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# Angel-resolved photoemission spectroscopy of Te-annealed 11-type iron-based superconductor $FeTe_{1-x}Se_x$

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## 1 Introduction

11-type iron-based superconductor has the simplest crystal structure in which layers consisting of Fe atoms tetrahedrally coordinated by chalcogens (e.g. Te, Se) stack altanatively. The parent compound FeTe has a bi-collinear double-stripe antiferromagnetic (AFM) order with the wave vector $\boldsymbol{Q}$=($\pi$/2, $\pi$/2) that is distinct from the usual collinear magnetic structure observed in other parent compounds like $BaFe_2As_2$ [1]. As tellurium is gradually replaced by selenium in the parent compound, AFM order is suppressed and superconductivity is observed. It has been reported that the superconductivity is disrupted by excess Fe atoms located at interstitial sites and is very sensitive to its stoichiometry. By annealing the specimen, the excess Fe can be removed and superconductivity is recovered [2,3]. Superconductivity with the superconducting transition temperature ($T_C$) of ~10 K is observed in a wide low-concentration region and the maximum bulk $T_C$ reaches 14.5 K at x~0.4. According to recent transport measurement on Te-annealed samples [4], The Hall coefficient ($R_H$) of annealed $FeTe_{0.6}Se_{0.4}$ changes from positive to negative with decreasing temperature, indicating that there are both hole and electron carriers in the optimally doped samples. On the other hand, for $FeTe_{0.8}Se_{0.2}$, $R_H$ is always positive similar to the case of as-grown samples. Thinking naively, this behavior indicates that contribution to transport properties predominantly comes from hole carriers in contrast to the optimally doped sample. Indeed, similar behavior in $R_H$ of FeSe under pressure has been reported [5]. However, the Fe-based superconductor has multi-orbital nature because 3*d* electrons of Fe form energy bands near the Fermi level. Therefore, it is not clear whether such simple picture is valid or not. Therefore, we have performed angle-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.

## 2 Experiment

High-quality single crystals of $FeTe_{1-x}Se_x$ were grown using the Bridgman method and annealed under 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 system (Quantum Design) 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 $k_z$ = 0. A SIENTA SES-2002 electron analyzer was used. The total energy resolution was ~ 20 meV. The crystals were cleaved *in situ* below $T$ =20 K, and the measurements were carried out in an ultrahigh vacuum of ~ $9\times10^{-11}$ Torr.

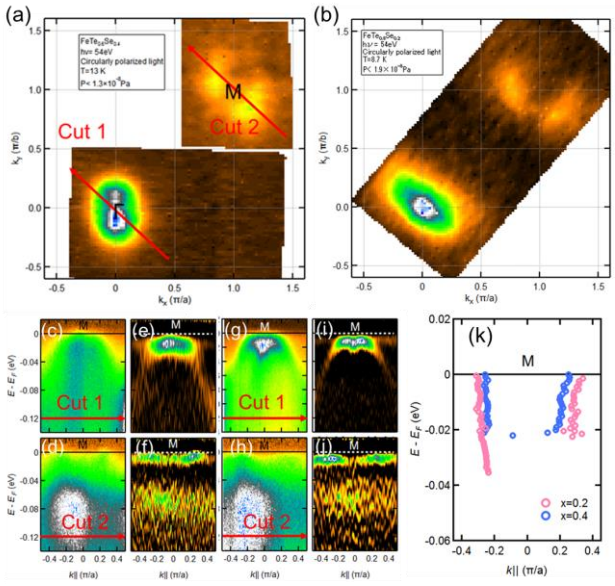


Fig. 1: ARPES results of $FeTe_{1-x}Se_x$. (a), (b) Fermi surface mapping for x=0.4 and 0.2, respectively. (c), (d) Energy-momentum plots for x=0.4 divided by Fermi-Dirac function along cut 1 and cut 2, respectively, depicted by red arrows in panel (a). (e), (f) Second derivatives of (c) and (d), respectively,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

## 3 Results and Discussion

ARPES results are shown in Fig. 1. The 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). An electron pocket around the M point and strong intensity around the Γ point coming from hole bands can be observed. ARPES data for $x$=0.4 can be seen in Figs. 1(c) and (d). We also show the second derivatives of them with respect to energy in Figs. 1(e) and (f), respectively, in order to

highlight the band dispersions. 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)). In order to compare the band structures between the two compositions, we plot the peak positions of the momentum distribution curves (MDC) peak in Fig. 1(k). We find that Fermi wave vector derived from the MDC peaks changes with varying Se contents, and that the electron pocket of the low Se-concentration sample is larger than that of the optimally doped sample. These results suggest that the low Se-doped compounds as well as the optimally doped compound can be regarded as a multiband system. It has been reported that Hall coefficient of some electron-doped cuprates undergoes a sign reversal as a function of temperature in spite of the fact that the Fermi surface of the electron-doped cuprates is composed of a large hole pocket. The phenomena is, therefore, attributed to the scattering of carriers by antiferromagnetic fluctuations, and a theoretical study taking into account the vertex correction due to antiferromagnetic fluctuations can reproduce the temperature dependence of $R_H$ for the electron-doped cuprates [6]. Actually, the parent compound has an AFM ordering, and spin correlations with the double stripe-type antiferromagnetic wave vector $\boldsymbol{Q}_{\mathbf{DSAF}}$=(π/2, π/2) and those with the stripe-type antiferromagnetic wave vector $\boldsymbol{Q}_{\mathbf{SAF}}$= (π, 0) has been observed by inelastic neutron scattering measurements [7]. Even in a previous study on FeSe under pressure, correlation between the tendency of $R_H$ with a hole-dominated character and enhanced spin fluctuations is pointed out. Thus the magnetic scattering may play an important role in the transport properties of $FeTe_{1-x}Se_x$. However, it should be mentioned that the sign reversal in the $R_H$ of $FeTe_{0.6}Se_{0.4}$ is opposite to that in the case of the electron-doped cuprates. Furthermore, although 11-type iron-based superconductors are the system close to a Mott insulator due to sizable renormalization factor of the compounds [8], $FeTe_{0.6}Se_{0.4}$ is different from the cuprate in that both electron and hole pockets exist. Hence, it is not clear whether magnetic fluctuations alone can explain the temperature dependence of the transport properties in $FeTe_{1-x}Se_x$. Further studies are needed to investigate the effects of magnetic fluctuations on the electronic states near the Fermi level related to the transport properties by estimating the scattering rate of quasi-particles.

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