Polarization-dependent three-dimensional angle-resolved photoemission study on LiFeAs

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Abstract. We report the orbital character of the Fermi surfaces for the Fe-based superconductor LiFeAs using the polarization-dependent three-dimensional angle-resolved photoemission spectroscopy and the angular-dependent dipole selection rule analysis. The evaluated orbital character is well reproduced by the band calculation. The results demonstrate that the polarization-dependent ARPES combined with the selection rule analysis is useful to identify the orbital character of the Fe-based unconventional superconductivity.

1. Introduction

Since the discovery of iron-based superconductors in 2008, a new research field of superconductivity has been established [1]. The common feature of the new superconductors is the existence of iron pnictide layers, similar to CuO$_2$ planes of cuprate superconductors. The phase diagram of iron pnictides is also similar to that of cuprates, i.e., the superconductivity appears by an electron/hole doping to parent compounds [2]. Although intensive research works have been performed, the relation of antiferromagnetic fluctuation to superconductivity is still under debate.

From previous theoretical [3] and experimental [4] works, it has been reported that the high critical temperature ($T_c$) of iron pnictides has relevance to a Fermi surface (FS) nesting with a sign reversal of the order parameter. However, the determination of the FS nesting condition in iron pnictides is intrinsically difficult due to three-dimensionality of their FSs [5]. In addition, the different orbital character at the "multi" FSs has been proposed [6]. Therefore, the direct investigation of the orbital character forming three dimensional (3D) FS as well as the relation between the FS nesting and the superconductivity has been anticipated. In this point of view, the polarization-dependent angle-resolved photoemission spectroscopy (ARPES) is a preferable probe. Particularly, 3D-ARPES using linearly polarized synchrotron radiation is a powerful tool.

The title compound LiFeAs, categorized into the family of 111-type iron pnictides, is an ideal system to investigate the unconventional superconductivity as well as the orbital character at the multi FSs, because the superconductivity of the pristine LiFeAs emerges at $T_c = 18$ K without the antiferromagnetic and structural transitions, unlike the other 1111-type and 122-type iron pnictides. It
should be noted that the FS nesting is expected to be small and the superconductivity is suggested to be assisted by ferromagnetic fluctuation [7].

In this work, we performed the polarization-dependent 3D-ARPES to determine the orbital character of band dispersions near the Fermi level ($E_F$) of LiFeAs. Furthermore, we analyse the orbital symmetry of the FSs by considering the angular-dependent dipole selection rule.

2. Experiment

High-quality single crystals of LiFeAs with $T_c = 18$ K were grown by a Bridgman method [8]. The polarization-dependent 3D-ARPES measurements were performed at the BL7U, namely "SAMRAI" beamline [9], of UVSOR-II, in the Institute for Molecular Science. At the beamline, the linearly [e.g., horizontally ($E_p$) and vertically ($E_s$)] polarized VUV light from the APPLE-II-type undulator is available. The experimental geometry of the present polarization-dependent ARPES is shown in Fig. 1 (a). The total energy and angular resolutions, and sample temperature were set to 6 ~ 15 meV, 0.17°, and 12 K, respectively. The experimental energy and angular resolutions are sufficiently high to detect orbital character of the multi FSs [10]. The clean surface was prepared by in situ cleaving under a vacuum of $8 \times 10^{-9}$ Pa. The photon energies corresponding to high-symmetry $k_z$ points were obtained by using normal-emission ARPES. The inner potential was 15.4 eV. The $E_F$ of samples was referred to that of evaporated gold film. LDA band structure calculation was performed by using the full potential linearized augmented plane wave plus local orbital (LAPW+lo) method including spin-orbit coupling implemented in the Wien2k code [11].

Figure 1. (a) Experimental geometry of the polarization-dependent ARPES. (b) The spatial symmetry of the 3$d$ orbitals with respect to the mirror plane, which are selectively excited with each polarization ($S$ or $P$) under normal-emission configuration. The mirror plane including the direction of incidence and emission is defined along the $x$ axis as shown by bold lines.

