Phase diagram of the SmCo$_{1-x}$Fe$_x$AsO system

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Abstract.

A series of SmCo$_{1-x}$Fe$_x$AsO ($x=0, 0.05, 0.1, 0.2, 0.3$) polycrystalline samples were synthesized, and their transport and magnetic properties were studied. Similar to the undoped SmCoAsO, these Fe-doped samples of SmCo$_{1-x}$Fe$_x$AsO also undergo successive magnetic phase transitions associated with the Co or Fe magnetic moments below 100 K, i.e., the system become ferromagnetic (FM) ordered first below the Curie temperature $T_c$, and then become antiferromagnetic (AFM) ordered with further decreasing temperature. With Fe doping on the Co site, both FM ($T_c$) and AFM ($T_N^1$) transition temperatures are rapidly suppressed and then both $T_c$ and $T_N^1$ approaching zero temperature at $x = 0.3$, whereas the AFM order ($T_N^2$) of Sm$^{3+}$ moments below 5.1 K is still robust. Based on these results, the magnetic phase diagram in the Co-rich side of the SmCo$_{1-x}$Fe$_x$AsO system is established.

1. Introduction

Recently, it has been reported that superconductivity can be induced with partial Co substitution on the Fe site in iron-based pnictide superconductors[1, 2]. Similar results have been realized for the Co-doped CeFeAsO[3], PrFeAsO[4], and SmFeAsO systems [2]. However, at the other end, i.e., Fe is completely replaced by Co, the LnCoAsO (Ln: rare earth) compounds (Co-based 1111 phase) do not show superconductivity. Instead, they exhibit rich magnetic properties at low temperature. The parent compounds LaCoAsO shows an itinerant ferromagnetic (FM) behaviors below 63 K[5]. As La is replaced by other rare earth element with magnetism, LnCoAsO (Ln=Ca, Pr, Nd, Sm, Gd)[6] compound undergoes multiple magnetic phase transitions with decreasing temperature. Furthermore, the antiferromagnetic (AFM) order of Ln ions (Ce, Nd, and Pr etc.) in those compounds is still observed at a very low temperature. For example, SmCoAsO[7, 8] undergoes ferromagnetic (FM) transition around $T_c$ of 80 K, and then becomes AFM order due to the magnetic coupling between the CoAs layers below 45 K, and finally shows a second AFM order of Sm moments at 5.6 K.

Up to now, there are few reports on the evolution of ground state in Co-based 1111 phase with Fe doping, and the phase diagram of magnetism versus Fe/Co ratio is unknown. In this paper, we investigated the magnetic properties of Fe-doped SmCo$_{1-x}$Fe$_x$AsO system ($0 \leq x \leq 0.3$) . Our studies reveal that both the FM and AFM transition temperatures associated with the Co/Fe layer are gradually suppressed with increasing Fe content, and then the magnetic orders in the Co/Fe layer disappear as $x \geq 0.3$, whereas the AFM order of Sm$^{3+}$ ions around 5.1 K is still robust. The AFM transition temperature ($T_N^2$) of Sm$^{3+}$ ions slightly varies with
the Fe content. Thus, a magnetic phase diagram of the SmCo$_{1-x}$Fe$_x$AsO system is established for the regime of $0 \leq x \leq 0.3$.

2. Experimental
The polycrystalline samples of SmCo$_{1-x}$Fe$_x$AsO were synthesized by two-step solid state reaction methods in vacuum, similar to our previous reports[2]. The pellets of SmCo$_{1-x}$Fe$_x$AsO were annealed in an evacuated quartz tube at 1423 K for 40 hours and furnace-cooled to room temperature.

The crystal structure and phase purity was checked by powder X-ray diffraction (XRD) at room temperature using a D/Max-rA diffractometer with Cu K$_\alpha$ radiation and a graphite monochromator. Lattice parameters were calculated by a least-squares fit using at least 20 XRD peaks. The electrical resistivity was measured by a standard four-probe method. The temperature dependence of d.c. magnetization was measured on a Quantum Design Magnetic Property Measurement System (MPMS-5) with an applied field of 10 Oe.

3. Results and Discussion

![Figure 1](image_url)

Figure 1. Temperature dependence of resistivity ($\rho$) for the SmCo$_{1-x}$Fe$_x$AsO ($x = 0, 0.05, 0.1, 0.2, 0.3$) samples.

