



Parent phase and superconductors in the fluorine derivative family

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ARTICLE INFO

Article history:

Available online 19 March 2009

PACS:

74.70.-b

74.25.Fy

73.43.Qt

Keywords:

Fluorine derivative

Parent phase

Superconducting phase

ABSTRACT

The family of fluoride–arsenide AeFeAsF compounds (Ae = alkaline earth elements or other divalent elements) with the ZrCuSiAs structure was successfully fabricated and investigated. The resistivity and magnetic anomalies associated with the structural phase transition or Antiferromagnetic (AF) transition were found to occur at about 173 K for SrFeAsF, 118 K for CaFeAsF and 153 K for EuFeAsF, respectively. The Hall coefficients R_H of two parent phases, SrFeAsF and CaFeAsF, were measured and found to be positive at low temperatures. By doping rare earth elements into the system, superconductivity was induced. Superconductivity at 29.5 K in Sr_{0.6}La_{0.4}FeAsF and beyond 50 K in Ca_{0.4}Re_{0.6}FeAsF (RE = rare earth elements) was successfully obtained. More superconductors are expectable in this family by doping charges into the system in multiple ways.

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1. Introduction

Since the discovery of superconductivity in LaFeAsO_{1-x}F_x [1], which is abbreviated as FeAs-1111 phase with the ZrCuSiAs structure, great interests have been stimulated in the fields of condensed matter physics and material science. Very soon, the superconducting transition temperature was promoted above 50 K by replacing lanthanum with other rare-earth elements. Meanwhile, the hole-doped superconductors were discovered both in the FeAs-1111 phase and the FeAs-122 phase (e.g., Ba_{1-x}K_xFe₂As₂) [2–5]. Experimental data accumulated so far indicate that the parent phases both in the FeAs-1111 system and the FeAs-122 system are intimately connected with an Antiferromagnetic order in the FeAs layers [6,7] in low temperature region. For example, in undoped LaFeAsO, an Antiferromagnetic-driven or structural phase transition occurs around 150 K [6]. By suppressing this low temperature AF order, the superconductivity will win eventually. Although the mechanism of superconductivity in the FeAs-based system remains unclear, it seems that replacing the REO-layers with some new building blocks may lead to new superconductors.

It has long been known that the SrZnPf [8] compound has the ZrCuSiAs structure. It is constructed by the alternative building series of SrF and ZnP, and the SrF has a total ionic state of “+1”. It is also known that the F-doped ReFeAsO (Re = rare earth elements) superconductors with ZrCuSiAs structure have the ReO series with an ionic state of “+1”. Therefore a natural idea is to replace the REO

layers in REFeAsO with AeF layers (Ae = alkaline earth elements or other divalent elements). Consequently the new family of fluoride–arsenide AeFeAsF (Ae = alkaline earth elements or other divalent elements) compounds was successfully synthesized [9–12]. By partially substituting the Fe with Co, superconductivity at 22 K was found in CaFe_{1-x}Co_xAsF [11] and 4 K in SrFe_{1-x}Co_xAsF [13]. However, it is known that this kind of substituting inevitably introduces disorders into the FeAs planes which may bring about difficulties in improving the transition temperature. Therefore different ways to introduce charges into the system are worthwhile to try [14].

In this paper, we report some recent work and developments on the fluorine derivative family. We successfully fabricated three parent compounds SrFeAsF, CaFeAsF, and EuFeAsF. We measured the resistivity and DC magnetic susceptibility of these samples. They have an structural/AF like transition at about 173 K for SrFeAsF, 118 K for CaFeAsF and 153 K for EuFeAsF, respectively. We also measured the Hall effect of the parent phases on SrFeAsF and CaFeAsF, and found that the Hall coefficients R_H were positive at low temperatures. This is very different from the case in REFeAsO and (Ba,Sr)Fe₂As₂. By doping electrons into the system, we have got new superconductors Sr_xLa_{1-x}FeAsF with $T_c = 29.5$ K, and Ca_xRe_{1-x}FeAsF (Re = rare earth elements) with the critical transition temperatures beyond 50 K [10,15].

