Two Fermi surface states and two $T_c$-rising mechanisms revealed by transport properties in $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ ($R$=La, Pr and Nd)

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We demonstrate the relation between critical temperature $T_c$ and transport properties in $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ ($R$=La, Pr and Nd). $T_c$ and resistivity power-law exponent $n$ form a universal line on the $T_c$ vs. $n$ plane for all the $R$-systems with $x$$\sim$0.6~0.8, indicating that $T_c$ increases with bosonic fluctuation. Transport properties show anomalies suggesting a change of Fermi surfaces around $x$$\sim$0.6~0.8. Above $x$$\sim$0.6~0.8, $T_c$ and $n$ approach the second $T_c$-$n$ line for the higher $T_c$ systems. A further increase of $T_c$ above $x$$\sim$0.6~0.8 indicates the presence of an additional $T_c$-rising mechanism in this system.

KEYWORDS: superconductivity, iron-based superconductor, transport properties

1. Introduction

Since the discovery of superconductivity (SC) in iron pnictides,$^1$ a lot of experimental and theoretical efforts have been paid to find key parameters for determining high critical temperature $T_c$ in this system. The pioneering work by Lee et al. demonstrated that the crystal structure, particularly the bond angle of (As,P)-Fe-(As,P) is strongly correlated with $T_c$. However, further experiments have shown that Lees’ conclusion is not applicable for all the iron based superconducting systems. Another parameter related to $T_c$ is the pnictogen height from the Fe-layer ($h_{pm}$).$^{3,4}$ However, it is not clear yet what electronic parameter is modified by this angle or $h_{pm}$. Although some theories suggest that the antiferromagnetic (AF) fluctuation plays an important role for the appearance of SC in the iron pnictides,$^{4,5}$ there is no direct experimental evidence that $T_c$ is correlated with the strength of AF fluctuation. Therefore, in order to clarify the mechanism of SC in this system, it is necessary to find a microscopic parameter that scales with $T_c$, comparing various physical properties of various iron pnictides with different $T_c$.

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In the present study, we focus on $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$, where $R$=La, Pr and Nd. One of the advantages of this system is that P and As are isovalent elements and thus a carrier number is kept constant in principle. The change in physical properties with $x$ is considered to be induced by a structural change due to chemical pressure. The second advantage is that we can cover a wide range of $T_c$ from $\sim$3 K to $\sim$50 K by changing $x$. This helps us to find a physical quantity that scales with $T_c$. Both end materials are rather well investigated. $R$FeAsO$_{1-y}$F$_y$ becomes an AF metal when F is not doped.$^{1,6–8}$ With increasing $y$, the AF order is suppressed and the SC emerges above $y$~0.08. Therefore, the end material in the present study, $R$FeAsO$_{0.9}$F$_{0.1}$ shows SC at low temperatures, but has large AF fluctuation. In contrast, the other end material $R$FePO$_{1-y}$F$_y$ are superconducting .$^9,10$ Even without F-doping, it is also superconducting below $\sim$ 4 K, and shows a paramagnetic metallic behavior in the normal state. Therefore, the AF fluctuation is expected to be controllable by changing $x$ in $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$.

![Powder X-ray diffraction patterns](image)

**Fig. 1.** (Color online) Powder X-ray diffraction patterns for PrFeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ with various $x$s. Almost all the diffraction peaks are indexed assuming the tetragonal structure with the $P4/nmm$ symmetry. The peaks indicated by triangles are due to impurities.

