Anomalous Transport Properties and Phase Diagram of the FeAs-Based $SmFeAsO_{1-x}F_x$ Superconductors

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We report the detailed phase diagram and anomalous transport properties of Fe-based high-T_c superconductors $SmFeAsO_{1-x}F_x$. It is found that superconductivity emerges at $x \sim 0.07$, and optimal doping takes place in the $x \sim 0.20$ sample with the highest $T_c \sim 54$ K. T_c increases monotonically with doping; the anomaly in resistivity from structural phase or spin-density-wave order is rapidly suppressed, suggesting a quantum critical point around $x \sim 0.14$. As manifestations, a linear temperature dependence of the resistivity shows up at high temperatures in the $x < 0.14$ regime but at low temperatures just above T_c in the $x > 0.14$ regime; a drop in carrier density evidenced by a pronounced rise in the Hall coefficient is observed below the temperature of the anomaly peak in resistivity. A scaling behavior is observed between the Hall angle and temperature: $\cot\theta_H \propto T^{1.5}$ for all samples with different x in $SmFeAsO_{1-x}F_x$ system.

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Since the discovery of high-transition temperature (T_c) superconductivity in layered copper oxides, extensive effort has been devoted to exploring the higher T_c superconductivity. Very recently, layered rare-earth metal oxypnictides LnMPnO (Ln = La, Pr, Ce, Sm; M = Fe, Co, Ni, Ru; Pn = P and As) with a ZrCuSiAs-type structure [1,2] have attracted great attention due to the discovery of superconductivity at $T_c = 26$ K in the iron-based $LaFeAsO_{1-x}F_x$ ($x = 0.05$–0.12) [3]. Immediately, T_c was drastically raised to 43 K in $SmFeAsO_{1-x}F_x$ [4], followed by reports of $T_c = 41$ K in $CeFeAsO_{1-x}F_x$ [5] and 52 K in $PrFeAsO_{1-x}F_x$ [6]. These discoveries have generated much interest for exploring a novel high-temperature superconductor and have provided a new material base for studying the origin of high-temperature superconductivity. The superconductivity in these materials appears to be unconventional, and much careful work will be required to elucidate the interesting physics here. It appears that the electron-phonon interaction is not strong enough to give rise to such high-transition temperatures [7], while strong ferromagnetic and antiferromagnetic fluctuations have been proposed to be responsible [8–10].

The magnetic fluctuations associated with a quantum critical point (QCP) are widely believed to cause the non-Fermi liquid behaviors and unconventional superconductivities, for example, in heavy-fermion systems and high-temperature cuprate superconductors. The superconductivity in a $LaFeAsO_{1-x}F_x$ system competes with spin-density-wave (SDW) order [11]. Neutron diffraction shows a long-rang SDW-type antiferromagnetic order at ~134 K in LaOFeAs [12]. Therefore, a possible QCP and its role in this system are of great interests.

The undoped material LaFeAsO shows an anomaly in resistivity at 150 K, which is associated with the structural transition and is 15–20 K higher than SDW transition [12]. The SDW and the anomaly in resistivity are suppressed, and superconductivity emerges with increasing F doping [11]. Systematical characterizations for evolution of the superconductivity and SDW (or structural transition) with F doping are important for understanding the underlying physics. Here we successfully prepared a series of $SmFeAsO_{1-x}F_x$ samples with $x = 0$–0.3 and systematically studied their resistivity and Hall coefficient and gave its phase diagram. The resistivity shows a clear anomaly, and the Hall coefficient increases sharply at temperature $T_s \sim 148$ K for SmFeAsO, indicating structural transition or the onset of SDW. T_s was found to decrease with increasing doping, manifesting suppression of structural transition or SDW order. A crossover occurs around $x \sim 0.14$ for T-linear dependence of resistivity from the high-temperature range to in low-temperature range (just above T_c) with increasing doping. The drop in carrier density and a T-linear dependence of the resistivity are two hallmarks of a quantum phase transition in metals [13], suggesting the existence of a QCP in this system.

