

# Controlling factors of $T_c$ dome structure in 1111-type iron arsenide superconductors

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We investigated the effects of phosphorus substitution on the shape of the  $T_c(x)$  dome in 1111-type  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  ( $0 < x < 0.5$ ). Hydride ion substitution of oxide sites ( $\text{O}^{2-} \rightarrow \text{H}^-$ ) exerts a chemical pressure effect, i.e., a structural reduction of the  $Pn\text{-Fe-P}n$  angle  $\alpha$  ( $Pn = \text{P, As}$ ) and also dopes electrons into the  $\text{FeP}n$  layer to induce superconductivity. Isovalent phosphorus substitution ( $\text{P}^{3-} \rightarrow \text{As}^{3-}$ ) can induce only a chemical pressure effect, i.e., an increase of  $\alpha$  for La substitution of Sm sites. As  $y$  increases from 0.0 to 0.5, the single  $T_c$  dome gradually splits into two domes, similar to those of  $\text{LaFeAsO}_{1-x}\text{H}_x$  with a  $T_c$  valley at  $x \approx 0.16$ . We found that the  $T_c$  valley is located around  $(x, \alpha) \approx (0.16, 113^\circ)$  for both of the  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  and  $\text{LaFeAsO}_{1-x}\text{H}_x$  series, irrespective of changes in the  $Pn$  anion and  $Ln$  cation species. This result suggests that suppression of  $T_c$  leads to the emergence of a  $T_c$  valley when both the shape of  $\text{FeP}n_4$  tetrahedra represented by  $\alpha$  and electron-doping level of  $x$  meet the above criterion in 1111-type iron oxyphosphide superconductors.

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## I. INTRODUCTION

1111-type  $Ln\text{FeAsO}$  ( $Ln = \text{lanthanide}$ ) composed of an  $\text{FeAs}$  conducting layer and a  $Ln\text{O}$  insulating layer is a prototypical parent compound of iron-based superconductors [1–8] showing a tetragonal-orthorhombic structural transition at 120–160 K accompanied by antiferromagnetic (AFM) ordering of Fe spins [9–11]. Superconductivity, with a critical temperature  $T_c$  of up to 57.8 K [12,13], emerges when the structural-magnetic transition is suppressed by electron doping of the  $\text{FeAs}$  layer via fluoride-ion ( $\text{F}^-$ ) substitution of oxygen sites. The doped electron concentration  $x$  vs temperature  $T$  diagram of  $Ln\text{FeAsO}_{1-x}\text{F}_x$  possess a superconducting (SC) region in the range  $x = 0.1\text{--}0.2$  adjacent to an AFM region at  $x \approx 0.0$  [14], although the whole shape of the SC region has not been determined because the solubility limit of  $\text{F}^-$  restricts  $x$  to  $< 0.2$ . The SC region adjacent to the AFM region is a common feature of the electronic phase diagram for many unconventional superconductors, including iron pnictides [15]. Therefore, the electronic structure of  $Ln\text{FeAsO}_{1-x}\text{F}_x$  has been well studied, and the material dependence of  $T_c$  on the spin-fluctuation-mediated pairing mechanism has been explained to some degree [16–19].

Recently, we found that the SC region continues beyond  $x \approx 0.4$  far from the AFM region by inducing high electron doping, with  $x$  up to 0.5, using hydride-ion ( $\text{H}^-$ ) substitution in place of  $\text{F}^-$  [20–23]. In particular,  $\text{LaFeAsO}_{1-x}\text{H}_x$  features a SC region with a unique shape that consists of a conventional dome (SC1) around  $x \approx 0.1$  with a maximum  $T_c$  ( $T_c^{\text{max}}$ ) of 29 K and an additional dome (SC2) around  $x \approx 0.3$  with a  $T_c^{\text{max}}$  of 36 K. Inelastic neutron-scattering measurements of  $\text{LaFeAsO}_{1-x}\text{H}_x$  revealed a switch of momentum transfer of magnetic excitation from  $1.14 \text{ \AA}^{-1}$  for the first dome to  $1.25\text{--}1.38 \text{ \AA}^{-1}$  for the second dome, indicating that the pairing channels for SC1 and SC2 regions are different from each other [24]. The split of the SC region, i.e., the emergence of double dome  $T_c$  structures, is an unexpected phenomenon in iron-based superconductors

from theoretical studies. Thus, we consider that the elucidation of its origin is key to understanding the SC mechanism and to exploring the pathway to improve  $T_c$ .

