

Superconductivity in $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$ ($x=0.0-0.30$)V. P. S. Awana,^{1,a)} Anand Pal,¹ Arpita Vajpayee,¹ R. S. Meena,¹ H. Kishan,¹ Mushahid Husain,² R. Zeng,³ S. Yu,⁴ K. Yamaura,⁴ and E. Takayama-Muromachi^{4,5}¹National Physical Laboratory (CSIR), Dr. K.S. Krishnan Marg, New Delhi 110012, India²Department of Physics, Jamia Millia Islamia, New Delhi 110025, India³ISEM, University of Wollongong, New South Wales 2522, Australia⁴Transformative Research-Project on Iron Pnictides (TRIP), JST, Tsukuba, Ibaraki 305-0044, Japan and Superconducting Materials Center, National Institute for Materials Science, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan⁵International Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science, Tsukuba, Ibaraki 305-0044, Japan

(Presented 21 January 2010; received 30 October 2009; accepted 10 December 2009; published online 11 May 2010)

We report synthesis, structural details, and magnetization of $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$ with x ranging from 0.0 to 0.30. It is found that Co substitutes fully at Fe site in SmFeAsO in an isostructural lattice with slightly compressed cell. The parent compound exhibited known as the spin density wave (SDW) character is below at around 140 K. Successive doping of Co at Fe site suppressed the SDW transition for $x=0.05$ and later induced superconductivity for $x=0.10, 0.15,$ and $0.20,$ respectively, at 14, 15.5, and 9 K. The lower critical field as seen from magnetization measurements is below 200 Oe. The appearance of bulk superconductivity is established by wide open isothermal magnetization $M(H)$ loops. Superconductivity is not observed for higher content of Co, i.e., $x \geq 0.30$. Clearly the Co substitution at Fe site in $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$ diminishes the Fe SDW character, introduces bulk superconductivity for x between 0.10 and 0.20 and finally becomes nonsuperconducting for x above 0.20. The Fe^{2+} site Co^{3+} substitution injects mobile electrons to the system and superconductivity appears; however direct substitution introduces simultaneous disorder in superconducting FeAs layer and thus superconductivity disappears for higher content of Co. © 2010 American Institute of Physics. [doi:10.1063/1.3366601]

I. INTRODUCTION

Soon after the discovery of superconductivity at 26 K in $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$, the substitution of La by other rare earth elements such as Ce, Pr, Sm, Nd, Gd, and Tb has led to a family of 1111 phase high- T_c superconductors.¹⁻⁹ The parent compound REFeAsO (RE=rare earth) itself is nonsuperconducting but shows an anomaly (spin density wave) at around 150 K. Doping of fluorine into insulating REO layer provides the electrons as charge carriers in FeAs layer, which suppresses the anomaly and superconductivity is obtained. Carrier doping plays a major role in the appearance of superconductivity by suppressing the magnetic order in these complex compounds. Recently it has been shown that superconductivity can be induced in LaFeAsO , CeFeAsO , and SmFeAsO by cobalt substitution¹⁰⁻¹³ in place of iron that results in direct injection of electrons in the conducting FeAs layers.

Here we report the synthesis and characterization of Co-doped SmFeAsO and obtained superconductivity for 0%–20% doping levels. Chemically, Co is a better mean of electron doping and carriers are doped directly into the FeAs planes.¹⁰⁻¹³ For higher Co content the superconductivity disappears. This is most probably due to over doping.

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II. EXPERIMENTAL DETAILS

Polycrystalline $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$; $x=0.0, 0.15, 0.20,$ and 0.30 samples were synthesized by single step solid-state reaction method, details are given in Ref. 11.

The x-ray diffraction (XRD) pattern of the compound was taken on Rigaku mini-flex II diffractometer. The resistivity measurements were carried out by conventional four-probe method on a Quantum Design Physical Property Measurement System (PPMS). Heat capacity measurements $C_p(T)$ were also carried out on the same PPMS. The magnetization measurements were carried out on Quantum Design Magnetic Property Measurement System (MPMS).

