BL-8B, AR-NE1A/2013S2-002, 2016S2-004

Peculiar response to pressure on iron-based superconductor $LaFeAsO_{1-x}H_x$

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1 Introduction

Iron pnictides are new family of high-temperature superconductors, whose charge carriers play an active role primarily on the two-dimensional iron plane [1]. Pnictide (*Pn*) atomic position acts major perturbations for the 3*d* multi-orbital bands of irons through the hybridization of Fe-3*d* and *Pn*-*p* electrons. As a consequence, chemical substitution along with carrier-doping/chemical-pressure or pressurizing crystal can induce drastic alteration for solid state properties such as emerging and developing the T_c. A lot of ensembles of chemical substitution to iron pnictides have provided an empirical guide that the T_c is maximized when the FePn_4 tetrahedron approaches the regular one. Meanwhile, although pressure is a direct and clean way to modify the local geometry of FePn_4 without carrier concentration and degrading crystal by chemical substitution, the correlation of the superconductivity and the structure remains explored under pressure.

One of the significant fascinating materials in iron pnictides is $LaFeAsO_{1-x}H_x$, composed of ZrCuSiAs-type structure with alternating stacking of the conducting $FeAs_4$ and the insulating (O,H)La_4 layers. $LaFeAsO_{1-x}H_x$ exhibits a peculiar phase diagram via hydrogen anion substitution i.e. doping the electron: two superconducting domes with $T_{c,max}$ = 26 K at $x \sim 0.08$ (SC1) and $T_{c,max}$ = 37 K at $x \sim 0.35$ (SC2) and two parent phases at $x \sim 0$ (PP1) and $x \sim 0.5$ (PP2) [2]. The second SC2 and PP2 phases are barely observed among iron-based as well as copper oxide superconductors because there is usually nothing in the heavy electron-doped region. Takahashi et al. has recently identified that the applying pressure to $LaFeAsO_{0.72}H_{0.18}$ induces a notable enhancement of T_c from 18 K at ambient pressure to 52 K at 6 GPa, along with forming single SC dome from two SC domes [3]. The maximum T_c under pressure is close to the highest class of T_c = 55 K in Sm1111 among the iron-based superconductors.

2 Experiment

The synchrotron X-ray diffractions (sXRD) for x = 0, 0.20 and 0.51 were carried out at room temperature up to ~8 GPa at NE1A of PF-AR, KEK. Pressure was generated by a diamond anvil cell (DAC) with 600 μm culet diamond anvils and a gasket with 300 μm hole. The two-dimensional images collected by RIGAKU R-AXIS on the curved imaging plate were integrated to yield 2θ-intensity data. Crystal structures were refined by Rietveld method based on the space group *P*4/*nmm*, and the reliable factors in the refinements are R_{wp} = 3.9-6.6%. We moreover traced the PP2 by means of the sXRD down to 8 K up to 3.2 GPa at BL-8B of PF, KEK, measured at λ = 0.8267 Å. A 4:1 methanol-ethanol mixture was used as a pressure transmitting medium in all the experiments.

3 Results and Discussion

Figure 1 illustrates the contour plots of T_c for $\alpha_{As\text{-}Fe\text{-}As}$ and $d_{Fe\text{-}As}$ of $LaFeAsO_{1-x}H_x$ under pressure. Pressure triggers to merge two SC domes at ambient pressure into the single SC dome, along with the T_c growing up to 52 K at 6 GPa for x = 0.2, and then the T_c gently go down. In the single T_c dome region, the ridge line of T_c runs along the variation for x = 0.20 as the pressure increased.

In iron-based superconductors, the relation between the optimum T_c and the structural parameters of $FeAs_4$ are hitherto proposed: the highest T_c realizes on the $\alpha_{As\text{-}Fe\text{-}As}$ = 109.5° as a regular tetrahedron of $FeAs_4$ or $h_{As} \sim 1.38$ Å. Actually, $SmFeAsO_{0.78}H_{0.22}$ with the highest class T_c = 55 K in iron pnictides gives nearly ideal values of the $\alpha_{As\text{-}Fe\text{-}As}$ = 109.3° and the h_{As} = 1.386 Å at ambient pressure. Moreover, the values of $\alpha_{As\text{-}Fe\text{-}As}$ for $BaFe_2As_2$ and LiFeAs act toward and away from the regular tetrahedron of $FeAs_4$ along with increasing and decreasing the T_c, respectively. Meanwhile, the present results that the values of the $\alpha_{As\text{-}Fe\text{-}A}$ and the h_{As} deviate from the optimum values with pressure reveal the inconsistent to the guides for developing the T_c. From the electronic state calculations of $LaFeAsO_{0.8}H_{0.2}$, the Fermi surface topologies are unaltered on the application of pressure. Additionally, the broadening of the band width with pressure is observed because of the shrinkage of $d_{Fe\text{-}As}$, resulting in the decrease of the spin-fluctuation hence the T_c going down. The aforementioned findings might be simply unaccountable for the rise of T_c under pressure. We consequently require an otherwise hint to the enhancement of T_c.

