Superconductivity in hole-doped (Sr$_{1-x}$K$_x$)Fe$_2$As$_2$

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A series of layered (Sr$_{1-x}$K$_x$)Fe$_2$As$_2$ compounds with nominal x=0 to 0.40 are synthesized by solid state reaction method. Similar to other parent compounds of iron-based pnictide superconductors, the pure SrFe$_2$As$_2$ shows a strong resistivity anomaly near 210 K, which was ascribed to the spin-density-wave instability. The anomaly temperature is much higher than those observed in LaOF$_2$As and BaFe$_2$As$_2$, the two prototype parent compounds with ZrCuSiAs- and ThCr$_2$Si$_2$-type structures. K-doping strongly suppresses this anomaly and induces superconductivity. Like in the case of K-doped BaFe$_2$As$_2$, sharp superconducting transitions at $T_c$ ~38 K was observed. We performed the Hall coefficient measurement, and confirmed that the dominant carriers are hole-type. The carrier density is enhanced by a factor of 3 in comparison to F-doped LaOF$_2$As superconductor.

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The recent discovery of superconductivity with transition temperature $T_c \sim 26$ K in LaO$_{1-x}$F$_x$FeAs has generated tremendous interest in scientific community[1]. Shortly after this discovery, the $T_c$ was raised to 41-55 K by replacing La by rare-earth Ce, Sm, Pr, Nd, etc, making those systems the first non-copper based materials with $T_c$ exceeding 50 K.[2, 3, 4, 5, 6] The undoped quaternary compounds crystallize in a tetragonal ZrCuSiAs-type structure, which consists of alternate stacking of edge-sharing Fe$_2$As$_2$ tetrahedral layers and La$_2$O$_2$ tetrahedral layers along c-axis. Except for a relatively high transition temperature, the system displays many interesting properties. Most remarkably, the superconductivity was found to be in the vicinity of a spin-density-wave (SDW) instability.[7, 8] The competition between superconductivity and SDW instability was first identified in LaO$_{1-x}$F$_x$FeAs system[7], and subsequently found in other rare-earth substituted systems.[2, 9, 10] Those observations reveal that the superconductivity was intimately related to the magnetic fluctuations.

Very recently, it is found that the ternary iron arsenide BaFe$_2$As$_2$ with a tetragonal ThCr$_2$Si$_2$-type structure, which contains identical edge-sharing Fe$_2$As$_2$ tetrahedral layers as LaOF$_2$As, exhibits a similar SDW instability at 140 K, which was characterized by strong anomalies in resistivity, specific heat, magnetic susceptibility, and structural distortion.[11] It is therefore suggested that BaFe$_2$As$_2$ could serve as a new parent compound for ternary iron arsenide superconductors. Indeed, soon after that, the superconductivity with $T_c \sim 38$ K was found in K-doped BaFe$_2$As$_2$, which was suggested to be a hole-doped iron arsenide superconductor, although a direct measurement of Hall coefficient is lacking.[12] Before K-doped BaFe$_2$As$_2$, the superconductivity induced by hole-doping was reported in (La$_{1-x}$Sr$_x$)FeAs system.[13]

In this work, we report fabrication of a series of layered (Sr$_{1-x}$K$_x$)Fe$_2$As$_2$ compounds with nominal x=0, 0.1, 0.2, and 0.40 by solid state reaction method. We expect to see similar phenomena as observed in (Ba$_{1-x}$K$_x$)Fe$_2$As$_2$ compounds. Indeed, the pure SrFe$_2$As$_2$ shows a strong SDW anomaly in resistivity. However, the anomaly occurs near 210 K, being substantially higher than that observed on LaOF$_2$As and BaFe$_2$As$_2$, the two prototype parent compounds with ZrCuSiAs- and ThCr$_2$Si$_2$-type structures. K-doping strongly weakens this anomaly and induces superconductivity. We performed the Hall coefficient measurement on K-doped superconducting sample, and confirmed that the dominant carriers are hole-type. Furthermore, the carrier density is much higher than F-doped LaO$_{0.5}$F$_{0.5}$FeAs superconductor.

The polycrystalline samples were synthesized by solid state reaction method using SrAs, KAs, Fe$_2$As$_2$ as starting materials. The synthesizing method is similar to that described in our earlier paper[2]. SrAs was prepared starting from reaction involving Sr chips and As pieces at 500 °C for 10 hours and then 750 °C for 20 hours. KAs was prepared by reacting K lumps and As pieces at 600 °C for 24 hours. In order to avoid Sr and K attack on the quartz tubes at elevated temperatures, the elements were put into alumina crucibles and finally sealed in quartz tubes under Ar gas atmosphere. The obtained materials SrAs, KAs, Fe$_2$As$_2$ were thoroughly mixed in a correct ratio and pressed into pellets. The pellets were wrapped with Ta foil and sealed in quartz tube under Ar gas atmosphere. They were then annealed at 750-900 °C for 24 hours. The resulting samples were characterized by a powder X-ray diffraction(XRD) method with Cu Kα radiation at room temperature. The electrical resistivity was measured by a standard 4-probe method and the Hall coefficient was measured by a 5-probe method. The ac magnetic susceptibility was measured with a modulation field in the amplitude of 10 Oe and a frequency of 333 Hz. These measurements were preformed in a Physical Property Measurement System(PPMS) of Quantum Design company.