2. Experiments

The samples of the fluorine derivative family were prepared using a two-step solid-state reaction method, as that used for preparing the LaFeAsO samples [16]. In the first step, by reacting La grains (purity 99.99%), Pr grains (purity 99.99%), Sr (or Ca, Eu,

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etc. purity 99%) pieces with As grains at 500 °C for 8 h and then 700 °C for 16 h, we successfully fabricated the powders of LaAs, PrAs, and SrAs (or CaAs, EuAs, etc.). Then they were thoroughly ground together with FeF₂ (purity 99%) and Fe powder (purity 99.99%) in the formula AeFeAsF or Ae_{1-x}Re_xFeAsF (Ae = Sr, Ca and Eu, Re = rare earth elements), and pressed into a pellet shape. Then the pellets were sealed in an evacuated silica tube and heated at about 1000 °C for 30 h, followed by cooling down slowly to room temperature. All the weighing, mixing, grinding and pressing procedures were finished in a glove box under argon atmosphere with the moisture and oxygen below 0.1 PPM.

The resistivity and Hall effect measurements were performed in a physical property measurement system (Quantum Design, PPMs) with magnetic fields up to 9 T. The six-lead method was used in the measurement on the longitudinal and the transverse resistivity at the same time. The resistivity was measured by either sweeping magnetic field at a fixed temperature or sweeping temperature at a fixed field. The temperature stabilization was better than 0.1% and the resolution of the voltmeter was better than 10 nV.

3. Results and discussion

3.1. Parent phase

In Fig. 1, the structure of AeFeAsF is shown. The Fe₂As₂ layers are well separated by the building blocks of Ae₂F₂. In Fig. 2, we present the X-ray diffraction patterns of the parent phases of SrFeAsF, CaFeAsF and EuFeAsF. It is found that all main peaks can be

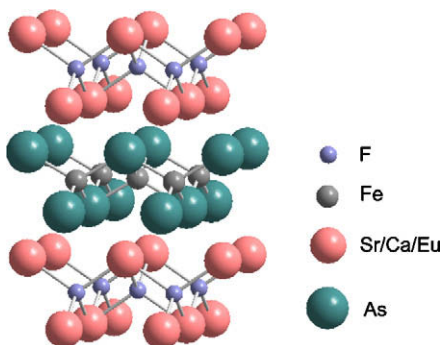


Fig. 1. Crystal structure of AeFeAsF (Ae = alkaline earth elements or other divalent elements). The Fe₂As₂ layers are well separated by the building block Ae₂F₂.

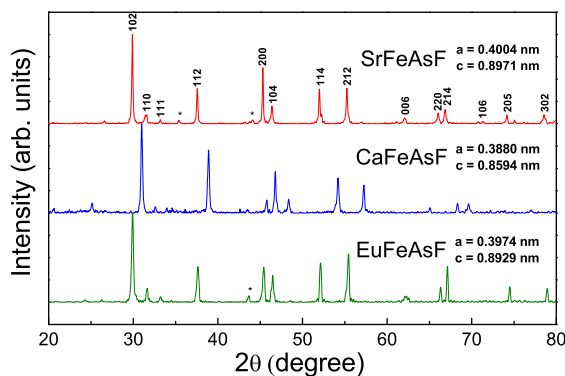


Fig. 2. X-ray diffraction patterns of the parent phases of SrFeAsF, CaFeAsF and EuFeAsF. All main peaks can be indexed by a tetragonal structure with $a = b = 4.004 \text{ \AA}$ and $c = 8.971 \text{ \AA}$ for the sample SrFeAsF, $a = b = 3.88 \text{ \AA}$ and $c = 8.594 \text{ \AA}$ for the sample CaFeAsF, $a = b = 3.974 \text{ \AA}$ and $c = 8.929 \text{ \AA}$ for the sample EuFeAsF, respectively.

indexed by a tetragonal structure very well, with $a = b = 4.004 \text{ \AA}$ and $c = 8.971 \text{ \AA}$ for the sample SrFeAsF, $a = b = 3.88 \text{ \AA}$ and $c = 8.594 \text{ \AA}$ for CaFeAsF, $a = b = 3.974 \text{ \AA}$ and $c = 8.929 \text{ \AA}$ for EuFeAsF, respectively. One can see that the lattice constants of AeFeAsF are very close to that of REFeAsO.