Both parent phases indicate tetragonal to orthorhombic structural transitions (T_s), subsequently an antiferro-magnetic orderings (T_N). The parent phase of PP2 is uniquely observed in the 1111 series. Inset of Fig. 2 shows the temperature dependence of the full width at

half maximum (FWHM) of 220 reflection for x = 0.51, which is a good indicator to pursuit the structural transition. The T_{S2}'s are indicated by arrows estimated from the power law fitting. At 0.5 and 0.9 GPa, the broadenings of FWHMs start moderately in high-temperature region as extensive fluctuations in vicinity of the structural transitions. At 1.5 GPa, the FWHM exhibits the slight broadening at low-temperature, not implying the structural transition but the precursory phenomenon near the phase boundary. Any broadening was not observed at 3.2 GPa. Takahashi et al. presented that an anomaly in resistivity, corresponding to T_{N2}, for x = 0.51 was suppressed near 2 GPa. This denotes that the T_{N2} as well as the T_{s2}, i.e. the PP2 was obscured by low-pressure although the PP1 being up to 20 GPa (T_{N1}) and 30 GPa (T_{s1}) is robust to pressure.

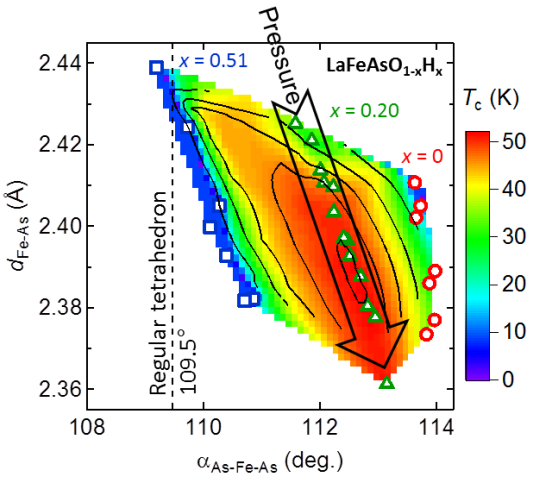

Fig. 1. Contour plots of T_c for $LaFeAsO_{1-x}H_x$ as a function of the As–Fe–As bond angle ($\alpha_{As\text{-}Fe\text{-}As}$) and the Fe–As distance ($d_{Fe\text{-}As}$). Broken lines represent the regular tetrahedron in $FeAs_4$ [4].

Figure 2 illustrates the phase diagram at ambient and high-pressure for $LaFeAsO_{1-x}H_x$. The distinct responses to pressure for both parent phases are unexpected and presumably arise due to their different origins. Because the origin of PP1 is interpreted as a spin density wave with Fermi surface nesting, its robust behavior of PP1 against pressure is consistent with insensitive change of the Fermi surface. In contrast, the origin of PP2 is seemingly less relevant to the Fermi surface topology. Iimura et al. performed an electronic state calculation based on the molecular orbital concept for $LaFeAsO_{1-x}H_x$ [5]. In the low x region, the bonding and antibonding states of the Fe-3d and As-4p orbitals give a large gap. This gap decreases rapidly with an increase in h_{As}, namely a decrease in the hybridization of Fe and As orbitals, making the non-bonding Fe-3d_{xy} orbital the half-filled state. The resulting state implies that the orbital selective Mott state on Fe-3d_{xy} is realized in the higher x region, i.e. in PP2, for $LaFeAsO_{1-x}H_x$. Contrary to electron doping with increasing x, the pressure induces the rapid decrease of h_{As}. Therefore, in contrast to PP1, PP2 can be easily broken by pressure.

Generally in high-T_c superconductors, the nature of the parent phase is strongly related to the superconducting state; that is, fluctuations derived from the parent phase may drive the pairing of the superconducting electrons. Thus, the origins of SC1 and SC2 adhering to PP1 and PP2, respectively, is considered as fluctuations from PP1 and PP2. The former is the itinerant spin-fluctuation, and the latter is another type spin-fluctuation with a strongly localized character of the orbital selective Mott phase. Because PP2 disappears easily with pressure, the fluctuation from PP2 might be sensitive to pressure. Consequently, we would suggest that the significant enhancement of T_c under pressure is due to the favorable change of the latter fluctuation.

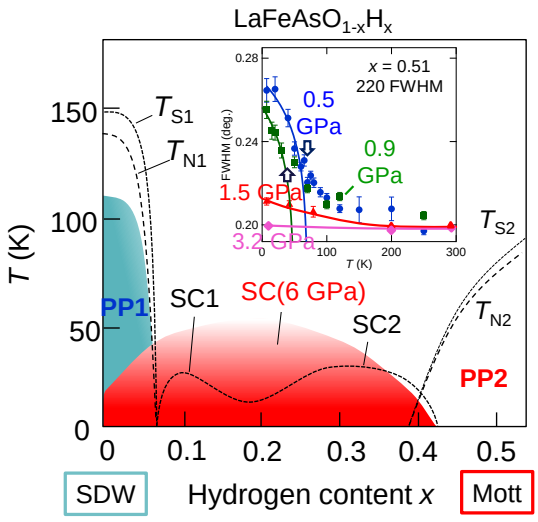

Fig. 2. T vs x phase diagram of the parent phases (PP) with structural (T_S) and magnetic (T_N) transitions, and superconducting phases (SC) at 0 and 6 GPa for $LaFeAsO_{1-x}H_x$. The PP1 and SC at 6 GPa are blue and red, respectively [4]. Inset shows the temperature dependence of FWHM on the 220 reflection under pressures.

References

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