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2010 J. Phys.: Condens. Matter 22 115701

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Effects of Co doping on the transport properties and superconductivity in $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$

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Received 10 November 2009, in final form 13 January 2010

Published 5 March 2010

Online at stacks.iop.org/JPhysCM/22/115701

Abstract

A series of $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ oxyarsenide compounds with Co doping on iron sites ($x = 0-0.2$) have been synthesized by a solid state reaction method. The effects of Co doping on the electrical transport properties and superconductivity were analyzed with a special emphasis on the analysis of thermopower. Undoped CeFeAsO shows an electrical resistivity anomaly at about 150 K, which was ascribed to a spin-density-wave (SDW) instability. This anomaly is suppressed and a superconducting transition occurs at $T_c = 3.2$ K in $\text{CeFe}_{0.95}\text{Co}_{0.05}\text{AsO}$, the maximum superconducting transition temperature (T_c) of 12.5 K is observed in $\text{CeFe}_{0.90}\text{Co}_{0.10}\text{AsO}$, and the thermopower is increased by the Co doping. As has been previously suggested, the emergence of superconductivity seems to be closely linked to the thermopower, and there is a close correlation between T_c and the thermopower, both showing a similar dome-like doping dependence.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Much attention has been paid to the layered rare-earth iron oxypnictides LnFePnO ($\text{Ln} = \text{La, Pr, Ce, Sm}$; and $\text{Pn} = \text{P}$ and As) with the ZrCuSiAs tetragonal structure (space group: $P4/nmm$) since superconductivity was discovered at $T_c = 26$ K in the iron-based $\text{LaFeAsO}_{1-x}\text{F}_x$ ($x = 0.05-0.12$) [1]. The superconductivity was induced by partial substitution of oxygen by fluorine in the parent compound LaFeAsO . Undoped LaFeAsO itself is not superconducting but shows an anomaly near 150 K in both electrical resistivity and magnetic susceptibility. This anomaly has been shown to be caused by a spin-density-wave (SDW) instability. Electron doping by fluorine suppresses the SDW instability and yields the superconductivity [2, 3]. The crystal structure of LnFeAsO related materials consists of insulating $[\text{La}_2\text{O}_2]^{2+}$ layers and conducting $[\text{Fe}_2\text{As}_2]^{2-}$ layers [4]. The ionic LnO layers act as a charge reservoir that transfers electrons to the FeAs layer, while the FeAs layer acts as conducting layer [5] and is thought to be responsible for superconductivity. Indeed,

several other compounds with FeAs layers were found to be superconductors [6]. The superconductivity can be obtained by high pressure [7], isoelectronic impurity doping [8], hole doping [9] or by electron doping [10] through suppression of the orthorhombic distortion and the SDW magnetism. Electron doping can be induced in several ways: partial substitution of fluorine for oxygen [1], oxygen deficiency [11] or partial substitution of thorium for the rare-earth [12]. All these ways of inducing electron carriers into the system are limited to the substitution performed in the charge reservoir LnO layer. However, electron doping can also be induced directly by doping into FeAs layers [13]. Indeed, the superconductivity is not destroyed by the induced disorder in FeAs layers through the substitution of cobalt [14], nickel [15] and rhodium [16] for the Fe site. The superconductivity at $T_c = 11.3$ K for non-superconducting cerium oxypnictides (CeFeAsO) has been recently observed by doping a magnetic ion (Co) in the FeAs layers [17]. This indicated a major difference from cuprate superconductors, where any substitution in the conducting CuO_2 planes by other metal ions always destroys

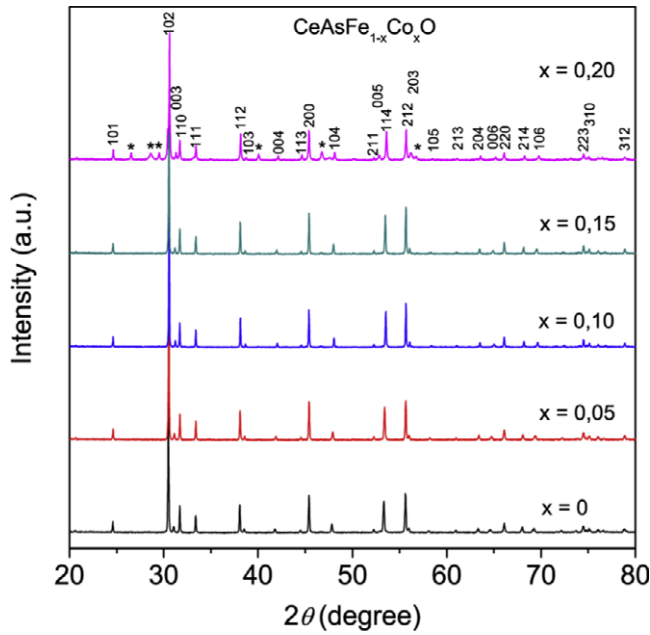


Figure 1. X-ray powder diffraction patterns for $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ samples.