Here we focus on the effects of physical and chemical pressure on the  $T_c$ -dome shape reported in Ref. [22]. When a physical pressure of up to 3 GPa was applied, the two SC regions for  $\text{LaFeAsO}_{1-x}\text{H}_x$  merged into a single dome with  $T_c^{\text{max}} = 47$  K. A similar phenomenon was also induced by replacement of La with Ce, Sm, and Gd. These results strongly suggest that the  $T_c$ -dome shape is controlled by the local structure of the  $\text{FeAs}_4$  tetrahedron. As for the  $Pn\text{-Fe-P}n$  angle  $\alpha$  of the  $\text{FeP}n_4$  tetrahedron ( $Pn = \text{P, As}$ ), Lee *et al.* have suggested that  $T_c$  increases as  $\alpha$  approaches that of a regular tetrahedron [25]. Further, Kuroki *et al.* proposed on the basis of theoretical calculations that  $T_c$  is raised as the  $Pn$  height between the Fe plane  $h_{Pn}$  [18,19] is increased. However, because both structural parameters are related to each other by the equation  $h_{Pn} = r_{\text{Fe-P}n} \cos(\frac{\alpha}{2})$ , where  $r_{\text{Fe-P}n}$  is the  $\text{Fe-P}n$  bond length, it is difficult to distinguish which parameters of  $\alpha$  and  $h_{Pn}$  have a dominant effect on the  $T_c$ -dome shape.

In this paper, we report the effects of phosphorus substitution on the  $T_c(x)$ -dome shape of 1111-type  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  ( $0 \leq x < 0.5$ ). Isovalent P substitution of the As site ( $\text{P}^{3-} \rightarrow \text{As}^{3-}$ ) induces a chemical pressure effect that changes  $\alpha$  and  $h_{Pn}$  in a similar manner to that induced by La-substitution of the Sm site. As a result, we found double dome-shaped SC regions in  $y = 0.45$  and  $0.53$ . Because P substitution changes  $r_{\text{Fe-P}n}$ , the resulting crystal structure can access the region in  $(\alpha, h_{Pn})$  space unexplored by  $Ln$  substitution. By considering the behavior of  $\alpha$  and  $h_{Pn}$  in  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  and  $Ln\text{FeAsO}_{1-x}\text{H}_x$  ( $Ln = \text{La, Ce, Sm}$ ) systems, we discuss the controlling factors for the  $T_c(x)$ -dome shape of 1111-type Fe pnictides.

## II. EXPERIMENTAL

$\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  with nominal  $y$  ( $y_{\text{nom}} = 0.3, 0.5$ , and  $0.6$ ) were synthesized by the solid-state reaction of  $\text{SmAs}$ ,  $\text{FeAs}$ ,  $\text{Fe}_2\text{As}$ ,  $\text{Fe}_2\text{P}$ ,  $\text{Sm}_2\text{O}_3$ , and  $\text{SmH}_2$  using a belt-type high

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pressure anvil cell. The metal arsenides and phosphides were prepared from their respective metals.  $\text{SmH}_2$  was synthesized by heating metal samarium in a  $\text{H}_2$  atmosphere. All starting materials and precursors for the synthesis were prepared in a glove box filled with purified Ar gas ( $\text{H}_2\text{O}$ ,  $\text{O}_2 < 1$  ppm). The mixture of starting materials was placed into a BN capsule with a mixture of  $\text{Ca}(\text{OH})_2$  and  $\text{NaBH}_4$  as an excess hydrogen source and then heated at 1473 K and 2 GPa for 30 min.

