover temperature in the superconducting doping region, which is above the structural and superconducting transitions. At high temperatures, the Hall angle, $\cot \theta_H$, reveals $T^3$-dependence for superconducting samples. Below a crossover temperature $T^*$, the resistivity and Hall angle deviate from power-law temperature dependence. In the electronic phase diagram, $T^*$ depicts a characteristic temperature similar to the pseudogap crossover in cuprate superconductors.

2. Experimental details

High-quality single crystals of NaFe$_{1-x}$Co$_x$As were grown by the NaAs flux method [22]. The accurate chemical composition of the single crystals was determined by energy-dispersive x-ray spectroscopy. The standard instrument error for this method is around 10%. The single crystals of $x = 0, 0.010, 0.014, 0.017, 0.028, 0.042, 0.047, 0.061, 0.075$ and 0.109 were measured, with a high quality characterized and demonstrated in our previous report [22]. Measurements of resistivity and the Hall effect were conducted by using the PPMS-9T (Quantum Design). The resistivity was measured using the standard four-probe method. The contacts cover the sides of the samples to ensure in-plane transport. The Hall coefficient was measured by sweeping the field from $-5$ to $5$ T at various temperatures. We firstly confirmed the linearity of Hall resistivity $\rho_{xy}$ to a magnetic field in the normal state. Excellent linearity can be observed for all the samples in the normal state except that a very weak deviation from linearity appears below $T_{SDW}$ in underdoped samples. Then the temperature dependence of Hall coefficients was obtained from the subtraction of the voltages measured at $5$ and $-5$ T, $R_H = [V_{xy}(5\,T) - V_{xy}(-5\,T)] \times d/2I$, where $d$ is the thickness of crystals and $I$ is the current. The cotangent of the Hall angle, $\cot \theta_H = \rho / \rho_{xy}$, was calculated at $5$ T, where $\rho_{xy}$ is Hall resistivity. It should be addressed that the Hall coefficient cannot be well measured at temperatures above $200$ K; the possible reason is that Na ions could move above $200$ K. All the samples we used are the same as those in our previous work [22] and $T_c$, $T_{SDW}$ and $T_s$ are summarized in table 1 and plotted in figure 6. All the superconducting samples show nearly full shielding fraction except for the parent compound, which has filamentary superconductivity with no jump in specific heat at $T_c$ and tiny superconducting shielding fraction down to 2 K. The filamentary superconductivity is due to multiply connected superconducting filaments.

| $x$  | $T_c$ (K) | $T_{SDW}$ (K) | $T_s$ (K) | $T^*$ (K) | $T^\dagger$ (K) |
|-----|-----------|----------------|----------|----------|-----------------|
| 0   | 40.6 ± 1.0 | 51.4 ± 1.0     | 105 ± 5  | 116 ± 4  |
| 0.010 | 13.8 ± 0.3 | 28.2 ± 1.0     | 115 ± 5  | 113 ± 4  |
| 0.014 | 16.3 ± 0.3 | 21.8 ± 1.5     | 120 ± 5  | 106 ± 4  |
| 0.017 | 17.1 ± 0.3 | 30.0 ± 1.8     | 120 ± 5  | 103 ± 4  |
| 0.028 | 20.1 ± 0.3 |              | 105 ± 5  | 92 ± 4   |
| 0.042 | 18.9 ± 0.3 |              | 95 ± 5   | 85 ± 6   |
| 0.047 | 16.2 ± 0.3 |              | 80 ± 5   | 70 ± 6   |
| 0.061 | 13.5 ± 0.3 |              | 60 ± 5   | 52 ± 6   |
| 0.075 | 10.7 ± 0.3 |              | 50 ± 5   | 40 ± 6   |
| 0.109 | 0          |              | 0        | 0        |
Figure 1. Temperature dependence of in-plane resistivity for NaFe$_{1-x}$Co$_x$As single crystals. The solid line is the fitting curve by power-law temperature dependence with the formula $\rho = A + B \times T^\alpha$ to the resistivity data. The power-law exponents, $\alpha$, are shown on the right of the panel, which decrease from 2.23 for $x = 0$ to 1.41 for the optimal doping level and then increase to 1.90 for the heavily overdoped sample. Obvious deviation from the high-temperature power-law behavior can be seen in resistivity and the deviation temperature ($T^*$) decreases with increasing Co concentration. In addition, the characteristic temperatures of the spin density wave (SDW) and structural transitions are also marked as $T_{SDW}$ and $T_s$.

which do not show bulk superconductivity. The filamentary superconductivity was also observed in other iron pnictide superconductors such as Ca$_{1-x}$RFe$_2$As$_2$ (R is rare earth) [23].