superconductivity or lowers the transition temperature [18, 19]. Therefore, it is of great interest to explore the effects of Co doping on CeFeAsO . However, a previous report about Co doped CeFeAsO only provided limited information [17]. In this paper, we report on a systematic study about the influence of the Co content in $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ on the electrical transport properties.

2. Experimental details

Samples with the chemical composition $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ ($x = 0, 0.05, 0.10, 0.15$ and 0.20) were prepared by using a two-step solid state reaction route. In the first step, CeAs was prepared by heating Ce and As chunks in a sealed silica tube under argon, at 500°C for 15 h, followed by 800°C for 5 h, and then 900°C for 5 h, with a ramp-up speed of 50°C h^{-1} . The obtained alloy was single phase CeAs , as confirmed by x-ray diffraction measurements. Next, the CeAs alloy was thoroughly ground together with Fe_2O_3 powder, Fe powder and Co powder in stoichiometric amounts as given by the formula $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$. The thoroughly mixed powders were pressed under 250 MPa to form $2\text{ mm} \times 3\text{ mm} \times 12\text{ mm}$ bars. The bars were wrapped in tantalum foils and sealed in a silica tube under argon. In order to ensure the optimum reaction of the reactants, the pellets were reacted sequentially first at 500°C for 10 h and then 850°C for 5 h before a final annealing step at 1150°C for 50 h, with a ramp-up speed of 100°C h^{-1} . The sample preparation processes including weighing the raw materials, grinding powders and pressing pellets were carried out in a pure argon-filled glove box with less than 1 ppm O_2 and H_2O .

X-ray diffraction patterns were obtained using a Panalytical X'Pert with a $\text{Ge}(111)$ incident monochromator and

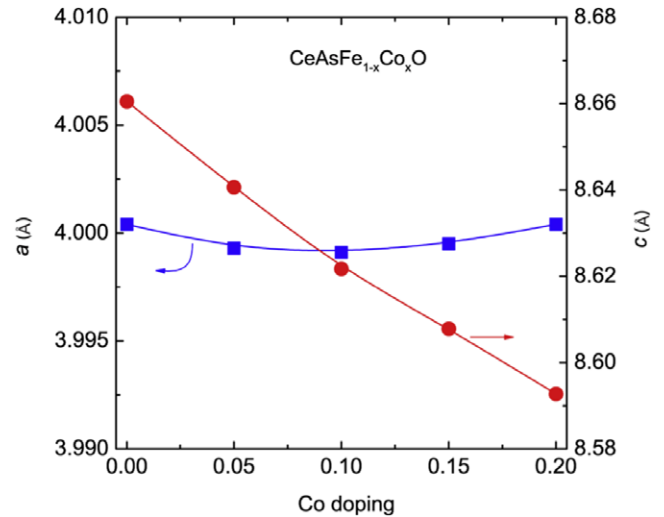


Figure 2. Lattice parameters as a function of Co doping content.

an X'celerator detector, with a rotating sample. Rietveld refinement was performed using FullProf software [20].

The thermoelectric power was measured by a method with two T-type thermocouples, by using the slope of the $\Delta V - \Delta T$ curve with gradients up to about 0.2 K mm^{-1} , in a closed cycle cryostat from 300 to about 20 K by using a laboratory made system. The resistivity was measured by a four-probe method from 2.5 to 300 K with magnetic fields up to 9 T using a Physical Property Measurement System (PPMS) from Quantum Design. All transport measurements were performed in a direction perpendicular to the pressing direction.

3. Results and discussion

Figure 1 shows the XRD patterns of the synthesized $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ samples. All the XRD peaks can be well indexed based on a tetragonal cell with space group $P4/nmm$, indicating that the samples are almost single phase. However, it should be noted that the sample with the largest amount of cobalt ($x = 0.20$) contains a few per cent of the impurity phases (Ce_2O_3 and CeO_2).

