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## FAST TRACK COMMUNICATION

# Superconductivity at 56 K in samarium-doped SrFeAsF

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### Abstract

We synthesized the samples  $Sr_{1-x}Sm_xFFeAs$  with a ZrCuSiAs-type structure. These samples were characterized by resistivity and susceptibility. It is found that substitution of rare earth metal for alkaline earth metal in this system suppresses the anomaly in resistivity and induces superconductivity. Superconductivity at 56 K in nominal composition  $Sr_{0.5}Sm_{0.5}FFeAs$  is realized, indicating that the superconducting transition temperatures in the iron arsenide fluorides can reach as high as that in oxypnictides with the same structure.

(Some figures in this article are in colour only in the electronic version)

The recent discovery of superconductivity in oxypnictides with the critical temperature  $(T_c)$  higher than the McMillan limit of 39 K (the theoretical maximum predicted by BCS theory) has generated great excitement [1-6] since the superconductivity is clearly unconventional compared with the cuprate superconductors. The high- $T_c$  iron pnictides with a ZrCuSiAs-type structure adopt a layered structure of alternating (FeAs)<sup>-</sup> and (LnO)<sup>+</sup> layers with eight atoms in a tetragonal unit cell. A structural phase transition from tetragonal to orthorhombic happens before the antiferromagnetic spin-density-wave (SDW) ordering [7, 8]. Such a transition leads to an anomaly in resistivity. Doping of charge into the system suppresses the structural and magnetic instabilities, and induces superconductivity. Recently, iron arsenide fluorides AEFeAsF (AE = Sr, Ca etc) with a ZrCuSiAs-type structure, where the  $(LnO)^+$ layers in LnFeAsO are replaced by (AEF)<sup>+</sup> layers have been reported [9–11]. Co doping in CaFeAsF leads to a maximum  $T_{\rm c}$  of 22 K [9], and a superconducting transition at about 36 K is reported in Sr<sub>0.8</sub>La<sub>0.2</sub>FeAsF [12].

Here we report the discovery of superconductivity at 56 K in nominal composition  $Sr_{1-x}Sm_xFeAsF$  (x = 0.5) with a ZrCuSiAs-type structure. The  $T_c$  is almost the same as the highest superconducting transition temperature observed in F-doped oxypnictide superconductors.

The polycrystalline samples with nominal composition of  $Sr_{1-x}Sm_xFeAsF$  were synthesized by a solid state reaction method by using SrF<sub>2</sub>, SrAs, SmAs, and Fe<sub>2</sub>As as starting materials:  $0.5\text{SrF}_2 + (0.5 - x)\text{SrAs} + x\text{SrAs} + 0.5\text{Fe}_2\text{As} \rightarrow 0.5\text{Fe}_2\text{As}$  $Sr_{1-x}Sm_xFeAsF$ . SrAs was pre-synthesized by heating the mixture of Sr powder and As powder in an evacuated quartz tube at 873 K for 10 h. SmAs and Fe<sub>2</sub>As were obtained by respectively reacting the mixture of Sm powder, Fe powder and As powder in evacuated quartz tubes at 1073 K for 10 h. The raw materials were accurately weighed according to the stoichiometric ratios of  $Sr_{1-x}Sm_xFeAsF$ , then the weighed powders were thoroughly ground and pressed into pellets. The pellets were wrapped with Ta foil and sealed in evacuated quartz tubes. The SrFeAsF was slowly heated to 1173 K, kept at this temperature for 40 h and then cooled down to room temperature. Then the resultant pellet was ground again and sealed in a quartz tube for a second sintering at 1273 K for 20 h.

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**Figure 1.** X-ray diffraction patterns at room temperature for the samples  $Sr_{1-x}Sm_xFeAsF$  with x = 0, 0.2, and 0.5. All diffraction peaks can be indexed with a tetragonal structure for the x = 0 sample. However, impurity phases of  $SrF_2$  and SmAs are observed in x = 0.2 and 0.5 samples.

The samples of  $Sr_{0.8}Sm_{0.2}FeAsF$  and  $Sr_{0.5}Sm_{0.5}FeAsF$  were slowly heated to 1273 K for 20 h, then the products were ground again and sealed in a quartz tube for a second sintering at 1273 K for 10 h. The sample preparation process except for annealing was carried out in glove box (O<sub>2</sub>, H<sub>2</sub>O < 1 ppm) in which high pure argon atmosphere is filled.

The crystal structure of these samples was characterized by x-ray diffraction (XRD) with a Rigaku D/max-A x-ray diffractometer using Cu K $\alpha$  radiation ( $\lambda = 1.5418$  Å) in the  $2\theta$  range of 5°–65° with steps of 0.02° at room temperature. Resistivity measurements were performed on an AC resistance bridge (Linear Research Inc., Model LR700) by the standard four-probe method. The measurement of susceptibility was performed in a Quantum Design PPMS system (Quantum Design).