Polycrystalline samples with nominal composition $SmFeAsO_{1-x}F_x$ ($x = 0$–0.3) were synthesized by conventional solid state reaction using high purity SmAs, SmF_3, Fe, and Fe_2O_3 as starting materials. SmAs was obtained by reacting Sm chips and As pieces at 600 °C for 3 h and then 900 °C for 5 hours. The raw materials were thoroughly grounded and pressed into pellets. The pellets were wrapped into Ta foil and sealed in an evacuated quartz tube. They are annealed at 1160 °C for 40 h, then cooled to 400 °C at 4 °C/min, and finally to room temperature by furnace. The sample preparation process except for annealing was carried out in glove box in which high pure argon atmosphere is filled. Figure 1(a) shows the x-ray diffraction

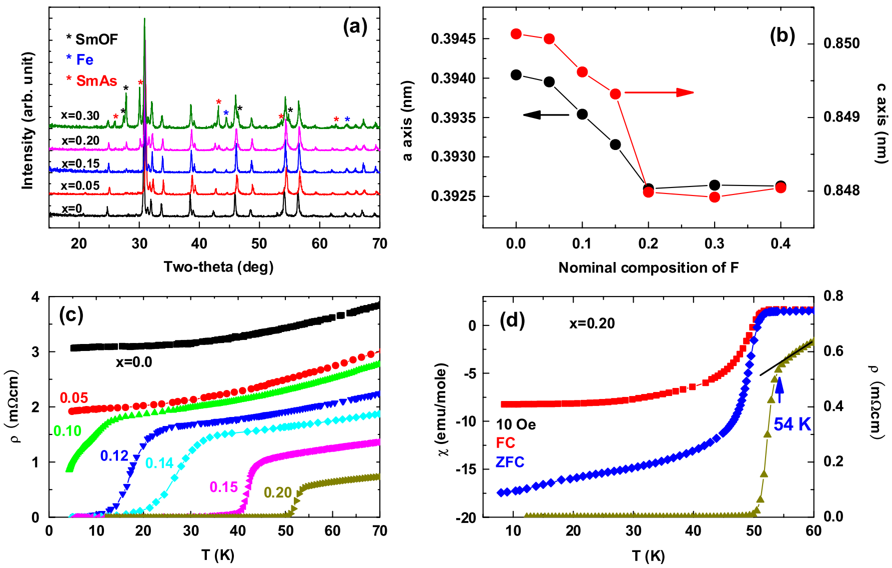

FIG. 1 (color online). (a) X-ray diffraction patterns at room temperature for the samples $SmFeAsO_{1-x}F_x$. (b) Variation of lattice parameters with x. (c) Superconducting transitions for the samples $SmFeAsO_{1-x}F_x$ with $x =$ 0–0.20. (d) The highest T_c of ~54 K and 51.8 K determined by resistivity and susceptibility is obtained in the $x =$ 0.20 sample. FC and ZFC are defined as field and zero-field cooling conditions in susceptibility measure, respectively.

(XRD) patterns for the polycrystalline samples $SmFeAsO_{1-x}F_x$ with different x. The main peaks in XRD pattern can be well indexed to the tetragonal ZrCuSiAs-type structure. The XRD patterns show that the samples with $x = 0$ and 0.05 are single phase. A tiny but noticeable trace of impurity phases SmOF, Fe, and SmAs is observed for $0.12 \leq x \leq 0.20$. Large amount of impurity phases are observed in the samples with $x > 0.2$. Figure 1(b) shows variation of a-axis and c-axis lattice parameters with F doping. It shows that both of a-axis and c-axis lattice parameters decrease systematically with nominal dopant concentration by substitution of F^- for O^{2-}. But the lattice constants do no change beyond $x = 0.2$, suggesting that the chemical phase boundary is reached at $x \sim 0.2$ in the $SmFeAsO_{1-x}F_x$ system.

Figure 1(c) shows the superconducting transition of the resistivity for the $SmFeAsO_{1-x}F_x$ system. No superconducting transition is observed down to 5 K for the samples with $x = 0$ and 0.05. The $x = 0.1$ sample shows an onset transition at ~12 K, but no zero resistance is observed down to 5 K. The emergence of superconductivity roughly occurs at $x \sim 0.07$. The superconducting transition temperature increases monotonically with increasing F content up to 0.2; optimal doping takes place in $x = 0.2$ sample with highest $T_c \sim 54$ K as shown in Fig. 1(d). The T_c determined by susceptibility is 17.1, 27.0, 41.8, and 51.8 K for $x = 0.12$, 0.14, 0.15, and 0.20, respectively. As shown in Fig. 1(a), a large amount of impurity phases shows up for the samples with x larger than 0.2, but these samples with x up to 0.5 still show superconductivity at ~50 K, which is nearly independent of x. It further evidences that the F doping is limited to be about 0.2.

