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Magnetic-field-induced log-T insulating behavior in the resistivity of fluorine-doped $SmFeAsO_{1-x}F_x$

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We report the resistivity of a series of nominally fluorine-doped $SmFeAsO_{1-x}F_x$ polycrystalline superconductors in magnetic fields up to 60 T. For samples where $x<0.15$, the low-temperature resistive state is characterized by pronounced magnetoresistance including an upturn at low temperatures. The "insulating behavior" is characterized by a log-T divergence observed over a decade in temperature, a behavior strikingly similar to the underdoped cuprates. The normal state for samples with doping $x>0.15$ behaves very differently: metallic behavior with little magnetoresistance, where intense magnetic fields broaden the superconducting transition rather than significantly suppressing T_c. The doping at which the insulating behavior disappears coincides with the reported collapse of the structural phase transition in the phase diagram for $SmFeAsO_{1-x}F_x$ series.

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The normal state underlying the superconducting regime in the $REFeAsO_{1-x}F_x$, where RE is a rare-earth element, is not well understood. A nearly uniform characteristic of the Fe-based superconductor quaternary and ternary parent compound is an structural phase transition (SPT) from tetragonal to orthorhombic upon decreasing temperature, while still lower in temperature exists a magnetically ordered phase.[1] Increasing the fluorine concentration in these materials kills both the SPT and static magnetism up to a critical concentration at which superconductivity emerges.[2] The RE earth plays an interesting role, for example, in the La series there is complete suppression of magnetic order before the onset of superconductivity, while for Sm there is a region in which both static magnetism and superconductivity coexist. For Sm the maximum T_c is roughly twice that than in the La family, suggesting the importance of strongly disordered static magnetism as a precursor to superconductivity.[3] Neutron data, on the other hand, show a complete suppression of magnetic order before the onset of superconductivity in $CeFeAsO_{1-x}F_x$.[4] Electrical transport measurements in high magnetic fields have proven valuable for studying this regime of phase space by suppressing superconductivity and revealing the behavior of the system well below the superconducting transition temperature T_c.

For the cuprates, transport measurements established key features of the phase diagram: the insulating Mott state of the undoped parent compound, the existence, amplitude, and extent of the superconducting "dome" as a function of doping, the yet-unexplained robust linear temperature dependence of the normal-state resistivity, and more recently, the shape of the Fermi surface in the underdoped[5] and overdoped regimes.[6]

The successes of these measurements in the cuprates, especially the direct observation of an insulator-to-metal crossover (IMC) in the in-plane *and* out-of-plane resistivities (ρ_{ab} and ρ_c, respectively) for both $La_{2-x}Sr_xCuO_4$ (LSCO) (Ref. 7) and $Bi_2Sr_{2?x}La_xCuO_{6+\delta}$ (BSLCO),[8] in the cuprates serve as motivation for transport measurements in the oxyarsenides. In these studies it was revealed that the resistive upturn in the insulating regime of the cuprate phase diagram is well characterized by a logarithmic temperature (log-T) dependence. The same phenomena have also been reported for the electron-doped cuprate $Pr_{2-x}Ce_xCuO_{6+\delta}$.[9]

These observed phenomena have left open such questions as what is the role of disorder and magnetism in shaping the superconductivity. Recent experiments suggest the log-T behavior may be disorder driven.[10,11] These experiments used electron irradiation to induce controlled amounts of disorder into $YBa_2Cu_3O_{7+\delta}$ samples, showing that the log-T behavior scales with the square of the density of impurities. In the underdoped regime, the phenomenology of the log-T temperature dependence is consistent with Kondo scattering,[12] although it has been pointed out that high magnetic fields would likely suppress conventional (spin-flip) Kondo scattering[9] and the origin of the log-T behavior remains unknown. The magnetic-field-induced normal-state IMC as well as the log-T behavior are three-dimensional low-temperature transport properties of the normal-state regime in cuprates, as both phenomena are observed when external magnetic fields are oriented either parallel or perpendicular to the c axis of the lattice.[7]

With the goal of understanding the low-temperature normal state of the pnictide superconductors, we report measurements on a series of four $SmFeAsO_{1-x}F_x$ samples with nominal fluorine doping (F doping) ranging from 0.05 to 0.20. The series of samples spans a large portion of the underdoped superconducting regime, possibly up to optimum doping.[13] Polycrystalline samples with nominal composition $SmFeAsO_{1-x}F_x$ ($x=0-0.2$) were synthesized by conventional solid-state reaction using SmAs powder, SmF_3 (99.99%, powder), Fe (99.5%, powder), and Fe_2O_3 (99.8%, powder) as starting materials. SmAs was obtained by reacting Sm (99.9%, chips) and As (99.9999%, pieces) in an evacuated quartz tube at 600 °C for 3 h and then 900 °C for 5 h. The raw powder materials were thoroughly grounded and pressed into pellets. The pellets were wrapped into Ta

