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## Effect of Co-doping on the resistivity and thermopower of $SmFe_{1-x}Co_xAsO$ (0.0 $\le x \le 0.3$ )

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We report structure, electrical resistivity and thermopower of polycrystalline SmFe<sub>1-x</sub> Co<sub>x</sub>AsO samples for  $0.0 \le x \le 0.3$ . The XRD data revealed full Co substitution at Fe-site with slight compression of the unit cell. Resistivity data showed that the spin-density wave observed at 130 K for x = 0 is suppressed when x = 0.05, above which superconductivity emerges due to injection of mobile electrons, supporting the substitution of Co<sup>3+</sup> at Fe<sup>2+</sup> site but disappears for x = 0.3. The thermopower (*S*) data indicate that the majority of charge carriers is electron-like and its value reaches  $-81 \mu$ V/K (at 300 K) for x = 0.3. Noticeable deviations from the expected linear behavior in *S*(*T*) at low temperatures and *S*/*T* against temperature curves at intermediate temperatures were observed. These observations were interpreted on the basis of nested Fermi surface and Umklapp processes. *Copyright 2012 Author(s). This article is distributed under a Creative Commons Attribution 3.0 Unported License*. [http://dx.doi.org/10.1063/1.4766936]

#### I. INTRODUCTION

Although cuprates have demonstrated wonders of high-temperature ( $T_c$ ) superconductivity by evading aniferrmagnetic Mott insulator state<sup>1</sup> way back in 1987,<sup>2</sup> the iron-based superconductors are known to be a class of glaring examples of coexisting superconductivity and magnetisms, known earlier to be impossible.<sup>3</sup> In the latter, both electronic correlations and multiorbital/ band effects are suggested to play important roles.<sup>4–10</sup> The parent compounds, LaFeAsO, exhibit structural phase transition<sup>11,12</sup> followed by a spin density wave (SDW) ordering at low temperatures.<sup>11,13</sup> Doping of electron/ hole into the FeAs layers suppresses the long-range SDW order leading to superconductivity.<sup>13,15</sup> This phenomenon is apparently analogous to that in cuprates, where superconductivity is induced by doping of charge carriers into an aniferrmagnetic Mott insulator.<sup>1</sup> Band structure calculations for *REOMAs* (*RE* = rare earth, *M* = Mn, Fe, Co and Ni)<sup>16</sup> suggest induction of electrons directly onto FeAs layers due to Fe-site doping with Co or Ni and hence inducing superconductivity. Later on, search for superconductivity started in similar compounds with complete substitution of Fe with other 3*d* metals such as Co.<sup>17–31</sup> Contrarily, the magnetic properties, including itinerant ferromagnetism<sup>17,26,27</sup> and interacting *RE*<sup>4f</sup> and Co<sup>3d</sup> moments<sup>28–32</sup> have been studied recently albeit the iso-structural *RE*CoAsO compounds do not superconduct.

In particular, the superconductivity with the maximum  $T_c = 17.2$  K for samarium oxypnictides (SmFe<sub>1-x</sub>Co<sub>x</sub>AsO) has been recently observed by doping a magnetic ion (Co) in the FeAs layers<sup>19–22</sup> for wide concentration range, although with lower than that of electron doping within *RE*-O charge reservoir.<sup>3,33–37</sup> Due to the Co-doping it is suggested that there is induced disorder in FeAs layers<sup>38,39</sup> and that there is possible reduction in the degree of nested Fermi surfaces (FS) of electrons and

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FIG. 1. Refined and observed XRD patterns with difference curves of  $SmFe_{1-x}Co_xAsO$  for x = 0.0 and 0.3.

holes cylinders.<sup>40</sup> In this context, there is poor understanding of the salient parameters governing the evolution of the physical properties. Particularly, thermopower is very sensitive to subtle changes in electronic structure due to doping, and can provide information on the ground state and band structure.

We have investigated the effects of Co doping (electron doping) on SmFeAsO to understand the evolution of physical properties. We found that the Co substitutes fully at Fe site in SmFeAsO in an iso-structural lattice with slightly compressed unit cell. Superconductivity emerges for Co concentrations x > 0.05 as substitution injects mobile electrons to the system. Our results suggest that nested FS and Umklapp processes play an important role for the observed anomalies in the transport properties.

