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Pressure-induced shift of T_c in $K_xSr_{1-x}Fe_2As_2$ ($x=0.2, 0.4, 0.7$): Analogy to the high-T_c cuprate superconductors

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The systematic pressure shifts of T_c were investigated in the whole phase diagram of the FeAs-based superconducting compound $K_xSr_{1-x}Fe_2As_2$. Different regions, arising from corresponding responses of T_c to pressure ($dT_c/dp>0$, $\simeq 0$, or <0), can be clearly distinguished. This reveals an interesting similarity of the FeAs superconductors and the high-T_c cuprates. This behavior is a manifestation of the layered structure of the FeAs compounds and the pressure-induced charge transfer between the (Fe_2As_2) and (K/Sr) layers. The coexistence of superconductivity and spin-density wave behavior were also observed, and the pressure effects on the latter is explored.

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The discovery of superconductivity in quaternary rare-earth transition-metal oxypnictides (*ROT*Pn, where *R*=rare earth, *T*=transition metal, and Pn=pnictogen) with transition temperatures up to 26 K in F-substituted LaOFeAs and even higher T_c's in compounds with smaller *R* ions have initiated immense excitement and stimulated renewed activities in high-T_c superconductivity research.[1–6] Only the copper oxide superconductors are known so far to reach or exceed the high-T_c values of the FeAs-based compounds, and it has been speculated that both classes of superconducting materials are structurally and electronically related. The structural similarities are obvious, both compounds form layered structures with active superconducting layers (Fe_2As_2 or CuO_2) separated by charge reservoir blocks providing control of the nature and density of charge carriers. The Fe_2As_2 layers are extremely rigid allowing for significant variations in the charge reservoir blocks which can even be a single plane of alkali or alkaline-earth metals.[7–9] It is very important to show that other high-T_c systems than the cuprate superconductors exist and to understand the common features of these compounds. This will facilitate the fundamental understanding of high-T_c superconductivity and possibly lead to the discovery of different materials with even higher critical temperatures.

To reveal the common properties of the FeAs superconductors and the cuprates and to verify their close relations we focus on the T_c dependence on the carrier density and the effect of pressure on T_c for different doping levels. For the high-T_c cuprates it is well established that T_c exhibits a universal doping dependence with a maximum at an optimal carrier density. Application of pressure transfers charges from the reservoir to the CuO_2 planes and, accordingly, results in a positive, close to zero, and negative pressure coefficient of T_c for underdoped, optimally doped, and overdoped regions in the phase diagram, respectively.[10] In the FeAs-based superconductors the control of carriers is realized through the replacement of O with F (in *R*OFeAs),[1,4,11–15] the control of oxygen deficiency,[16] or through the substitution of the alkaline earth (Ae) with a monovalent cation, K, Cs, etc. (in $AeFe_2As_2$).[17,18] While the maximum degree of F substitution in *R*OFeAs is limited by the chemical stability and the region of highest carrier concentration is difficult to achieve[1,4,11–13,15] the $K_xSr_{1-x}Fe_2As_2$ system was recently shown to be stable in the whole doping range from $x=0$ ($T_c=0$) through optimal doping, $x_{opt}\simeq 0.45$ ($T_c=37$ K), to $x=1$ ($T_c=3.7$ K).[18] Similar phase diagrams have been reported for Ba replacing Sr.[19,20] This is the perfect system to be compared to the high T_c cuprates and to allow for a thorough investigation of the pressure shift of T_c in different regions of the phase diagram for low, optimal, and high carrier densities.

Due to the complex Fermi surface of the FeAs superconductors the effects of changes in carrier concentration on the electronic and superconducting properties are not as clear as, e.g., in the single-band high-T_c cuprates. Band structure calculations reveal that the Fermi surface includes electron as well as hole pockets.[21] For superconducting $K_xSr_{1-x}Fe_2As_2$ near the optimal chemical substitution (x_{opt}) the Hall coefficient was found to be positive hinting that the majority carriers are holes.[22] However the negative Hall coefficient of the nonsubstituted Sr-122 indicates that electrons dominate the transport properties.[23] This redistribution of states is expected since increasing x reduces the number of electrons in the Fe_2As_2 layer and the majority carriers in the K-doped compounds are holes. These results are consistent with recent angle resolved photoemission spectroscopy measurements[24] and band-structure calculations.[21]

