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Thermoelectric power of RFeAsO (R = Ce, Pr, Nd, Sm and Gd)

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1. Introduction

The recent discovery of superconductivity at 28 K in fluorine doped LaFeAsO [1], and the subsequent reports of superconducting transition temperatures even up to 50 K in some of the related rare-earth (R) materials [2-7], have stimulated a great interest in these rare earth oxypnictides. These compounds crystallize in the tetragonal phase (space group P4/nmm) at room temperature, and their structure is composed of two kinds of layers stacked along *c*-axis. One layer is composed of tetrahedra, centered by "O" with the R at vertices as a block, and in the other layer Fe is coordinated by As tetrahedron. While the R-O chemical bond is ionic. Fe-As has a predominantly covalent nature. The conductive carriers are confined two-dimensionally in the Fe-As layer causing strong interactions among the electrons in RFeAsO series of compounds [1,8]. Doping of O²⁻ by F⁻ provides an extra positive charge in the insulating layer (charge reservoir layer) and a negative charge in the conduction layer. Later, it has also been observed that superconductivity can even be introduced by hole doping [9] of the non-superconducting parent compounds RFeAsO as well. The superconductivity in such doped RFeAsO compounds appears to be unconventional [8,10]. An extensive work has been done on these doped compounds to have a clear understanding of the related physics as well as the effect of doping. However, studies of

ABSTRACT

Thermoelectric powers of a series of compounds RFeAsO (R = Ce, Pr, Nd, Sm and Gd) have been reported for temperatures ranging from 77 K up to room temperature. The behavior of S(T) in this temperature range can be divided into three regions. Every region has been fitted with mathematical functions of T. The physical significance of separate terms in the mathematical functions has been discussed. Some kind of universality has been observed between different members of the series.

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the physical properties of the un-doped parent compounds are also necessary to understand the superconductivity in the doped compounds.

One interesting observation in the un-doped parent compounds RFeAsO is the presence of long-range spin density wave. The spin density wave (SDW) is an antiferromagnetic (AFM) ground state for which the density of the conduction electron spins is spatially modulated, and is a many-particle phenomenon of an itinerant magnetism. An antiferromagnetic phase transition occurs at a lower temperature accompanied by a tetragonal to orthorhombic structural distortion on cooling down below 200 K. The magnetic structure of the oxypnictides within the *a*-*b* plane consists of chains of parallel Fe spins. These chains are coupled antiferromagnetically in the orthogonal direction, with an ordered moment less than 1 μ_B . This again suggests that such systems are itinerant with magnetism arising from a nesting-induced spin density wave (SDW) [11].

LaFeAsO shows an anomaly near T = 150 K both in the resistivity and in the magnetic susceptibility [12–14]. This is attributed to a spin-density-wave (SDW) instability. It has also been reported, that the system undergoes a structural phase transition from tetragonal to orthorhombic structure on cooling below 155 K. The structural distortion has been suggested to occur over a wide range of temperature starting from 200 K, and sufficient distortion takes place around 160 K. This distortion results in carrier localization and local moment formation on the Fe atoms. These local moments order antiferromagnetically near 145 K [15]. Local probe



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measurements of the magnetic properties of LaFeAsO by ⁵⁷Fe Mossbauer spectroscopy and muon-spin relaxation, in absence of external field suggest a static magnetic order below $T_N = 138$ K with a strongly reduced ordered moment at the Fe site [14]. Assuming that the full sample volume is contributing to the magnetic scattering an ordered moment of $\sim 0.35 \ \mu_B$ is predicted from the weak superlattice reflections in powder neutron diffraction [15]. Neutron scattering study of CeFeAsO [16] suggests that CeFe-AsO undergoes a structural lattice distortion from tetragonal to orthorhombic structure near 155 K followed by a commensurate AFM ordering on the Fe sublattice below \sim 140 K. Neutron scattering studies of PrFeAsO suggest antiferromagnetic ordering below 127 K, with an ordered moment of $0.48(9) \mu_B$ [17]. This iron magnetic ordering occurs below the structural transition, that takes place around 153 K. The magnetic moments on the Pr sites are also antiferromagnetically ordered below 14 K, similar to the parent compounds of the other rare earth FeAs-based superconductors such as CeFeAsO and NdFeAsO [17]. Polarized neutron diffraction measurements in NdFeAsO suggest the antiferromagnetic ordering below 141(6) K, with an ordered moment of 0.25(7) $\mu_{\rm B}$ [11]. However, there are also reports of the absence of the SDW antiferromagnetic order of the type discovered in LaFeAsO, along with the occurrence of the structural transition at 150 K [18]. Rietveld refinement [19] of high resolution synchrotron powder diffraction data on SmFeAsO collected at 300 K and 100 K suggests that, it crystallizes in the tetragonal phase at 300 K and in the orthorhombic phase at 100 K. Both resistive and magnetic measurements reveal that the tetragonal to orthorhombic phase transition is located at $T \sim 140$ K. GdFeAsO is a less studied system in which role of oxygen deficiency [20] in facilitating superconductivity has also been proved.

