Evidence of quantum criticality in the phase diagram of $K_xSr_{1-x}Fe_2As_2$ from measurements of transport and thermoelectricity

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The electrical transport and thermoelectric properties of $K_xSr_{1-x}Fe_2As_2$ are investigated for $0 \leq x \leq 1$. The resistivity $\rho(T)$ shows a crossover from Fermi-liquid-like temperature dependence at small x to linear $\varrho \sim T$ dependence at $x_c \simeq 0.4$. With further increasing of x, $\varrho(T)$ becomes nonlinear again. The thermoelectric power $S(T)$ exhibits a similar crossover with increasing x with a logarithmic T dependence, $S/T \sim \ln(T)$, near the critical doping x_c. These results provide evidence for a quantum critical behavior due to the coupling of low-energy conduction electrons to two-dimensional spin fluctuations.

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The discovery of superconductivity in quaternary rare-earth transition-metal oxypnictides with transition temperatures up to 55 K and the existence of a spin-density wave (SDW) phase in the nonsuperconducting parent compounds[1–6] has stimulated extensive discussions about the coexistence of both states of matter, the role of magnetic order, its competition and mutual interaction with superconductivity, and the possible existence of unusual normal-state properties and a quantum critical point in the phase diagram of FeAs-based compounds.[7–15] The oxygen-free "122" compounds $A_xAe_{1-x}Fe_2As_2$, with A=alkali metal and Ae =alkaline earth element, have been shown recently to form a solid solution over the whole compositional range $0 \leq x \leq 1$.[16–18] The $AeFe_2As_2$ compounds are not superconducting but they exhibit a SDW transition at 172, 140, and 205 K for Ae=Ca, Sr, and Ba, respectively.[19–21] With alkali-metal doping the SDW transition temperature decreases and superconductivity is observed above a critical concentration of about 0.17 in $K_xSr_{1-x}Fe_2As_2$ (Ref. 16) and $K_xBa_{1-x}Fe_2As_2$.[17,18] With further increasing of x (hole doping) the superconducting T_c passes through a maximum at 37 K and decreases to 3.7 K for $x=1$ (KFe_2As_2). The $K_xAe_{1-x}Fe_2As_2$ superconductor is therefore the ideal system in probing the complete superconducting phase diagram and the mutual correlation with the SDW order and magnetic fluctuations.

It is not clear, however, if the magnetic and superconducting orders coexist and how the SDW phase boundary extends or extrapolates into the superconducting phase with increasing doping. Careful investigations of the phase diagrams of $K_xSr_{1-x}Fe_2As_2$ (Refs. 22 and 23) and $K_xBa_{1-x}Fe_2As_2$ (Refs. 17 and 18) suggest a narrow region between $x \simeq 0.17$ and $x \simeq 0.25$ where a SDW transition is followed by a superconducting transition upon decreasing temperature. Although the magnetic order is apparently suppressed with the onset of superconductivity, the extrapolation of the SDW phase boundary to $T=0$ would locate the quantum phase transition from the SDW phase to the paramagnetic phase near a critical doping of $x_c \simeq 0.4$. In order to follow the magnetic phase to $T=0$ and to investigate the possible quantum critical point, the superconductivity has to be suppressed, for example, by large external magnetic fields. However, this is difficult to achieve since the critical field of the superconducting phase is very large.[16,23] Alternatively, the physical properties can be investigated in the temperature range above the suspected quantum critical point for the typical crossover signature that is expected in a quantum critical regime.

We have therefore studied the normal-state electrical and thermoelectric transport properties in the whole phase diagram of $K_xSr_{1-x}Fe_2As_2$ for $0 \leq x \leq 1$. We observe a crossover from Fermi-liquid behavior ($x=0$) to non-Fermi-liquid temperature dependence of resistivity and thermoelectric power close to the critical doping x_c. The results are consistent with the expected temperature dependencies near a magnetic quantum critical point.