III. RESULTS AND DISCUSSION

The room temperature XRD pattern for $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$ samples with $x=0.0, 0.15, 0.20,$ and 0.30 along with their Rietveld analysis are shown in Fig. 1(a). The structure of $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$ at room temperature for all compositions of x was refined with the tetragonal space group $P4/nmm$ (space-group no. 129). Both Sm and As atoms are located at Wyckoff positions $2c$, O is situated at $2a$, and Fe/Co are shared at site $2b$. Good fits to the reported structural model are observed. It is observed that all main peaks can be well indexed based on the space group $P4/nmm$, indicating that the studied samples are essentially single phase. Some extra peaks at around 30° and 43° having very low intensity are also seen in the XRD pattern of pure SmFeAsO and

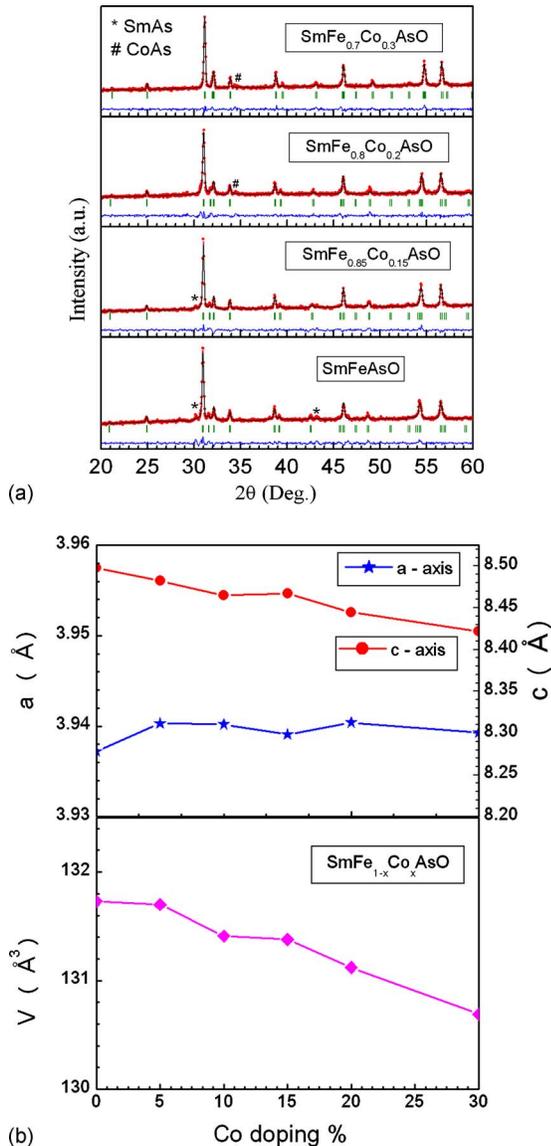


FIG. 1. (Color online) (a) Fitted and observed room temperature XRD patterns of $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$; $x=0.0, 0.15, 0.20$, and 0.30 compounds; impurity peaks are marked with * and “#” in the figure. (b) Effect of Co content on lattice parameters and unit cell volume.

$\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$ sample, which are due to the minor presence of SmAs. Another small peak at around 34.5° shows the minute presence of CoAs for $\text{SmFe}_{0.80}\text{Co}_{0.20}\text{AsO}$ and $\text{SmFe}_{0.70}\text{Co}_{0.30}\text{AsO}$ sample. The exact final compositions of all the studied samples are not determined. Detailed compositional, structural, and microstructural analysis on these samples is already underway and will be reported later.

The Lattice parameters for $\text{SmFe}_{0.9}\text{Co}_{0.1}\text{AsO}$ are found to be $a=3.9398(7)$ Å and $c=8.4639(4)$ Å, while for the parent compound the lattice parameters are $a=3.9375(6)$ Å and $c=8.5021(4)$ Å. Clearly, Co doping leads to decrement in the c -axis lattice, while the a -axis remains nearly unaltered [Fig. 1(b)]. The decrease in c lattice parameter suggests toward the successful chemical substitution of Co in the trivalence Co^{3+} state at the place of Fe^{2+} . The shrinkage of c -axis suggests the strengthening of interlayer Coulomb attraction, implying the increase in density of negative charge in FeAs