The amount of hydrogen incorporated into the resulting samples was evaluated by thermal desorption spectroscopy (TDS; ESCO TDS1200). Approximately 5 mg of the sample was heated to 1373 K at a heating rate of 60 K/min under vacuum  $< 10^{-6}$  Pa. Hydrogen released from the sample, in the form of  $\text{H}_2$  molecules, was ionized and detected by a quadrupole mass spectrometer as an ion with mass-to-charge ratio ( $m/z$ ) = 2. Other nonhydrogen elemental compositions (Sm: Fe: As: P: O) were determined by an electron-probe microanalyzer (EPMA; JEOL model JXA-8530F) equipped with a field-emission-type electron gun and wavelength dispersive x-ray detector. The micrometer-scale compositions within the main phase were probed on five to 10 focal points, and the results were averaged.

Phase purity of the resulting samples was determined by powder x-ray diffraction (XRD) using a Bruker diffractometer model D8 ADVANCE (Cu rotating anode). The crystallographic parameters of the synthesized 1111-type compounds were determined by Rietveld analysis of XRD patterns using TOPAS code [26], assuming a tetragonal symmetry (space group of  $P4/nmm$ , lattice parameters  $a = b \approx 0.39$  nm and  $c \approx 0.84$  nm) with atomic positions of Sm ( $1/4, 1/4, z_{\text{Sm}}$ ), Fe ( $3/4, 1/4, 1/2$ ), Pn ( $1/4, 1/4, z_{\text{Pn}}$ ), and O ( $3/4, 1/4, 0$ ). During the refinement of structural parameters, site occupancies of Pn and O sites were fixed to the values estimated by the EPMA.

Four-probe dc resistivity ( $\rho$ ) and magnetic susceptibility ( $\chi$ ) were measured in the temperature range 2–300 K, using a physical properties measurement system (Quantum Design, Inc.) with a vibrating sample magnetometer attachment.

### III. RESULTS AND DISCUSSION

#### A. P-substitution effect on $\alpha$ and $h_{\text{Pn}}$

Using high pressure techniques, we obtained polycrystalline pellets containing >80 weight% of the 1111-type phase of  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  ( $0 \leq x < 0.5$ ). The elemental composition analysis indicates that actual phosphorous content  $y$  tended to be less than the nominal content  $y_{\text{nom}}$  of the starting mixtures. As a result, we obtained samples with  $y$  values of  $\approx 0.23$ ,  $\approx 0.45$ , and  $\approx 0.53$  from starting mixtures with  $y_{\text{nom}} = 0.3, 0.5$ , and  $0.6$ , respectively. The composition analysis is described in detail in the Supplemental Material [27]. The amount of oxide vacancies measured almost matched the hydrogen content, indicating that the oxide ions are successfully replaced by hydride to form  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$ . Previous neutron diffraction analysis and density functional theory calculations demonstrated that the hydrogen exclusively replaces  $\text{O}^{2-}$  sites in the form of  $\text{H}^-$  ions and supplies an electron to the  $\text{FePn}$  layer to maintain charge neutrality [21].

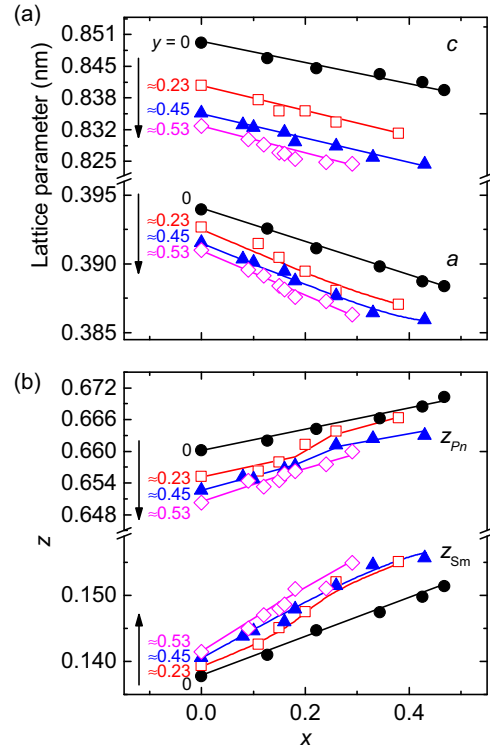


FIG. 1. (Color online) Crystallographic parameters of  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  with  $y = 0, \approx 0.23, \approx 0.45$ , and  $\approx 0.53$  as a function of  $x$ . (a) Lattice parameters  $a$  and  $c$ . (b)  $z$  coordinate of Sm and Pn sites ( $z_{\text{Sm}}$  and  $z_{\text{Pn}}$ ). Parameters for  $y = 0$  are taken from previous literature [20].