Polycrystalline samples of $K_xSr_{1-x}Fe_2As_2$ have been synthesized from the high-purity ternary compounds KFe_2As_2 and $SrFe_2As_2$, as described earlier.[16] The careful mixing of the two end members at nominal ratios allows for a sensitive control of the composition (doping). The x-ray analysis confirms the phase purity of the solid solutions. Characteristic spectra and their corresponding Rietveld refinements are shown in Fig. 1. All observed peaks are assigned to the $I4/mmm$ structure. The continuous and nearly linear progression of the lattice parameters with increasing x (with c increasing and a decreasing; inset of Fig. 1) proves that a uniform solid solution forms for all values of x. The resistivity was measured in a four probe configuration using the low-frequency (19 Hz) ac resistance bridge LR700 (Linear Research). The thermoelectric power was measured employing a highly sensitive ac (0.1 Hz) method with a typical resolution of 0.05 μV/K.[24] The probe and the wires have been calibrated using a Hg high-T_c superconductor below 130 K, and high-purity lead between 130 and 300 K.

The resistivities of the complete series of $K_xSr_{1-x}Fe_2As_2$ are shown in Fig. 2. Different curves are offset by a constant unit of 0.25 for better clarity. Both ternary compounds, KFe_2As_2 and $SrFe_2As_2$, show a strongly nonlinear temperature dependence of the resistivity, $\rho(T)$. However, approaching the critical doping of $x_c \simeq 0.4$, the low-T part of the resistivity curve becomes increasingly linear, indicating a