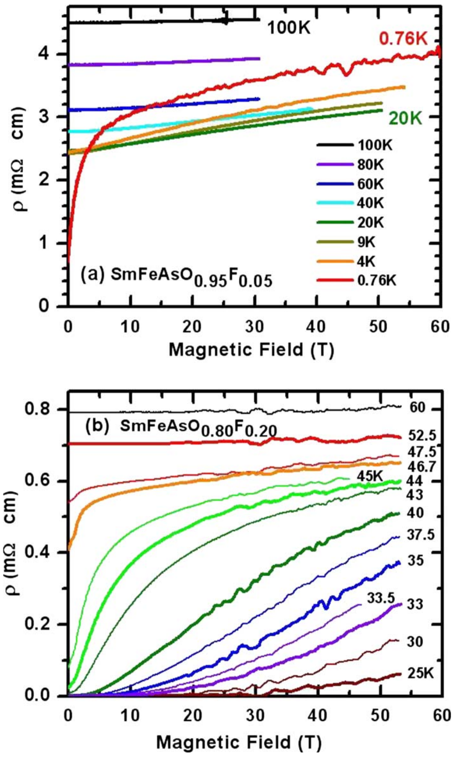

FIG. 1. (Color online) Pulsed field measurements of resistivity vs magnetic field for fluorine doping (a) $x=0.05$ and (b) $x=0.20$ in $SmFAsO_{1-x}F_x$. The insulating behavior can clearly be seen in the $SmFeAsO_{0.95}F_{0.05}$ sample at temperatures below 20 K, while $x=0.20$ remains metallic at all temperatures.

foil and sealed in an evacuated quartz tube. They are heated to 1160 °C at the rate of 3 °C/min and then annealed for 40 h. After 40 h, the samples were furnace cooled to room temperature. The sample preparation process except for annealing was carried out in glove box in which high pure argon atmosphere is filled.[14,15] The resulting polycrystals were cut into rectangular prisms with a typical size of $1.5\times 1\times 0.1\ \mathrm{mm}^3$. The resistivity ρ transverse to the applied magnetic fields was measured using the standard four-terminal digital ac lock-in technique in continuous fields up to 35 T and in pulsed fields up to 60 T at the National High Magnetic Field Laboratory. The T_c values as measured at the midpoint of the SC transition for $x=0.05$, $x=0.12$, $x=0.15$, and $x=0.20$ are ~2, 18, 40, and 46 K with transition widths of 8, 14, 7, and 8 K, respectively.

Figure 1(a) shows the resistivity versus magnetic field B for our least-doped sample, $SmFeAsO_{0.95}F_{0.05}$, at selected temperatures. Note that at 10 T the magnetic field suppresses the superconductivity at $T=0.76$ K, revealing the normal-state resistivity at higher magnetic fields. Also note that for low temperatures ($T<20$ K) the normal-state resistivity is increasing as temperature decreases (insulating behavior). Figure 1(b) contains the resistivity for our most-doped sample, $SmFeAsO_{0.80}F_{0.20}$, in which there is no insulating behavior and the effectiveness of the high magnetic field in suppressing superconductivity is greatly reduced.[16]

