

## 論文の内容の要旨

### **Angle-resolved photoemission spectroscopy studies of electronic structure and its relationship to transport properties of iron-based superconductors**

(角度分解光電子分光による鉄系超伝導体の電子状態と輸送特性の関連の研究)

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Since superconductivity in Fe-based compounds was discovered in 2008, many efforts have been made to understand the mechanism of the unconventional superconductivity. Iron-based superconductors (FeSCs) are mainly classified into six categories according to their structures. One of them is the “122”-type FeSCs, which have been studied most extensively. In addition, ”11”-type FeSCs are also one of the most extensively studied FeSCs because of their simple crystal structures and the absence of antiferromagnetic order. So far, in these compounds, these phenomena have been found and to investigate their origins and the relationship between the non-trivial phenomena and superconductivity is one of the main issues in order to understand the mechanism of the unconventional superconductivity in FeSCs. In the present thesis, we have studied the electronic structures of both classes of FeSCs, the ”122”- and ”11”-type FeSCs, by photoemission spectroscopy in order to clarify their electronic states and relationship to the non-trivial transport properties.

Photoemission spectroscopy is the most powerful technique, which allows us to directly investigate the electron states in the strongly correlated materials. In this thesis, we have investigated the electronic structures of FeSCs by angle-resolved

photoemission spectroscopy (ARPES), resonant photoemission spectroscopy (RPES), and X-ray photoemission spectroscopy (XPS).

It is known that Cr substitution in the 122-type  $\text{BaFe}_2\text{As}_2$  does not induce superconductivity in spite of the fact that, according to the rigid-band model, the Cr substitution corresponds to hole doping as in the case of Mn doping, in contrast to K doping. Its electronic structure has not been studied and the origin of the absence of the superconductivity is still unclear. Furthermore, the Cr-doped Ba122 shows sign change of the in-plane resistivity anisotropy with Cr content. The mechanism of the non-trivial sign change of the in-plane resistivity anisotropy is also uncertain.

In Chapter 3,  $\text{Ba}(\text{Fe}_{0.81}\text{Cr}_{0.19})_2\text{As}_2$  has been studied by RPES in order to investigate the electronic structure of Cr 3*d* electrons. Since RPES is an element-specific measurement, RPES is very useful to investigate the doped Cr 3*d* states. The Fermi edge has been observed in the Cr 3*d* PDOS and the Cr 3*d* electrons have itinerant features. Therefore,  $\text{Ba}(\text{Fe}_{0.81}\text{Cr}_{0.19})_2\text{As}_2$  is considered to be an effectively hole-doped system. However, the Cr 3*d* PDOS is mainly located at  $\sim 1$  eV below  $E_F$ , indicating the localization of the Cr 3*d* electrons. Then the high-spin state at the Cr site is realized by the on-site Coulomb interaction and Hund's coupling. In Chapter 4, we have studied the electronic structure of  $\text{Ba}(\text{Fe}_{1-x}\text{Cr}_x)_2\text{As}_2$  with varied Cr concentration ( $x=0.06, 0.09, \text{ and } x=0.19$ ) by ARPES in order to investigate the Fe 3*d* band affected by the Cr substitution and its relationship to the sign change of the in-plane resistivity anisotropy. Upon Cr substitution, the Fermi level moves downward exhibiting upward band dispersion shift without significant changes of dispersion, indicating sufficient hole doping with the Cr substitution. Moreover, the Fermi surface topology changes from that of the parent compound. We concluded that the changes of the Fermi surface topology play an important role in the sign reversal of the in-plane resistivity anisotropy.

For "11"-type FeSCs,  $\text{FeTe}_{1-x}\text{Se}_x$  have been studied. It is well established that as-grown  $\text{FeTe}_{1-x}\text{Se}_x$  does not exhibit superconductivity due to interstitial Fe atoms. After annealing, the excess Fe atoms are removed and the superconductivity recovers. Interestingly, depending on the Se content, annealed samples show dramatically different temperature dependence of the Hall coefficient. Especially, in this study, the samples were annealed in Te vapor in order to remove the excess Fe atoms completely.

In chapter 5, we have studied the doping-evolution of the electronic structure in

Te-annealed  $\text{FeTe}_{1-x}\text{Se}_x$  ( $x=0.15, 0.2, 0.3, \text{ and } 0.4$ ) at low temperature by ARPES in order to clarify the electronic states of  $\text{FeTe}_{1-x}\text{Se}_x$ , in which the excess Fe atoms are completely removed. Upon increasing Se content, the  $\gamma$  band shifts downward and induces disappearance of the outer large hole pockets around the  $\Gamma$  point. We concluded that the changes of the Fermi topology and mass renormalization play an important role in the sign reversal of the Hall coefficient. In chapter 6, the temperature dependence of the electronic states of Te-annealed  $\text{FeTe}_{1-x}\text{Se}_x$  ( $x=0.2$  and  $0.4$ ) has been studied by ARPES in order to investigate the relationship between the electronic structure and the non-trivial magneto-transport properties. Upon increasing temperature, the coherent peak of the electron band around the zone corner weakens and disappears above 100 K in the optimally-doped compound in contrast to the temperature independent behavior of it for Se-underdoped compound, indicating the disappearance of the electron pockets. Moreover, the possible changes of the Fermi surface topology predicted from present results can explain the temperature dependence of the Hall coefficient.