

Analysis on Photoemission Spectrum of Superconducting FeSe

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Abstract

In this paper, we present the result of soft x-ray photoemission spectroscopy and its comparison with the density functional calculation. Although local density approximation seems to be a good starting point for describing the electronic structure of FeSe, the simulated spectrum poorly reproduced the structure around $E_B = 2$ eV. This result suggests the necessity of theoretical treatment beyond local density approximation.

Key words: iron chalcogenide superconductor, FeSe, photoemission spectroscopy, band calculation

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1. Introduction

FeSe has the simplest crystal structure and the least number of elements among the recently-found iron based superconductors [1] although it still composes of tetrahedrally-bounded (Fe-Se) layers. While there are several reports on the electronic correlations in the family of iron based superconductors [2, 3], one may consider that the simplicity in FeSe yields the easiness of theoretical handling, and studying the electronic structure of FeSe can provide the further insights into the correlation effects. Based on this motivation, we studied the electronic structure of FeSe using soft x-ray photoemission spectroscopy. Since we present the basic information of electronic structure in the previous work [4], we focus on the comparison of experimental spectra with the result of band calculations.

2. Experimental

We synthesized our FeSe samples using the standard solid state reaction method as described elsewhere [5]. Here we comment on the quality and the stoichiometry of our samples in reference to the validity of our

photoemission data. Although we previously assumed the selenium deficiency in our samples [4, 6], there exist the lack of consensus on the stoichiometry of FeSe and the presence of excess iron in the form of oxide or iron metal has been claimed for off-stoichiometric samples [7]. However, while our sample may contain such impurity phases, the effect of such trace impurity phases on our photoemission data should be insignificant. In fact, signal of O 1s was not detected in the wide scan.

Our photoemission measurements were performed at two different sites: BL-5 (Okayama University Beamline) of Hiroshima Synchrotron Radiation Center ($h\nu=140$ eV, $\Delta E=200$ meV, @ Room Temp.) and BL27SU of SPring-8 ($h\nu=1100$ eV, $\Delta E=180$ meV, @ 13 K). Samples were fractured *in situ*, and binding energies of the spectra were calibrated in reference to the Fermi edge of a gold film (or molybdenum) evaporated near the sample. The base pressure of main chamber was kept better than 5×10^{-10} torr in these experiments.

In order to obtain some insight on the effect of electronic correlations, we simulated the experimental photoemission spectrum using the calculated electronic partial densities of states (DOS) for FeSe [4], where local density approximation (LDA) is employed. The calcu-

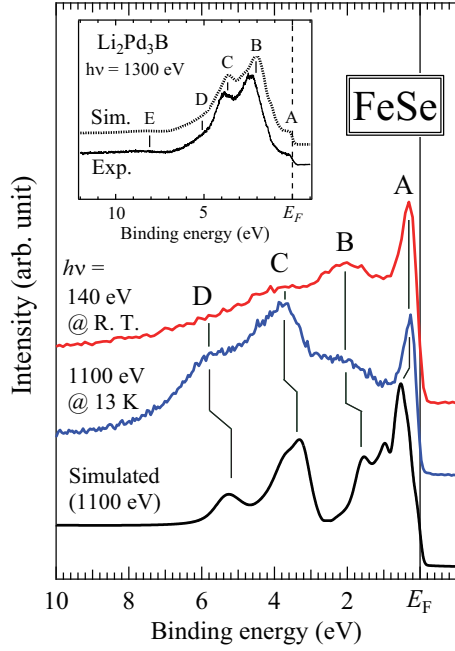


Figure 1: (Color online) Valence band spectrum of FeSe measured at room temperature with photon energy of 140 eV and the one measured at 13 K with soft x-rays of 1100 eV. Comparison of experimental photoemission spectrum with the one simulated from the calculated partial density of states is also presented. (Inset) Similar comparison for the case of $\text{Li}_2\text{Pd}_3\text{B}$. [9]

lated partial DOS were summed taking photoionization cross sections [8] into account. Then, we convoluted the sum with energy-dependent Lorentzian function ($\alpha = 0.10$) and Gaussian function (FWHM = 180 meV) to incorporate reasonable life-time broadening and experimental resolution, respectively.