Figure 2 plots the lattice parameters as a function of Co doping content. The a axis remains nearly unchanged while the c axis shrinks significantly with increasing Co content. Thus the cell volume decreases almost linearly, which is related to the radius of the Co^{2+} ions being smaller than that of the Fe^{2+} ions. These changes of cell parameters, according to Vegard's law, suggest that Co atoms were successfully incorporated into the lattice. The shrinkage of the c axis is consistent with an increase of the density of negative charge in the FeAs layers induced by the Co doping, which leads to the strengthening of the interlayer Coulomb attraction. This lattice parameter evolution is different from the one observed by Prakash *et al* [17], who reported a simultaneous decrease of both a and c with increasing Co content. Nevertheless, our results are similar to the ones observed in Co doped $\text{LaFe}_{1-x}\text{Co}_x\text{AsO}$ and $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$ samples by two other groups [13, 14], and are consistent with the ionic radius in this system.

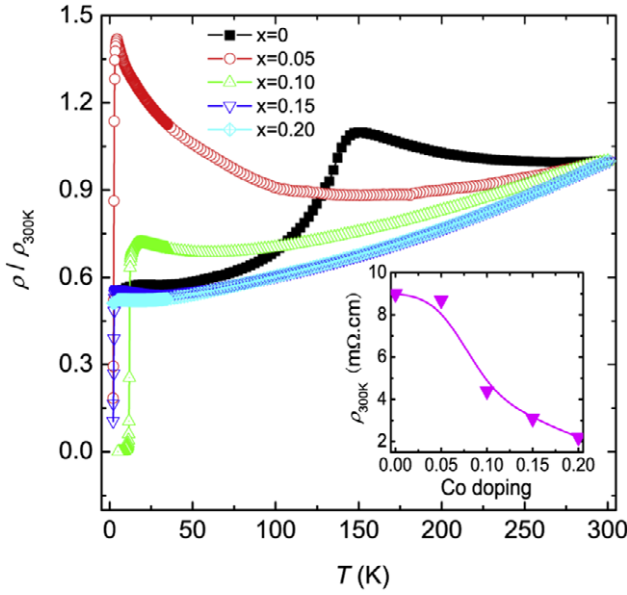


Figure 3. Temperature dependence of electrical resistivity (ρ) for $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ samples in zero field, the ρ data are normalized to $\rho_{300\text{K}}$. The inset is $\rho_{300\text{K}}$ as a function of Co doping content.

Figure 3 shows the temperature dependence of the electrical resistivity (ρ) in zero field. The resistivity data are normalized to $\rho_{300\text{K}}$, as the resistivity measured on polycrystalline samples is often higher than the intrinsic value, due to the grain boundaries and porosity effects. The resistivity for undoped CeFeAsO increases slightly with decreasing temperature, peaking at about 150 K, and then it decreases with decreasing temperature. The present resistivity anomaly in CeFeAsO has most probably a similar physical origin to that observed in LaFeAsO [21]. As reported before [22], this upturn in resistivity is probably associated with a structural phase transition and/or an antiferromagnetic SDW transition. It can also be considered as an increase of the charge-carrier scattering by lattice fluctuations related to the onset of the structural transition [12, 13]. This anomaly was not observed, and a superconducting transition occurred at $T_c = 3.2\text{K}$ for the sample with Co doping $x = 0.05$. This superconducting transition has been realized by suppressing the upturn in resistivity through increasing the number of carriers through Co doping (thermopower measurements indicate that the majority carriers are electrons, see later). It should be noted that the superconducting transition temperature increases to the highest value, $T_c = 12.5\text{K}$, for the sample with $x = 0.10$, and then decreases to $T_c = 2.9\text{K}$ with a further increase in the Co doping content to $x = 0.15$. No superconductivity has been observed with higher doping levels ($x = 0.2$) down to 2 K and $\text{CeFe}_{0.8}\text{Co}_{0.2}\text{AsO}$ is metallic. The highest T_c value (12.5 K), observed in $\text{CeFe}_{0.9}\text{Co}_{0.1}\text{AsO}$ in the present work, is a bit higher than the ($T_c = 11.3\text{K}$) reported by Prakash *et al* [17], even though both samples have the same nominal composition. This most probably originates from small differences in the actual composition for Co doping resulting from different heating processes. The inset of figure 3 shows the Co doping dependence of the electrical resistivity (ρ) at 300 K for $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$. The resistivity