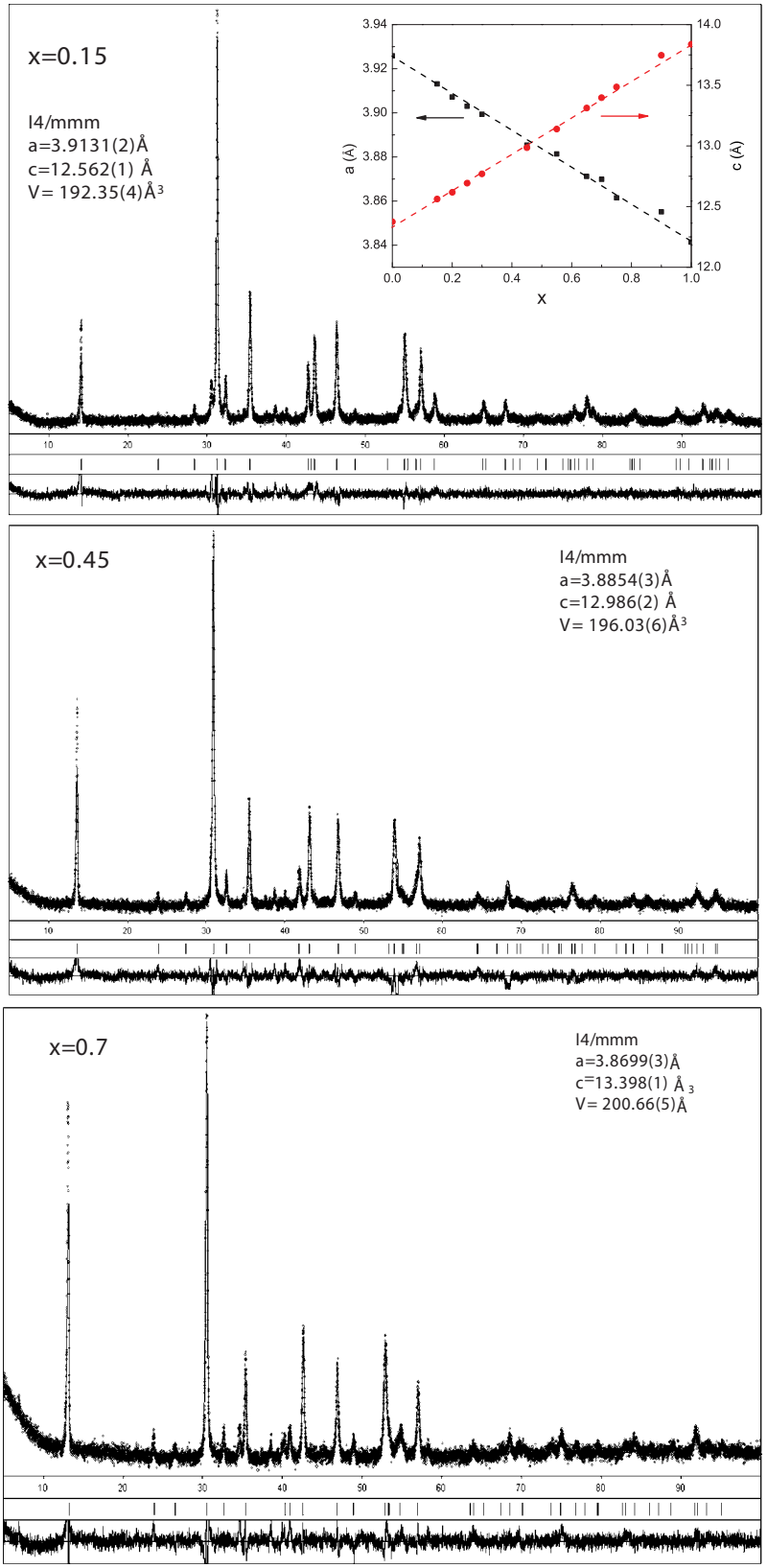

FIG. 1. (Color online) Selected x-ray spectra of the system $K_xSr_{1-x}Fe_2As_2$ for $x=0.15, 0.45, 0.7$. The inset shows the change in lattice parameters a and c with x.

crossover into a non-Fermi-liquid regime. Fitting the low-temperature part (above T_c) of $\rho(T)$ to a simple power law, $\rho(T)=\rho_0+AT^n$, an effective exponent n can be extracted from the data of Fig. 2. Although the fitted exponent may have an error of the order of 10%, it provides a qualitative picture of the crossover of the transport properties. The power-law dependence of $\rho(T)$ with the exponent n is shown by dotted lines in Fig. 2. It also defines the temperature range for which the power law is valid. The exponent n decreases with x from about two to one near the critical x_c and then in-

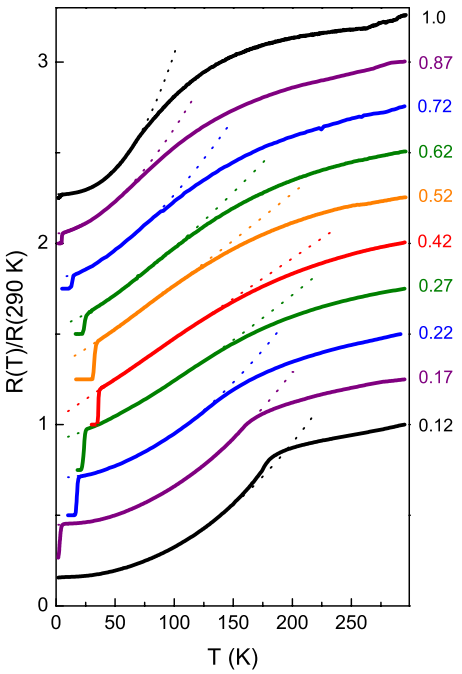

FIG. 2. (Color online) Resistivity of $K_xSr_{1-x}Fe_2As_2$ vs temperature for $0 \leq x \leq 1$. For clarity different curves are vertically offset by 0.25 units. The labels mark the values of x. The dotted lines show the fit to a power law, $\rho(T)=\rho_0+AT^n$, with the exponent n shown in Fig. 3.

creases again to values close to two, as shown in Fig. 3. This crossover behavior is consistent with the transport property expected at temperatures above a magnetic quantum critical point.[25] For example, in the high-T_c cuprate superconductors, the linear $\rho(T)$ was observed above the superconducting dome and the non-Fermi-liquid property was associated with the quantum critical point defined by the extrapolation of the pseudogap temperature to $T=0$. In heavy Fermion compounds, e.g., $CeCu_{6-x}Au_x$, the deviations from Fermi-liquid properties have been observed frequently and the linear $\rho(T)$ was explained as a characteristic transport behavior in the quantum critical regime next to a magnetic quantum phase transition.[26] The main difference between the heavy Fermion compounds and the $K_xSr_{1-x}Fe_2As_2$ system is the energy scale of the magnetic fluctuations. Whereas in $CeCu_{6-x}Au_x$ the typical magnetic transition temperatures are of the order of 1 K, the SDW transition in $SrFe_2As_2$ takes place at 200 K, two orders of magnitude higher. Therefore, the quantum critical region in which clear deviations from the Fermi-liquid model can be observed extends to higher temperatures.