Figure 2 shows the temperature dependence of resistivity in normal state with temperature up to 475 K for the samples $SmFeAsO_{1-x}F_x$ ($x =$ 0–0.2). The undoped sample shows a similar behavior to that observed in LaFeAsO [3]. An anomalous peak associated with structural transition shows up at $T_s \sim 150$ K [12]. Below T_s, the resistivity drops steeply. The temperature corresponding to the peak in resistivity is defined as T_s. F doping leads to a suppression of the anomaly peak and to the shift of T_s to lower

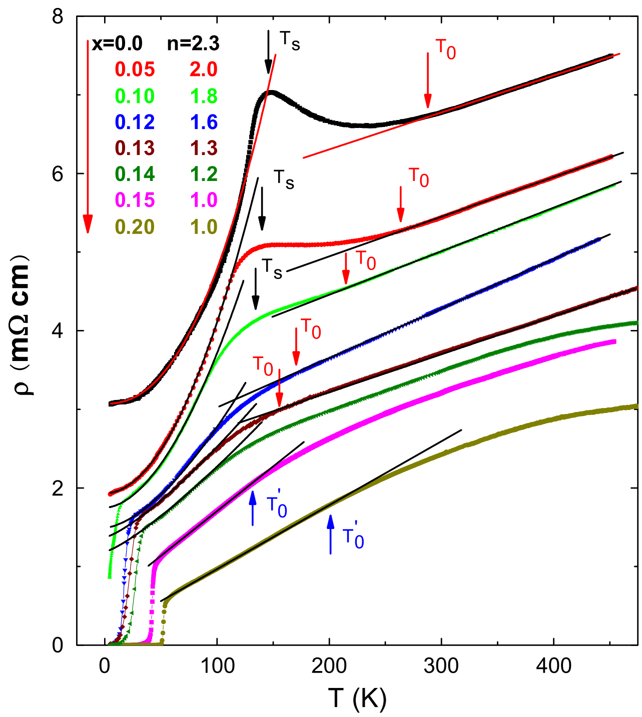

FIG. 2 (color online). Temperature dependence of the resistivity in the normal state up to 475 K for the samples $SmFeAsO_{1-x}F_x$ with $x = 0$, 0.05, 0.10, 0.12, 0.13, 0.14, 0.15, and 0.20. Arrows denote the anomaly temperature (T_s) of resistivity and deviating temperatures from T-linear behavior (T_0 and T_0'). The low-temperature resistivity can be well fitted with $a + bT^n$ for all the samples with $x \leq 0.14$, and the fitting parameter n decreases monotonically from 2.3 to 1.2 with increasing x from 0 to 0.14, while the n is fixed at 1 for $x \geq 0.15$.

temperature. At 10% F doping, a weak anomaly peak is still observed. Another striking feature is that a linear temperature dependence of the resistivity persists from high temperatures to a characteristic temperature (T_0) in the $x \leq 0.10$ range, at which the resistivity deviates from T-linear behavior and increases with decreasing temperature. This could arise from magnetic correlation before formation of SDW. The T_0 of undoped sample is about ~290 K, much higher than $T_s \sim 150$ K of the anomaly peak. Such a behavior is very similar to that observed in the "stripe" phase of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$, where deviation of resistivity from T-linear behavior also occurs at 150 K above the stripe formation temperature [14]. In contrast to the case of the samples with $x \leq 0.10$, no anomaly peak is observed in the $x \geq 0.12$ sample, and the resistivity decreases more quickly than the linear behavior below T_0, being similar to the pseudogap behavior in high-T_c cuprates [15]. The steep drop in resistivity below T_s has been ascribed to the occurrence of SDW [11].

Remarkably, the low-temperature resistivity can be well fitted with $a + bT^n$, and the fitting parameter n shows a systematical change from 2.3 to 1 with increasing F content from $x = 0$ to 0.15 (Fig. 2). An intriguing observation is that a crossover in the temperature dependence of the resistivity happens between the samples with $x = 0.14$ and $x = 0.15$. In contrast to the samples with $x \leq 0.13$, the high-temperature resistivity for the samples with $x > 0.14$ does not follow a T-linear behavior but tends to be saturated. However, the temperature dependence of the low-temperature resistivity just above T_c changes to T-linear dependence, and the resistivity deviates from the T-linear behavior in high temperatures at certain temperature (T_0'). T_0' increases from 130 K for the $x = 0.15$ sample to 205 K for the $x = 0.20$ sample. These results indicate that a profound change in resistivity takes place around $x \sim 0.14$, suggesting that the complete suppression of SDW (or structural transition) occurs and the QCP appears around $x = 0.14$. Particularly, a possible explanation for T-linear resistivity, which is widely used to explain the T-linear resistivity in heavy-fermion metals [13], is the scattering of charge carriers by fluctuation associated with the QCP.

