Resistivity saturation in PrFeAsO$_{1-x}$F$_y$ superconductor: An evidence of strong electron-phonon coupling

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Abstract

We have measured the resistivity of PrFeAsO$_{1-x}$F$_y$ samples over a wide range of temperature in order to elucidate the role of electron-phonon interaction on normal- and superconducting-state properties. The linear T dependence of ρ above 170 K followed by a saturationlike behavior at higher temperature is a clear signature of strong electron-phonon coupling. From the analysis of T dependence of ρ, we have estimated several normal-state parameters useful for understanding the origin of superconductivity in this system. Our results suggest that Fe-based oxypnictides are phonon mediated BCS superconductors like Chevrel phases and A15 compounds.

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I. INTRODUCTION

The discovery of iron-based layered oxypnictide $LaFeAsO_{1-x}F_x$ with high superconducting transition temperature (T_c) 26 K has stimulated intense experimental and theoretical activities in the field of superconductivity [1, 2]. Following the initial report, attempts have been made to enhance T_c mainly by changing the sample composition. It has been shown that T_c increases significantly when La is replaced by other rare earth elements (R) of smaller ionic size such as Ce, Pr, Nd, Sm, etc [3, 4, 5, 6, 7]. T_c as high as 55 K has been reached in this class of materials, which is the highest after cuprate superconductors. The stoichiometric compound RFeAsO is a nonsuperconducting metal, undergoes a structural phase transition around T_s=150 K and exhibits a long-range antiferromagnetic ordering of Fe moments slightly below T_s[1, 8]. Partial substitution of fluorine for oxygen or creating oxygen vacancy suppresses both structural and magnetic phase transitions, and drives the system to a superconducting ground state. Besides high transition temperature, this system exhibits many interesting phenomena possibly due to its layered structure as in the case of high-T_c cuprates and the presence of iron. At room-temperature, the crystal structure of RFeAsO is tetragonal with space group $P4/nmm$ and it consists of RO and FeAs layers which are stacked along the c axis. In spite of high transition temperature and layered structure, there are some important differences with respect to cuprate superconductors. For oxypnictides, the parent compound is not a Mott insulator and the superconducting-state properties are insensitive to oxygen or fluorine content over a wide range. Another important difference between high-T_c cuprates and oxypnictides is the dependence of T_c on the ionic size of rare earth ion. Unlike high-T_c cuprates, the present system shows that T_c increases with the decrease of ionic radius of rare earth ion [7]. Moreover, the symmetry and temperature dependence of gap in these two systems are not same. The gap function of the latter is s-wave and is consistent with the BCS theory [9, 10].

Several theoretical and experimental attempts aimed at identifying the possible superconducting mechanism in oxypnictides have been made [9, 10, 11, 12, 13, 14, 15, 16, 17]. The high transition temperature, structural similarities with cuprates and the occurrence of superconductivity on the verge of a ferromagnetic instability led to speculate that the superconductivity in this system is unconventional. Most of the theoretical reports suggest

that superconductivity in oxypnictides is not mediated by phonon [12, 13, 14, 15, 16]. Boeri *et al.* [15] calculated the value of electron-phonon (e-ph) coupling constant λ_{tr}=0.21 for pure LaFeAsO using the standard Migdal-Eliashberg theory and concluded that λ_{tr} should be even smaller for the fluorine-doped superconducting samples. According to them, the maximum value of T_c due to the e-ph coupling should not be above 0.8 K and a 5-6 times larger coupling constant would be needed to reproduce the experimental value of T_c in this system. In contrast to this, Eschrig [17] argued in favor of strong e-ph coupling of the Fe inplane breathing mode for the high transition temperature in this system. Chen *et al.* [9] from the Andreev reflection spectroscopy and Kondo *et al.* [10] from the angle resolved photoemission spectroscopy, have shown s-wave symmetry of the order parameter as in the case of phonon mediated conventional superconductors. The temperature dependence of energy gap is also consistent with the BCS theory. To shed some light on the mechanism of superconductivity, we have measured the temperature dependence of resistivity up to 550 K for PrFeAsO$_{1-x}$F$_y$ samples. From the analysis of high-temperature resistivity, the e-ph coupling strength for the superconducting and nonsuperconducting samples has been estimated. Indeed, we observe that λ_{tr} is quite small (0.24) for the nonsuperconducting sample in accordance with the theoretical prediction but it is 5-6 times larger for the superconducting samples.

II. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUES

Good quality single phase samples are necessary for understanding the normal-state transport properties and the mechanism of superconductivity in this system. Usually, two methods have been followed for the preparation of iron-based oxypnictides. Using high-pressure synthesis technique, one can obtain fluorine-free and oxygen-deficient RFeAsO$_{1-\delta}$ superconducting samples [18, 19]. However, samples prepared in this method contain appreciable amount of impurity phases resulting from the unreacted ingredients. In the other method, oxygen is partially replaced by fluorine and the sample is prepared either in high vacuum or in the presence of inert gas. Though samples prepared in this method are superior in quality, they are not always free from impurity phases [20, 21]. We use a slightly different method for the preparation of superconducting PrFeAsO$_{1-x}$F$_y$ samples. The nominal composition for these samples is oxygen deficient and at the same time oxygen

is partially replaced by a small amount of fluorine ($x \neq y \neq 0$). We observe that single phase samples with $x \leq 0.4$ can be prepared in this method. Polycrystalline samples of nominal compositions $PrFeAsO$, $PrFeAsO_{0.6}F_{0.12}$ and $PrFeAsO_{0.7}F_{0.12}$ were prepared by conventional solid-state reaction method. High purity chemicals Pr (99.9%), Fe (99.998%), Fe_2O_3 (99.99%), As (99.999%), PrF_3 (99.9%) and Pr_6O_{11} (99.99%) from Alfa-Aesar were used for the sample preparation. Finely grounded powders of $Pr_{0.96}As$, Fe, Fe_2O_3, Pr_6O_{11} (pre-heated at 600 ^oC) and PrF_3 were thoroughly mixed in appropriate ratios and then pressed into pellets. The pellets were wrapped with Ta foil and sealed in an evacuated quartz tube. They were then annealed at 1250 ^oC for 36 h. $Pr_{0.96}As$ was obtained by slowly reacting Pr chips and As pieces first at 850 ^oC for 24 h and then at 950 ^oC for another 24 h in an evacuated quartz tube. The product was reground, pressed into pellets and then sealed again in a quartz tube and heated at 1150 ^oC for about 24 h. The phase purity and the room-temperature structural parameters were determined by powder x-ray diffraction (XRD) method with Cu K_α radiation. dc magnetization and electrical resistivity measurements up to 300 K were done using a Quantum Design Physical Property Measurement System (PPMS). High temperature resistivity above 300 K was measured in a home-made set-up. Resistivity was measured by standard four-probe technique. In order to avoid oxidation, resistivity measurements at high temperatures were done in vacuum.

III. EXPERIMENTAL RESULTS AND DISCUSSION
A. Powder x-ray diffraction analysis

Figure 1 shows the XRD pattern for the fluorine-doped $PrFeAsO_{0.6}F_{0.12}$ sample at room temperature. No impurity phase was observed for $x=0.3$ and 0.4 samples. The diffraction pattern can be well indexed on the basis of tetragonal ZrCuSiAs-type structure with the space group P4/nmm. The lattice parameters obtained from the Rietveld refinements are $a=3.9711$ $\mathring{A}$ and $c=8.5815$ $\mathring{A}$. These values of lattice parameters are comparable with those reported for fluorine-doped and oxygen-deficient PrFeAsO samples [5, 18]. As expected, the lattice parameters of the present samples are located between $CeFeAsO_{1-x}F_x$ and $NdFeAsO_{1-x}F_x$ [4, 19]. The deduced bond lengths are d(Pr-O/F)=2.3405, d(Pr-As)=3.3008, d(As-Fe)=2.3828, and d(Fe-Fe)=2.8081 $\mathring{A}$. The values of several bond angles

are $\langle$O/F-Pr-As$\rangle$=76.57, $\langle$O/F-Pr-O/F$\rangle$=73.73, $\langle$Pr-As-Fe$\rangle$=77.83, $\langle$Fe-As-Fe$\rangle$=72.20, and $\langle$As-Fe-As$\rangle$=107.79^o. We would like to mention here that the Fe-Fe distance in the present system is about 1.4 % smaller than the Fe-Fe distance 2.8481 $\mathring{A}$ in LaFeAsO$_{1-x}$F$_x$ [20].

