BL-28A/2015S2-003, 2016G096

ARPES study of Te-annealed 11-type iron-based superconductor $FeTe_{1-x}Se_x$

Keisuke KOSHIISHI[1,*], Takumi. OTSUKA[2], Suguru. NAKATA[1], Kenta. HAGIWARA[1], Chun. LIN[1], Yuxuan. WAN[1], Hiroshi. KUMIGASHIRA[3], Kanta. ONO[3], Koji. HORIBA[3], Takao. WATANABE[2] and Atsushi. FUJIMORI[1]

[1] Department of Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo, 113-0033, Japan

[2] Graduate School of Science and Technology, Hirosaki University, 3, Bunkyo-cho, Hirosaki-shi, Aomori-ken, 036-8561, Japan

[3]Photon Factory, High Energy Accelerator Research Organization, 1-1 Oho, Tsukuba, Ibaraki, 305-0801, Japan

1 Introduction

11-type iron-based superconductors have the simplest crystal structure in which layers consisting of Fe atoms tetrahedrally coordinated by chalcogens (e.g. Te, Se) stack. The parent compound FeTe shows a bi-collinear double-stripe antiferromagnetic (AFM) order with the wave vector $\boldsymbol{Q}$=(π/2, π/2), which is distinct from the usual collinear magnetic structure observed in other parent compounds like $BaFe_2As_2$ [1]. As tellurium in the parent compound is gradually replaced by selenium, the AFM order is suppressed and superconductivity is observed. It has been reported that the superconductivity is affected by excess Fe located at interstitial sites and very sensitive to its stoichiometry. By annealing a specimen, the excess Fe can be removed, and then the superconductivity can be improved. [2,3] The superconductivity with the superconducting transition temperature (T_C) of ~10 K is observed in a wide low Se-concentration region from x = 0.05 up to 0.5 and the maximum bulk T_C reaches 14.5 K at x ~ 0.4. According to recent transport measurement for Te-annealed sample [4], although Hall coefficient (R_H) of annealed $FeTe_{0.6}Se_{0.4}$ is almost independent of temperature above 50 K, it decreases with decreasing temperature below 50 K and eventually changes their sign from positive to negative around 30 K, indicating multi-carrier features of holes and electrons in optimally doped samples. Moreover, the strong temperature dependence with the sign change suggests that band-specific pseudo gaps may exist. On the other hand, for $FeTe_{0.8}Se_{0.2}$, R_H is always positive similar to the case of as-grown samples. Naively, this behavior indicates that contribution to the transport properties dominantly comes from hole carriers in contrast to the optimally doped sample. However, since Fe-based superconductor has multi-orbital nature because the 3*d* electrons of Fe form bands near the Fermi level, it is not clear whether such simple speculation is valid or not. Therefore, we have performed angle-resolved photoemission spectroscopy (ARPES) measurements on $FeTe_{1-x}Se_x$ in order to investigate differences in the electronic structure between the optimally doped and low Se-doped samples and the temperature dependence of their electronic structure.

2 Experiment

High-quality single crystals of $FeTe_{1-x}Se_x$ were grown using the Bridgman method and annealed in a tellurium vapor ("Te-anneal"). To check the T_c and the susceptibility, magnetization measurements were carried out by using a Magnetic Property Measurement System-5S (Quantum Design, Co., Ltd.) at The University of Tokyo Cryogenic Research Center. ARPES measurements were performed at beamline 28A of Photon Factory using circularly polarized light with the photon energy of 54 eV corresponding to $k_z \approx 0$. A SIENTA SES-2002 electron analyzer was used with the total energy resolution of ~ 20 meV. The crystals were cleaved *in situ* below T = 20 K, and the measurements were carried out in an ultrahigh vacuum of ~ 9×10^{-11} Torr.