The resistivity data presented in Fig. 2 and discussed above have been acquired for dense polycrystalline samples. It is not known whether the grain-boundary contributions could affect the T dependence of ρ and the exponent n in Fig. 3. The thermoelectric power S, however, is a zero current property and does not depend on grain boundaries as much as the resistivity. The temperature-dependent data of $S(T)$ throughout the phase diagram of $K_xSr_{1-x}Fe_2As_2$ are summarized in Fig. 4. $S(T)/T$ is shown as a function of $\ln(T)$. All data can be separated into two groups with each group showing a consistent and characteristic T dependence: (i) for x

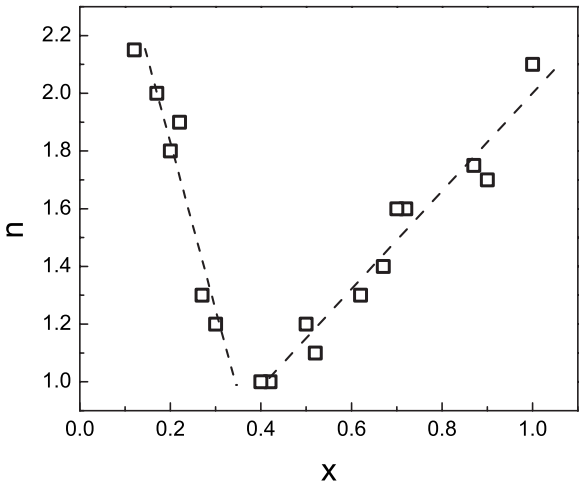

FIG. 3. The resistivity exponent n as a function of x.

<0.3 the thermoelectric power S/T exhibits an almost identical T dependence above the SDW transition. This is obvious from the data shown in Fig. 4(a). The dashed circle indicates the relevant T range above T_{SDW}. (ii) For $x>0.5$ the temperature dependence of S/T is similar for all x above the superconducting transition, as shown in Fig. 4(b). The dashed circle emphasizes on this temperature range and the closeness of all data in the high doping range. $S(T)/T$ as a function of $\ln(T)$ is nonlinear in both cases: for $x<0.3$ and $x>0.5$. However, between the dopings of 0.3 and 0.5, the thermoelectric power shows a distinct crossover between the characteristic T dependence of groups 1 [Fig. 4(a)] and 2 [Fig. 4(b)]. For $x \simeq 0.4$ S/T varies linearly with $\ln(T)$ over a large temperature range, from the superconducting $T_c \simeq 36$ K to about 180 K. The data in the crossover range are shown in more detail in Fig. 4(c). The dashed line attached to the $x=0.42$ data shows that $S/T \sim \ln(T)$ at this critical doping. The scaling property of the thermoelectric power, as revealed in Fig. 4(c), is a strong signature of quantum criticality and it is consistent with the non-Fermi-liquid-like temperature dependence of the resistivity. It should be noted that a similar logarithmic dependence of S/T has been reported very recently in the hole-doped high-T_c cuprate superconductor, $La_{1.4-x}Nd_{0.4}Sr_xCuO_4$.[27]

Thermodynamic and transport properties near magnetic quantum phase transitions have been investigated mostly in heavy Fermion systems like the above mentioned $CeCu_{6-x}Au_x$. Whereas the undoped parent compound, $CeCu_6$, is nonmagnetic, the substitution of Au induces incommensurate antiferromagnetic order for $x>0.1$. Right at the critical concentration of $x=0.1$, the $CeCu_{6-x}Au_x$ system exhibits quantum critical scaling at finite temperatures as expressed in a linear T dependence of resistivity and a logarithmic temperature dependence of the heat capacity, C/T.[26] Similar scaling properties have been found in other heavy Fermion compounds with a magnetic quantum critical point.[25] Since the typical scaling range in heavy Fermion compounds is of the order of 1 K, the heat capacity is the entropy-related quantity that is experimentally accessible with the precision required to prove the quantum critical behavior (note that at higher temperatures the lattice contribu-