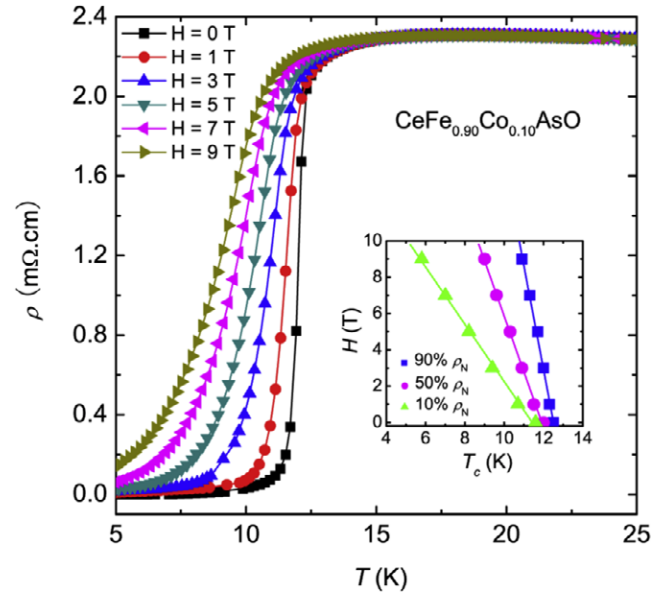


Figure 4. Temperature dependence of electrical resistivity (ρ) in different magnetic fields for $\text{CeFe}_{0.90}\text{Co}_{0.10}\text{AsO}$. The upper critical field H_{c2} is found from 10%, 50%, and 90% estimates of the normal state value ρ_N and plotted versus the critical temperature as an inset.

at 300 K decreases significantly with increasing Co doping content, which suggests that the carrier concentration has been increased by the Co doping.

The upper critical field H_{c2} is one of the important parameters to characterize superconductivity. To get information about the H_{c2} of the $\text{CeFe}_{0.90}\text{Co}_{0.10}\text{AsO}$ sample, we measured the electrical resistivity under selected magnetic fields up to 9 T. The in-field resistivity transitions for the sample $\text{CeFe}_{0.90}\text{Co}_{0.10}\text{AsO}$ are shown in figure 4. The transition shifts to lower temperatures on applying a magnetic field, and its width increases with increasing H , which is a characteristic of type-II superconductivity [13, 23]. The offset of the transition shifts by about 5 K in an applied field of 9 T, and the offset of transition shifts by about 3.5 K in an applied field of 5 T, which is similar to the reported values [17]. Here, we define a transition temperature $T_c(H)$ that satisfies the condition that $\rho(T_c, H)$ equals a fixed percentage of the normal state value (ρ_N) for each field H . The $T_c(H)$ values for $\rho = 10\%$, 50% , and 90% are shown in the inset of figure 4, which are represented by the upper critical field $H_{c2}(T)$. In all cases, we find that $H_{c2}(T)$ has a linear dependence, with no sign of saturation. The slope $(dH_{c2}/dT)|_{T=T_c} = -1.58\text{TK}^{-1}$ for $\rho_N = 10\%$, -3.06TK^{-1} for $\rho_N = 50\%$, and -5.73TK^{-1} for $\rho_N = 90\%$. The upper critical field $H_{c2}(0)$ values can be extrapolated using the Werthamer–Helfand–Hohenberg (WHH) formula, $H_{c2}(0) = -0.693T_c(dH_{c2}/dT)|_{T=T_c}$ [24]. $H_{c2}^{\text{WHH}}(0) = 12.7\text{T}$ for $\rho_N = 10\%$, 25.5T for $\rho_N = 50\%$, and 49.6T for $\rho_N = 90\%$. Here, it should be noted that this upper critical field (H_{c2}) of 49.6 T is of the same order as that (45.22 T) reported by Prakash *et al* with the same nominal composition [17]. This value is much lower than the one (94 T) observed in $\text{CeFeAsO}_{1-x}\text{F}_x$ [25], which may be correlated to the strong disorder induced by the substitution of Fe by Co in the conducting layer.