3. Results & Discussion

Figure 2(a) shows the polarization-dependent 3D-ARPES images obtained near high-symmetry points. On the each images, the image of left (right) side represents the energy bands excited by the $S$ ($P$) polarized light. We found that the FSs of LiFeAs mainly consist of the three and two hole-pockets at the $\Gamma$ and $Z$ points, respectively and the two electron-pockets at the $M$ and $A$ points. These findings are consistent with previous reports [12]. For all images, the prominent features as indicated by solid and dotted curves are clearly changed dependent on the polarization.

When the linear polarization dependence of the electronic band structure is identified, the dipole selection rule can be exploited to determine the orbital symmetry with respect to the mirror planes of the crystal surface. In the case of the normal emission, the final state $|f\rangle$ has an even symmetry with respect to the mirror plane [13]. Then the nonvanishing condition of the dipole transition $< f |A p | i \rangle$ is satisfied when the initial state $|i\rangle$ has the same symmetry (even/odd) as the dipole operator $A p$, where $A$ and $p$ are the vector potential of electromagnetic field and the momentum operator, respectively. For example, if $A p$ is even (odd) corresponding to the $P$ ($S$) polarization in Fig. 1 (a),
then the initial states with even (odd) symmetry should be reflected in ARPES spectra. In Fig. 1(b), we summarized the polarization-dependent sensitivity of Fe 3$d$ orbitals under the normal-emission geometry [13].

![Figure 2](image_url)

**Figure 2.** (a) Polarization-dependent ARPES images at some symmetry points. The left and right images in each panel are obtained by the $S$ and $P$ polarizations, respectively. (b) The schematic band dispersions and their orbital characters. See the text for detail.

Figure 2 (b) shows the resultant orbital character for each band near $E_F$ estimated by using the present analysis. At $\Gamma$ point ($\phi = 0^\circ$), the emission with the $P$ polarization provides the $d_{xz}$ orbital band (the smallest hole pocket), while the $d_{yz}$ orbital band (the medium hole pocket) is obtained by using the $S$ polarization. The $d_{xy}$ orbital band is also observed at $Z$ point. The outer band is observed with both polarizations at $\Gamma$ and $Z$ points. It is attributed to the $d_{xy}$ orbital band with the $S$ polarization and the $d_{xz}$ orbital band with the $P$ polarization. Therefore the outer band at the $\Gamma$ and $Z$ points should be the combination of the the $d_{xy}$ and $d_{xz}$ orbitals. Since the intensity of $d_{xy}$ orbital is expected to be smaller than that of $d_{xy}$ orbital on the basis of the present analysis, the $d_{xy}$ orbital is not clearly observed. Similarly, the intensity of $d_{xz}$ orbital is also smaller than that of the $d_{xz}$ orbital.

For $M$ and $A$ points, the electron band observed with $S$ polarization is attributed to the $d_{xy}$ orbital. Regarding $P$ polarization, the $d_{xy}$ orbital is naturally expected. In addition, considering the matrix element in $\phi = 30^\circ$, the $d_{xy}$ orbital should be also observed. As a result, the electron pocket observed with $P$ polarization is attributed to $d_{xz} + d_{yz}$ orbitals. Compared to the band calculations for LiFeAs using LMTO method [12], our characterization of $d_{xz} + d_{yz}$ orbitals is consistent, but $d_{xy}$ orbital is not so. On the other hand, our LDA calculations as shown in Fig. 3 give a good agreement even for the $d_{xy}$ orbital.

**4. Summary**

We investigated the orbital character of band dispersions near $E_F$ of LiFeAs by using the polarization-dependent 3D-ARPES. By applying the angular-dependent dipole selection rule analysis, we succeeded to clarify the orbital symmetry of the Fermi surfaces. The evaluated orbital character was well reproduced by the LDA band calculation, suggesting that the present analysis combined with the
polarization-dependent 3D-ARPES is a powerful tool to clarify the relation between multi orbital 3D-FSs nesting and superconductivity.

![Character-dependent LDA band calculations for LiFeAs](image)

**Figure 3.** Character-dependent LDA band calculations for LiFeAs. The size of circles is proportional to the $d_{xy}$ and $d_{x^2-y^2}$ partial weights.

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