Figures 1(a) and 1(b) show the variations in the lattice parameters ( $a$  and  $c$ ) and  $z$  coordinate of Sm and Pn sites ( $z_{\text{Sm}}$  and  $z_{\text{Pn}}$ ) as a function of  $x$  in  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  specified by  $y = 0, \approx 0.23, \approx 0.45$ , and  $\approx 0.53$ . With increases in  $x$ , the lattice parameters decrease, and the  $z$  coordinates increase, i.e.,  $\text{Sm}_4\text{O}$  and  $\text{FePn}_4$  tetrahedra become stretched along the  $c$  axis upon compression of the crystal lattice. With increases in  $y$ ,  $a(x)$  and  $c(x)$  curves are shifted down, and the  $z_{\text{Sm}}(x)$  curve is shifted up, indicating that the effects of P substitution on these parameters are the same as those of H substitution. In contrast, the effect of P substitution on  $z_{\text{Pn}}$  is the opposite to that of H substitution. Because P-substitution effects are the same as those of La and Ce substitution of Sm sites, the ranges of the geometric parameters of  $\text{FePn}_4$  tetrahedron, i.e.,  $\alpha$  and  $h_{\text{Pn}}$  in  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$ , partially overlap with those of the  $\text{LnFeAsO}_{1-x}\text{H}_x$  ( $\text{Ln} = \text{La-Sm}$ ) system.

Figure 2 shows  $\alpha$  vs  $h_{\text{Pn}}$  plots for  $\text{LnFeAsO}_{1-x}\text{H}_x$  and  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$ . All the points of  $\text{LnFeAsO}_{1-x}\text{H}_x$  are located within a narrow bandlike region corresponding to  $r_{\text{Fe-Pn}} = 0.239\text{--}0.243$  nm. In contrast to  $\text{Ln}$  or H substitution, which increases  $r_{\text{Fe-Pn}}$  (+2 pm for 50% H substitution and full substitution of La to Sm), P substitution decreases  $r_{\text{Fe-Pn}}$  (−7 pm for 50% P substitution) and expands the observable range in ( $\alpha, h_{\text{Pn}}$ ) space to allow investigation of the structural dependence of the  $T_c$ -dome shape.

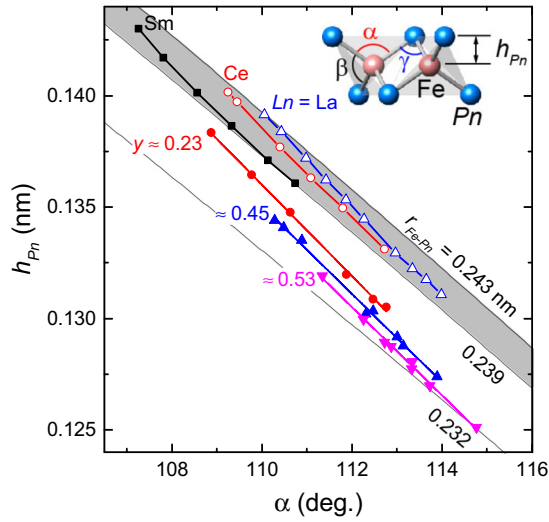


FIG. 2. (Color online)  $\alpha$  vs  $h_{Pn}$  plots of  $LnFeAsO_{1-x}H_x$  ( $Ln = La, Ce,$  and  $Sm$ ) [22] and  $SmFeAs_{1-y}P_yO_{1-x}H_x$  ( $y = 0.23, 0.45,$  and  $0.53$ ). Inset is a drawing of edge-shared  $FePn_4$  tetrahedra in  $LnFePnO$  with notation of  $\alpha$  ( $Pn-Fe-Pn$ ),  $\beta$  (another  $Pn-Fe-Pn$ ), and  $\gamma$  ( $Fe-Pn-Fe$ ) angles and pnictogen height  $h_{Pn}$ . Lines are guides to the eye, tracing the  $\alpha$ - $h_{Pn}$  relation for specified  $y$  and  $Ln$ . In  $LnFeAsO_{1-x}H_x$ , the change in Fe-As bond length is small ( $r_{Fe-Pn} = 0.239$ – $0.243$  nm), and  $h_{Pn}$  is strongly linked to  $\alpha$ . By replacing As by P,  $r_{Fe-Pn}$  clearly decreases.