We have measured systematically the thermopower of a number of parent compounds RFeAsO (R = Ce, Pr, Nd, Sm and Gd) over a range (77–300 K) of temperature in which AFM ordering and structural distortion have been reported for these compounds. Among various transport properties, thermopower is a simple and sensitive one for analyzing charge carrier dynamics. Thermopower can predict the nature of the charge carriers, and is also sensitive to local moments. Moreover, as the thermoelectric response is driven by the temperature gradient rather than by the electric field, it is insensitive to grain boundaries [21].

2. Experimental

The samples were prepared by the solid-state reaction route using the elements R (Ce, Pr, Nd, Sm and Gd), Fe, Fe₂O₃ and As. Firstly, RAs were prepared by taking stoichiometric amounts of R (99.99%) and As (99.99%) chips in 1:1 ratio, pressed into pellets and sealed in evacuated quartz tube. With repeated heat treatment, attaining a maximum temperature of 900 °C, and grinding inside a glove box filled with inert Ar gas, single phase RAs were obtained. RAs were then mixed thoroughly with Fe₂O₃ powder (purity 99.995%) and Fe powder in stoichiometry and pressed into pellets. The pellets were wrapped with Ta foil and sealed in an evacuated quartz tube. They were then annealed at 1150-1200 °C for 40-45 h to obtain the final samples of RFeAsO (R = Ce, Pr, Nd and Sm). In case of R = Gd, stoichiometric amounts of GdAs, Gd₂O₃, FeAs and Fe were used. FeAs was prepared using Fe powder (99.99%) and As chips (99.99%) mixed in 1:1 ratio by the procedure similar to that of RAs. The phase purity of each sample at room temperature was checked by powder X-ray diffraction (XRD) method with Cu Ka radiation. XRD patterns of all the compounds suggest tetragonal ZrCuSiAs-type structure at room temperature with space group P4/nmm. The lattice parameters of the samples are summarized in Table 1.

Table 1

Lattice parameters of RFeAsO obtained from XRD measurements.

Sample	a (Å)	<i>c</i> (Å)
CeFeAsO	3.997	8.652
PrFeAsO	3.977	8.606
NdFeAsO	3.961	8.572
SmFeAsO	3.939	8.499
GdFeAsO	3.908	8.446

The thermoelectric power of each sample has been measured using a differential technique where a temperature gradient is created across the sample. The voltage developed (ΔE) between the hot and cold ends is measured. In our apparatus, we have used two heaters (one at the top and the other at the bottom of the sample) so that either end of the sample may be heated with respect to the other. The temperature difference ΔT between two ends of the sample is kept in the range $\sim \pm 0.5$ K throughout the measured temperature range (77–300 K). The temperature as well as the temperature difference across the sample were measured with Chromel–Alumel thermocouples. The sample temperature is controlled by a manganin heater, and monitored by a Si-diode sensor. At a particular temperature the values of ΔE and ΔT were recorded and TEP (S) of the sample is calculated from the relation $\Delta E / \Delta T = (S_{cu} - S)/(S_{chromel} - S_{alumel})$.