3. Results and discussion

We carefully measured the resistivity on single crystals of the NaFe$_{1-x}$Co$_x$As system with $x = 0$–0.109 ranging from the parent compound to the heavily overdoped non-superconducting composition as shown in figure 1. For the parent compound and the underdoped crystals, there exists an upturn in resistivity below 50 K due to the structural and SDW transitions [22]. An evident feature is that the resistivity curvature changes well above the $T_i$ in the parent compound and all underdoped samples. For the optimally doped and overdoped crystals, there seems to be no anomaly in the normal-state resistivity. In the high-temperature cuprate superconductors, the mapping of in-plane resistivity curvature is a useful way to determine electronic phase diagrams. In particular, the pseudogap crossover line can be conveniently obtained by this method [24].
Figure 2. \((\rho - A)/T^\alpha\) as a function of temperature for all the crystals of \(\text{NaFe}_{1-x}\text{Co}_x\text{As}\). It clearly shows the temperature at which the deviation develops from the high-temperature power-law dependence of in-plane resistivity \(\rho\). The dashed lines are a guide to the eye for the \(A + B \times T^\alpha\) dependence. The deviation at \(T^\dagger\) is marked by arrows.

Here, we made power-law fitting on the resistivity data with the formula \(\rho = A + B \times T^\alpha\) and show them in figure 1. One can see that the resistivity follows power-law dependence at high temperature and starts to deviate at a characteristic temperature, \(T^\dagger\), for all the samples. The doping dependence of \(T^\dagger\) will be shown in figure 6. For the heavily overdoped non-superconducting sample \((x = 0.109)\), no deviation from the power-law dependence can be observed. As shown in figure 1, the power-law exponent \(\alpha\) decreases with increasing Co doping from the parent compound to the optimally doped crystals, and then increases with Co concentration in the overdoped region. The \(\alpha\) reaches the smallest value of 1.41 in the optimally doped sample.

To accurately determine \(T^\dagger\), we plot \((\rho - A)/T^\alpha\) versus temperature, as shown in figure 2. The characteristic temperature \(T^\dagger\) can be well defined, at which \(\rho\) starts to deviate from the power-law temperature dependence. As shown in figure 2, the heavily overdoped crystal with \(x = 0.109\) shows a deviation from the power-law temperature dependence due to a tiny superconducting transition around 6 K, so that we take \(T^\dagger = 0\) K. The deviation temperature, \(T^\dagger\), determined by our fitting is highly repeatable. Table 1 summarizes the \(T^\dagger\), which monotonically decreases with increasing Co concentration and goes to zero in the heavily overdoped non-superconducting compound \((x = 0.109)\). We should address that the variation of the temperature region used for fitting does not change the deviation temperature \(T^\dagger\) significantly, which suggests that our results are reliable.

Figure 3 shows the temperature dependence of Hall coefficients, \(R_{\text{H}}\), for various single crystals of \(\text{NaFe}_{1-x}\text{Co}_x\text{As}\). The Hall coefficients show a systematic evolution with increasing Co doping with a strong temperature dependence. The magnitude of Hall coefficients at 200 K increases upon Co doping, and then sharply decreases for the heavily overdoped non-superconducting sample with a value smaller than those of superconducting samples.
Figure 3. Temperature dependence of the Hall coefficient, $R_H$, for various single crystals of NaFe$_{1-x}$Co$_x$As.

It is worth noting that the Hall coefficient in the non-superconducting $x = 0.109$ compound is nearly independent of temperature, bearing a feature that is usually found in a conventional Fermi-liquid metal. This behavior suggests that NaFe$_{1-x}$Co$_x$As becomes a traditional metallic material at very high doping levels. The abrupt increase in magnitude of Hall coefficients below 50 K for the underdoped samples is due to the structural transition.

The complicated properties of temperature-dependent Hall coefficients can be expressed in a simple manner by looking at the cotangent of the Hall angle, \( \cot \theta_H = \rho / \rho_{xy} \) \[25\] as shown in figure 4, where $\rho_{xy}$ is Hall resistivity. It is interesting to notice that \( \cot \theta_H \) shows power-law temperature dependence for all the single crystals of NaFe$_{1-x}$Co$_x$As: $T^3$ for the parent compound, approximately $T^3$ for all the superconducting crystals and $T^2$ for the heavily overdoped non-superconducting sample. $T^\beta$-dependent $\cot \theta_H$ with $\beta = 2.5–3.0$ has been reported in BaFe$_{2−x}$Co$_x$As$_2$ single crystals \[26\]. In cuprates, $\cot \theta_H$ behaves approximately as $T^2$ \[25\], regardless of materials and doping level, except that $T^4$-dependence is found in the electron-doped cuprates \[27\], which is interpreted by the multi-band effect with different contributions from various bands.