The application of high pressure and its effects on the superconducting T_c were first investigated in the *R*OFeAs system. The increase of T_c with pressure initially observed in $LaO_{0.89}F_{0.11}FeAs$ has raised the hope of achieving higher transition temperatures with chemical or physical pressures in other *R*(O/F)FeAs superconductors.[25] However, it was quickly shown that the pressure coefficient of T_c strongly depends on the doping level, and that the T_c of $SmO_{1-x}F_xFeAs$ ($x=0.3$, $T_c=43$ K) did actually decrease with pressure.[26] Related work on $LaO_{1-x}F_xFeAs$ came to a similar conclusion. After the increase in T_c at low pressure (p), for compositions $x\leq 0.1$, T_c dropped precipitously with

further increase in p.[2] The negative pressure coefficient of R(O/F)FeAs compounds with high T_c's was also confirmed for $LaO_{0.9}F_{0.1}FeAs$, $CeO_{0.88}F_{0.12}FeAs$, and $RO_{0.85}FeAs$ (R =Sm or Nd).[27–29] However, for a deeper understanding of the physics of the FeAs superconductors and the suspected similarities to the high-T_c cuprates, a more systematic investigation of the effect of pressure for different carrier concentrations covering the complete phase diagram is warranted.

We have therefore investigated the pressure dependence of T_c in the phase diagram of $K_xSr_{1-x}Fe_2As_2$ and compared our results with those of the cuprate superconductors. We conclude that, similar to the high-T_c cuprates, the main pressure effect can be explained by a p-induced charge transfer between the reservoir and the active (Fe_2As_2) layer. In a limited range of x the superconducting state below T_c was observed together with the spin-density-wave (SDW) state that is stable below a higher temperature T_S. The p dependence of T_S is also studied.

Polycrystalline samples of $K_xSr_{1-x}Fe_2As_2$ for x=0.2, x =0.4, and x=0.7 were prepared by high-temperature solid-state reactions, as previously described.[18] The three samples are located in the phase diagram as shown in the inset of Fig. 2(a) (vertical arrows). Hydrostatic pressure up to 18 kbar was generated in a piston-cylinder clamp cell using a Pb gauge for *in situ* pressure measurements[30] and liquid Fluorinert 70/77 as pressure transmitting medium. The ac magnetic susceptibility of two samples (x=0.4 and 0.7) was measured through a dual-coil inductance transformer wrapped to the sample's surface. For x=0.2 the resistivity was measured under pressure since this also allows for extracting the p dependence of T_S. The low-frequency (19 Hz) ac bridge LR700 (Linear Research) was employed for inductance and resistivity measurements.

The x=0.2 sample deserves special attention since it shows two phase transitions easily detected in the resistivity data of Fig. 1(a). At T_S=139 K the SDW transition is clearly resolved as a peak of the derivative $d\rho/dT$. At lower temperature, T_c=13 K, a superconducting transition is defined by the drop of $\rho(T)$ or another sharp peak of $d\rho/dT$. We use this peak position [inflection point of $\rho(T)$] to define T_c at ambient and high pressures. At ambient pressure the so determined critical temperature coincides with the onset of the diamagnetic signal, as shown in the inset to Fig. 1(b). The region of coexistence of both transitions, SDW and superconductivity, extends from $x \simeq 0.17$ to $x \simeq 0.25$ in the phase diagram shown in Fig. 2(a). This coexistence was also observed in $K_xBa_{1-x}Fe_2As_2$.[19] The high-pressure resistivity data and their derivative are shown in Fig. 1(b). T_c apparently increases with p, but it saturates or approaches a maximum at the highest pressures obtained in this investigation. This is shown as the normalized shift, $T_c(p)/T_c(0)$, in Fig. 3 (top curve). Note that the resistivity drop broadens at the highest pressure, possibly due to pressure-induced stress in the polycrystalline sample. The choice of the inflection point (which roughly corresponds to a 50% drop in ρ) is therefore a reasonable criterion in assigning T_c. While T_c increases with p, the SDW transition temperature T_S (shown in the inset of Fig. 3) decreases from 139 K (p=0) to 129 K at 15 kbar. The decrease in T_S is consistent with a p-induced increase in the hole density in the Fe_2As_2 layers. A similar suppression of the SDW state was observed in R(O/F)FeAs (Ref. 4) and $K_xBa_{1-x}Fe_2As_2$.[19]