Figure 1 shows x-ray powder diffraction patterns of SrFeAsF, Sr<sub>0.8</sub>Sm<sub>0.2</sub>FeAsF and Sr<sub>0.5</sub>Sm<sub>0.5</sub>FeAsF, respectively. All diffraction peaks in the XRD pattern of the sample SrFeAsF can be indexed by a tetragonal structure with a =0.3995 nm and c = 0.8961 nm, where no impurity peak is observed. It indicates that the sample is single phase. For the Sr<sub>0.8</sub>Sm<sub>0.2</sub>FeAsF sample, all the main peaks can be indexed to a ZrCuSiAs-type structure with a = 0.3929 nm and c =0.8958 nm. Only small amount of an impurity phase of SrF<sub>2</sub> is detected. For the Sr<sub>0.5</sub>Sm<sub>0.5</sub>FeAsF sample, the diffraction peaks except for the diffraction peaks from impurity phases of SrF2 and SmAs can be indexed by a tetragonal structure with a = 0.3918 nm and c = 0.8956 nm. It is found that Sm doping leads to an apparent decrease in the *a*-axis and a slight contraction of the *c*-axis. With increasing Sm doping, the impurity phase of SrF<sub>2</sub> apparently increases. The impurity phase of SmF shows up in the Sr<sub>0.5</sub>Sm<sub>0.5</sub>FeAsF sample.

Figure 2 shows temperature dependence of resistivity for the samples SrFeAsF,  $Sr_{0.8}Sm_{0.2}FeAsF$  and  $Sr_{0.5}Sm_{0.5}FeAsF$ . The resistivity of SrFeAsF shows a clear anomaly in resistivity at about 173 K. While cooling down from higher temperature,

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**Figure 2.** Temperature dependence of resistivity for the samples  $Sr_{1-x}Sm_xFeAsF$  with x = 0, 0.2, and 0.5. The anomaly in resistivity associated with structural and SDW transitions is suppressed with Sm doping. A sharp superconducting transition at 56 K is observed in the x = 0.5 sample.

the sample's resistivity grows slightly before 173 K, then drops sharply as the temperature continues decreasing. At low temperature, the resistivity shows a semiconducting behavior. This is consistent with previous reports [10, 11]. The anomaly in resistivity is ascribed to the formation of an SDW order or a structural transition, similar to what is observed in the LnFeAsO (Ln = rare earth elements) and MFe<sub>2</sub>As<sub>2</sub> (M = Ba, Sr) systems. With Sm doping, the anomaly in resistivity is suppressed, and the transition temperature shifts down to 163 K for Sr<sub>0.8</sub>Sm<sub>0.2</sub>FeAsF, while the semiconducting behavior at low temperature is also suppressed. As shown in figure 2, a sharp superconducting transition at about 56 K occurs in resistivity for the sample Sr<sub>0.5</sub>Sm<sub>0.5</sub>FeAsF. The resistivity shows a metallic behavior in the whole temperature range and no anomaly appears. It suggests that the SDW order or the structural transition is completely suppressed for the superconducting sample. The behavior of resistivity in the normal state is exactly the same as that observed in superconducting samples SmFeAsO<sub>1-x</sub> $F_x$  (x = 0.15 and 0.2) [5]. It should also be emphasized that no superconducting transition is observed in the sample Sr<sub>0.8</sub>Sm<sub>0.2</sub>FeAsF.

To confirm the superconductivity observed in resistivity for the sample with nominal composition Sr<sub>0.5</sub>Sm<sub>0.5</sub>FeAsF, the susceptibility is measured under 10 Oe in zero-fieldcooled and field-cooled cycles. The temperature dependence of susceptibility is shown in figure 3. A clear diamagnetic transition occurs at 53.5 K corresponding to the mid-transition temperature in resistivity, indicating a bulk superconductivity. It should be pointed out that the superconducting volume is small, and the apparent Meissner fraction is less than 10%, while the resistance goes to zero. The reason could be that there are some magnetic impurities (such as FeAs) in this sample. The magnetic ordering of these impurities may depress the Meissner fraction. The actual Meissner fraction must be larger. The next step is how to synthesize a sample of high purity and improve the superconducting volume fraction. The superconducting phase should be Sm-doped SrFeAsF



**Figure 3.** Temperature dependence of DC magnetization for the sample with nominal composition  $Sr_{0.5}Sm_{0.5}FeAsF$  measured at a DC field of 10 Oe in the zero-field-cooled (ZFC) and field-cooled cycles. It indicates a bulk superconductivity.

because only impurity phases  $SrF_2$  and SmAs are observed in the x-ray diffraction pattern of  $Sr_{0.5}Sm_{0.5}FeAsF$ . These impurity phases are not superconducting.

In summary, partial substitution of Sr with Sm leads to the suppression of the structural and SDW transition,

and meanwhile induces superconductivity. The electrical conductivity and magnetization measurements demonstrate a bulk superconductivity at 56 K in a nominal composition of  $Sr_{0.5}Sm_{0.5}FeAsF$ . Our results indicate that it is possible to find superconductivity in other members of the fluorine–arsenide family.

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