A careful examination of figure 4 indicates slight curvatures in the plot, suggesting that the best power laws in a wide temperature range deviate slightly from an integer $\beta$. Figure 5 shows plots of $(\cot \theta_H - C) / T^\beta$ versus $T$ for all the samples, in which the power-law temperature dependence is cancelled out, so that one can easily see the temperature range in which the $T^\beta$ behavior holds well. Here, $C$ is the offset value and $\beta$ is the best power. In figure 5, the power-law temperature-dependent $\cot \theta_H$ holds very well down to a characteristic temperature, $T^*$, for all the samples. Below $T^*$, $\cot \theta_H$ departs from the power-law $(T^\beta)$ behavior. This characteristic temperature decreases with increasing Co doping and goes to zero in the heavily overdoped non-superconducting sample. One may notice that the deviation goes in different directions for various doping, which is probably related to the details in the quasiparticle scattering processes.
Figure 4. The cot $\theta_H$ plotted in power-law temperature scale for NaFe$_{1-x}$Co$_x$As single crystals. All the superconducting samples show approximately $T^3$-dependent cot $\theta_H$ except for the parent compound with filamentary superconductivity.

that varies with Co concentrations. We should address that the variation of the temperature region used for fitting does not change the deviation temperature $T^*$ significantly, which suggests that our results are reliable.

Figure 6 shows the phase diagram and we plot $T^+$ and $T^*$ as a function of doping. The two characteristic temperatures are highly consistent with each other, although they are obtained by different methods. Generally, both $T^+$ and $T^*$ decrease with increasing Co doping, and go to zero at the doping level $x = 0.109$ where $T_c$ goes to zero. The phase diagram in figure 6 is quite similar to the pseudogap phase diagram of the high-temperature cuprate superconductors. Indeed, the deviation of cot $\theta_H$ from $T^2$-dependence has been used to characterize the onset of the pseudogap in hole-doped cuprate superconductors [28], and the pseudogap crossover line can be conveniently determined by resistivity curvature mapping [24]. Here, these methods were used to determine $T^+$ and $T^*$ and reveal the crossover line above the structural and superconducting transitions. Our data strongly suggest that a crossover occurs at the $T^*$ in
Figure 5. (cot $\theta_H - C)/T^\beta$ as a function of temperature for all the crystals of NaFe$_{1-x}$Co$_x$As. It clearly shows the temperature at which the deviation develops from the high-temperature power-law dependent cot $\theta_H$. The dashed lines are a guide to the eye for the $C + D \times T^\beta$ dependence. The deviation at $T^*$ is marked by arrows.

Figure 6. Phase diagram of the NaFe$_{1-x}$Co$_x$As system. The superconducting transition temperature, $T_c$, SDW transition temperature, $T_{SDW}$, and structural transition temperature, $T_s$, were determined from resistivity, susceptibility and specific heat measurements by our group [22]. For the parent compound, the specific heat shows the structural and SDW transitions with no anomaly at $T_c$, suggesting a filamentary superconductivity. $T^\dagger$ and $T^*$ were determined in figures 2 and 5, respectively.

ferropnictide superconductors. We are careful that our data do not indicate any 'gapping' behavior as observed in cuprates, although we stress the similarity between the characteristic $T^*$ temperature and the pseudogap crossover temperature.
A natural question is what happens below $T^*$? Without the driving forces from structural or magnetic transitions, $T^*$ indicates a crossover with purely electronic origin. A possible scenario is that the electronic nematic state sets in below the characteristic temperature. Indeed, a similar crossover induced by electronic nematicity has been observed in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ well above the structural and superconducting transitions [18]. Recently, in the in-plane and interplane resistivity of NaFe$_{1-x}$Co$_x$As, some anomalies were observed, which was thought to possibly be the feature of a pseudogap [29]. Hereby we presented clear and precise pictures for such a crossover line in the phase diagram. Another intriguing fact is that the Hall angle exhibits approximate $T^3$-dependence for all the superconducting crystals despite the multiband nature, suggesting a peculiar balance between the motions of carriers on electron-type and hole-type Fermi sheets. However, the parent compound with filamentary superconductivity and the heavily overdoped non-superconducting crystal exhibit a different power-law behavior. Such distinct power-law temperature dependences of $\cot \theta_H$ between samples with bulk superconductivity and those without bulk superconductivity were actually observed in doped Ba-122 systems, as reported in our other paper [30]. This suggests that some balance between the motions of carriers on different types of Fermi sheets may be connected with the occurrence of bulk superconductivity.

4. Conclusion

In conclusion, our electronic transport studies reveal the existence of a crossover temperature well above structural and magnetic transitions. In the phase diagram of NaFe$_{1-x}$Co$_x$As, the crossover line resembles the pseudogap phase diagram in cuprate superconductors. An interesting phenomenon is that the Hall angle reveals approximate $T^3$-dependence of $\cot \theta_H$ in the whole superconducting regime, suggesting that this behavior is associated with bulk superconductivity in ferropnictides. These findings shed light on the mechanism of the high-temperature superconductivity in ferropnictides and potentially the superconductivity in cuprates as well.

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