Figure 1 shows the XRD patterns for the parent compound. The diffraction peaks could be well indexed on the basis of tetragonal ThCr$_2$Si$_2$-type structure with the
space group I4/mmm. A little impurity phases detected from the measurements were attributed to the unstable behavior of SrFe$_2$As$_2$ in air. The lattice constants were $a = 0.3920$ nm and $c = 1.240$ nm, consistent with the reported values.

Figure 2(a) shows the temperature dependence of the resistivity for (Sr$_{1-x}$K$_x$)Fe$_2$As$_2$. The pure SrFe$_2$As$_2$ sample exhibits a strong anomaly near 210 K; the resistivity drops steeply below this temperature. This is a characteristic feature related to SDW instability. The anomalous feature is observed for K-doped samples with $x = 0.2$ and 0.4. The temperature dependence of resistivity for the reannealed sample Sr$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ is shown in Figure 2(b). The superconductivity with $T_c$ $\approx$ 38 K was observed for K-doped samples with $x = 0.2$ and 0.4. The residual anomaly at high temperature comes from the inhomogeneity of samples. It can be removed by further annealing process (powered and pressed again), but this treatment decreases the superconducting transition temperature. Figure 2(c) shows the real and imaginary parts of ac susceptibility in a temperature range near $T_c$ for the reannealed sample Sr$_{0.6}$K$_{0.4}$Fe$_2$As$_2$.

In (Sr$_{1-x}$K$_x$)Fe$_2$As$_2$, the replacement of Sr$^{2+}$ with K$^+$ adds extra holes to the system, the conducting carriers are expected to be hole-type. To confirm this, we performed Hall coefficient measurement in the normal state. Figure 3 shows the Hall coefficient versus $T$ between 30 and 200 K of the reannealed Sr$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. The inset shows the Hall voltage driven by magnetic field at 100 K in field up to 5T. A linear dependence of the transverse voltage against applied magnetic field is observed. The experiment indicates that the Hall coefficient is positive in the measured temperature, confirming the hole-type conducting carriers for (Sr$_{1-x}$K$_x$)Fe$_2$As$_2$. This is strong contrast to that of La(O$_{1-x}$F$_x$)FeAs where the conducting carriers are found to be electron-type. It is interesting that the Hall coefficient shows
temperature-dependent with a broad peak at about 100K. It is possible that the T-dependence of $R_H$ comes from the multiple bands effect. In LaOFeAs, the band calculations show that all the five Fe d-orbital energy levels are not fully occupied, they cross the Fermi level $E_F$, leading to five Fermi surfaces. Here for $(\text{Sr}_{1-x}\text{K}_x)\text{Fe}_2\text{As}_2$, we would expect to have similar five Fermi surfaces. When the carrier scattering rates change with temperature at different rates for different bands, $R_H$ can become T-dependent. The carrier density is estimated to be $5.4 \times 10^{21}$ cm$^{-3}$ at 200K if we simply adopt a single-band formula $n=1/R_H$. The carrier density is more than 3 times as large as that of electron-doped single FeAs layer LaO$_{0.9}$Fe$_{0.1}$FeAs with $n \sim 1.8 \times 10^{21}$ cm$^{-3}$ at 200K.

To conclude, we have synthesized a series of layered (Sr$_{1-x}$K$_x$)Fe$_2$As$_2$ compounds with nominal x=0 to 0.40 by solid state reaction method. We found that the overall behavior of (Sr$_{1-x}$K$_x$)Fe$_2$As$_2$ series is very similar to Re-OFeAs (Re=La, Ce, Pr, Nd, Sm). The observed anomaly at 210 K is attributed to a spin-density-wave (SDW) instability. K-doping suppresses this anomaly and induces superconductivity. This strongly suggests that the competing orders are the common feature for Fe$_2$As$_2$ tetrahedral layers being present in both ZrCuSiAs- and ThCr$_2$Si$_2$-type structures. We performed the Hall coefficient measurement, and confirmed that the dominant carriers are hole-type with much higher carrier density compared with that of single FeAs layer LaO$_{0.9}$Fe$_{0.1}$FeAs.

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