

Supplementary Information : Importance of d_{xy} orbital and electron correlation in iron-based superconductors revealed by phase diagram for 1111-system

Tsuyoshi Kawashima¹, Shigeki Miyasaka^{1,*}, Hirokazu Tsuji¹, Takahiro Yamamoto¹, Masahiro Uekubo¹, Akira Takemori¹, Kwing To Lai^{1,2}, and Setsuko Tajima¹

¹Department of Physics, Osaka University, Toyonaka, Osaka, 560-0043, Japan

²Department of Physics, The Chinese University of Hong Kong, Shatin, Hong Kong

*miyasaka@phys.sci.osaka-u.ac.jp

Structural Parameters

All the polycrystalline samples of $R\text{FeAs}_{1-x}(\text{P/Sb})_x\text{O}_{1-y}(\text{F/H})_y$ ($R = \text{La}$ and Nd) were characterized by synchrotron X-ray diffraction. Figure S1 shows the x -dependence of the lattice constants a and c , the pnictogen (Pn) height from the Fe-layer (h_{Pn}), the Pn -Fe- Pn bond angle (α), and the Fe- Pn bond distance ($d_{\text{Fe}-Pn}$) in $R\text{FeAs}_{1-x}(\text{P/Sb})_x\text{O}_{1-y}(\text{F/H})_y$ ($R = \text{La}$ and Nd). The x -dependence of the lattice constants for $\text{LaFeAs}_{1-x}\text{P}_x\text{O}$ and $\text{NdFeAs}_{1-x}\text{P}_x\text{O}_{0.95}\text{F}_{0.05}$ is also plotted in Figs. S1(a) and (b) for reference. The crystallographic parameters for other samples with different y -values have been shown in Fig. 1 and in our previous report¹.

In all samples of $R\text{FeAs}_{1-x}(\text{P/Sb})_x\text{O}_{1-y}(\text{F/H})_y$, the lattice constants show linear x -dependence, indicating that P or Sb was successfully substituted for As. In addition, the structural parameters have been systematically changed by F- and H-doping. The local structural parameters α and $d_{\text{Fe}-Pn}$ were used when the contour plot of superconducting transition temperature T_c was generated in Fig. 6.

Temperature Dependence of Resistivity

Figures S2 and S3 present the temperature (T -) dependence of resistivity for $\text{NdFeAs}_{1-x}\text{P}_x\text{O}_{1-y}(\text{F/H})_y$ and $\text{LaFeAs}_{1-x}\text{Sb}_x\text{O}_{1-y}(\text{F/H})_y$, respectively. The values of T_c were determined from T for zero resistivity. The power n in $\rho(T) = \rho_0 + AT^n$ was estimated by fitting $\rho(T)$. These T_c and n values were shown in Figs. 4, 5 and 6. The T -dependent resistivities for other compositions were shown in Fig. 2 and our previous reports¹⁻³.

References

1. Miyasaka, S. *et al.* Three superconducting phases with different categories of pairing in hole- and electron-doped $\text{LaFeAs}_{1-x}\text{P}_x\text{O}$. *Phys. Rev. B* **95**, 214515 (2017).
2. Miyasaka, S. *et al.* Two Fermi Surface States and Two T_c -Rising Mechanisms Revealed by Transport Properties in $\text{RFeP}_{1-x}\text{As}_x\text{O}_{0.9}\text{F}_{0.1}$ ($R = \text{La}, \text{Pr}, \text{and Nd}$). *J. Phys. Soc. Jpn.* **82**, 124706 (2013).
3. Lai, K. T. *et al.* Evolution of the phase diagram of $\text{LaFeP}_{1-x}\text{As}_x\text{O}_{1-y}\text{F}_y$ ($y = 0 - 0.1$). *Phys. Rev. B* **90**, 064504 (2014).