B. Electrical resistivity analysis

The measured temperature dependence of resistivity for PrFeAsO$_{0.6}$F$_{0.12}$ (S1) and PrFeAsO$_{0.7}$F$_{0.12}$ (S2) samples in the temperature range of 45-550 K is illustrated in Fig. 2(a). For both the samples, ρ decreases initially at a very slow rate and then at a faster rate with decreasing temperature until the superconducting onset temperature ($T_c^{on} \sim$50 K) is reached. Below T_c^{on}, ρ drops sharply and becomes zero at around 47 K. The inset displays the enlarged view of the onset of superconductivity for sample S1. For this sample, the transition width ΔT_c is 2.7 K, where ΔT_c is defined as the width of a transition between 10% and 90% of the normal-state resistivity. For S1 sample, we have measured field-cooled (FC) and zero-field-cooled (ZFC) magnetic susceptibilities at H=50 Oe. The values of shielding and Meissner fractions determined from FC and ZFC are 65 and 23 %, respectively at 5 K. The resistivity ratio ρ(300 K)/ρ(54 K) of these samples is about 13. The resistivity ratio and the transition width are respectively larger and narrower as compared to that reported for other fluorine-doped and oxygen-deficient PrFeAsO samples [5, 18]. The larger resistivity ratio and narrower transition width are the indications of high sample quality. We have also determined the approximate value of residual resistivity for S1, ρ(0)$\sim$0.10 mΩ cm, by extrapolating the ρ vs T curve between 150 and 60 K to T=0. It may be mentioned that we have prepared another sample with nominal oxygen content 0.50. The value of resistivity ratio for this sample is also large ($\sim$9) and its T_c is almost same as that for S1 and S2. However, x-ray diffraction shows the presence of impurity phases in this sample. Kito *et al.* [19] prepared Nd-based superconducting samples using high-pressure synthesis technique and found that the amount of impurity phases increase rapidly when the oxygen vacancy exceeds 0.4. This suggests that samples with oxygen deficiency more than 0.4 can not be prepared in single phase. Though both the magnitude and T dependence of normal-state resistivity depend to some extent on composition and purity of the samples, T_c is more or less insensitive to oxygen content. This observation is consistent with the

reported phase diagram T_c vs fluorine or oxygen content [1, 19].

The analysis of T dependence of ρ reveals three different temperature regimes. ρ exhibits a quadratic temperature dependence, $\rho=\rho(0)+AT^2$, in the range 70 K$\leq T \leq$150 K while it is linear in T in the intermediate region, 170 K$\leq T \leq$270 K. The T^2 behavior of ρ below 150 K indicates a strong electronic correlation and is consistent with the formation of a Fermi-liquid state. Above 270 K, the resistivity behavior shows marked departure from the usual temperature dependence of simple metals and high-T_c superconductors. Normally, in good metals, ρ is linear in T at high temperature due to the e-ph scattering. For the present samples, the slope of $\rho(T)$ curve decreases continuously with increasing temperature above 270 K and a saturationlike behavior appears at higher temperature [Fig. 2(b)]. For sample S1, the increase of ρ is small while ρ is effectively T-independent for sample S2 above 500 K [Fig. 2(c)]. The saturationlike behavior of ρ at high temperature at once draw our attention to the T dependence of ρ for Nb, Chevrel phases, several intermetallic and A15 compounds [22]. This phenomenon has been explained on the basis of the conduction-electron mean free path approaching a lower limit with the consequent breakdown of the classical Boltzmann theory [23]. The breakdown has been interpreted in terms of the Ioffe-Regel criterion [24]. The electron mean free path can not be shorter than the interatomic distance. The resistivity of poor metals at high temperatures tends to saturate to a temperature-independent value when the mean free path l approaches the wavelength $\lambda_F=2\pi/k_F$ associated with the Fermi level, where k_F is the Fermi wave vector. The Ioffe-Regel criterion for the onset of this saturation is $k_F l\leq$1. In the latter section, we will show that this criterion is indeed satisfied in the present case.