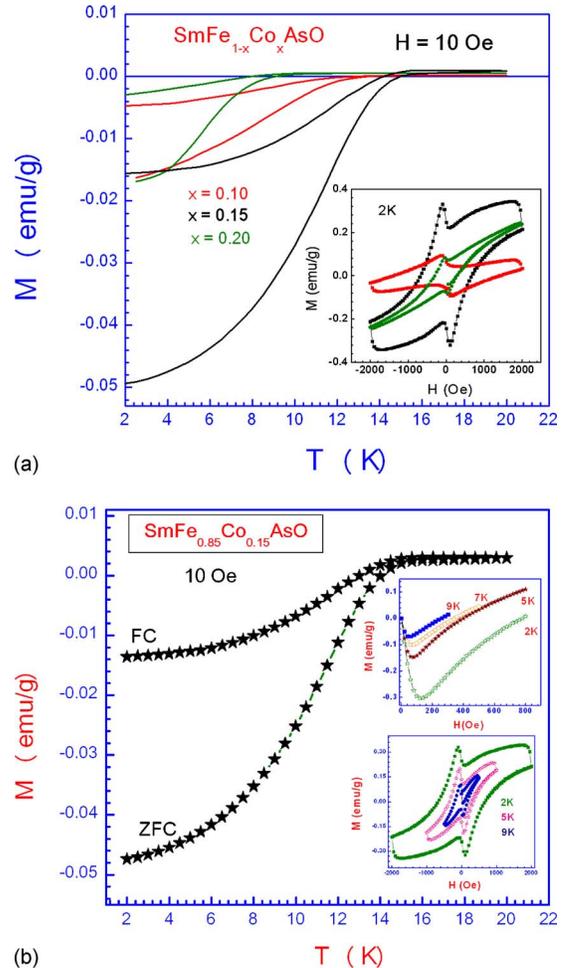


FIG. 2. (Color online) (a) Temperature variation in magnetic susceptibility $M(T)$ in FC and ZFC conditions for $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$; $x=0.10, 0.15$, and 0.20 compounds. Inset shows complete magnetization loops $M(H)$ at 2 K for the same. (b) Temperature dependence of magnetic susceptibility $M(T)$ in FC and ZFC conditions for $\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$ sample. Inset (i) shows the complete magnetization loops $M(H)$ at 2, 5, and 9 K for the same and inset (ii) shows the first quadrant of $M(H)$ loop at 2, 5, 7, and 9 K for $\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$ sample.

layers by the Co doping. The incorporation of Co in the Fe site reduces the cell volume due to contraction of the c -axis lattice constant.

Figure 2(a) shows the temperature dependence of the dc magnetization measured under zero-field-cooled (ZFC) and field-cooled (FC) conditions at 10 Oe for $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$; $x=0.10, 0.15$, and 0.20 samples. The susceptibility becomes negative (diamagnetic) below temperature 14, 15.5, and 9 K for $x=0.10, 0.15$, and 0.20 , respectively. This shows that $x=0.15$ possess the highest transition temperature (T_c) among the three superconducting samples. The $\text{SmFe}_{0.70}\text{Co}_{0.30}\text{AsO}$ does not show superconductivity in magnetization measurement. This is most probably due to over doping of carriers by Fe^{2+} substitution with Co^{3+} . Another reason could be the extent of disorder due to the substitution of Fe^{2+} by Co^{3+} in FeAs superconducting layers. The magnitude of the diamagnetic signal confirms the bulk superconductivity in our samples. The inset of Fig. 2(a) shows M versus H at $T=2$ K for all the three superconducting samples. The $M(H)$ loop opening at 2K is maximum for $x=0.15$ sample, coinciding with its highest T_c of 15.5 K.