3. Results and discussion

Fig. 1 shows thermopower *S* as a function of temperature for the parent compound CeFeAsO. In the higher temperature region (200–300 K), *S* is negative, and its value increases linearly as a function of temperature. This negative value suggests that the electrons dominate in the electrical conduction. It may be mentioned that negative Hall coefficient has also been observed in SmFeAsO [22]. A linear dependence of *S* on *T* is a signature of metallic behavior. Thus in contrast to the Cuprates, parent compound is not a Mott–Hubbard insulator but a metal. This can be explained within the framework of the nearly free electron (NFE) model [22]. In the absence of magnetic ordering, thermoelectric power consists of two components, viz., "diffusion component" (*S*_d) and "phonondrag component (*S*_g). *S*_d is associated with only the electron gas and its dispersion whereas *S*_g is related with the phonon dispersion and the electron–phonon interaction. NFE model [22] gives an



Fig. 1. Thermoelectric power S as a function of temperature for CeFeAsO.

expression of the characteristic diffusion thermopower of a metal as

$$S(T) = \frac{\pi^2 k_{\rm B}^2 T}{3e} \left[\frac{N(E_{\rm F})}{n} + \frac{1}{\tau} \frac{d\tau}{dE_{\rm F}} \right]_{E=E_{\rm F}}$$
(1)

where $k_{\rm B}$ is the Boltzmann's constant, $E_{\rm F}$ is the Fermi energy, N(E) is the density of states, τ is the relaxation time and e is the charge of the carriers. The second term contributes mainly below the transition where it is related with the changes in mobility of the carriers with doping and temperature. In the absence of any phase transition, the contribution from the second term can be neglected. The first term scales as $1/E_{\rm F}$, and we have

$$S(T) = \frac{\pi^2 k_{\rm B}^2 T}{3eE_{\rm F}} \tag{2}$$

Thus S(T) is a linear function of T for such a metal, and the slope can give an estimate for the Fermi energy $E_{\rm F}$. For CeFeAsO the estimated $E_{\rm F}$ is 0.19 eV.

The structural distortion in these compounds starts around 200 K, where S(T) deviates from its linear behavior. Then it reaches a minimum (T_{min}) around 170 K. Similar feature of minima in S(T) curve has also been reported recently in (Nd/Sm)FeAsO polycrys-



Fig. 2. Thermoelectric power S as a function of temperature for LaFeAsO [24].



Fig. 3. Thermoelectric power S as a function of temperature for PrFeAsO.



Fig. 4. Thermoelectric power S as a function of temperature for NdFeAsO.



Fig. 5. Thermoelectric power S as a function of temperature for SmFeAsO.



Fig. 6. Thermoelectric power S as a function of temperature for GdFeAsO.

talline samples [23]. The existence of this T_{min} might be considered as a signature of the development of antiferromagnetic correlation

Table 2

 T_{min} and values of different coefficients obtained by fitting thermopower curves of RFeAsO for different temperature ranges using Eq. (4).