Figure 3 shows the temperature dependence of resistivity for the undoped PrFeAsO sample. One can see that the behavior of $\rho(T)$ for the parent sample is very different from that of its superconducting counter part. An anomalous peak at $T_s\sim$155 K in ρ is associated with the structural phase transition from tetragonal to orthorhombic with decreasing temperature. Below T_s, resistivity data can be fitted well with an expression, $\rho=\rho(0) + aT^n$, with n close to 1.5. However, ρ increases linearly with T from 250 K up to 525 K at the rate of $\sim$ 1.5 $\mu\Omega$ cm/K. No saturationlike behavior is observed for this sample

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in the measured temperature range. Thus, the behavior of ρ at high temperature for the nonsuperconducting sample is quite different from that of the superconducting sample. In the linear region of ρ vs T curves, the slope for the nonsuperconducting parent compound is about 6 times smaller than that for the superconducting sample (~ 8.6 $\mu\Omega$ cm/K).

For the quantitative understanding of the behavior of $\rho(T)$ at high temperature, we estimate several normal-state transport parameters related to the e-ph scattering and examine its effect on superconductivity as it was done in the case of cuprate superconductors [25]. Expressing resistivity in terms of plasma energy $(\hbar\omega_p)$, $\rho(T) = 4\pi/(\omega_p^2\tau) = 4\pi v_F/(\omega_p^2 l)$, where v_F is the Fermi velocity [26]. At high temperature, where resistivity is linear in T, τ is the e-ph scattering time τ_{ep}, and is given by $\hbar/\tau_{ep} = 2\pi\lambda_{tr}kT$ [26]. λ_{tr} is closely related to the coupling constant λ that determines the superconducting transition temperature. In the case of Nb and Pd it is found that λ_{tr} differs from λ by about 10% [22]. From the above relations one can deduce

$$\lambda_{tr} = \frac{\hbar\omega_p^2}{8\pi^2 k}\frac{d\rho}{dT} \tag{1}$$

and

$$l = \frac{\hbar v_F}{2\pi\lambda_{tr}kT}. \tag{2}$$

We can calculate λ_{tr} using the values of resistivity slope (8.6 $\mu\Omega$ cm/K) in the linear region and plasma energy (~ 0.8 eV) determined from the inplane penetration depth [27]. Eq. (1) reveals $\lambda_{tr} = 1.3$. Such a large value of λ_{tr} suggests that the e-ph coupling strength in this system is quite strong. Using this value of λ_{tr} and Fermi velocity $v_F = 1.3 \times 10^7$ cm/s calculated from band theory [12], we find $l \sim 2.5$ $\mathring{A}$ at 500 K. This value of l is comparable to the Fe-Fe separation. We observe that the Ioffe-Regel criterion for the onset of saturation $k_F l \leq 1$ is satisfied in this case. k_F was deduced using the values of Fermi energy ($E_F = 0.4$ eV) and Fermi velocity, as well as from the experimental value of carrier density ($n = 10^{21}$ cm^{-3}) reported for $LaFeAsO_{1-x}F_x$ with free electron approximation [12, 20]. In the case of layered system, one can also check the Ioffe-Regel criterion solely from the resistivity data using the relation $k_F l \sim (hc/4e^2)/\rho$, where c is the interlayer distance. If we use the value of ρ at 500 K and $c = 8.6$ $\mathring{A}$, then also $k_F l < 1$. Thus, the e-ph scattering is dominating

the high temperature resistivity and the large value of λ_{tr} is consistent with the resistivity saturation.

Having acquired the qualitative and quantitative knowledge on normal-state transport properties, we now discuss the role of e-ph coupling on superconductivity. In the limit of strong e-ph coupling, one can use the McMillan equation [28] to estimate the superconducting transition temperature

$$T_c = \frac{\Theta_D}{1.45} exp[-\frac{1.04(1+\lambda)}{\lambda - \mu(1+0.62\lambda)}]. \tag{3}$$