A similar study was reported in BaFe$_2$(As$_{1-x}$P$_x$)$_2$,$^{11}$ where the AF interaction is modified
Fig. 2. (Color online) Temperature dependence of resistivity below room temperature ((a)-(c)) and below 120 K ((d)-(f)) for $RFeP_{1-x}As_xO_{0.9}F_{0.1}$ ($R$=La, Pr and Nd) with various $x$s, respectively. In the panels (d)-(f), the dots and the broken lines indicate the experimental results and the fitting curves by using $\rho=\rho_0+AT^n$, respectively. The fitting of $\rho(T)$ was performed between the onset $T$ of resistive transition ($T_{\text{onset}}$) and 100 K.

by P/As-substitution. With increasing $x$, AF order is suppressed and SC manifests itself near $x=0.33$, giving a quantum critical behavior. Although we have preliminarily investigated F-free $RFeP_{1-x}As_xO$ which shows an AF order at $x=1.0$, we observed neither SC with $T_c$ higher than 10 K nor any anomalous behavior due to a magnetic quantum criticality. By contrast, $RFeP_{1-x}As_xO_{0.9}F_{0.1}$ does not have any magnetic order, but shows a drastic change with $x$ in physical properties. In the present study, we have investigated the resistivity ($\rho(T)$) and the Hall effect in $RFeP_{1-x}As_xO_{0.9}F_{0.1}$ with various $T_c$, lattice constants and presumably AF fluctuation strength to find a relationship among $T_c$, crystal structure and electronic properties in iron pnictides.

2. Experimental procedures

Polycrystalline $RFeP_{1-x}As_xO_{0.9}F_{0.1}$ ($x=0\sim1.0$) were synthesized by solid state reaction. The mixtures of $RA$, $RP$, $Fe_2O_3$, Fe and $FeF_2$ in the stoichiometric ratio were pressed into pellets in a pure Ar filled glove box and annealed at 1100 $^\circ$C for 40 h in evacuated silica tubes.
All the samples were prepared by the same careful procedure. The result of EDX (Energy Dispersive X-ray spectroscopy) indicates that the actual F concentration is about 0.03 ∼ 0.04, which is smaller than the nominal one. Since there are peaks for R and Fe near the peak for F in the EDX spectrum, we could not exactly determine the actual F concentration. Therefore, we show the nominal F concentration (0.1) in this paper.

The samples were characterized by powder X-ray diffraction using Cu $K_{\alpha}$ radiation at room temperature. In Fig. 1, we show the powder X-ray diffraction pattern for PrFeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ as an example. Almost all the diffraction peaks can be assigned to the calculated Bragg peaks for the tetragonal $P4/nmm$ symmetry. The in-plane ($a$) and out-of-plane lattice constants ($c$) were obtained by the least squares fitting of the X-ray diffraction data. The values of $a$ and $c$ for $x=0$ and 1.0 well agree with the reported data. As shown in Fig. 1, the peak position of the powder X-ray diffraction data is systematically changed with increasing $x$, and both of $a$ and $c$ linearly increase with $x$, as indicated later in Fig. 3(a). This proves that solid solutions of the present system have been successfully prepared, and the actual F concentrations are almost constant in the whole $x$-range.

The magnetic susceptibility was measured in a magnetic field of 10 Oe. The superconducting volume fractions estimated from the diamagnetic susceptibility at 2 K are over 80% for all the samples. The temperature ($T$) dependence of electrical resistivity ($\rho(T)$) was measured by a standard four-probe method from room $T$ down to 4.2 K. The Hall coefficient $R_H$ was measured in magnetic fields up to 7 T at various $T$s.

3. Results and discussion

Figures 2(a)-(c) show the $\rho(T)$ with various $x$s for $R=$La, Pr and Nd, respectively. In almost all the samples, the superconducting transitions are sharp enough to determine $T_c$ from the midpoint of $T$ of the resistive transition. (In the $x=0$ samples of $R=$Pr and Nd, $T_c < 4.2$ K is defined by an onset transition $T$ in magnetic susceptibility.) In contrast to the linear $x$-dependence of $a$ and $c$ (Fig. 3(a)), $T_c$ does not monotonically change with $x$ (Fig. 3(b)). In all the systems, $T_c$ gradually increases with $x$ up to $x=0.60$, while the behavior changes above $x=0.60$. For $R=$La, $T_c$ saturates at $x=0.6$ and slightly decreases above $x=0.6$, while for $R=$Pr and Nd, $T_c$ is more rapidly enhanced above $x=0.80$ than that for $x<0.6$.