#### **II. EXPERIMENTAL DETAILS**

Polycrystalline samples with SmFe<sub>1-x</sub>Co<sub>x</sub>AsO (x = 0.0, 0.1, 0.15, and 0.30) nominal compositions were synthesized by single step solid-state reaction method.<sup>24</sup> Stoichiometric amounts of Sm, Fe, As, Co<sub>3</sub>O<sub>4</sub> and Fe<sub>2</sub>O<sub>3</sub> were grounded thoroughly. It is to be noted that weighing and grinding was done in the glove box under high purity argon atmosphere. The powder was then palletized and vacuum-sealed in a quartz tube. The vacuum was of the order of  $10^{-4}$  Torr. Subsequently, this sealed quartz ampoule was placed in box furnace and heat-treated at 550 °C for 12 hours, 850 °C for 12 hours, and then finally at 1150 °C for 33 hours in continuum. Then furnace was allowed to cool naturally. The sintered sample was obtained by breaking the quartz tube. The obtained sample was black in color and a bit brittle, in fact it was nearly in powder form. The phase purity and structural identification were made via x-ray diffraction (XRD) using a Bruker D8 Advance X-ray diffractometer (Cu K $\alpha$  radiation). Electrical resistivity and thermopower of the samples were obtained by a standard dc four-terminal method and differential technique during the warming process using our homemade setups.<sup>41</sup> Seebeck coefficient of the Cu reference was subtracted to obtain the absolute values of the samples.

#### **III. RESULTS AND DISCUSSION**

#### A. X-ray diffraction

The room temperature XRD patterns for x = 0.0 and 0.30 as representative along with their Rietveld analysis are shown in Fig. 1. The structure of SmFe<sub>1-x</sub>Co<sub>x</sub> AsO at room temperature for all compositions of x was refined using least squares fitting with the tetragonal space group *P4/nmm* and the fitting parameters are listed in Table I. All main peaks can be well-indexed based on the space group *P4/nmm*, indicating that the samples are essentially single phase. Some extra peaks at

$SmFe_{1-x}Co_xAsO$ x =	a (Å)	b (Å)	c (Å)	Volume (Å)
0	3.9372(3)	3.9372(3)	8.4980(11)	131.73(2)
0.05	3.9403(3)	3.9403(3)	8.4823(10)	131.70(2)
0.1	3.9402(4)	3.9402(4)	8.4647(13)	131.41(3)
0.15	3.9391(2)	3.9391(2)	8.4672(6)	131.38(1)
0.3	3.9393(2)	3.9393(2)	8.4217(6)	130.69(1)

TABLE I. Crystallographic data of  $SmFe_{1-x}Co_xAsO$  with x = 0.0, 0.1, 0.15 and 0.3.



FIG. 2. Temperature dependence of the electrical resistivity of SmFe<sub>1-x</sub>Co<sub>x</sub>AsO. Inset shows the close up for  $x \ge 0.1$  samples. The line is a fit to  $\rho(T) = \rho_0 + AT^2$  below 100 K.

around 30° and 43° having very low intensity are also seen in the XRD pattern of pure SmFeAsO sample, which are due to the minor presence of SmAs. It is inferred from the Table I that the *c*-axis lattice constant decreases marginally with increasing *x*, without any significant change in *a or* b lattice parameter within experimental accuracy. The decrease in *c* lattice parameter suggests that the successful chemical substitution of Co in the Co<sup>3+</sup> state in place of Fe<sup>2+</sup>. Hence, incorporation of Co in the Fe-site reduces the cell volume marginally due to contraction of c parameter.

#### **B. Electrical resistivity**

Figure 2 shows the temperature dependence of the electrical resistivity  $\rho(T)$  of SmFe<sub>1-x</sub> Co<sub>x</sub>AsO samples for  $0.0 \le x \le 0.3$ . For pure SmFeAsO, the room temperature resistivity is found to be in below 2 m $\Omega$  cm, less than that of reported for other parent compounds,<sup>13,42-44</sup> indicating good quality of the sample. Further,  $\rho(T)$  increases slightly with decreasing temperature, local metallic maximum at about 130 K ( $T_0$ ), and then it increases rapidly with decreasing temperature further for parent sample. The present anomaly observed has most probably a similar physical origin to that observed in LaFeAsO and is perhaps associated with a structural phase transition and/or an aniferromagnetic SDW transition.<sup>11,13</sup> It is, thus, quite tempting to interpret the results as being due to the simultaneous occurrence of a structural and magnetic phase transition. For Co substitution at the Fe-site, the anomaly associated with  $T_0$  shifts to lower temperature (arrows in Fig. 2) finally disappearing for x > 0.1. We define the value of  $T_0$  as the mid point of upper and lower inflection points in the curve of the derivative of resistivity versus temperature and presented its evolution with Co concentration in Fig. 3. This has been attributed to a shift in the balance between the magnetic ordering and carrier concentration.<sup>45,46</sup>