where μ is the Coulomb pseudopotential and Θ_D is the Debye temperature. Assuming $\mu \sim 0$ and $\lambda = \lambda_{tr} = 1.3$ and using the reported value of $\Theta_D = 355$ K for PrFeAsO [29] we find that $T_c = 41$ K. This value of T_c is only few Kelvin lower than the observed T_c for PrFeAsO$_{0.6}$F$_{0.12}$. Nevertheless, the estimated and observed values of T_c are comparable in this class of materials. A more accurate estimation of T_c can be made using the Allen-Dynes equation [30] where the prefactor $\Theta_D/1.45$ in Eq. (3) is replaced by $\omega_{ln}/1.2$. As we do not have any knowledge on the value of ω_{ln} for PrFeAsO, T_c can not be deduced using this equation for the present system. If we assume that e-ph interaction does not change significantly from system to system then using the same value 1.3 for λ_{tr} along with theoretically derived $\omega_{ln} = 206$ K for LaFeAsO [15], surprisingly the observed value of $T_c = 27$ K for LaFeAsO$_{0.89}$F$_{0.11}$ can be reproduced. Now, we would like to comment on the dependence of T_c on the rare earth ionic size in RFeAsO$_{1-x}$F$_x$. In oxypnictide superconductors, it has been reported that the lattice parameter a decreases while T_c increases when La is replaced by smaller rare earth ions [18]. The decrease of a means the decrease of Fe-Fe distance. Thus, one expects an increase in characteristic phonon frequency and hence T_c with the decrease of ionic radius of R. This observation is consistent with the phonon-mediated superconductivity but quite different from that of cuprate superconductors where T_c is not sensitive to the rare earth ionic size.

It may be interesting to compare λ_{tr} deduced from the resistivity data with the theoretically predicted values for understanding the role of e-ph interaction in Fe-based oxypnictides and similar materials. In most of the reports, these Fe-based materials are commonly viewed as unconventional superconductors because of their high transition temperature in

spite of the predicted weak e-ph coupling and the occurrence of superconductivity in close proximity to magnetism [12, 13, 14, 15, 16]. For the undoped compound, the value (0.21) of the e-ph coupling strength calculated from band theory [15] is comparable with that (0.24) obtained from the resistivity data. However, there is a large disagreement in the value of λ_{tr} determined from the resistivity data with the theoretical prediction for the superconducting sample. According to the band structure calculation, the value of λ_{tr} does not change significantly as one goes from the undoped to the doped sample, though, a 5-6 times larger coupling constant (λ_{tr}=1.0-1.3) is needed to reproduce the experimental value of T_c in this system [15]. On the contrary, if λ_{tr} does not change significantly, it is not clear why the behavior of resistivity changes so dramatically with a small amount of fluorine doping. Eschrig [17] pointed out several shortcomings of the standard band structure calculations and argued that the e-ph coupling in this system is quite strong due to the Fe inplane breathing mode. Drechsler *et al.* [11] also cast doubt on the calculated weak value of e-ph coupling. The gap function is the single most important quantity that can be used to reveal the pairing mechanism of a superconductor. Chen *et al.* [9] studied Andreev spectroscopy on $SmFeAsO_{0.85}F_{0.15}$ and observed a single nodeless gap with nearly isotropic in size across different sections of the Fermi surface. The value of the normalized gap parameter ($2\Delta/kT_c$) determined by them was 3.68 which is slightly larger than the BCS prediction of 3.52 in the weak coupling regime but close to that observed in many conventional superconductors with strong e-ph coupling such as Nb, V_3Si, etc [9, 31]. Also, the T dependence of the gap is consistent with the BCS prediction, but dramatically different from that of the pseudogap in the cuprate superconductors. From these observations, they concluded that the structure of gap is not compatible with theoretical models involving antiferromagnetic fluctuations, strong electronic correlations, the t-J model, and that proposed for the cuprate superconductors. Isotropic s-wave symmetry of the gap has also been established from the angle resolved photo emission spectroscopy [10]. It has been argued that the isotropic nature of gap and the appearance of superconductivity in close proximity to a suppressed structural phase transition together with the observation of superconductivity in $CaFe_2As_2$ with application of a modest pressure bring the role of phonon in these Fe-based samples to the forefront [10].

The strong deviation of resistivity from linearity to saturationlike in superconducting oxypnictides immediately draw our attention to conventional superconductors where e-ph coupling is strong such as in A15 compounds and Chevrel phases [22]. In these cases, ρ shows a saturationlike behavior at high temperatures and the values of λ_{tr}, in general, are quite large and comparable with that of the present system [31]. It may be important to mention that apart from λ_{tr}, other parameters related to superconductivity such as the density of states at the Fermi level, the magnitude and T dependence of $2\Delta/kT_c$ and the symmetry of gap-parameter of these systems are comparable to oxypnictides [31]. The T dependence of gap parameter is compatible with the BCS prediction in both the cases. So by comparing these systems with the present one it is apparent that oxypnictides belong to a class of strongly coupled BCS superconductors.