### B. P-substitution effect on $T_c$ -dome shape

Figure 3(a) shows the temperature dependence of resistivity for  $y \approx 0.23$ . At  $x = 0$ , a kink in resistivity due to structural-magnetic transitions was observed around 90 K. As  $x$  increases, a sudden drop in resistivity to zero was observed at  $x > 0.11$ . The maximum onset  $T_c$  was 44.5 K at  $x = 0.20$ , and this decreased to  $< 2$  K at  $x = 0.38$ . For  $y \approx 0.45$ , as shown in Fig. 3(b), superconductivity was also observed at  $x > 0.08$ . The  $T_c$  decreased from 22.5 K at  $x = 0.08$  to 18 K at  $x = 0.16$  but subsequently increased to 24 K at  $x = 0.21$ . Finally,  $T_c$  decreased to  $< 2$  K at  $x = 0.43$ . A similar drop in  $T_c$  at  $x = 0.16$  was also observed for  $y \approx 0.53$  [see Fig. 3(c)].  $T_c = 4$  K was observed at  $x = 0.15$  between  $x = 0.12$  and  $0.18$ , showing  $T_c^{\max} = 14$  K [27]. It is notable that a drop of resistivity was observed at  $\approx 5$  K in the  $x = 0.0$  sample with  $y \approx 0.53$ . Because a drop in magnetic susceptibility was not observed in this temperature region, it is likely to originate from an AFM transition of  $Sm^{3+}$  spins similar to that observed in  $SmFePO$  [28]. Behavior observed in  $\chi$ - $T$  profiles also reflected the development of bulk superconductivity, as shown in Figs. 3(d)–3(f). A drop in susceptibility due to SC transitions was observed at the magnetic onset  $T_c^{\text{mag}}$ , which was slightly lower than the resistivity onset  $T_c$ . As shown in Figs. 3(e) and 3(f), a drop of  $T_c^{\text{mag}}$  was observed at  $x = 0.15$ – $0.16$  for  $y \approx 0.45$  and  $0.53$ . Figure 4 summarizes the magnetic onset  $T_c^{\text{mag}}$  as a function of  $x$ . A  $T_c^{\text{mag}}(x)$  curve for  $y = 0$  taken from a previous paper [22] is also shown in this