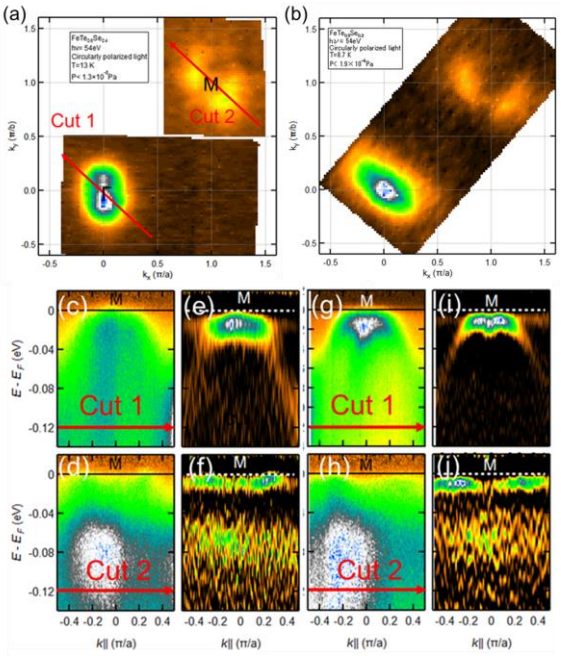

Fig. 1: ARPES results for $FeTe_{1-x}Se_x$ (a), (b) Fermi surface mapping for x = 0.4 and 0.2, respectively. (c), (d) Energy-momentum plots for x = 0.4 divided by the Fermi-Dirac function along cut 1 and cut 2 depicted by red arrows in panel (a), respectively. (e), (f) Second derivative of (c) and (d) with respect to energy, respectively. (g), (h), (i), (j) Same as (c), (d), (e) and (f), respectively, except for x = 0.2.

3 Results and Discussion

ARPES results are shown in Fig. 1. Intensity map at the Fermi level for x = 0.4 integrated from E_B = -10 meV to E_B=10 meV is shown in Fig. 1(a). The electron pocket

around the M point and the strong intensity around the Γ point coming from hole bands can be observed. ARPES data for x = 0.4 are shown in Figs. 1(c) and (d). We also show their second derivative images with respect to energy in Figs. 1(e) and (f), respectively, in order to highlight the band structure. Three hole bands and one electron band can be observed. At least one of the three hole bands seems to cross the Fermi level, forming a tiny hole pocket around the Γ point as mentioned above. For x = 0.2 (Figs. 1(g)-(j)), one can observe not only the hole bands around the Γ point but also the electron band around the M point similar to the band structure for x = 0.4, and the electron band forms a sizable electron pocket at the corner of the Brillouin zone (Fig. 1(b)). Interestingly, the heaviest hole band assigned to the d_{xy} band by the previous calculation [5] is crossing the Fermi level only in the case of x = 0.2, and then it is supposed to make a large Fermi surface around the Γ point although it is difficult to observe that Fermi surface in the intensity map of Fig. 1(b). This results suggest that the large hole pocket appears and that mainly hole carriers play a role in conductivity only in the case of x = 0.2, which is consistent with the tendency of R_H. Moreover, considering that the superconducting transition temperature scarcely depends on Se concentration over a wide doping region although there are large differences in the Fermi surfaces, one can suggest that the large hole pocket consisting of the d_{xy} orbital may possibly make only minor contribution to the superconductivity in $FeTe_{1-x}Se_x$.

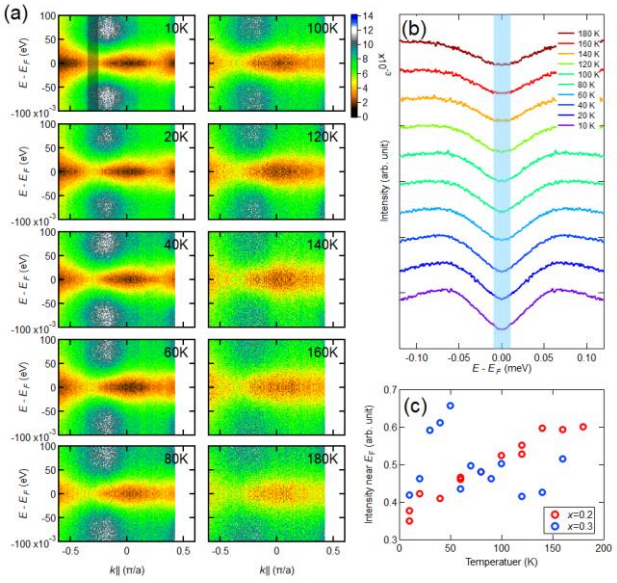

Fig. 2: Temperature evolution of ARPES spectra of $FeTe_{1-x}Se_x$ (a) Energy-momentum plots for x = 0.2 along cut 2 depicted by red arrows in Fig.1 (a). They are symmetrized with respect to the Fermi level. (b) Temperature evolution of EDC around k_F of x = 0.2. They were obtained by integrating EDCs in the shaded area in panel (a). (c) Temperature dependence of the intensity around the Fermi energy (blue area in panel (b)). Red and blue markers correspond to x = 0.2 and 0.3, respectively.