Fig.1 shows the plots of temperature dependent electrical resistivity ($\rho$) of SmCo$_{1-x}$Fe$_x$AsO samples. For the undoped SmCoAsO, the resistivity monotonically decreases with decreasing temperature, followed a distinguishable upturn around 45 K which is associated with the FM-AFM transition temperature $T_{N1}$, as in our previous report [8]. As Fe content increases to $x = 0.1$, this kink becomes more pronounced and moves to lower temperatures, and no anomaly is observed below $T_{N1}$. For $x = 0.2$, the resistive anomaly related with $T_{N1}$ is no longer observed. But another tiny kink is still distinguished at $T = 5.1$ K, which is due to the AFM transition ($T_{N2}$) of the magnetic sublattice of Sm$^{3+}$ ions. As $x$ increases to 0.3, although the kink in resistivity related to $T_{N1}$ is not observed, the change around $T_{N2}$ becomes more remarkable. Those transitions can be clearly observed by the measurement of heat capacity in SmCoAsO[8]. On the other hand, the resistivity increases as $x$ increases, which may be attributed to the increasing disorder in Fe-doped samples and also that the Fe 3d electron may not as itinerant as the Co 3d electrons.

Fig.2 shows the temperature dependence of magnetic susceptibility for the SmCo$_{1-x}$Fe$_x$AsO samples. With decreasing temperature, several magnetic transitions of undoped SmCoAsO...
Figure 2. Temperature dependence of magnetic susceptibility ($\chi$) for the SmCo$_{1-x}$Fe$_x$AsO samples.

sample are revealed. Below $T_c$ of 80 K, the magnetic susceptibility under $H = 1$ kOe strongly increases, suggesting that the Co sublattice forms ferromagnetic order. This magnetic behavior is similar to that reported for LnCoAsO[6]. As Co is replaced by Fe, FM transition temperature($T_c$) is suppressed and shifts to low temperature. For $x=0.3$, FM transition in $\chi(T)$ is not observed.

The second magnetic transition in SmCoAsO ($T_{N1}$) associated with the FM-AFM order of cobalt sublattice is observed about 45 K. Such a transition has been investigated in previous reports for Ln = Nd, Sm, and Gd [6]. As $x$ increases, $T_{N1}$ gradually decreases to 6.5 K for $x = 0.2$. Corresponding to the resistivity data, a clear upturn is observed at this transition temperature. For $x = 0.3$, $T_{N1}$ cannot be observed above 1.8 K. Instead, a sharp peak at 5.1 K is detected, which is attributed to the AFM ordering of Sm$^{3+}$ sublattice[8]. For other samples, it is notable that the AFM transition ($T_{N2}$) in their magnetization curves is distinguished or is absent. The reason is that the relative small moment of Sm$^{3+}$ is masked by the Co ions magnetization.

Based on above data, a magnetic phase diagram of the SmCo$_{1-x}$Fe$_x$AsO system is plotted in Fig.3. $T_c$ associated with the FM transition of the Co/Fe layer is sharply suppressed and disappear as $x$ increases to 0.3. Meanwhile, $T_{N1}$ associated the FM to AFM transition of Co/Fe sublattice is also moved to a lower temperature and disappears for $x = 0.3$. Finally, the AFM ordering of Sm moments at $T_{N2}$ varies slightly with Fe content, and is still robust at $x = 0.3$.

4. Conclusions

In summary, the Co-rich side of the SmCo$_{1-x}$Fe$_x$AsO system have been investigated and the magnetic phase diagram is established. The system undergoes successive magnetic phase transitions with cooling temperature. $T_c$ is suppressed more rapidly than $T_{N1}$ with increasing Fe content. Furthermore, both magnetic transitions disappear near the same Fe-doping critical point of $x = 0.3$. However, the AFM order of Sm moments at $T_{N2}$ is robust for $x$ as large as 0.3, indicating that the AFM order of Sm moment is hardly affected by Fe doping in the Co-rich side.

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Figure 3. Magnetic phase diagram for the SmCo_{1-x}Fe_xAsO system. The $T_{N2}$ (indicated by open symbols) is taken from Ref.[8]. Paramagnetic (PM), Ferromagnetic (FM), and Antiferromagnetic (AFM).

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