IV. CONCLUSIONS

In conclusion, we have analyzed the temperature dependence of resistivity of $PrFeAsO_{1-x}F_y$ samples. For nonsuperconducting PrFeAsO, ρ below 155 K shows a power-law behavior whereas in the high temperature region (250-525K), ρ is linear in T. For superconducting samples, ρ above T_c crosses over from T^2 dependence due to the electron-electron interaction to linear in T and then to a saturationlike behavior at higher temperature due to the e-ph interaction. The resistivity saturation indicates that the e-ph interaction is strong and the conduction-electron mean free path approaching a lower limit with the consequent breakdown of the classical Boltzmann theory. We have estimated the e-ph coupling parameter λ_{tr} from the linear dependence of ρ to be approximately 0.24 and 1.3, respectively for nonsuperconducting and superconducting samples. The small value of λ_{tr} for the former is consistent with band structure calculations, while for the latter, it is 5-6 times larger than the theoretical value. The present resistivity results along with the structure, value and temperature dependence of the gap-parameter $2\Delta/kT_c$, and other important parameters related to superconductivity suggest that Fe-based oxypnictides are a class of BCS superconductors with strong e-ph coupling similar to Chevrel phases and A15 compounds.

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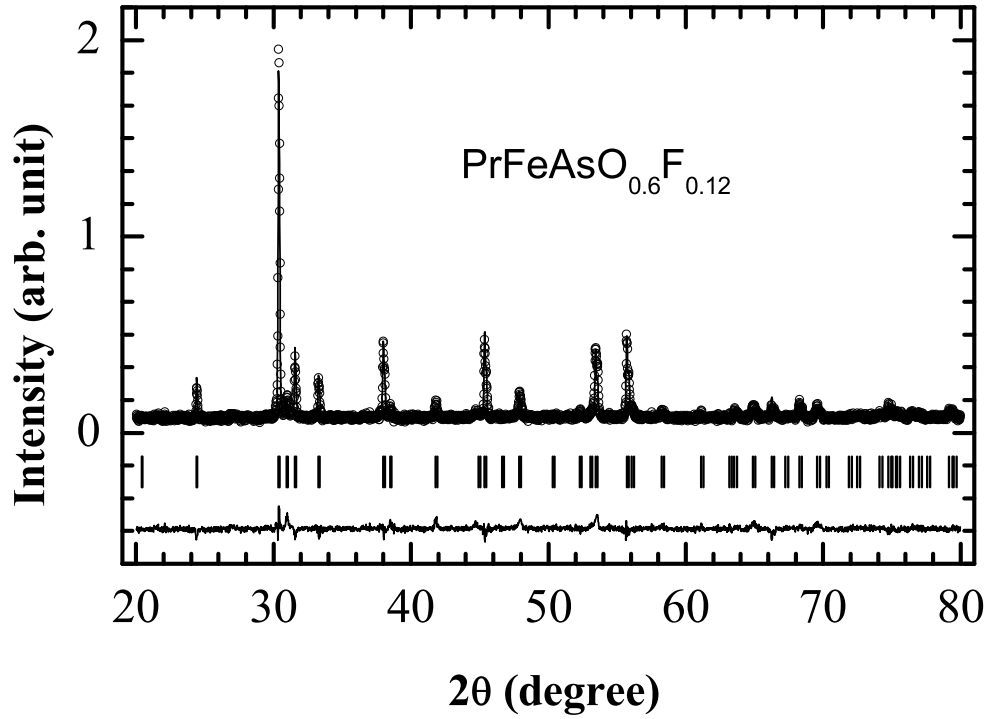[31] C. P. Poole, Jr. , H. A. Farach, R. J. Creswick, Superconductivity (Academic Press, Inc. San Diego, 1995)