System	Parameters obtained from literature	Temperature range and T-dependence of $S(T)$	<i>T_{min}</i> and values of different coefficients
CeFeAsO	AFM ordering below 140 K	77–134 K	<i>T_{min}</i> = 172 K
	Structural transition at 155 K	$\downarrow \\ S(T) = S_0 + S_1 T + S_2 T^2 + S_4 T^{4_a}$	$S_0 = 13.25 \pm 0.05,$ $S_1 = -0.49600,$ $S_2 = 0.00650,$ $S_4 = -1.989 \times 10^{-7}$
		135–172 K ↓	$S_0 = 2276.92 \pm 0.06,$ $S_1 = -37.255,$
		$S(1) = S_0 + S_1 1 + S_2 1^2 + S_4 1^{-4}$ 200–300 K \rightarrow	$S_2 = 0.17000,$ $S_4 = -1.050 \times 10^{-6}$ S = -45.53 + 0.132T
LaFeAsO [24]	AFM ordering below 138 K	77–135 K	$T_{min} = 169 \text{ K}$
	Structural transition at 156 K	$\downarrow \\ S(T) = S_0 + S_1 T + S_2 T^2 + S_4 T^{4a}$	$S_0 = -42.34 \pm 0.06,$ $S_1 = -0.07300,$ $S_2 = 0.00650,$ $S_4 = -2.285 \times 10^{-7}$
		138–169 K ↓ $S(T) = S_0 + S_1T + S_2T^2 + S_4T^{4a}$	$S_0 = 3672.87 \pm 0.16,$ $S_1 = -58.045,$ $S_2 = 0.25515,$
		200–304 K →	$S_4 = -1.456 \times 10^{-6}$ S = -72.03 + 0.188T
PrFeAsO	AFM ordering below 127 K	77–134 K	<i>T_{min}</i> = 178 K
	Structural transition at 153 K	$\overset{\downarrow}{S(T)} = S_0 + S_1 T + S_2 T^2 + S_4 T^{4a}$	$S_0 = -17.93 \pm 0.04,$ $S_1 = -0.21500,$ $S_2 = 0.00650,$ $S_4 = -2.168 \times 10^{-7}$
		137–173 K ↓ $S(T) = S_0 + S_1T + S_2T^2 + S_4T^{4a}$	$S_0 = 1624.04 \pm 0.07,$ $S_1 = -24.870,$ $S_2 = 0.10550,$ $S_1 = -5.647 \times 10^{-7}$
		200–307 K →	$S_4 = -5.547 \times 10$ S = -55.5 + 0.142T
NdFeAsO	AFM ordering below 141 K	77–134 K	<i>T_{min}</i> = 177 K
	Structural transition at 150 K	$\hat{S(T)} = S_0 + S_1 T + S_2 T^2 + S_4 T^{4a}$	$S_0 = -10.22 \pm 0.01,$ $S_1 = -0.33700,$ $S_2 = 0.00650,$ $S_4 = -2.024 \times 10^{-7}$
		135–177 K ↓ $S(T) = S_0 + S_1T + S_2T^2 + S_4T^{4a}$ 200–307 K →	$\begin{split} S_0 &= 1614.29 \pm 0.05, S_1 = -26.0, \\ S_2 &= 0.11640, \\ S_4 &= -6.925 \times 10^{-7} \\ S &= -39.56 + 0.0996T \end{split}$
SmFeAsO	Resistivity anomaly around <i>T</i> = 135–140 K	77–133 K	<i>T_{min}</i> = 174 K
	Drop in electron density below 150 K	$\downarrow \\ S(T) = S_0 + S_1 T + S_2 T^2 + S_4 T^{4a}$	$S_0 = 12.10 \pm 0.04,$ $S_1 = -0.42560,$ $S_2 = 0.00650,$ $S_4 = -2.314 \times 10^{-7}$
		134–173 К	$S_0 = 1240.34 \pm 0.06$,
		$\downarrow \\ S(T) = S_0 + S_1 T + S_2 T^2 + S_4 T^{4a}$	$S_1 = -20.184,$ $S_2 = 0.09120,$ $S_4 = -5.550 \times 10^{-7}$
		200–303 K→	S = -38.66 + 0.0989T
GdFeAsO		77–123 K	<i>T_{min}</i> = 177 K
		$\overset{\vee}{S(T)} = S_0 + S_1 T + S_2 T^2 + S_4 T^{4a}$	$S_0 = -6.54 \pm 0.03,$ $S_1 = -0.28032,$ $S_2 = 0.00650,$
		126–172 K	$S_4 = -2.673 \times 10^{-7}$ $S_0 = 722.10 \pm 0.05$,
		$\downarrow \\ S(T) = S_0 + S_1 T + S_2 T^2 + S_4 T^{4a}$	$S_1 = -12.569,$ $S_2 = 0.06050,$
		206–304 K →	$S_4 = -4.177 \times 10^{-7}$ S = -27.92 + 0.066T

 $^a~S_0,\,S_1,\,S_2$ and S_4 are expressed in $\mu V~K^{-1},\,\mu V~K^{-2},\,\mu V~K^{-3}$ and $\mu V~K^{-4},$ respectively.

[23]. In the region below this minimum *S* increases with lowering of temperature and finally crossing the zero-line it shows a peak (T_{peak}) around 100 K.

This can be compared with the previous measurement of S(T) in LaFeAsO (Fig. 2) [24], in which they observed *S* to be negative over the entire temperature range. Besides this, |S| has a similar overall behavior in the temperature range (77–300 K) of measurement. The thermopower as a function of temperature shows similar overall behavior for all our measured samples (RFeAsO; R = Ce, Pr, Nd, Sm and Gd). The details are given in Figs. 1–6 and Table 2.

Below T_{min} the absolute value of *S* decreases. A similar decrease in |*S*| along with a decrease in the carrier density '*n*' is observed below about 155 K in LaFeAsO [24]. They suggest, that the observed decrease in both |*S*| and *n* through the transition indicates, that the second term in Eq. (1) is dominant in this temperature regime. In fact, the charge carrier scattering mechanism is changed significantly as the material passes through the phase transition region, and suggests reduction of electron–phonon interactions in the orthorhombic phase. Finally, we will see that the electron–electron scattering plays the dominant role below T_{min} . Further evidence of such reduction has been obtained from the analysis of carrier mobility and thermal conductivity. This may be considered as an evidence of strong electron–phonon coupling in such systems, which are metals with conduction dominated by electrons and with no local magnetic moment at high temperatures [24].