Non-monotonic $x$-dependence was also observed in $\rho(T)$. As shown in Figs. 2(a)-(f), $\rho(T)$ for all the samples exhibits a metallic behavior. In all the $R$-systems, the resistivity value is the lowest at $x=0$. With increasing $x$, the residual resistivity $\rho_0$ and the slope of $\rho(T)$ are rapidly enhanced, showing a maximum at $x=0.60$~0.80. Such a non-monotonic but
systematic change of $\rho(T)$ with $x$ was observed in all the $R$-systems, which indicates that the observed change is intrinsic, but not due to a grain boundary effect.

As shown in Figs. 2(d)-(f), the $\rho(T)$ can be expressed as $\rho(T) = \rho_0 + AT^n$ at low $T$s, where $n$ is the power of $T$ and $A$ the coefficient. The fitting of $\rho(T)$ was performed between the onset $T$ of resistive transition ($T_{c \text{onset}}$) and 100 K. Figure 3(c) shows the $x$ dependence of $n$. For $x=0$, $n$ is close to 2 in all the $R$-systems, suggesting that the end materials with $x=0$ are a conventional Fermi liquid. As $x$ increases, $n$ decreases and reaches about unity at $x=0.60 \sim 0.80$. Above
Fig. 4. (Color online) Temperature dependence of Hall coefficient ((a)-(c)) for $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ ($R$=La, Pr and Nd) with various $x$s, respectively. The dots indicate the experimental results and the lines are guides for eyes.

$x=0.60$~$0.80$, $n$ slightly varies, but is still below 1.6. The $T$-linear $\rho(T)$ ($n=1$) is observed also in high-$T_c$ cuprates$^{15}$ and heavy fermion compounds$^{16}$ near the quantum critical point, which suggests that the conduction mechanism is governed by strong bosonic fluctuation such as AF fluctuation.$^{17}$

The $x$ dependence of power $n$ indicates that the chemical pressure induced by the P/As substitution rapidly increases bosonic fluctuation with $x$ up to $x=0.60$~$0.80$. In fact, the NMR study in $R$=La system detected the strong AF fluctuation around $x=0.6$, while almost no AF fluctuation at $x=1.0$. As shown in Fig. 3(d), $A$ is enhanced at $x=0.60$~$0.80$ and decreased toward $x=1.0$. The $A$ depends on the power $n$, and it is difficult to extract the physical origin only from the behavior of $A$. But all the $R$-systems show similar and systematic $x$-dependence of $A$, and the behavior of $A$ may be related with the enhancement of the AF fluctuation at $x=0.60$~$0.80$. Here we note that $\rho_0$ is also enhanced near $x=0.6$ in all the $R$-system, which cannot be explained by spin fluctuation theory. Although it is hard to discuss absolute values of $\rho(T)$ for polycrystalline samples, the change of $\rho_0$ in Fig. 2 is quite systematic and common in all the $R$-systems.

Figures 4(a)-(c) represent $T$-dependence of Hall coefficient $R_H$ with various $x$s for $R$=La, Pr and Nd, respectively. $R_H$ at $x=0$ is almost $T$-independent and has a small value ($\sim 2 - 3$ C/cm$^3$), while at $x=1.0$ $R_H$ is also small but shows a weak $T$-dependence. Our new finding is that the magnitude and the $T$-dependence of $R_H$ are strongly enhanced around $x=0.60$~$0.80$ in all the $R$-systems. Above $x=0.60$~$0.8$, they are suppressed with $x$. The $x$ dependence of $R_H$ at 50 K is plotted in Fig. 3(e). $R_H$ has a broad minimum around $x=0.60$~$0.8$ in all the systems. All the non-monotonic $x$-dependences of $T_c$, $n$, $A$ and $R_H$ seen in Fig. 3 demonstrate a critical
change in the electronic state around $x=0.6\sim 0.8$. This critical concentration $x$ may be slightly dependent on the $R$ element.