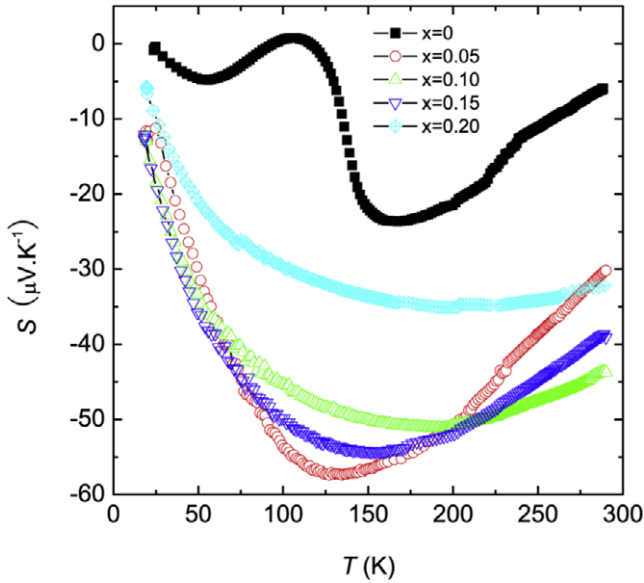


Figure 5. Temperature dependence of thermopower (S) for $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$.

Figure 5 shows the temperature dependence of thermopower (S) for $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ in the measurement temperature ranges from 20 to 300 K. The parent compound CeFeAsO exhibits a complex temperature dependence, which is similar to the LnFeAsO ($\text{Ln} = \text{La}, \text{Sm}$ and Nd) behavior reported previously [23, 26, 27]. The negative values of S indicate that, although oxypnictides are multiband materials, electrical transport properties in $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ are dominated by electrons. The steep upturn just below 150 K is associated with the structural phase transition and/or SDW. This upturn of S disappears just after increasing the Co doping content to $x = 0.05$. The room temperature $|S|$ values increase from $6.2 \mu\text{V K}^{-1}$ for CeFeAsO to $30.2 \mu\text{V K}^{-1}$ for $\text{CeFe}_{0.95}\text{Co}_{0.05}\text{AsO}$, peaks at $43.8 \mu\text{V K}^{-1}$ for $\text{CeFe}_{0.90}\text{Co}_{0.10}\text{AsO}$ and decreases for higher Co content. Since S can be considered as the macroscopic electric response of a conductor to the external temperature field, both the electrons and holes will give a contribution according to a two-band theory [28, 29]. For simplification, considering two bands with electron and hole conduction respectively, if the electrical conductivity contributions from the electrons and holes are represented by σ_e and σ_h , the total S can be given by

$$S = \frac{\sigma_h |S_h| - \sigma_e |S_e|}{\sigma_h + \sigma_e}, \quad (1)$$

where $|S_h|$ and $|S_e|$ represent the contributions from holes and electrons to the thermopower, respectively. Therefore, the relatively small S value for the parent compound is, to some extent, due to the compensation effect of electron and hole conduction. With electron doping, the hole contribution in equation (1) becomes even smaller, explaining the increase in $|S|$ with Co doping. In other words, the present thermopower and electrical resistivity measurement suggests that electrons are indeed introduced by Co doping through the substitution of Co for Fe (leading to the observed decrease in electrical resistivity), while the hole pockets are gradually suppressed