Temperature dependence of Hall coefficient (R_H) for the $SmFeAsO_{1-x}F_x$ ($x = 0$–0.2) system is shown in Figs. 3(a) and 3(b). In order to obtain a good and uniform Hall voltage signal, the sample was cut into a rectangle shape with a thickness of ~200 μm. The longitudinal and Hall voltages were measured by using the standard dc 6-probe method. The Hall voltage was found to be linear with the magnetic field. The Hall coefficient is negative and decreases with increasing x, indicating that F doping leads to an increase in carrier concentration, and the dominated carrier is electron. The Hall coefficient for the samples with $x = 0$, 0.05 and $x = 0.10$ shows a strong temperature dependence at low temperatures. As shown in Fig. 3(a), the Hall coefficient shows a pronounced rise at a certain temperature which coincides with T_s of the anomaly peaks observed in Fig. 2. It indicates a drop in carrier concentration at T_s due to the occurrence of SDW. It has been revealed before that the Hall coefficient is prominently enhanced if strong antiferromagnetic (or SDW) fluctuations exist in heavy-fermion system [16]. The evolution of R_H with x is very similar to the R_H behavior near a antiferromagnetic QCP in the heavy-fermion system

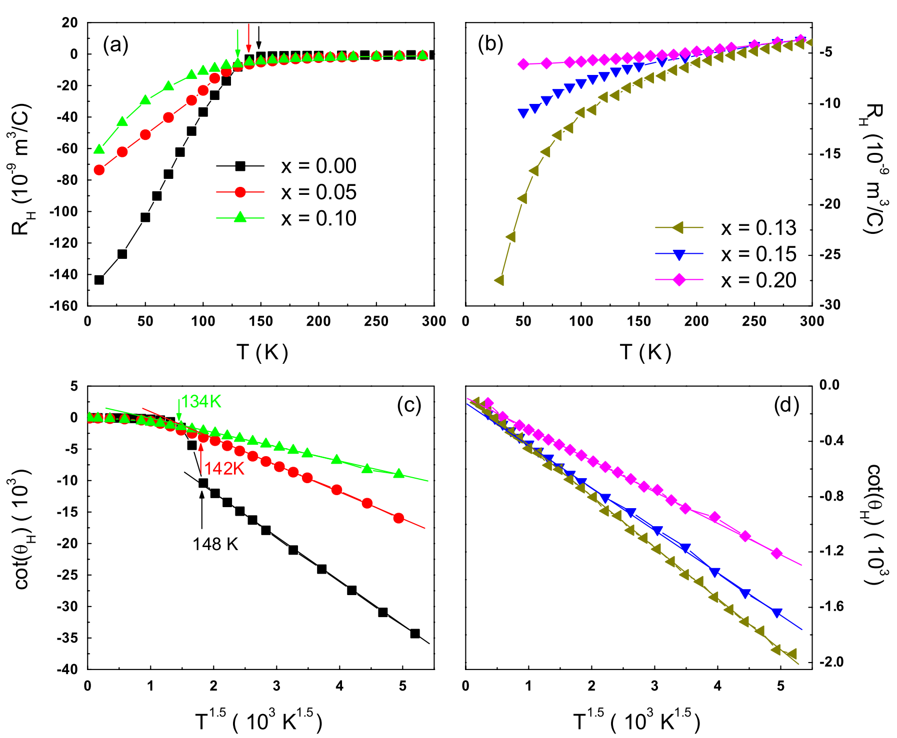

FIG. 3 (color online). (a) Temperature dependence of the Hall coefficient for the samples $SmFeAsO_{1-x}F_x$ with $x = 0$, 0.05, and 0.10. (b) Temperature dependence of the Hall coefficient for the samples with $x = 0.13$, 0.15, and 0.20. A pronounced rise in the Hall coefficient is observed for the $x = 0$, 0.05, and 0.10 samples. (c) The Hall angle is plotted as $\cot\theta_H$ vs $T^{1.5}$ for the samples with $x = 0$, 0.05, and 0.10; (d) $\cot\theta_H$ vs $T^{1.5}$ for the samples with $x = 0.13$, 0.15, and 0.20.