![Fig. 5.](image.png)

**Fig. 5.** (Color online) Relation between $T_c$ and the power $n$ of temperature in $\rho(T)$ for $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ ($R=$La, Pr and Nd) and other ion pnictides. The closed symbols with broken lines and open ones indicate the present results for $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ ($R=$La, Pr and Nd) and the previous ones for $R$FeAsO$_{1-y}$ in ref.,$^{19}$ respectively. The broad grey lines represent the two different correlations between $T_c$ and $n$ in $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ ($x<0.6\sim 0.8$) and $R$FeAsO$_{1-y}$. The results for (Ba,K)Fe$_2$As$_2$, $^{19}$ BaFe$_2$(As,P)$_2$,$^{11}$ and SrFe$_2$(As,P)$_2$,$^{30,31}$ can also be plotted on these two lines.

Another piece of evidence for the electronic change around $x=0.6\sim 0.8$ can be seen in the relation between $T_c$ and $n$ in Fig. 5. The samples with $x<0.6\sim 0.8$ show the almost linear relationship between $T_c$ and $n$, which is universal for all the $R$-systems. The $x=0$ samples exhibit the lowest $T_c$ and $T^2$ resistivity, while those with $x=0.6\sim 0.8$ show $T_c\sim 30$ K and almost $T$-linear $\rho(T)$. This distinct relation suggests that what causes the $T$-linear $\rho(T)$ is strongly involved in the mechanism of high $T_c$ SC in this system. A similar correlation between $T_c$ and $n$ has been observed in $R$FeAsO$_{1-y}$,$^{19}$ although the scaling line is shifted in parallel about 20 K from the present one.

In contrast to the samples below $x=0.6\sim 0.8$, the larger $x$ samples show no clear relation between $T_c$ and $n$. For $R=$Pr and Nd, $T_c$ is continuously increased with $x$, while $n$ is almost unchanged ($n=1\sim 1.4$) at $x>0.6\sim 0.8$. It suggests that the $T_c$-rising mechanism for $x>0.6\sim 0.80$ is different from that for $x<0.6\sim 0.80$. It is interesting that the data point for the $x=1.0$ samples with $R=$Pr and Nd are on another linear correlation for $R$FeAsO$_{1-y}$,$^{19}$ as shown in Fig. 5. It means that the As 100% compounds of $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ show the $T_c$-$n$ values sitting on the right correlation line for higher $T_c$, irrespective of F-content, while the values for $x<0.6\sim 0.80$
are on the left line and those for \( x=1.0 \) are not located on the right line in Fig. 5, the data points for \( x>0.6 \) approach those for LaFeAsO\(_{1-y}\) with increasing \( x \). One may consider that the left \( T_c \)-n line is sifted by 20 K because of the disorder induced pair-breaking effect due to As/P substitution. However, this is unlikely because the disorder effect on \( T_c \) is not strong in the present system. It is supported by the fact that \( h_{pn} \) determined by a precise Rietveld analysis for PrFeP\(_{1-x}\)As\(_x\)O\(_0\) exactly follow the universal \( h_{pn} \)-\( T_c \) curve.\(^{20}\)