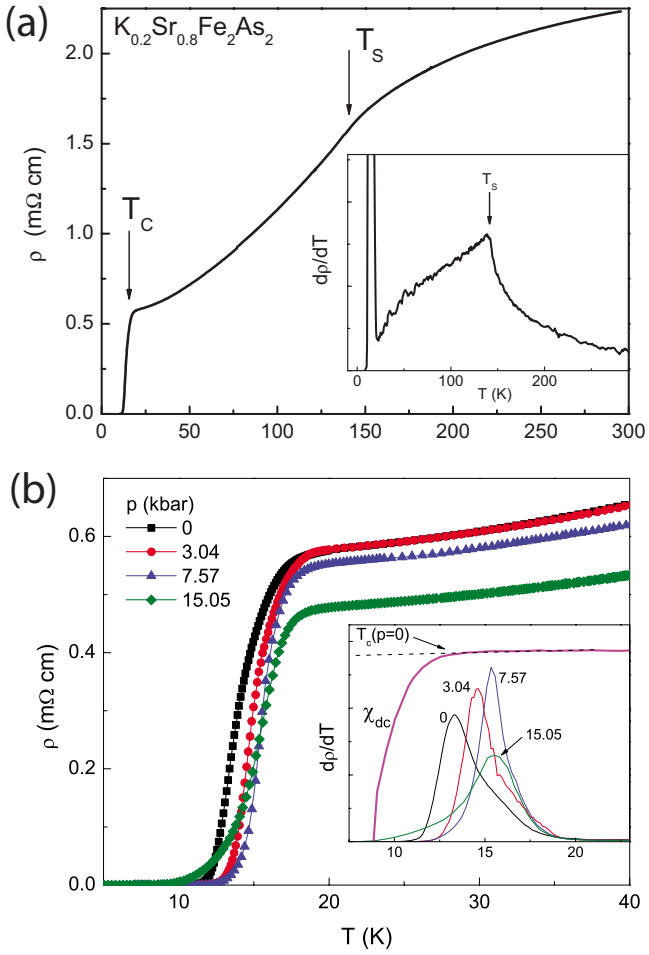

FIG. 1. (Color online) Resistivity of $K_{0.2}Sr_{0.8}Fe_2As_2$ vs temperature at (a) ambient pressure and (b) high pressures. The insets show the derivative with the anomalies at T_S (a) and T_c (b). The dashed line in the inset of (b) is the ambient pressure magnetic susceptibility.

The ac susceptibility of a sample with maximum T_c (x=0.4, T_c=37 K) is shown in Fig. 2(a). At zero pressure the diamagnetic response at 37 K is indicated by a sharp drop of the inductance but develops a shoulderlike feature at lower T (~32 K). This two-step magnetic transition is a characteristic of polycrystalline superconducting samples where intergrain coherence is established only at a lower temperature than the intragrain superconducting transition. With increasing pressure the shoulder is diminished because of p-induced improvements of the grain-grain contacts. At the highest pressure the inductance drop exhibits an overall broadening of the transition similar to the resistance drop in the Sr-rich (x=0.2) sample. In addition, the change in T_c with pressure is very small. In order to extract values of T_c we determined the onset temperature of the diamagnetic signal at different pressures. The normalized $T_c(p)$ is plotted in Fig. 3 (center curve). This data show that the pressure shift of T_c of this sample is negligibly small within the pressure range of this investigation. The observed pressure behavior is similar to that of the cuprate superconductors near optimal doping.

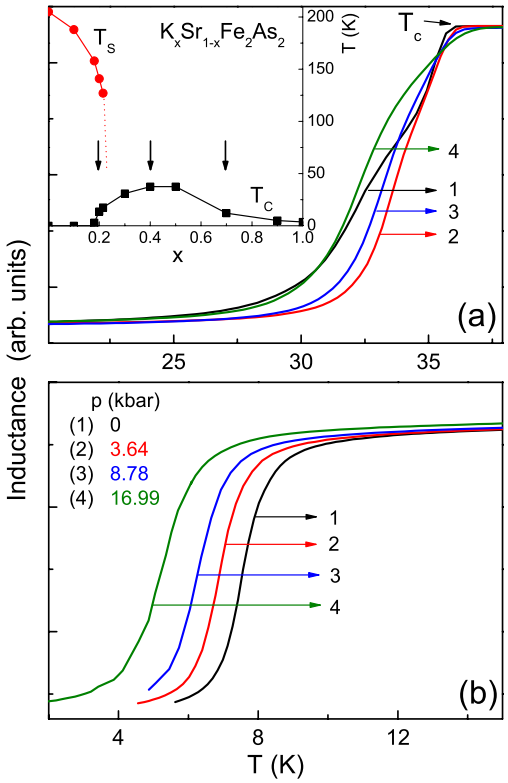

FIG. 2. (Color online) ac susceptibility of (a) $K_{0.4}Sr_{0.6}Fe_2As_2$ and (b) $K_{0.7}Sr_{0.3}Fe_2As_2$ near the superconducting transitions at different pressures. The pressure values from 1 to 4 are labeled in Fig. 2(b). The inset to Fig. 2(a) shows the phase diagram of $K_xSr_{1-x}Fe_2As_2$.