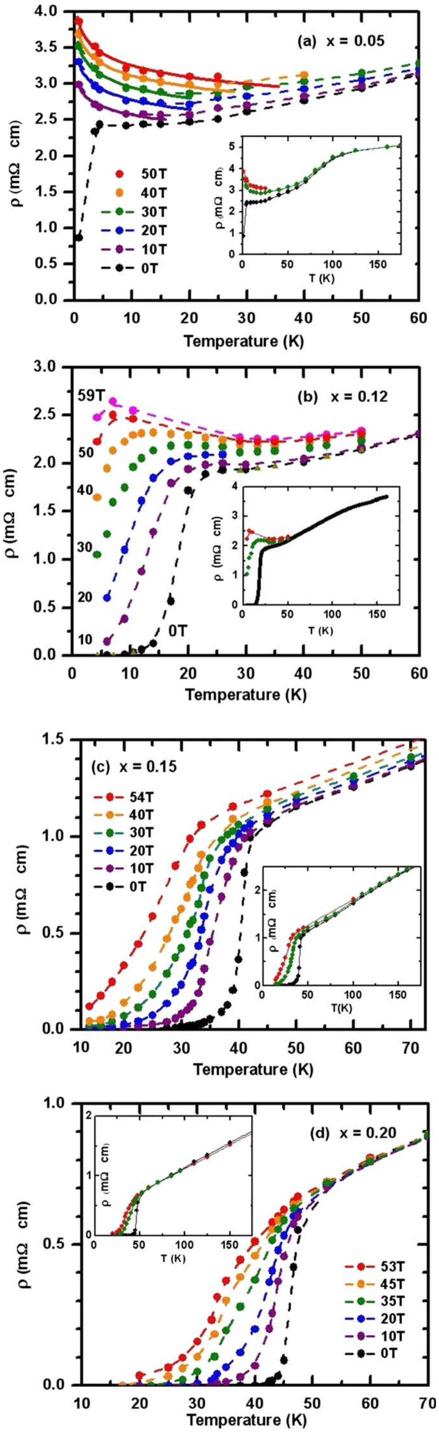

FIG. 2. (Color online) Resistivity vs temperature of the $SmFeAsO_{1-x}F_x$ samples with four different fluorine dopings studied in magnetic fields up to 60 T. Note the insulating behavior at low temperatures for samples with $x<0.15$. Note also that the samples with low doping show higher magnetoresistance above T_c, despite having higher resistivities (discussed in text). Solid lines in panel (a) are logarithmic fits to the low-temperature insulating behavior.

Figure 2 shows the evolution of the resistivity versus temperature with doping $x=0.05$, 0.12, 0.15, and 0.20. Plotted are measurements from fixed-temperature magnetic field sweeps of pulsed magnets (discrete points). Dotted lines are guides to the eyes connecting pulsed field data points. The most striking result is the insulating behavior of the $x=0.05$

and 0.12 samples compared to the metallic $x=0.20$ sample.

At $x=0.20$, the superconducting state is robust under a field of 55 T, which suppresses the onset of the superconducting transition by roughly 7% [Fig. 2(d)]. The midpoint of the superconducting transition is suppressed by 20%, in effect broadening the resistive transition, as is observed in $YBa_2Cu_3O_{7+\delta}$ (YBCO). This is not surprising as it has been reported that the Ginzberg parameter for $SmFeAsO_{0.80}F_{0.20}$ is similar to that for YBCO.[17]

Careful examination of Fig. 2(d) reveals that the magnetoresistance above T_c is negligible at highest doping: using the characteristic value of $\rho_{xx} \sim 1$ mΩ cm [Fig. 2(d)] and a Hall coefficient, $R_H \sim -6 \times 10^{-9}$ m^3/C for $SmFeAsO_{0.80}F_{0.20}$,[13] we estimate $\omega_c \tau \sim R_H / \rho_{xx} \sim B(T)/1700$ T, which equals 0.035 at our highest fields of 60 T. Thus orbital magnetoresistance, which is of the order of $(\omega_c \tau)$ within a Fermi-liquid picture for the $SmFeAsO_{0.80}F_{0.20}$ sample, is expected to be small in the resistive normal state.

The anomalous magnetoresistance is observed in the samples with $x < 0.15$ in which the magnetoresistance represents a larger proportion of the total resistivity than is observed at higher doping even though these samples have much higher resistivity than the most-doped sample. This is opposite to the trend expected from orbital effects. Note that the magnetoresistance extends both below and well above the zero-field T_c in the very underdoped regime, persisting to temperatures as high as ~90 K for $x=0.05$, a temperature which is in the vicinity of the reported SPT.[18] Also, the apparent temperature at which the magnetoresistance becomes negligible in the underdoped regime corresponds to the same temperature where the Hall resistivity becomes nonlinear in high magnetic fields.[19] The implication is that the large magnetoresistance in the underdoped regime may not be linked to the superconducting state, rather that it is a property of the normal state.

The contrast between the very underdoped samples and the most-doped sample is most dramatic at low temperatures and in high fields. At $x=0.15$ [Fig. 2(c)], the response to magnetic fields appears to be a transition between the two limiting behaviors: the magnetic field greatly broadens the resistive transition as it does at optimum doping, but it also reveals a large magnetoresistance extending well above the zero-field T_c. However, as is seen at optimum doping, the normal state at $x=0.15$, revealed by the suppression of T_c, remains metallic, unlike the insulating behavior seen in the very underdoped regime. The normal state appears to have monotonic resistivity in both the $x=0.15$ and $x=0.20$ cases. This is in direct contrast to the $x < 0.15$ case, for which magnetic-field-induced upturn in the resistivity near 50 K, as well as an apparently consistent temperature value at which a resistive minimum occurs, for a given value of field. This suggests the two higher doping cases remain metallic below T_c in large fields. We thus conclude that the log-T behavior disappears at a fluorine doping in the underdoped regime near $x=0.15$. Superconductivity, nevertheless, is quite robust at this doping ($T_c \sim 40$ K in zero magnetic field).