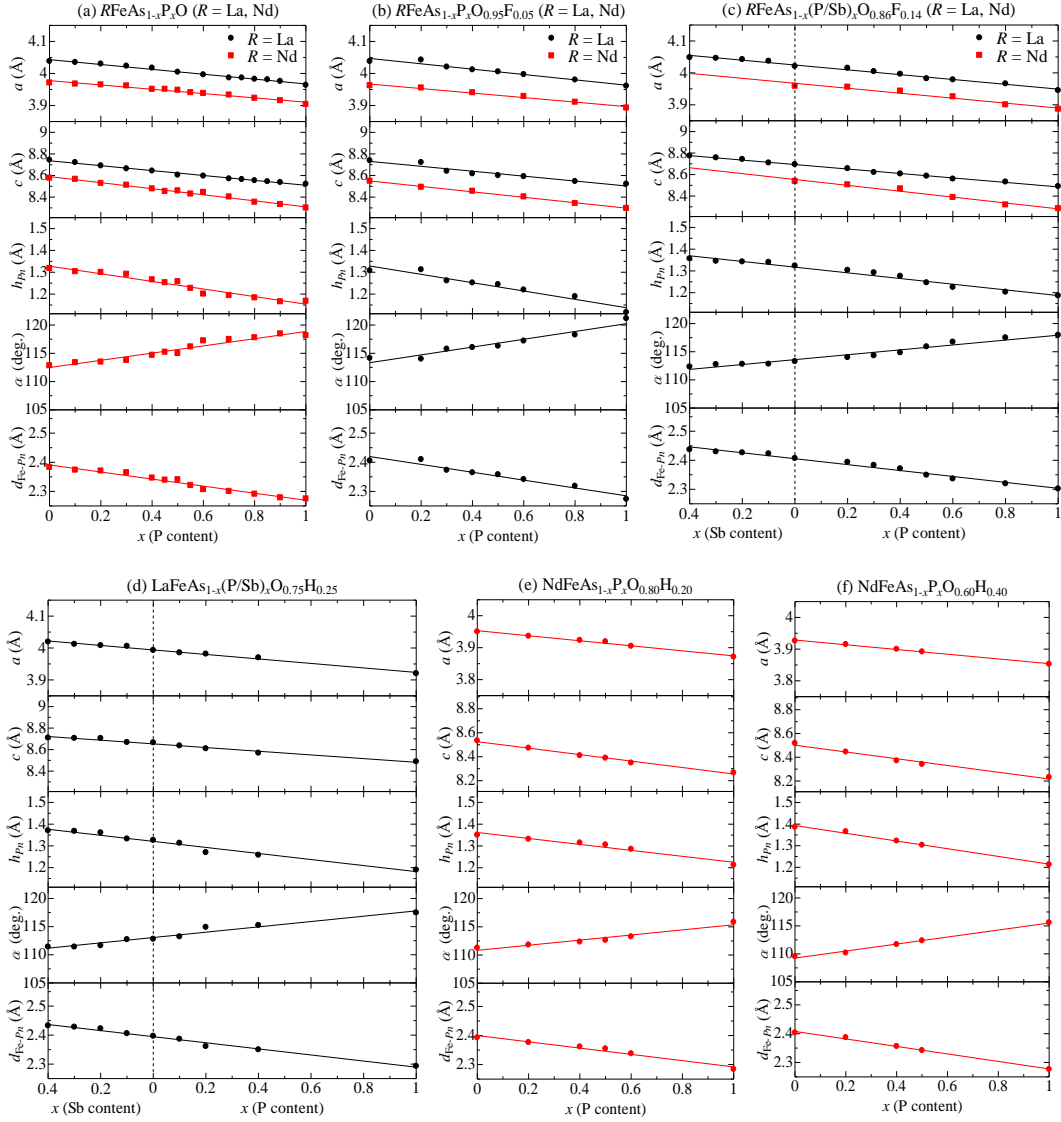


Figure S1. x -dependence of structural parameters: lattice constants a and c , pnictogen height from the Fe plane h_{Pn} , Pn-Fe-Pn bond angle α , and Fe-Pn bond length d_{Fe-Pn} , in (a) $RFeAs_{1-x}P_xO$ ($R = La$ and Nd), (b) $RFeAs_{1-x}P_xO_{0.95}F_{0.05}$, (c) $RFeAs_{1-x}(P/Sb)_xO_{0.86}F_{0.14}$, (d) $LaFeAs_{1-x}(P/Sb)_xO_{0.75}H_{0.25}$, (e) $NdFeAs_{1-x}P_xO_{0.80}H_{0.20}$, and (f) $NdFeAs_{1-x}P_xO_{0.60}H_{0.40}$.