FIG. 3. Doping dependence of superconducting transition temperature  $(T_c)$  and electrical resistivity anomaly  $(T_0)$  for SmFe<sub>1-x</sub> Co<sub>x</sub>AsO samples (left axis). S(300) is also shown for comparison (right axis).

Moreover,  $\rho(T)$  continues to increase at low temperature (well below  $T_0$ ) for x = 0 and 0.05 without any trace of superconductivity down to the temperature we measured. However, 10% Co doping sample begins to show superconductivity as evidenced by a sharp drop in  $\rho(T)$  at  $T_c = 10$  K (defined as the midpoint in the resistive transition) as shown in Fig. 2. Note that a weak anomaly maximum is still present (inset, Fig. 2), indicating the coexistence of magnetic fluctuations with superconductivity.<sup>25</sup> Further, superconducting transition temperature increases to the highest value  $T_c = 14.5$  for the sample with x = 0.15. No superconductivity has been observed with higher doping level and SmFe<sub>0.7</sub>Co<sub>0.3</sub>AsO is metallic. It should be noted that the observed  $T_c$  has been realized by suppressing the long-range SDW order through increasing carrier concentration.<sup>11,13</sup> Our samples showed  $T_c$  transition width somewhat broader, often observed in the polycrystalline samples due to the granular nature combined with the possible inhomogeneity of dopant ion distribution.<sup>24</sup> Fig. 3 presents the phase diagram representing the variation in  $T_0$  and  $T_c$  as a function of Co content x. Even though we have not studied large number of samples, nevertheless, the superconducting window forms its domelike regime that appears qualitatively similar to other reports.<sup>19,20</sup>

Shown in inset of Fig. 2 is the temperature dependence of resistivity up to 100 K for  $x \ge 0.1$ . Resistivity can be fitted by  $\rho = \rho_0 + AT^2$  in the temperature range of 5 < T < 50 K for x = 0.1. Here, the residual resistivity ( $\rho_0$ ) may arise from scattering due to grain boundaries and/or perturbations to the periodic potential felt by electrons, such as defects. On the other hand, samples with concentration x = 0.15 and 0.3 are found to follow  $T^2$  characteristic in a much broader temperature range. Therefore, it is instructive to mention that the electron-electron scattering rate should grow as  $T^2$  with increase in x. This shows that the present system is Fermi liquid (FL) system.<sup>14,47–49</sup>

#### C. Thermopower

The nature of the charge carriers, thus, in this material was probed with thermopower *S* as a function of temperature (Fig. 4). The room temperature value of thermopower ( $S_{300 \text{ K}}$ ) is found to be in the range of -36 to  $-81 \ \mu\text{V/K}$  and temperature variation nearly identical for all the samples with negative sign, indicating the majority charge carriers essentially are electrons. The temperature variation and absolute value of *S* are somewhat different from those of other Fe-based superconductors.<sup>19–21</sup> However, they are very much similar to those of usual metals suggesting perhaps that the preparation conditions maintained with these samples mimic the usual metals in their thermopower characteristics. At low temperatures, *S* exhibits a linear temperature variation (inset to the Fig. 4) followed by a dip at about 50 K, which is typical of the phonon-drag effect for undoped sample. Considering the value of Debye temperature ( $\theta_D$ ) from literature<sup>17</sup> about 342 K, the

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FIG. 4. Thermopower versus temperature for  $SmFe_{1-x}Co_xAsO$  samples as indicated. Inset shows expanded S(T)-T curves at low temperatures.



FIG. 5. Diffusion and phonon drag contributions of various  $SmFe_{1-x}Co_xAsO$  samples. *S/T* is found to be linear with  $1/T^2$  above 165 K.

appearance of the dip at 50 K is reasonable, as phonon-drag ( $S_g$ ) in metals leads to a prominent peak/ dip in  $S_g$  at  $\sim \theta_D / 5$ .<sup>50</sup> This is caused by a crossover between different phonon scattering mechanisms at higher and lower temperatures. The height of dip is substantially enhanced and shifted towards higher temperatures upon Co-doping. For normal semiconductor such a strong sensitivity of *S* on the doping is not expected, which however, for strongly correlated electron systems is not unusual.