In contrast to the two previously discussed samples, the ac susceptibility of the $x=0.7$ compound (largest hole concentration) exhibits a significant decrease in the transition temperature of the diamagnetic signal [Fig. 2(b)]. The inductance curves at different pressures are nearly parallel, with T_c decreasing rapidly and linearly with increasing p. The normalized pressure shifts of T_c for all three samples are shown in Fig. 3. From the data we conclude that, similar to the high-T_c cuprate superconductors, the pressure effects on T_c of the system $K_xSr_{1-x}Fe_2As_2$ depend strongly on carrier-concentration levels. Moreover, considering the phase diagram $T_c(x)$ shown in the inset to Fig. 2(a), we further conclude that the major effects of pressure is to increase the hole density in the Fe_2As_2 layers through charge transfer to the charge reservoir block.

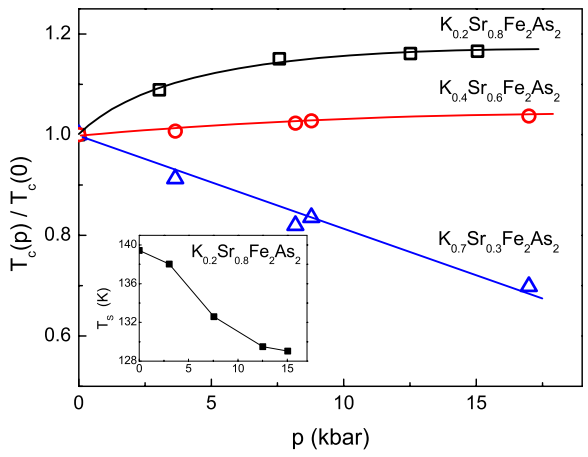

FIG. 3. (Color online) The shift $T_c(p)/T_c(0)$ with pressure for $K_{0.2}Sr_{0.8}Fe_2As_2$, $K_{0.4}Sr_{0.6}Fe_2As_2$, and $K_{0.7}Sr_{0.3}Fe_2As_2$. Inset: p dependence of T_S of $K_{0.2}Sr_{0.8}Fe_2As_2$.

However, comparing the $RO_{1-x}F_xFeAs$ and $K_xSr_{1-x}Fe_2As_2$ superconductors, there are significant differences regarding the pressure-induced charge control in the Fe_2As_2 layers. High pressure appears to increase the electron count in $RO_{1-x}F_xFeAs$ (Ref. 26) but it increases the hole density in $K_xSr_{1-x}Fe_2As_2$. The two classes of FeAs superconducting compounds are distinguished structurally by different stacking arrangements of the Fe_2As_2 layers along the c axis. In the $ThCr_2Si_2$-type structure ("122"), adjacent Fe_2As_2 layers are stacked in a way that two As atoms along the c axis are related by a mirror plane resulting in closer As-As distances and a doubling of the c axis with respect to the unit cell of the "1111" compounds.[18] In the ZrCuSiAs-type structure ("1111"), the adjacent Fe_2As_2 layers are stacked "in phase" along the c axis resulting in a smaller unit cell than in the "122" compounds. The application of pressure on the "122" structure compresses the lattice, moves the As atoms to approach the K/Sr layers and leads to shorter interlayer distances. The shortening of the interlayer distance results in an increased hybridization of the Sr $5s/5p$ orbitals with the c axis component of the hybridized Fe/As orbitals, and electrons are dispersed from the active Fe_2As_2 layer. This mechanism of pressure-induced charge transfer is confirmed by recent band-structure calculations.[31] As a consequence the T_c of the optimally doped sample is expected to decrease with further increase in pressure, similar to the pressure dependence of T_c derived from the onset of the resistivity drop in $LaO_{0.89}F_{0.11}FeAs$.[2] The small T_c increase observed at low hydrostatic pressures in $K_{0.4}Sr_{0.6}Fe_2As_2$ can be understood as a response of a slightly underdoped sample. According to the phase diagram [inset, Fig. 2(a)] x_{opt} lies between $x=0.4$ and $x=0.5$.[18]

The systematic investigation of the pressure effects on the superconducting T_c of the system $K_xSr_{1-x}Fe_2As_2$ has revealed surprising similarities with the effects of pressure on the high-T_c cuprate superconductors. This provides convincing evidence that the FeAs superconductors are not only structurally but also electronically similar to the cuprates. From our results we also conclude that high pressure increases the hole density in the Fe_2As_2 superconducting layers of the "122" compounds. This effect should also contribute to the suppression of the SDW or structural transitions and the stabilization of superconductivity at high pressures in the nonsuperconducting compounds $AeFe_2As_2$ (Ae=Ca, Sr, Ba).[32–34] Accurate band-structure calculations will be very useful in revealing the details of the Fermi surface and, by varying the lattice parameters, in elucidating the effects of pressure on the charge distribution and carrier densities in the Fe_2As_2 layers of the "122" family of superconducting FeAs-based compounds.

Note added. Recently we became aware of a pressure study of the related superconducting compound ($K_{0.45}Ba_{0.55}Fe_2As_2$) showing a decrease of T_c with pressure.[35]

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