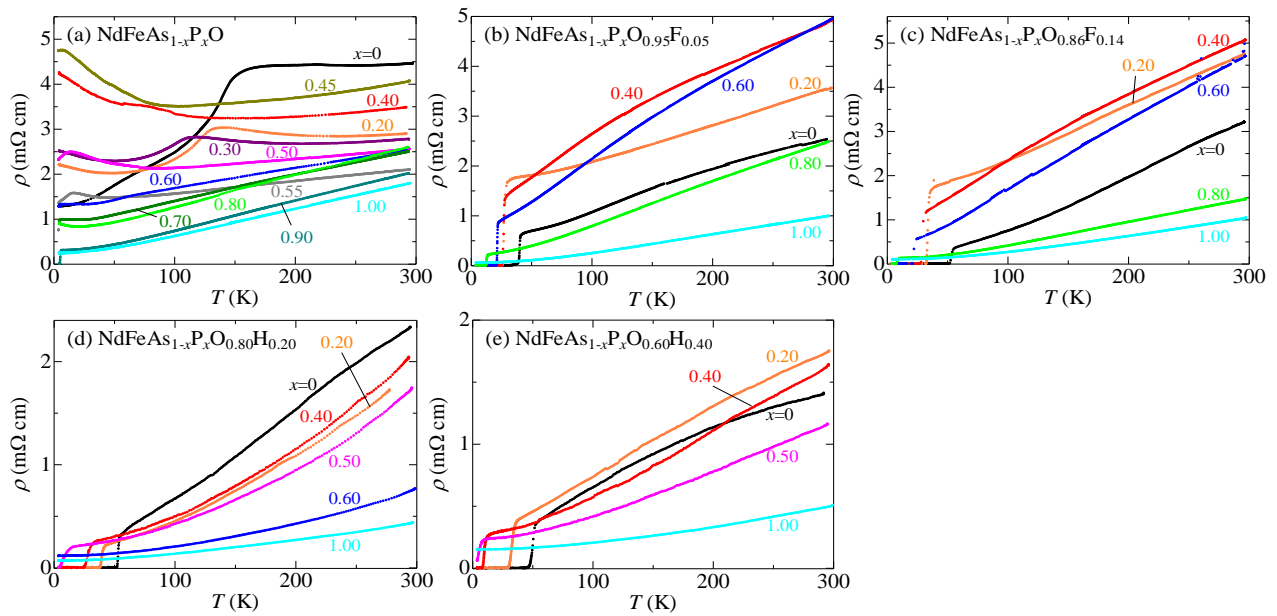


Figure S2. Temperature (T -) dependence of resistivity ρ for (a) $\text{NdFeAs}_{1-x}\text{P}_x\text{O}$, (b) $\text{NdFeAs}_{1-x}\text{P}_x\text{O}_{0.95}\text{F}_{0.05}$, (c) $\text{NdFeAs}_{1-x}\text{P}_x\text{O}_{0.86}\text{F}_{0.14}$, (d) $\text{NdFeAs}_{1-x}\text{P}_x\text{O}_{0.80}\text{H}_{0.20}$, and (e) $\text{NdFeAs}_{1-x}\text{P}_x\text{O}_{0.60}\text{H}_{0.40}$ for various values of x .

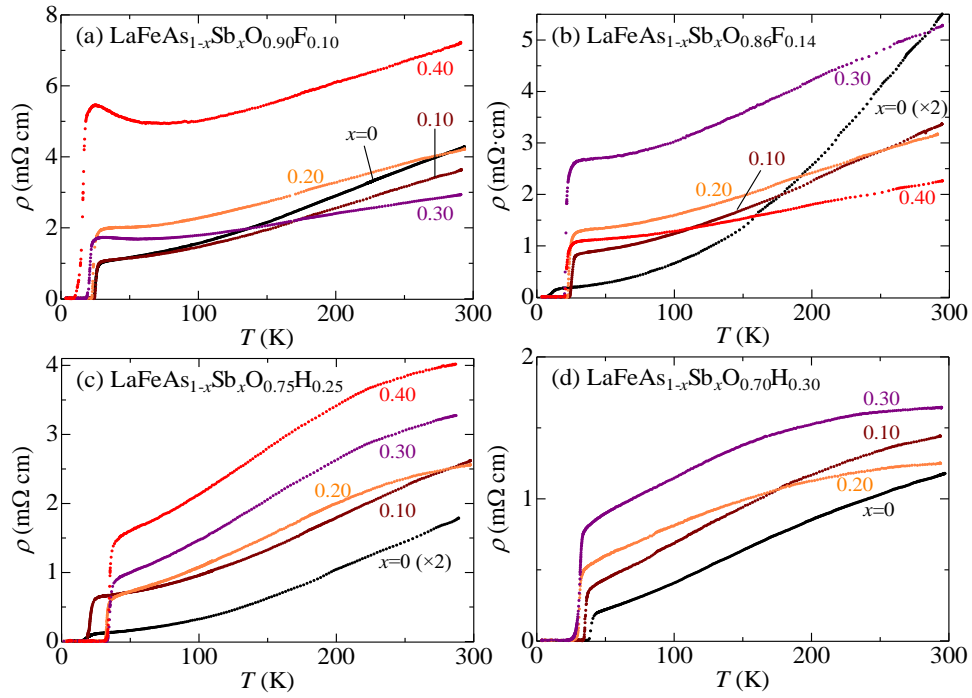


Figure S3. Temperature (T -) dependence of resistivity ρ for (a) $\text{LaFeAs}_{1-x}\text{Sb}_x\text{O}_{0.90}\text{F}_{0.10}$, (b) $\text{LaFeAs}_{1-x}\text{Sb}_x\text{O}_{0.86}\text{F}_{0.14}$, (c) $\text{LaFeAs}_{1-x}\text{Sb}_x\text{O}_{0.75}\text{H}_{0.25}$, and (d) $\text{LaFeAs}_{1-x}\text{Sb}_x\text{O}_{0.70}\text{H}_{0.30}$, with various values of x .