In the high temperature regime, on the other hand, the magnitude of *S* increases monotonically as the temperature rises, indicating that the diffusion thermoelectric transport prevails. In this regime, conventional diffusion and phonon drag components are involved in *S* such that S = aT + b/T.<sup>50</sup> Here, *a* and *b* are diffusion coefficient and phonon drag coefficient, respectively. We have tried to fit our experimental results and found that *S/T* is linear with  $1/T^2$  above 170 K (Fig. 5). However, in the intermediate temperature regime, observed complicated variation in *S* could not be quantitatively explained by the simple FL picture. As mentioned above, we have seen that FL is a reasonable assumption at very low temperatures for standard FS, where the interaction between phonons is negligible. On the other hand, Umklapp processes become very frequent at high temperatures and topology of FS should be important.<sup>40,51</sup> Moreover, the band structure value of *S* for usual metals only 6.8  $\mu$ V/K is much smaller than the values observed here. Therefore, this scenario

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is believed to be due to Umklapp processes and nested Fermi Surfaces. The role of Umklapp processes gets additional support from the observation of sharp low temperature peak in the thermal conductivity.<sup>52,53</sup> Qualitatively such effect could be corroborated in Mott's picture while considering the energy dependence of the relaxation time ( $\tau$ ). It is sensitive to changes in band structure or FS topology and, hence, evolution of observed temperature dependence of Seebeck coefficient (Fig. 5) as the material passes through the phase-transition region.<sup>50</sup>

Spin entropy could be another likely source of the anomalously large thermopower in these FeAs-base superconductors as spin fluctuations prevails therein,<sup>12</sup> comparing well with the Na<sub>x</sub>CoO<sub>2</sub> system.<sup>54</sup> This indicates that the large absolute value of *S* can be realized when the thermal energy  $(k_{BT})$  is much less than the on-site Coulomb repulsion *U*. If we apply it to our data, the *S* could be approximately  $-k_B \ln 2/|e|$ . Here, the ln2 term comes from the spin fluctuations of Fe<sup>+2</sup> ions and estimated value of  $|S| \sim 60 \ \mu V/K$  could account for about 75 % of the observed  $S_{max}$ . The under estimation of *S* from spin entropy is quite obvious as one may expect extra contribution from nested Fermi surface. In fact, the spin entropy term of *S* may also be affected by the magnetic ordering, which could indeed limit the energetically allowable spin configurations of carriers due to the exchange interaction between the magnetic ions and the carriers themselves. In this context, room temperature thermopower S(300K) values are worth attention, which are increased with x (Fig. 3). This increase is against the possible main contribution from spin entropy as spin entropy drops as *x* increases.<sup>12</sup> Therefore, these peculiar thermopower features are attributed to nested FS and Umklapp processes.

The S(300 K) increase, and  $T_c$  evolution into a domelike feature as x increases (Fig. 3) show that the emergence of superconductivity is related to the thermopower. This correlation is consistent with the earlier reports on FeAs based superconductors<sup>19,20</sup> and on high- $T_c$  cuprates.<sup>55,56</sup> Thus, even with our limited data points, the universal feature of the correlation between  $T_c$  and thermopower in these iron-based arsenide superconductors could be established.

#### **IV. CONCLUSIONS**

In summary, we report structure, electrical resistivity and thermopower of polycrystalline  $SmFe_{1-x}Co_xAsO$  samples for  $0.0 \le x \le 3$ . XRD data revealed Co substitution at Fe site in an isostructural lattice with slightly compressed unit cell. The SDW observed in pure sample is suppressed on x = 0.05 Co-doping. Superconductivity prevails for 0.05 < x < 0.3 at which it got completely suppressed, suggesting for a direct consequences of Co substitution on Fe sites and also disorder in superconducting FeAs layer. The sign of thermopower indicate that the majority charge carriers are electron-like and its absolute value reaches as high as  $-81 \mu V/K$  (at 300 K) for x = 0.3. Behavior of S(T) is quite similar to the usual metals but with much larger magnitude, which is attributed to the nested Fermi Surface and Umklapp processes.

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