What happens at the critical As-content (\( x=0.6\sim0.8 \))? The theoretical calculations have predicted that the Fermi surfaces (FSs) of \( R \)FeAsO (\( x=1.0 \)) and \( R \)FePO (\( x=0 \)) are very similar in many aspects but do have some differences.\(^4\),\(^{21}\) Experimentally, the FSs around \( \Gamma \) and M points of these systems have been confirmed by the angle-resolved photoemission spectroscopy.\(^{22–24}\) The most prominent difference predicted by theories is that the hole-type FS around \( (\pi,\pi,0) \) is missing in \( R \)FePO, while it is present in \( R \)FeAsO.\(^4\),\(^{21}\) Since this hole FS appears when the \( d_{x^2-y^2} \) band crosses Fermi energy (\( E_F \)), it is likely that the \( d_{x^2-y^2} \) band touches \( E_F \) at \( x=0.6\sim0.8 \), which gives a critical change in the electronic state. According to this picture, the electronic properties for \( x<0.6\sim0.8 \) should be discussed without the cylindrical hole FS around \( (\pi,\pi,0) \) but with two cylindrical FSs near \( \Gamma \) and M points and one three dimensional FS near \( (\pi,\pi,\pi) \) originating from \( d_{z^2} \) orbital. A systematic change in \( n \) of \( \rho(T) \) might be caused by the gradual enhancement of bosonic fluctuation due to the change of FS topology (size and shape) around \( \Gamma \) and M points with increasing \( x \). The maximum \( T_c \) is about 30 K in this configuration of FS. On the other hand, for \( x>0.6\sim0.8 \), the hole FS around \( (\pi,\pi,0) \) originating from \( d_{x^2-y^2} \) orbital provides an additional FS nesting channel. This additional channel may contribute to a further increase in \( T_c \).

The different lattice constants for different \( R \)-systems should give different FSs. Nevertheless, the critical \( x \)-value (\( x=0.6\sim0.8 \)) is only a little dependent on the \( R \) element. In these systems, the electronic structure and the FSs are closely dependent on the local structure around Fe ions. The present results indicate the P/As substitution linearly changes not only the lattice constants but also perhaps the local structure around Fe ions, and resultantly modifies the electronic state.\(^{20}\) The local structural parameter such as \( h_{pn} \) is a more important parameter that determines the electronic properties. A relevant experimental report was made for CeFeP\(_{1-x}\)As\(_x\)O that the AF order in the As-rich compositions disappeared at \( x=0.6 \).\(^{25}\)

Next, we discuss the origin of observed anomalies around \( x=0.6\sim0.8 \). In most of the iron pnictides, AF phase is close to the superconducting one. Therefore, AF fluctuation via the FS nesting is a strong candidate for a pairing interaction that may also govern the transport...
properties. However, among the observed anomalies around $x=0.6 \sim 0.8$, the increase of $R_H$, $\rho_0$ and $A$ cannot be explained by the spin fluctuation theory, $^{17}$ and requires something others.

Another candidate is charge fluctuation. We point out that similar enhancements of $R_H$, $\rho_0$ and $A$ and $T_c$ together with $T$-linear $\rho(T)$ were observed in CeCu$_2$(Si,Ge)$_2$. $^{26-28}$ Apart from a magnetic quantum critical point in the pressure-$T$ phase diagram, this heavy fermion compound shows another critical behavior at a higher pressure where the $T_c$ reaches the highest value. The observed anomalies were interpreted as a result of the rapid change of the Ce valence. Watanabe $et$ $al.$ successfully explained these anomalies by the microscopic theory for valence fluctuation based on an extended Anderson model. $^{29}$

In the case of $R$FeP$_{1-x}$As$_x$(O,F), P/As-substitution is an isovalent substitution in a chemicalsense. However, it is likely that the exchange of band energy with $x$ (the $d_{x^2-y^2}$ band is lifted up above $E_F$ and the $d_{z^2}$ band shifts down below $E_F$) causes valence (charge) fluctuation near the critical composition $x=0.6 \sim 0.8$. Below $x=0.6 \sim 0.8$, this charge fluctuation gradually increases with $x$ and causes the enhancement of $T_c$.