FIG. 1: The x-ray diffraction pattern for the superconducting $PrFeAsO_{0.6}F_{0.12}$ sample. The solid line corresponds to Rietveld refinement of the diffraction pattern with $P4/nmm$ space group.
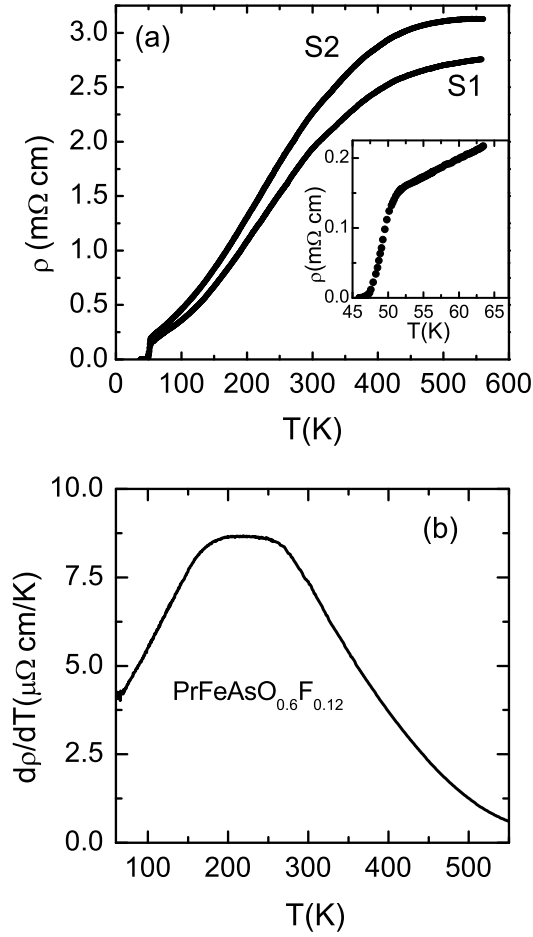
Fig. 2 (Bhoi et al)

FIG. 2: Temperature dependence of (a) resistivity (ρ) for the superconducting PrFeAsO$_{0.6}$F$_{0.12}$ (S1) and PrFeAsO$_{0.7}$F$_{0.12}$ (S2) samples, and (b) $d\rho/dT$ for PrFeAsO$_{0.6}$F$_{0.12}$ sample. Inset of (a) is the enlarged view of resistivity change close to the superconducting transition temperature for PrFeAsO$_{0.6}$F$_{0.12}$.
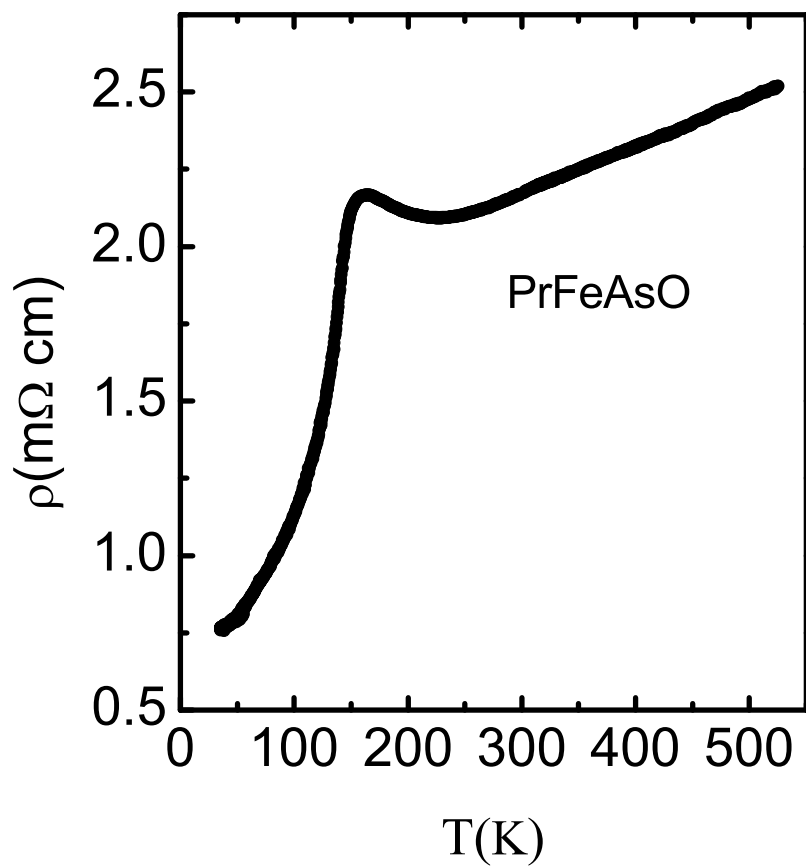
FIG. 3: Temperature dependence of resistivity for the nonsuperconducting PrFeAsO sample.