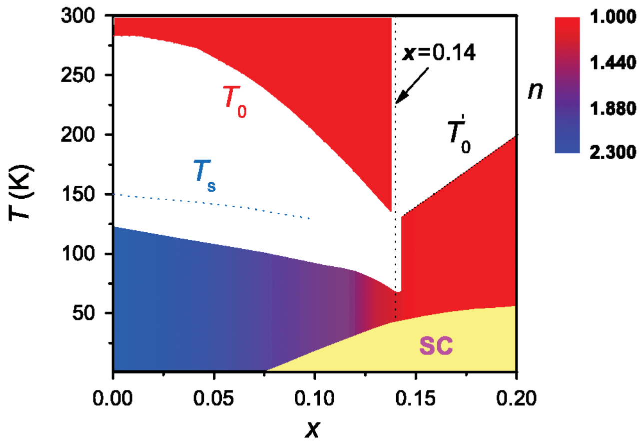

FIG. 4 (color). Electronic phase diagram for a $SmFeAsO_{1-x}F_x$ system. T_s indicates the temperature of the anomaly peak in resistivity. T_0 and T_0' represent the deviating temperature from a T-linear dependence of resistivity in low and high temperatures, respectively. The different color regions represent different n in the formula $\rho = a + bT^n$ shown in Fig. 2. The dotted line of $x = 0.14$ clearly shows a boundary for different behavior of T-dependent resistivity below and above $x = 0.14$, suggesting a QCP around $x \sim 0.14$. The superconductivity is abbreviated as SC.

$CeMIn_5$ (M = Co, Rh) [17]. Compared to the behavior of $x = 0$, 0.05, and 0.10 samples, the Hall coefficient of the $x = 0.15$ and 0.20 samples shows much weaker temperature dependence, and no clearly pronounced rise at low temperature is observed. Especially, the $x = 0.2$ sample shows very weak temperature dependence at low temperature. The Hall angle is plotted as $\cot\theta_H = \rho/R_H$ vs $T^{1.5}$ in Figs. 3(c) and 3(d). It is remarkable that the data make a straight line in the entire temperature range for the $x =$ 0.13, 0.15, and 0.20 samples, while the straight line shows up just above T_s for the $x = 0$, 0.05, and 0.10 samples. These results present that there exists a scaling law between the Hall angle and temperature: $\cot\theta_H \propto T^{1.5}$. The deviation of the Hall angle from $T^{1.5}$ dependence arises from the occurrence of SDW because its characteristic temperature is exactly the same as the temperature of the anomaly peak in resistivity. Such behavior is very similar to that observed in high-T_c cuprates where the Hall angle is proportional to T^2 [18], and deviation of the Hall angle from T^2 dependence occurs at the onset temperature of pseudogap [19]. The results of resistivity and the Hall coefficient show that a linear temperature dependence of the resistivity and a drop in carrier density as evidence by a pronounced rise in Hall coefficient are associated to the occurrence of spin density wave. Since the T-linear dependence of the resistivity and a drop in carrier density is the characteristic of a quantum phase transition in metals [13], it suggests a QCP around $x = 0.14$ due to competing of the SDW state and superconductivity.

Our findings are summarized in the electronic phase diagram shown in Fig. 4, where the characteristic temperatures of T_s, T_0, and T_0' are also shown. With increasing F doping, the T_s in a SmFeAs(O,F) system is driven down in temperature, and the superconducting state emerges at $x \sim 0.07$, reaching a maximum T_c of ~54 K at $x = 0.20$. Compared to the phase diagram of high-T_c cuprates, no decrease of T_c with increasing doping has been observed. It is because the chemical phase boundary is reached at $x \sim$ 0.20. Moreover, there is a large intermediate regime where superconductivity and SDW coexist for SmFeAs(O,F). Based on the evolutions of the resistivity with T and x in Fig. 2, the different power law dependent behaviors of the resistivity are summarized in Fig. 4. One could find drastically different temperature dependencies of resistivity at two sides of $x = 0.14$. On the left side, the n, as that used in the $a + bT^n$ to fit the low-temperature resistivity, continuously decreases from 2.3 to 1.2 when x is raised from 0 to 0.14, respectively, while on the right side, the n is fixed at 1 for $x > 0.15$. More importantly, together with the results of the Hall coefficient, it suggests the existence of a QCP, which may be crucial for the mechanism of superconductivity in these iron-based high-T_c superconductors, as being suggested before for the superconductivity in the copper-based ones.

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