Finally, we address the issue of $T_c$-rising mechanism. Although the enhancement of $R_H$, $\rho_0$ and $A$ is the largest and $n$ is close to 1 near $x=0.6 \sim 0.8$, $T_c$ is not a maximum at this composition but it further increases for larger $x$. This is because, as shown in Fig. 5, there exists another $T_c-n$ line (high $T_c$ line), and the data above $x=0.6 \sim 0.8$ seem to approach towards this line. The samples with $0.6 < x < 1.0$ have FSs with $d_{x^2-y^2}$ and $d_{z^2}$ orbital characters, and the two $T_c$-rising mechanisms perhaps by different nesting conditions and/or different bosonic fluctuations coexist. As a result, the samples with $0.6 < x < 1.0$ show a crossover behavior and their results are located between two $T_c-n$ lines in Fig. 5. We also plot the data for other iron pnictides such as (Ba,K)Fe$_2$As$_2$, $^{19}$ BaFe$_2$(As,P)$_2$, $^{11}$ and SrFe$_2$(As,P)$_2$ $^{30,31}$ (A-122 system where A=Ba, Sr and K). At a glance, we find that all the compounds are classified into two groups with the two universal $T_c-n$ relations, namely, the compounds which obey the left $T_c-n$ relation (low $T_c$ line) and those which obey the right one (high $T_c$ line). On the low $T_c$ line, $T_c$ is enhanced with $x$ owing to a gradual increase of spin or charge fluctuation. The As/P concentration seems to be a crucial parameter to control the pairing interaction, while the lattice constant controlled by the $R$-element in $R$FeP$_{1-x}$As$_x$O$_{0.5}$F$_{0.5}$ or the A-element in 122-systems does not play an important role. The maximum $T_c$ value in this class of compounds is about 30 K.

By contrast, all the compounds on the high $T_c$ line are P-free. $T_c$ varies with the lattice parameters controlled by the oxygen content and/or the size of $R$-element in $R$FeAsO$_{1-y}$ or the A-element in 122-systems. The maximum $T_c$ reaches 55 K in this class of compounds,
but there was reported no clear correlation between AF fluctuation and $T_c$ (or $n$). Therefore, the $T_c$-rising mechanism along this line is unclear. It is also unknown why the high $T_c$ line is shifted by 20 K from the low $T_c$ line.

It may be worth to note here that a nodal superconducting gap was reported for many compounds on the low $T_c$ line, while a full gap for the compounds on the high $T_c$ line. A qualitative difference in the FSs as revealed in the present study could contribute to this symmetry difference of the superconducting gap. All these facts related to the two $T_c$-$n$ lines suggest that there exist two different $T_c$-rising mechanisms in the iron pnictide superconductors and in some cases the two may act additively.

4. Conclusion

In summary, we have clarified the relation between $T_c$ and the transport properties by changing the As/P ratio in $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ with $R$=La, Pr and Nd. It has been revealed that there are two distinct regions of $x$. In the low $x$-region ($x<0.6$--$0.8$), $T_c$ linearly increases from 3 K to 30 K with decreasing the power $n$ in $\rho(T)=\rho_0+AT^n$ from 2 (at $x=0$) to 1 (around $x=0.6$--$0.8$). This strongly suggests that some bosonic fluctuation is a primary factor to enhances $T_c$. The universal $T_c$-$n$ relation holds for all the $R$FeP$_{1-x}$As$_x$O$_{0.9}$F$_{0.1}$ with $x<0.6$--$0.8$ in the present study as well as BaFe$_2$(As,P)$_2$ and SrFe$_2$(As,P)$_2$. In addition to the $T$-linear $\rho(T)$, $R_H$, $\rho_0$ and $A$ are strongly enhanced near $x=0.6$--$0.8$, suggesting some critical change of the electronic state. In the high $x$-region ($x>0.6$--$0.8$), on the other hand, $T_c$ becomes strongly $R$-dependent and further increases with $x$, but shows no clear correlation with $n$. The compounds with $x>0.6$--$0.8$ seem to approach another universal $T_c$-$n$ relation which holds for $R$FeAsO$_{1-y}$ and (Ba,K)Fe$_2$As$_2$. The presence of two distinct $T_c$-$n$ relations could be the evidence that there are two $T_c$-rising mechanisms in the iron pnictides.

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