

## Bipartite magnetic parent phases in the iron oxypnictide superconductor

Jun-ichi Yamaura<sup>1</sup>, Reiji Kumai<sup>2</sup>, Youichi Murakami<sup>2</sup><sup>1</sup> Materials Research Center for Element Strategy, Tokyo Institute of Technology, Yokohama, Kanagawa 226-8503, Japan,<sup>2</sup> Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), Tsukuba, Ibaraki 305-0801, Japan

A new class of high- $T_c$  superconductor iron-based materials has been widely studied since the discovery in 2008. In the first obtained iron-based superconductor, ZrCuSiAs-type LaFeAsO, the superconductivity emerges via doping the carrier to the non-doped parent compound with magnetic- and structural-ordered states. An advanced doping method using a hydrogen anion instead of fluorine in LaFeAsO surpassed the doping limit of fluorine, and uncovered the second superconducting phase (SC2) ( $T_{c,max} = 36$  K at  $x \sim 0.35$ ), following to the first dome (SC1) ( $T_{c,max} = 26$  K at  $x \sim 0.1$ ) [1]. To determine whether a certain hidden phase exists beyond the SC2 region, we performed X-ray powder diffraction measurements for LaFeAsO<sub>1-x</sub>H<sub>x</sub> on the beamlines of 8A/8B at KEK-PF.

Figure 1(a) shows the X-ray profile of the (2, 2, 0)<sub>T</sub> reflection in the non-superconducting state with  $x = 0.51$ . On cooling, the peak of (2, 2, 0)<sub>T</sub> was broadened, while, any broadening of the (0, 0, 6)<sub>T</sub> and (2, 0, 0)<sub>T</sub> peaks were not observed [2]. The experimental findings suggest that the tetragonal-orthorhombic structural transition emerges clearly in  $x = 0.51$ . Figure 1(b) shows the temperature dependence of the lattice parameters for  $x = 0.51$ . The  $a$ -axis length splits in two below the structural transition of  $T_s \sim 95$  K, and the  $c$ -axis shows an upturn at  $T_s$ . The X-ray powder-diffraction structural analysis with RIETAN-FP program [3] using high-resolution synchrotron radiation source provides the interesting result that the  $x = 0.51$  compound crystallizes in an orthorhombic non-centrosymmetric  $Aem2$  structure below  $T_s$  in contrast to the centrosymmetric  $Cmme$  structure for  $x = 0$ . Across the structural transition, only the As atoms moves slightly in  $x = 0$ , while, the Fe and As atoms move in antiphase in  $x = 0.51$  as shown in the inset of Fig. 1(c).

In the neutron powder diffraction measurement for  $x = 0.51$ , we observed magnetic peaks below  $T_N = 89$  K with the propagation vector of  $\mathbf{q} = (1/2, 1/2, 0)$ . The magnetic structure analysis reveals an exceptional stripe-type arrangement among the iron-pnictides as shown in the inset of Fig. 1(d). The magnetic moment  $1.2 \mu_B$  per iron atom is significantly larger than the value of  $0.63 \mu_B$  for  $x = 0$ . In the muon spin relaxation measurements, we observed that the value of  $T_N$  and the magnetic volume fraction decrease in unison with decreasing the hydrogen contents from  $x = 0.51$ .

Figure 1(d) illustrates the comprehended phase diagram of LaFeAsO<sub>1-x</sub>H<sub>x</sub> [2]. We note a new antiferromagnetic phase with the structural transition next to the superconducting phase that can be regarded as the

doped parent phase by the analogy of the non-doped antiferromagnetic ordered phase with the structural transition. This is unexpected result because the magnetic and electronic interactions are usually perceived as being weak by highly carrier-doping. We would account that in the iron-pnictides the parent compound not only refers to the non-doped compound but also more generally indicates a certain critical phase of the magnetic and electronic correlations. We can, moreover, definitely state that the two SC domes come from carrier-doping to the left- and right-hand parent compounds towards the intermediate region of the phase diagram.

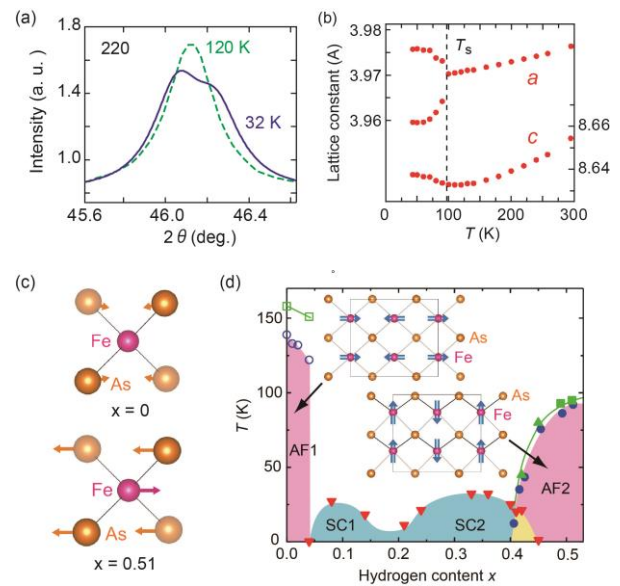


Fig.1 (a) X-ray profiles of (2, 2, 0)<sub>T</sub> reflections for LaFeAsO<sub>0.49</sub>H<sub>0.51</sub> at 120 K and 32 K. (b) Temperature dependence of the lattice constant for LaFeAsO<sub>0.49</sub>H<sub>0.51</sub>. (c) Schematic of the atom displacements across the structural transition for  $x = 0$  and 0.51. (d) Phase diagram of LaFeAsO<sub>1-x</sub>H<sub>x</sub>. AF1 and AF2 represent the original and the advanced parent phases, respectively.

## References

- [1] S. Iimura *et al.*, *Nature Commun.* **3**, 943 (2012).
- [2] M. Hiraishi *et al.*, *Nature Phys.* **10**, 300 (2014).
- [3] F. Izumi and K. Momma, *Solid State Phenom.* **130**, 15 (2007).

\* jyamaura@lucid.msl.titech.ac.jp