After crossing the zero line *S* becomes positive. This can be explained by the appearance of hole-like conduction in addition to electron like conduction. This supports the multi-band nature of the oxypinctides. For two bands, one electron- and the other hole-like, *S* becomes:

$$S = \frac{(\sigma_H | S_H | - \sigma_e | S_e |)}{(\sigma_H + \sigma_e)}$$
(3)

where $\sigma_e(h)$ and $S_e(h)$ are the contributions of electrons (holes) to the electrical conductivity and Seebeck coefficient, respectively.

At around 130 K, S(T) suffers a distinct change in slope. This may be associated with the transition to SDW state. In the region (130– T_{min} K), as well as in the temperature range (77–130 K), the behavior of S(T) can be well described by the expression

$$S(T) = S_0 + S_1 T + S_2 T^2 + S_4 T^4$$
(4)

In the temperature range (130– T_{min} K), S_1 , the coefficient of the linear term becomes large and negative. For our measured samples (R = Ce, Pr, Nd, Sm and Gd), it is minimum for Gd ($-12.569 \ \mu V \ K^{-2}$) and maximum for Ce ($-37.255 \ \mu V \ K^{-2}$). Being negative its absolute value again decreases below 130 K.

Electron–electron scattering in metals shows a T^2 dependence of S(T) [25]. From (77– T_{min} K), we observe a T^2 dependence of S(T). In the region (130– T_{min} K) the value of S_2 is minimum for Gd $(0.0605 \ \mu V \ K^{-3})$, and maximum for Ce $(0.17 \ \mu V \ K^{-3})$. However, below 130 K the coefficient S₂ reduces drastically, and its value remains same $(0.0065 \,\mu\text{V}\,\text{K}^{-3})$ for all the samples. This indicates that below T_{min} electron–electron scattering plays a significant role in these systems, and the scattering reduces drastically below 130 K. This reduction in electron-electron scattering suggests the enhanced mobility of the carriers. The undoped SmFeAsO exhibits a pronounced anomaly in the resistivity [22] behavior around T_{SDW} \sim 135–140 K. The Hall mobility of SmFeAsO has been found to increase by two orders of magnitude below T_{SDW} (135–140 K), the temperature at which an anomaly in resistivity has been observed and has been associated with the SDW transition. A similar behavior has also been observed in LaFeAsO [24]. Below T_{SDW} charge carriers, which are gapped out, disappear and the mobility of the carriers that do not condensate in the SDW state abruptly increases. This is also an indication of the fact that electron-electron scattering is the mechanism that mainly limits the carrier mobility above the SDW transition [22].

For SmFeAsO, an anomaly in molar susceptibility has also been observed in this temperature region 135–140 K [26]. These anomalies suggest an intimate coupling between the structural phase transition and the electronic as well as magnetic properties of the system. For LaFeAsO [14], a sharp decrease of χ near the structural transition (around 160 K) has been suggested as a signature of the enhancement of antiferromagnetic correlations. This is reflected in *S*(*T*) behavior by the *S*₄ term. This term may be associated [27] with the spin-wave fluctuation in the higher temperature region associated with antiferromagnetic correlations and shows the strong impact of structural transition on magnetism. This *S*₄ also reduces significantly below 130 K.

4. Conclusions

In summary, the qualitative behavior of S(T) is the same for all studied RFeAsO (R = Ce, Pr, Nd, Sm and Gd). In the temperature range (200–300 K) all the samples show metallic behavior with $S(T) = S_0 + S_1T$. S_1 is in the vicinity of 0.1 μ V K⁻² suggesting an E_F of about 0.25 eV. All the samples show a minimum in S(T) around $T_{min} = 170-180$ K and a change in slope around 130 K. The region below T_{min} is described by $S(T) = S_0 + S_1T + S_2T^2 + S_4T_1^4$. Strong electron–electron scattering plays an important role in the systems, and we observe a reduction in that scattering below 130 K, that is consistent with SDW picture. Spin fluctuations associated with anti-ferromagnetic correlations give S_4 term in the expression of S(T).

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