3. Discussion

Figure 1 shows the photoemission spectra taken at two different photon energies together with the simulated spectrum for $h\nu = 1100$ eV. It is noted here that feature **A** and **B** should correspond to Fe d -state; feature **C** shows hybridized state of Fe d -state and Se p -state; and feature **D** is mainly Se p -state. This assignment has been confirmed by the photon-energy dependence of photoemission spectra presented above where feature **A** and **B** enhances in $h\nu = 140$ eV spectrum [4].

One can notice that positions of these four major peaks slightly deviated from the ones of the simulation, from which weak or moderate renormalization of band structures is implied. Moreover, the structure around $E_B = 2$ eV is poorly reproduced by the simulation. This re-

sult is in sharp contrast with the case of $\text{Li}_2\text{Pd}_3\text{B}$ [9] or $\text{Mg}_{10}\text{Ir}_{19}\text{B}_{16}$ [10] where LDA calculation successfully reproduces the experimentally observed spectrum (For $\text{Li}_2\text{Pd}_3\text{B}$, see the inset). Therefore, our result suggests that we may need to consider the treatment beyond LDA to successfully explain the electronic structure of FeSe. A different approach in treating electronic correlations, such as LDA+DMFT, is encouraged.

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References

- [1] F.-C. Hsu, J.-Y. Luo, K.-W. Yeh, T.-K. Chen, T.-W. Huang, P. M. Wu, Y.-C. Lee, Y.-L. Huang, Y.-Y. Chu, D.-C. Yan, M.-K. Wu, *Proc. Natl. Acad. Sci. U.S.A.* **105** (2008) 14262.
- [2] W. L. Yang, A. P. Sorini, C.-C. Chen, B. Moritz, W.-S. Lee, F. Vernay, P. Olalde-Velasco, J. D. Denlinger, B. Delley, J.-H. Chu, J. G. Analytis, I. R. Fisher, Z. A. Ren, J. Yang, W. Lu, Z. X. Zhao, J. van den Brink, Z. Hussain, Z.-X. Shen, T. P. Devereaux, *Phys. Rev. B* **80** (2009) 014508.
- [3] S. L. Skornyakov, A. V. Efremov, N. A. Skorikov, M. A. Korotin, Yu. A. Izyumov, V. I. Anisimov, A. V. Kozhevnikov, D. Vollhardt, *arXiv:0906.3218v1*
- [4] R. Yoshida, T. Wakita, H. Okazaki, Y. Mizuguchi, S. Tsuda, Y. Takano, H. Takeya, K. Hirata, T. Muro, M. Okawa, K. Ishizaka, S. Shin, H. Harima, M. Hirai, Y. Muraoka, T. Yokoya, *J. Phys. Soc. Jpn.* **78** (2009) 034708.
- [5] Y. Mizuguchi, F. Tomioka, S. Tsuda, T. Yamaguchi, Y. Takano, *Appl. Phys. Lett.* **93** (2008) 152505.
- [6] S. Margadonna, Y. Takabayashi, M. T. McDonald, K. Kasperkiewicz, Y. Mizuguchi, Y. Takano, A. N. Fitch, E. Suard, K. Prassides, *Chem. Commun.* (2008) 5607.
- [7] T. M. McQueen, Q. Huang, V. Ksenofontov, C. Felser, Q. Xu, H. Zandbergen, Y. S. Hor, J. Allred, A. J. Williams, D. Qu, J. Checkelsky, N. P. Ong, R. J. Cava, *Phys. Rev. B* **79** (2009) 014522.
- [8] J.J. Yeh, I. Lindau, *Atomic Data and Nuclear Tables* **32** (1985) 1.
- [9] T. Yokoya, T. Muro, I. Hase, H. Takeya, K. Hirata, K. Togano: *Phys. Rev. B* **71** (2005) 092507.
- [10] R. Yoshida, H. Okazaki, K. Iwai, K. Noami, T. Muro, M. Okawa, K. Ishizaka, S. Shin, Z. Li, J. L. Luo, G.-q. Zheng, T. Oguchi, M. Hirai, Y. Muraoka, T. Yokoya, *J. Phys. Soc. Jpn.* **78** (2009), 034705.