We have also measured the temperature dependence of electronic states near the Fermi level. Figure 2(a) shows the temperature evolution of symmetrize ARPES spectra around the M point for x = 0.2. One can see the electron band near the Fermi level and identify the position of k_F at all temperatures. To see the energy gap features, EDCs integrated around k_F are plotted in Fig. 2(b). Due to the small size of the superconducting gap of ~1.6 meV at 2.5 K [6] compared to the energy resolution of ~17 meV, the coherence peak cannot be distinguished even at 10 K. However, the intensity proportional to the density of states around E_F seems to be enhanced with increasing temperature, indicating an energy gap closure. For more qualitative comparison, we have plotted the intensity near E_F as a function of temperature in Fig. 2(c). Red and Blue circles correspond to x=0.2 and 0.3, respectively. The RH of x = 0.3 shows temperature dependence similar to x = 0.4 although R_H does not change sign at low temperatures. As for x = 0.2, the intensity grows monotonically with increasing temperature, and eventually saturates around 150 K. This result indicates that pseudo gap opens above T_c for x = 0.2. On the other hand, behavior for x = 0.3 is not monotonic. The intensity increases with temperature in the low temperature region but drastically drops around 50 K, suggesting that somehow a pseudo gap on the electron pocket opens only above ~50 K. Assuming a pseudo gap opening on the electron Fermi surface above ~50 K, the strong temperature dependence of R_H can be explained. Namely, if a pseudo gap opens only on the electron Fermi surface above ~50 K, hole carriers would be dominant in the high temperature region and electron carriers may recover with the closure of the pseudo gap below ~50 K. Thus one can understand why R_H takes positive values above ~50 K and undergoes strong temperature dependence below ~50 K.

The same temperature dependence of R_H has been reported even for FeSe under high pressure [7]. In that case, the dominant carriers can be changed from electrons to holes by applying pressure even though the application of pressure does not introduce extra carriers into the system, and the authors have concluded that the temperature dependence of Hall resistivity is due to Fermi surface reconstruction with antiferromagnetic ordering. However, in the present case, we could not observe any evidences for antiferromagnetic order such as band folding, and so far antiferromagnetic nature has been observed by ARPES only in FeTe [8]. Therefore, it might be difficult to conclude that the pseudo gap on the electron pockets of superconducting compounds is derived from antiferromagnetic order. On the other hands, antiferromagnetic spin correlations with stripe-type antiferromagnetic wave vector $\boldsymbol{Q}_{SAF}$ = (π, 0) in $FeTe_{1-x}Se_x$ has been observed by inelastic neutron scattering measurements [9]. In order to clarify the origin of pseudo gap and investigate effects of magnetic fluctuation on electronic state near Fermi level in $FeTe_{1-x}Se_x$, further work is necessary

References

[1] S. Li *et al.*, Phys. Rev. B **79**, 054503 (2009).

[2] Y. Sun *et al*., Sci. Rep. **6**, 32290 (2016)
[3] Y. Koshika *et al*., J. Phys. Soc. Jpn. **82**, 023703 (2013)
[4] T. Otsuka *et al*., 2016 JPS autumn meeting
[5] L. C. C. Ambolode, II *et al*., Phys. Rev. B **92**, 035104 (2015).
[6] K. Okazaki *et al*., Phys. Rev. Lett. **109**, 237011 (2012).
[7] J. P. Sun *et al*., Phys. Rev. Lett. **118**, 147004 (2017).
[8] Y. Zhang *et al*., Phys. Rev. B **82**, 165113 (2010).
[9] Z. Xu *et al*., Phys. Rev. B **93**, 104517 (2016).

* keisuke@wyvern.phys.u-tokyo.ac.jp