In Fig. 3, the temperature dependence of resistivity for the parent samples of AeFeAsF (Ae = alkaline earth elements or other divalent elements) was shown. Meanwhile, we also show the zero field cooled DC magnetization of SrFeAsF and CaFeAsF at 5000 Oe in the inset of Fig. 3. A clear resistivity anomaly was observed at about 173 K for SrFeAsF, 118 K for CaFeAsF, and 153 K for EuFeAsF, respectively. Above the anomaly point T_{an} , the resistivity increases slightly with decreasing temperature, but it drops sharply below T_{an} . As we can see, a magnetic anomaly measured by DC magnetization was found to be corresponding very well to the resistivity anomaly in SrFeAsF and CaFeAsF. The clear anomaly in the magnetization curve confirms the structural and Antiferromagnetic transition observed in the resistivity data, which resembles the case in the REFeAsO and AeFe₂As₂ system. The reason for this resistivity and magnetic anomaly is still an open question. One possibility is that the spin-related scattering rate depresses sharply when the long range AF order is formed. In this case, the itinerant electrons

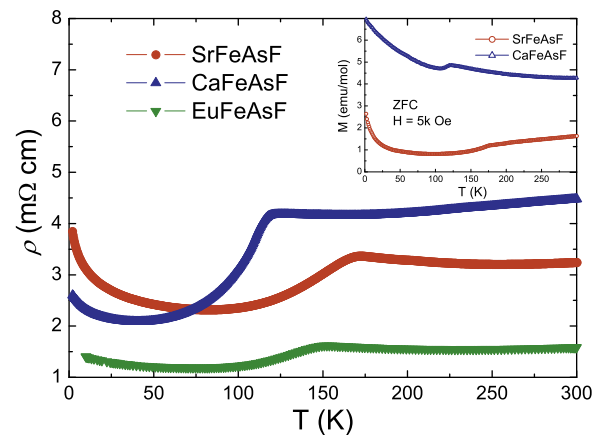


Fig. 3. Resistivity versus temperature in SrFeAsF, CaFeAsF and EuFeAsF. The inset shows the zero field cooled DC magnetization of SrFeAsF and CaFeAsF at 5000 Oe. An anomaly was observed both in the resistivity and magnetization curves which may be induced by the structural and AF transition. The anomaly temperature T_{an} is about 173 K for SrFeAsF, 118 K for CaFeAsF and 153 K for EuFeAsF, respectively.

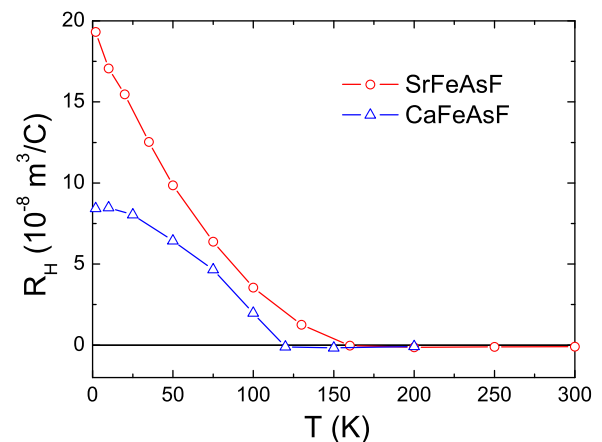


Fig. 4. Temperature dependence of Hall coefficients R_H determined on the present samples SrFeAsF and CaFeAsF. One can see a monotonic decrease in R_H in the temperature region below about 160–170 K for SrFeAsF and 110–120 K for CaFeAsF.

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