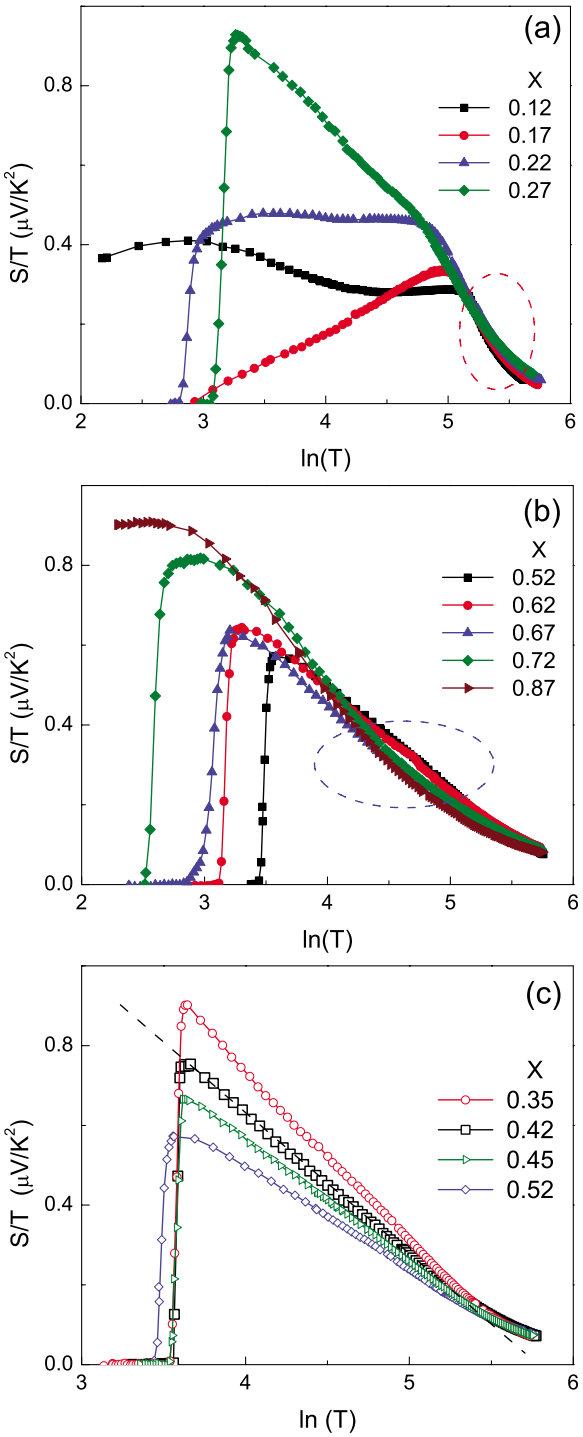

FIG. 4. (Color online) Scaling plot S/T vs $\ln(T)$ of the thermoelectric power in the phase diagram of $K_xSr_{1-x}Fe_2As_2$. (a) S/T for $x<0.3$, (b) S/T for $x>0.5$, and (c) S/T in the crossover region between $x=0.3$ and $x=0.5$. The dashed line in (c) shows the logarithmic scaling at the critical doping x_c.

tion to the heat capacity masks the electronic part, and makes it far more difficult, if not impossible, to extract accurately the electronic heat capacity that is expected to follow the scaling laws). In our high-T_c $K_xSr_{1-x}Fe_2As_2$ system, however, the low-temperature region is not accessible because the superconducting state is stabilized and dominates the physical properties below T_c. Therefore, other physical quantities, like resistivity or thermoelectric power, had to be investigated with respect to quantum critical scaling properties.