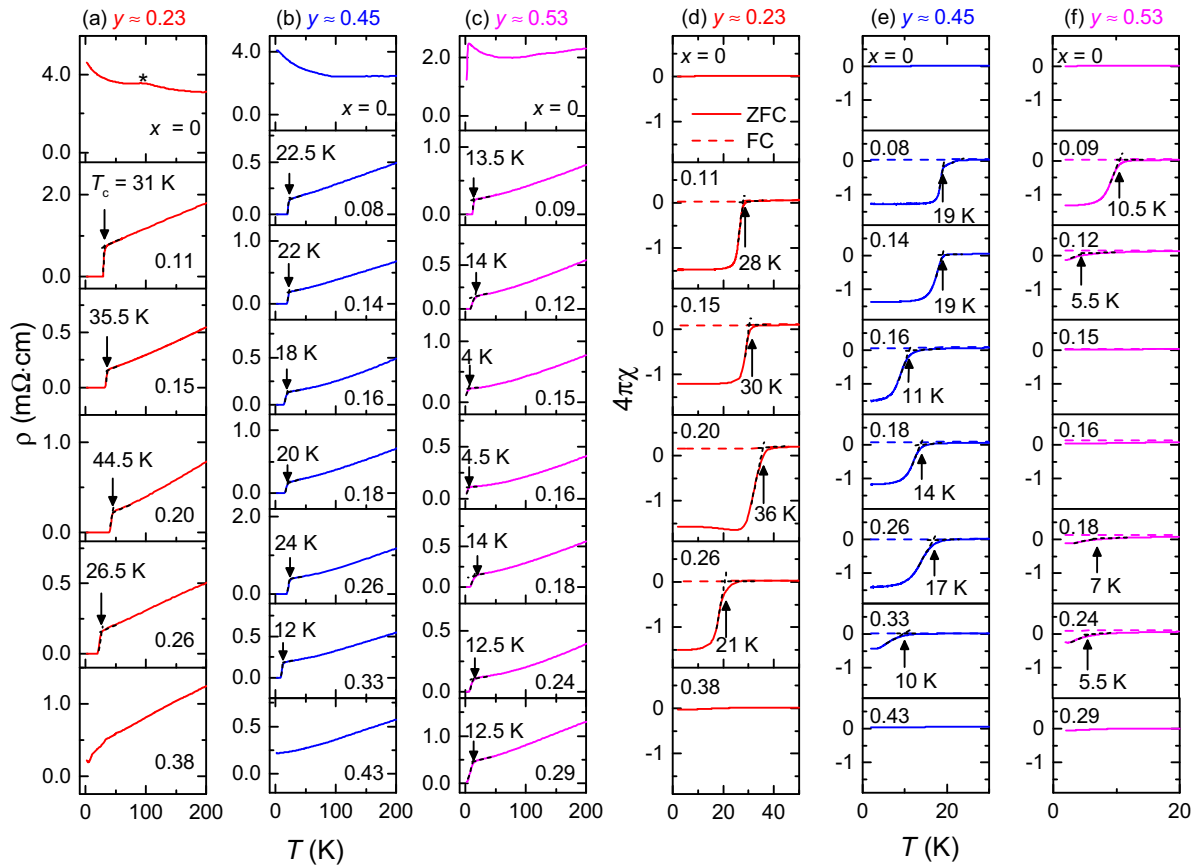


FIG. 3. (Color online) Temperature dependence of electrical resistivity (a)–(c) and magnetic susceptibility (d)–(f) in  $SmFeAs_{1-y}P_yO_{1-x}H_x$  with  $y \approx 0.23, \approx 0.45,$  and  $\approx 0.53$ .

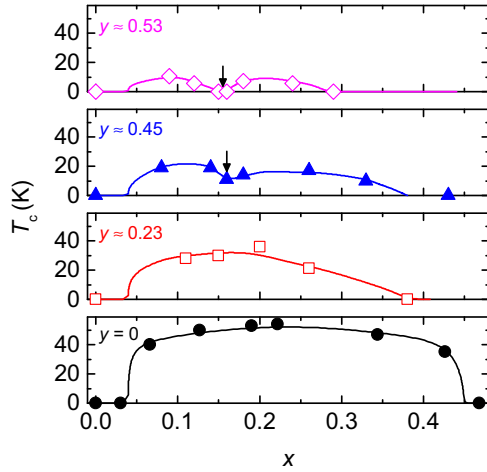


FIG. 4. (Color online) Magnetic onset  $T_c$  of  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  as a function of  $x$  with  $y = 0, \approx 0.23, \approx 0.45$ , and  $\approx 0.53$ .  $T_c$  values of  $y = 0$  samples were taken from Ref. [20].

figure. In contrast to the single dome-shaped SC region for  $y = 0$  and  $\approx 0.23$ , a small valley is found at the center of the SC region in the  $y \approx 0.45$  system. The split of the SC region was enhanced by further increase in  $y$  and the non-SC region ( $T_c < 2$  K) was found at  $x \approx 0.16$  in the  $y \approx 0.53$  series.

