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Search for new iron oxypnictide superconductors by using high pressure synthesis technique

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Abstract:

Here we report on the invention of the Co-doped induced superconductivity in $\text{PrFe}_{1-x}\text{Co}_x\text{AsO}$ ($x=0$ and 0.075). Polycrystalline samples were prepared by high-pressure synthesis method. The $\text{PrFe}_{1-x}\text{Co}_x\text{AsO}$ ($x=0$ and 0.075) crystallizes with the ZrCuSiAs type crystal structure (P4/nmm). The PrFeAsO is a parent compound with non-superconducting phase which can be transformed to superconductor by replacement 7.5 % of Fe by Co. The magnetic susceptibility measurement shows the onset of superconducting transition temperature at 15 K.

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1. Introduction

The recent discovery of high superconducting transition temperatures in FeAs based compounds has generated great interest in the scientific community. In the oxypnictide system $LnFeAsO$ ($Ln = \text{Pr}, \text{Sm}, \text{Nd}, \text{Gd}$), electron- or hole-doping has produced critical temperatures (T_c) as high as 56 K [1–14]. Furthermore our present results on the substitution effect in LaFeAsO_y showed that T_c can be increased to 43 K by replacing high ionic radius La^{3+} (1.16 Å) by smaller ionic radius Y^{3+} (1.019 Å) [15]. The high pressure synthesis technique paves a unique pathway to tune the higher T_c values as well as to search for the new superconductors [15–17]. High pressure synthesis technique is a powerful technique to search for new oxypnictide superconductors, as it suppresses evaporation of toxic elements like As, Hg, Tl etc., requires short reaction time and has ability to form stable phases with high density. Here we show that bulk superconductivity at 7.5 % Co doping level at Fe sites in PrFeAsO , giving onset transition temperature up to $T_{c\text{-onset}} = 15$ K.

2. Experimental

Polycrystalline samples were prepared by high-pressure synthesis method using a cubic-anvil-type apparatus (Riken CAP-07). Powders of PrAs, Fe, Co, Fe_2O_3 were used as the starting materials. PrAs was synthesized by reacting Pr and As at 500 °C for 15 hours and then 850 °C for 5 hours in an evacuated quartz tube. The starting materials were mixed with nominal compositions of $\text{PrFe}_{1-x}\text{Co}_x\text{AsO}$ ($x=0$ and 0.075) and ground by an agate mortar in a glove box filled with dry nitrogen gas. The samples were synthesized by heating the mixtures in BN crucibles under a pressure of about 2 GPa at 1150 °C for 2 h. Powder X-ray diffraction (XRD) patterns were measured using CuK_α radiation (Rigaku RINT 1100). The lattice parameters were

calculated by the least-square fit method. In order to characterize the superconducting properties, zero field cooled (ZFC, shielding signal) and field cooled (FC, Meissner signal) magnetization measurements were performed using a quantum design MPMS magnetometer with applied field of 5 Oe.

3. Results

Figure 1 shows the powder XRD patterns of the samples with nominal compositions of $\text{PrFe}_{1-x}\text{Co}_x\text{FeAsO}$ ($x=0$ and 0.075). The ZrCuSiAs type crystal structure ($P4/nmm$), as expected for PrFeAsO is formed as the main phase for all samples. All the apparent peaks can be indexed based on the LaFeAsO [18] type crystal structure. Accordingly, the notation used in the paper simply means the nominal value of the sample composition and does not necessarily agree with the real composition.

The refined lattice parameters show a decrease with increase in Co-substitution which is expected since the ionic size of Co^{2+} is smaller than that of Fe^{2+} . This indicates the substitution of cobalt in place of iron in PrFeAsO . The lattice parameters of PrFeAsO were $a = 3.9880 \text{ \AA}$ and $c = 8.6150 \text{ \AA}$, changed to $a = 3.9834 \text{ \AA}$ and $c = 8.6102 \text{ \AA}$, respectively by 7.5 % Co doping at Fe site. The change in the lattice parameters are smaller in comparison with the substitution of Y at La site, which changed the lattice parameter $\sim 0.9 \%$ [15].

The temperature (T) - dependence of the magnetic susceptibility (χ) of $\text{PrFe}_{1-x}\text{Co}_x\text{FeAsO}$ ($x=0$ and 0.075) is shown in Fig. 2. The $\text{PrFe}_{1-x}\text{Co}_x\text{FeAsO}$ ($x=0$) is the parent compound and no superconducting transition temperature was observed as shown in the Fig. 2(a). The clear drop in χ , corresponding to the onset of superconductivity (indicated by arrows), is observed at 15 K when 7.5 % of Fe is replaced by Co, as shown in Fig.2 (b). The superconducting volume fraction

estimated at 5K for the zero field cooled (ZFC, shielding signal) is $\sim 50\%$, which is considerable large as bulk superconductivity.

Conclusions

High pressure synthesis technique is a powerful technique to search for new oxypnictide superconductors, as it suppresses evaporation of toxic elements like As, Hg, Tl etc., requires short reaction time and has ability to form stable phases with high density. This method can be used for the study of doping effects as our findings shows that PrFeAsO system can be transformed to superconductor by doping 7.5 % Co at Fe sites. The superconducting transition temperature (T_c) on-sets at 15 K.

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Figure captions:

Fig. 1 X-ray diffraction pattern of (a) PrFeAsO and (b) $\text{PrFe}_{0.925}\text{Co}_{0.075}\text{AsO}$ samples.

Fig. 2 Temperature dependence of the susceptibility for of (a) PrFeAsO and (b) $\text{PrFe}_{0.925}\text{Co}_{0.075}\text{AsO}$ samples. The field cooled and zero field cooled samples are represented by FC and ZFC respectively. Inset of fig. b shows the magnified graph from 0-20K scale.

Figure 1

Figure 2a

Figure 2b