(leading to the initial increase in the absolute value of the thermopower). It should be noted that $\text{CeFe}_{0.95}\text{Co}_{0.05}\text{AsO}$ shows the maximum S value at about 125 K, and the maximum absolute S values decrease with increasing Co doping content. An anomalous peak of S has also been observed in LnFeAsO ($\text{Ln} = \text{La}, \text{Sm}$ and Nd) with F doping [27] and it has been suggested that this enhancement of S is correlated to the onset of superconductivity [14]. However, the origin of this maximum absolute of S value is unclear and the Co-doping dependence of the maximum value of S cannot be explained easily, due to the multiband nature of the electronic structure of these materials. Nevertheless some interesting relationships between S and T_c have been found in some superconductors. It is well established that there is a universal doping (hole concentration) dependence of T_c for high- T_c cuprates [30]. Furthermore, it has been found that there exists a close correlation between the room temperature thermopower, $S_{290\text{ K}}$, and the hole concentration, p , and thus a universal correlation between T_c and $S_{290\text{ K}}$ is observed [31, 32] even in the metallic region. As for the iron-based arsenide superconductor family, Wang *et al* [14] proposed a simple model assuming two distinct contributions for the thermopower of $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$. The first one, ($S_{300\text{ K}}^0$), is a classical diffusion contribution, which would be observed if the compounds were not superconducting. The second one, ($S_{300\text{ K}}^{T_c}$), is closely linked to the appearance of the superconductivity and can be obtained by subtracting the classical contribution to the total room temperature value, following $S_{300\text{ K}} = S_{300\text{ K}}^0 + S_{300\text{ K}}^{T_c}$. That ‘abnormal enhanced part’, $S_{300\text{ K}}^{T_c}$, only appears in the superconducting window. Therefore, it seems that a close correlation could exist between the superconducting state and $S_{300\text{ K}}^{T_c}$. The same group also reported a similar relationship between thermopower and T_c for $\text{SmFe}_{1-x}\text{Ni}_x\text{AsO}$ [15]. However, it is unclear why this relationship would be valid only using the thermopower at room temperature. In this paper, the thermopower at 175 K (above 155 K for the SDW transition), 300 K and 400 K (extrapolated values according to the slope of the curves around room temperature) were selected. Figure 6 shows the Co doping content dependence of thermopower (S) at different temperatures and critical temperature (T_c^{90}) for $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$. It can be seen that T_c exhibits something of a dome-like behavior and reaches a maximum at a Co content $x = 0.10$; the thermopower at different temperatures also show dome-like behavior. The thermopower values deviate from the straight dashed line (the classical diffusion contribution for thermopower with no superconductivity) only in the superconducting window, and show a dome-like doping dependence as does T_c . The emergence of superconductivity seems to be closely linked to the abnormally enhanced part of the thermopower, which is similar to the reports of $\text{SmFe}_{1-x}\text{Co}_x\text{AsO}$ [14], $\text{SmFe}_{1-x}\text{Ni}_x\text{AsO}$ [15], $\text{LaFe}_{1-x}\text{Zn}_x\text{AsO}_{0.9}\text{F}_{0.1}$ [33] and $\text{Sm}_{1-x}\text{Th}_x\text{FeAsO}_{1-y}\text{F}_y$ [34]. Whether the enhanced thermopower is related to strong electron correlations, magnetic fluctuations or SDW is an open issue. It would be an interesting issue whether such a correlation between T_c and thermopower is a universal feature for all the iron-based arsenide superconductors. Moreover,

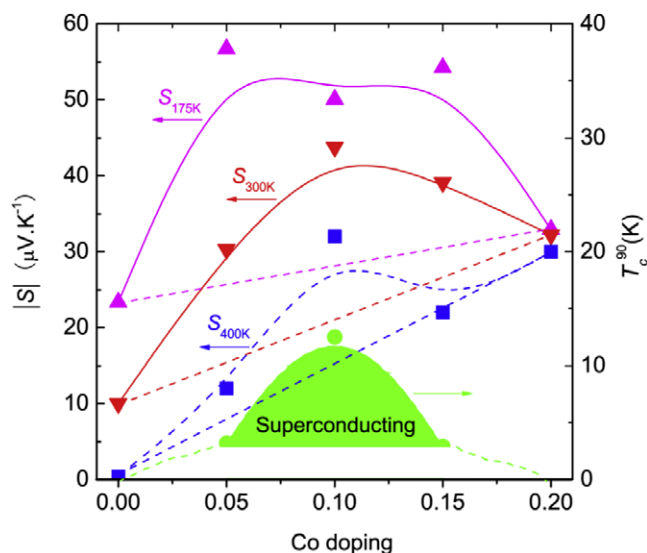


Figure 6. Co doping content dependence of thermopower (S) and critical temperature (T_c^{90}) for $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$. The thermopowers at 400 K are the extrapolated values according to the slope of the curves around room temperature. The straight dashed lines correspond to the normal part of the thermopower, which is proposed as the classical diffusion contribution for thermopower with no superconductivity. See the text for details.

further insight to the reasons for enhanced thermopower for all the iron-based arsenide superconductors may give a clue to the superconducting transport mechanism.

4. Conclusions

$\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ compounds with different Co doping content have been successfully synthesized by a solid state reaction method. The superconductivity was realized by Co doping in the FeAs plane of non-superconducting cerium oxypnictides (CeFeAsO). The superconducting transition occurs at $T_c = 3.2$ K in $\text{CeFe}_{0.95}\text{Co}_{0.05}\text{AsO}$ and the spin-density-wave instability is destroyed. The changes of superconducting temperature, resistivity and Co doping content indicated that the highest superconducting transition temperature can be obtained at an optimized carrier concentration, and the maximum superconducting transition temperature of 12.5 K and an upper critical field of 49.6 T have been observed in $\text{CeFe}_{0.90}\text{Co}_{0.10}\text{AsO}$. The abnormal enhanced thermopower only occurred in the superconducting window, which evidences a connection between thermopower enhancement and superconductivity in this system.

Acknowledgment

This work was supported by the *Triangle de la Physique* under project STP 2008-095T.

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