### C. $T_c$ valley on $x$ - $\alpha$ and $x$ - $h_{Pn}$ spaces

Figure 5(a) is an  $\alpha$  vs  $x$  plot for  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  and  $\text{LnFeAsO}_{1-x}\text{H}_x$ . The symbol size of each point represents the  $T_c$  value. Because of the chemical pressure effect of H substitution, the  $\alpha(x)$  curve of  $\text{SmFeAsO}_{1-x}\text{H}_x$  has a negative slope, and its intercept  $\alpha_0$  at  $x = 0$  increases with P or La/Ce substitution of Sm sites. Therefore, the ranges of  $\alpha(x)$  curves for  $y = 0.23$ – $0.53$  overlap with those for  $\text{Ln} = \text{La}, \text{Ce}$ . The regions of the  $T_c$  valleys for  $y = 0.45$  and  $0.53$  are denoted by a light blue area and those for  $\text{Ln} = \text{La}$  by a light pink area. Both regions are clearly located at the same area around  $(x, \alpha) = (\approx 0.16, \approx 113^\circ)$ . In other words, double dome-shaped  $T_c(x)$  curves are observed when the  $\alpha(x)$  curve has a relatively large  $\alpha_0$  ( $> 114^\circ$ ) and passes through the region around  $(\approx 0.16, \approx 113^\circ)$ . Figure 5(b) is an analogous  $x$  vs  $h_{Pn}$  plot. In  $x$ - $h_{Pn}$  space, the  $T_c$  valley for  $y = 0.45$ – $0.53$  is located at a position with different  $h_{Pn}$  (0.130 nm for  $y = 0.45$ – $0.53$ ; 0.134 nm for  $\text{Ln} = \text{La}$ ). In addition, a drop of  $T_c$  is not observed in the  $y \approx 0.23$  series, while their  $h_{Pn}(x)$  curve crosses the region of the  $T_c$  valley of  $\text{LaFeAsO}_{1-x}\text{H}_x$ . These results indicate that the emergence of a  $T_c$  valley depends on  $\alpha$  and not  $h_{Pn}$ , irrespective of the  $Pn$  anion and  $\text{Ln}$  cation species.

Finally, we discuss the effects of  $\alpha$  on  $T_c$ . Parallel to this study, we have discovered the AFM ordering of iron spins with Néel temperature  $T_N$  up to 89 K in electron over-doped  $\text{LaFeAsO}_{1-x}\text{H}_x$  ( $x = 0.45$  and  $0.51$ ) [29]. The magnetic phase has a unique noncentrosymmetric structure different from that of the conventional AFM phase located at  $x \sim 0.0$ . This result suggests that the first and the second  $T_c$  domes in  $\text{LaFeAsO}_{1-x}\text{H}_x$  come from different parent phases. Based on this idea, the result of the present study indicates that the  $T_c$  valley is a position where two SC phases encounter with

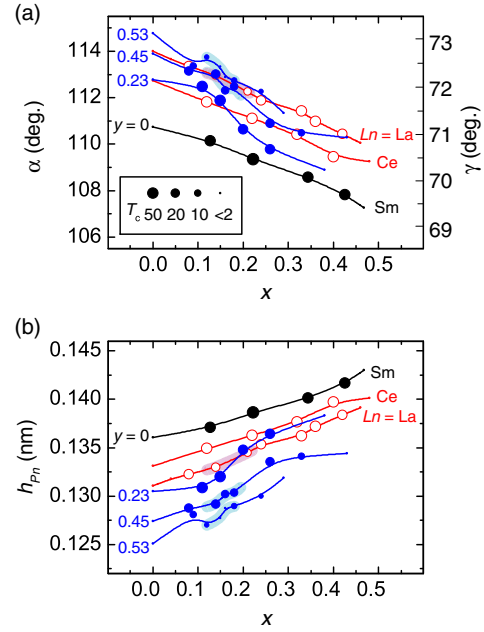


FIG. 5. (Color online) Correlation between the emergence of the  $T_c$  valley and structural or doping parameters in  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  ( $y = 0, \approx 0.23, \approx 0.45$ , and  $\approx 0.53$ ; blue solid symbols) and  $\text{LnFeAsO}_{1-x}\text{H}_x$  ( $\text{Ln} = \text{La}$  and  $\text{Ce}$ ; red empty symbols). (a)  $\alpha$  angle of  $\text{FePn}_4$  tetrahedron as a function of  $x$ . Lines are guides to the eye, tracing  $\alpha(x)$  for specified  $y$  and  $\text{Ln}$ . The symbol size indicates the  $T_c$  value. The region of the  $T_c$  valley for  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  is denoted by a light blue area around  $(x, \alpha) = (0.16, 113^\circ)$ , which overlaps with that of  $\text{LaFeAsO}_{1-x}\text{H}_x$  (denoted by a light pink colored area). (b)  $h_{Pn}$  vs  $x$  plot. The regions of the  $T_c$  valleys in each system do not overlap.