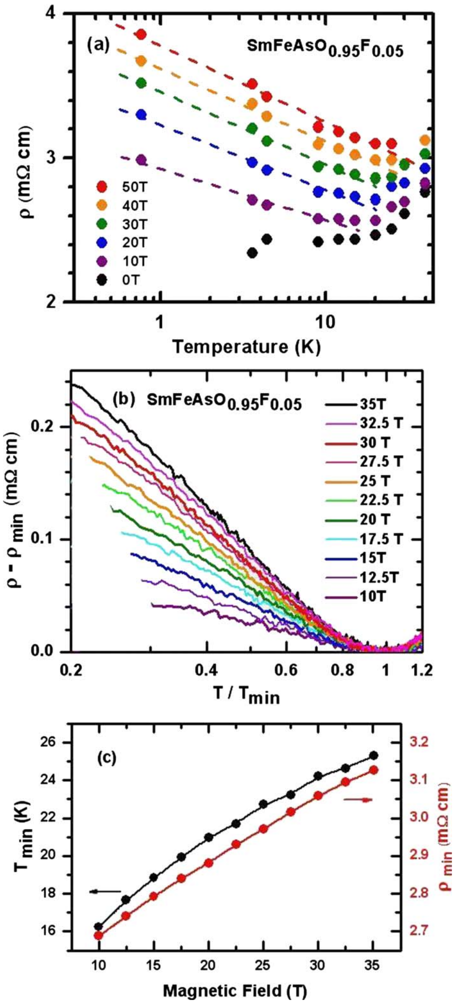

FIG. 3. (Color online) Resistivity vs logarithm of temperature for two different samples of $SmFeAsO_{0.95}F_{0.05}$ from (a) magnetic field pulses up to 60 T at fixed temperatures and (b) temperature sweeps in fixed magnetic fields up to 35 T. Data show a weak log-T divergence of resistivity over roughly a decade in temperature. Data in (b) are scaled by ρ_{min} and T_{min}, the resistivity and temperature at which the resistivity is a minimum. The magnetic field dependences of the normalization factors T_{min} (right axis) and ρ_{min} (left axis) are given in panel c.

The $x=0.05$ and $x=0.12$ samples exhibit insulating behavior in the low-temperature normal state that can be characterized by a resistance increasing as the logarithm of temperature. Figures 3(a) and 3(b) show the log-T resistivity of two different $SmFeAsO_{1-x}F_x$ samples with nominal $x=0.05$ doping. Magnetic fields not only increase the minimum value of the resistivity, ρ_{min}, but also shift the temperature at which the minimum resistivity occurs, T_{min}, upward. Figure 3(b) normalizes the log-T behavior seen in the $x=0.05$ sample by subtracting ρ_{min} and dividing temperature by T_{min}. Figure 3(c) shows the magnetic field dependence of the two parameters.

As stated above, insulating behavior has been observed in single crystals of cuprate superconductors in the underdoped regime.[7,9] Insulating behavior has also been observed in granular superconductors and is discussed in connection with the nanoscale phase separation observed in the cuprates.[20] This is one similarity we find between $SmFeAsO_{1-x}F_x$ and the cuprates. We also find that the insulating behavior of the resistivity follows a logarithmic dependence at temperatures below $T_{\min}$ for $SmFeAsO_{0.95}F_{0.05}$, consistent with the behavior seen in the low-temperature normal-state properties of underdoped cuprates. For each family of materials, high magnetic fields are required to reveal the log-T behavior. The cuprates are unique from the oxyarsenides in two notable ways, however: (a) a log-T divergence of resistivity is seen only once superconductivity is suppressed, revealing the underlying log-T normal state at low temperatures and (b) the log-T behavior for all magnetic fields is the same. For the Sm oxyarsenide, the onset of the insulating behavior in high magnetic fields occurs as much as 1 decade in temperature above the value of T_c in zero magnetic field. This behavior seems to originate from the large magnetoresistance in the extremely underdoped oxyarsenides that extends to temperatures well above the log-T regime, giving rise to log-T divergences that are magnetic field dependent. This is consistent with the metallic zero-field behavior of oxyarsenide parent compounds at low temperatures.[13,21]

In conclusion, the resistivity of $SmFeAsO_{1-x}F_x$ exhibits a doping dependence with two key three-dimensional properties. There is an observed log-T divergence of the resistivity for the less-doped samples in high magnetic fields. Increasing doping leads to the disappearance of the magnetic-field-induced insulating behavior. Future work using higher fields to further suppress superconductivity as well as more samples with doping values in the range of interest should illuminate these features further.

Recently, work was published reporting insulating behavior in the underdoped regime in the 122 family of pnictide superconductors.[22] The same insulating regime has now been reported in polycrystalline $LaFeAsO_{1-x}F_x$,[23] LaFeAsO, and $LaFe_{0.96}Ni_{0.04}AsO$, as well as a single-crystal sample from another pnictide material, $Sr_3Sc_2O_5Fe_2As_2$.[24] In each of these three systems, the insulating behavior is also shown to be logarithmic in temperature.

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