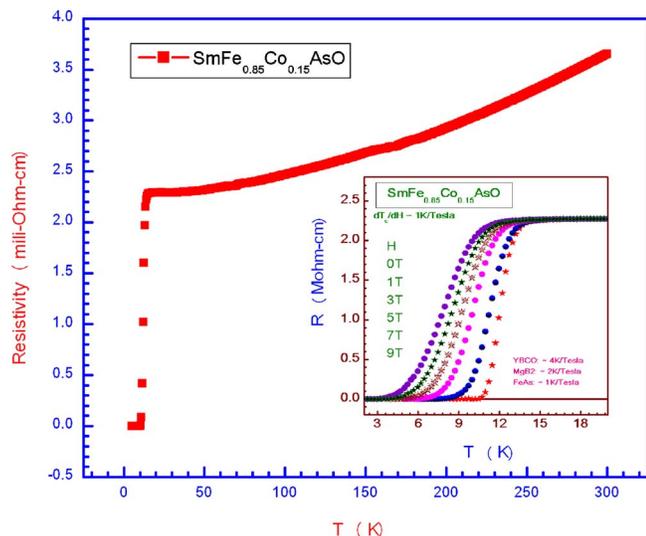


FIG. 3. (Color online) Resistivity behavior with temperature variation $\rho(T)$ of $\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$ sample at zero field. Inset shows the variation in resistivity in the presence of applied magnetic field $\rho(T)H$ up to 9 T.

Temperature dependence of dc susceptibility of highest T_c $\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$ sample is further shown in Fig. 2(b) separately and its magnetization loops at various temperatures of 9, 7, 5, and 2 K are depicted in insets of the same figure. Although transition is broad, the compound undergoes into superconducting state at below 15.5 K. Upper inset shows the first quadrant of $M(H)$ loop at temperature 9, 7, 5, and 2 K for $\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$, where linear response of the Meissner state up to less than 180 Oe is followed by a decrease in magnitude of M with increasing H , as is typical of flux penetration in the vortex state of a type-II superconductor. This gives the lower critical field (H_{c1}) of around 180 and 50 Oe at 2 and 9 K, respectively. As expected, the H_{c1} decreases with increase in temperature from 2 to 9 K. Lower inset shows the complete $M(H)$ loops for $\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$ at 2, 5, and 9 T field in applied fields of 2000 to -2000 Oe. Although wide open loops are seen right up to 9 T field, but with a positive background, which arise from paramagnetic contributions from Sm^{3+} , Fe^{2+} , and Co^{3+} spins.

The temperature dependence of electrical resistivity $\rho(T)$ in zero field is shown in main panel of Fig. 3 for $\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$ sample and inset of Fig. 3 shows the variation in resistivity under magnetic field $\rho(T)H$ up to 9 T field. At room temperature the resistivity value is 3.8 m Ω cm and it decreases with temperature as expected for metallic behavior. Finally at 14 K, the $\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$ sample becomes superconducting. The superconducting transition temperature being determined from resistivity measurements as $\rho=0$ is at 14 K; though the diamagnetic transition temperature (T_c^{dia}) is slightly higher at 15.5 K, see Fig. 2(a). This is because T_c^{dia} coincides roughly with the superconducting onset. The parent compound SmFeAsO does not show superconductivity and the same rather exhibits the SDW characteristic at around 140 K.¹⁴ $\rho(T)H$ measurements reveal that the resistive transitions for this superconducting composition shift to lower tempera-

tures by applying a magnetic field. The transition width for the sample becomes wider with increasing H . If we focus on the temperature at which resistivity becomes negligible ($\rho \rightarrow 0$) we find that rate of decrease in transition temperature with applied magnetic field is 1 K/T $\{dT_c/dH \sim 1$ K/T $\}$ for Co substituted $\text{SmFe}_{0.85}\text{Co}_{0.15}\text{AsO}$ sample. This value is far less than that of YBCO $\{dT_c/dH \sim 4$ K/T $\}$ and MgB_2 $\{dT_c/dH \sim 2$ K/T $\}$ samples; it suggests toward a high value of upper critical field (H_{c2}) in these compounds.¹⁵ We can say that oxypnictides are emerging as a new class of high field superconductors surpassing the H_{c2} of Nb_3Sn , and MgB_2 . In fact, FeAs seems surpassing the 100 T field benchmarks of the high- T_c cuprate superconductors.

We observed that by doping the magnetic ion Co^{3+} in the conducting Fe-As plane of SmFeAsO , the SDW state of Fe^{2+} is suppressed and superconductivity is induced by induction of mobile electron carriers in the system. This is unlike the cuprate superconductors, in which substitution of Cu with its neighbors in the periodic table (Ni, Zn, Co, Fe, etc.) in CuO_2 plane severely destroys the superconductivity.¹⁶

ACKNOWLEDGMENTS

The work was supported by Indo-Japan (DST-JSPS) bilateral exchange research program. V.P.S.A. further thanks NIMS for providing him with the MANA visiting scientist position for three months.

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