The thermoelectric property near a quantum phase transition is less well investigated as compared to resistivity or heat capacity. For $CeCu_{6-x}Au_x$ ($x=0.1$) at low temperatures $S(T)$ was reported to vary nonlinearly with T in contrast to the linear dependence expected for a Fermi liquid.[28] Theoretically, logarithmic scaling in the quantum critical regime of, for example, C/T is expected if the dimension of the critical fluctuations (d) is equal to the dynamical critical exponent (z).[25] The critical scaling properties of $CeCu_{5.9}Au_{0.1}$ have been explained based on a scaling theory for $d=2$ and $z=2$.[29] Alternatively, based on a spin-Fermion model proposed by Abanov and Chubukov,[30] it has been shown that low-energy conduction electrons interacting with quasi-two-dimensional (2D) spin fluctuations give rise to a linear with temperature resistivity, a logarithmic T dependence of the heat capacity C/T, and a similar logarithmic scaling of the thermoelectric power, $S/T \sim \ln(T)$.[31] The $\ln(T)$ dependence of S/T for $K_{0.42}Sr_{0.58}Fe_2As_2$ shown in Fig. 4(b) as well as the T-linear resistivity (Figs. 2 and 3) are consistent with the model of low-energy conduction electrons interacting with quasi-2D spin fluctuations. The lower dimensionality of the magnetic fluctuations finds its natural origin in the layered structure of the FeAs compounds with the magnetic Fe ions confined to the Fe_2As_2 layers.[32]

One question remaining is the origin of the magnetism and free carriers in FeAs compounds. While in typical heavy Fermion systems (e.g., $CeCu_6$) magnetic moments are introduced through localized f electrons of the rare-earth ions and conduction electrons are provided by transition metals, this is not necessarily the case in FeAs compounds since the electrons at the Fermi surface are mainly from hybridized orbitals of Fe and As with mainly d-electron or p-electron character. In the present $K_xSr_{1-x}Fe_2As_2$ system there is no alternative source for magnetic moments than the Fe ions. A recent study of the "undoped" parent compounds proposed a separation of the electronic excitations into an "incoherent" part, further away from the Fermi surface and giving rise to local magnetic moments interacting with each other through frustrated superexchange coupling, and a "coherent" part in the vicinity of the Fermi surface.[9] Recent electron-spin-resonance experiments on La(O,F)FeAs seem to support the existence of local magnetic moments, their coupling to itinerant electrons, and the presence of strong magnetic frustration.[33] The coherent carriers couple to the local moments and compete with the SDW order. Increasing the carrier concentration by doping can tune the system to a magnetic quantum phase transition. The theoretical treatment within a low-energy Ginzburg-Landau theory indeed describes an antiferromagnetic quantum phase transition with the dynamical critical exponent $z=2$ and the effective dimension $d+z=4$.[9] However, the dimension and the nature of the spin fluctuations are important since logarithmic scaling is expected only for $d=2$ and $z=2$ or $d=3$ and $z=3$.[25] For three-dimensional magnetic fluctuations ($d=3$), the dynamical critical exponent has to be $z=3$ to explain the logarithmic scaling, as, for example, in the case of a metallic ferromagnet. The existence of ferromagnetic spin fluctuations in

$LaO_{1-x}F_xFeAs$ has recently been proposed based on the large Wilson ratio.[15] Which scenario applies to the system $K_xSr_{1-x}Fe_2As_2$ remains an open question and has to be resolved in future investigations.

Our results obtained for the $K_xSr_{1-x}Fe_2As_2$ system, linear T dependence of resistivity and logarithmic T dependence of S/T, are consistent with the expected scaling behavior of various thermodynamic and transport properties in the critical regime near a magnetic quantum phase transition.[25] The crossover properties of $\rho(T)$ and S/T throughout the phase diagram of $K_xSr_{1-x}Fe_2As_2$ and the scaling behavior near the critical doping x_c provide strong experimental support for the existence of a magnetic quantum critical regime above the superconducting T_c. This regime needs to be investigated further by extending the accessible temperature range to lower T. To this end the superconducting phase has to be suppressed. Since the critical fields appear to be too high to suppress the superconducting state, other control parameters such as pressure or chemical substitution may be considered as an alternative.

Note added in proof. The current data have been acquired for polycrystalline samples. Single crystals have recently become available[34] and a systematic investigation of the quantum critical effects in single crystalline samples is suggested. However, special care has to be taken to ensure a uniform composition and dopant distribution that is particularly difficult to achieve in the growth of single crystals.

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