each other, and their relationship becomes noncompetitive with the decrease of  $\alpha$ . Because of the relationship between  $\alpha$  and  $T_c$  in  $\text{LnFePnO}_{1-x}\text{H}_x$ , the “pairing glue” that induces superconductivity appears to depend on  $\alpha$ . A candidate for this glue is the electron-phonon interaction associated with  $\alpha$ . Saito *et al.* suggested in their orbital fluctuation theory that electron-phonon interactions are strongest when the  $\text{FeAs}_4$  tetrahedron has a regular shape ( $\alpha = 109.5^\circ$ ) [30]. This theory can empirically explain the maximum  $T_c$  occurring when  $\alpha$  is  $109.5^\circ$ . However, in comparing the  $\alpha$  and  $x$  dependence of  $T_c$ , the resulting  $T_c$ - $\alpha$  relation is not monotonic but does depend on  $x$ . Another possibility is spin fluctuation enhanced by hopping between Fe  $3d$  orbitals via As  $4p$  orbitals. The hopping integral, i.e., the overlap integral of Fe  $3d$  and As  $4p$  orbitals, is directly determined by the Fe-As-Fe angle  $\gamma$  having a one-to-one relation with  $\alpha$ :  $\sin(\gamma/2) = 1/\sqrt{2} \cdot \sin(\alpha/2)$ . In Fig. 5(a), the right vertical axis represents the  $\gamma$  value corresponding to  $\alpha$ , and we can confirm that the  $T_c$  valley is located at  $\gamma \sim 72^\circ$ .

Very recently, a calculation using the fluctuation exchange approximation successfully reproduced the shape variation of  $T_c$ - $x$  with  $\alpha$  in a  $\text{LnFeAsO}_{1-x}\text{H}_x$  system from a double dome shape at large  $\alpha$  to a single dome shape at small  $\alpha$  [31]. The primary suggestion by this calculation is that the spin-fluctuation enhancement mechanism is different between low  $x$  and high  $x$  regions. The spin fluctuation in the former



region is caused by nesting of a Fermi surface composed of Fe  $3d_{xz,yz}$  and  $3d_{xy}$  orbitals, while that of the latter originates from real space motion of electrons within the  $3d_{xy}$  orbitals, i.e., next-nearest-neighbor diagonal hopping ( $t_2$ ) dominates the nearest-neighbor hopping ( $t_1$ ). In this scheme,  $\alpha$  plays an important role in determining the magnitude of spin fluctuations and  $T_c$  because both  $t_2$  and indirect  $t_1$  ( $t_1^{\text{indirect}}$ ) are controlled by hopping via As  $4p$  orbitals. Given the experimental findings and the theoretical real-space picture of electronic spin,  $\alpha$  is the decisive factor for the shape variation of  $T_c$ - $x$  in  $\text{LnFeAsO}_{1-x}\text{H}_x$  systems.

#### IV. SUMMARY

We found a double dome-shaped SC region in the  $x$ - $T$  diagram of  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  with  $y = 0.45$  and  $0.53$ .

This finding agrees with previous results on  $\text{LaFeAsO}_{1-x}\text{H}_x$ . By investigating the correlation between  $T_c$  and structural parameters of the  $\text{FePn}_4$  tetrahedron, we confirmed that the  $T_c$  valley separating the two SC domes is located at  $x \approx 0.16$  and  $\alpha \approx 113^\circ$  in both  $\text{SmFeAs}_{1-y}\text{P}_y\text{O}_{1-x}\text{H}_x$  and  $\text{LaFeAsO}_{1-x}\text{H}_x$  systems. These results indicate that the emergence of two SC domes is generally induced in 1111-type iron oxypnictides when the electron-doping level matches the shape of the  $\text{FePn}_4$  tetrahedron.

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