

Electronic Structure of the Newly-Discovered Iron-Pnictide Superconductors Revealed by Photoemission Spectroscopy

We have investigated the electronic structure of the newly discovered iron-based superconductors $\text{LaFeAsO}_{1-x}\text{F}_x$ and $\text{LaFePO}_{1-x}\text{F}_x$ by photoemission spectroscopy (PES). The valence-band (VB) spectra show good agreement with band-structure calculations, and indicate that Fe 3d states are dominant near the Fermi level (E_F). Also, we have obtained the experimental Fe 3d partial density of states (PDOS) from the spectra taken in the Fe 3p \rightarrow 3d core-absorption region which could be explained in terms of a band-structure calculation with a phenomenological self-energy correction, yielding a mass renormalization factor of ~ 2 . This is an indication of the moderate electron correlations in these compounds in contrast to the strongly-correlated high- T_c cuprates.

At the early stage of the iron pnictides research [1], it was an urging interest to reveal the basic features of the electronic structure of these newly-discovered materials. Moreover, it was important to check whether and to what extent these high- T_c superconductors are similar to or different from the famous high- T_c cuprates. Therefore, we have performed photoemission spectroscopy measurements on $\text{LaFeAsO}_{1-x}\text{F}_x$ and $\text{LaFePO}_{1-x}\text{F}_x$, and compared our results with those from band-structure calculations [2].

High-resolution ultraviolet photoemission spectroscopy measurements of LaFeAsO (non-superconducting bad metal), $\text{LaFeAsO}_{0.94}\text{F}_{0.06}$ ($T_c \approx 26$ K) and $\text{LaFePO}_{0.94}\text{F}_{0.06}$ ($T_c \approx 5$ K) polycrystals were performed at BL-28A. The energy resolution was ~ 20 meV at 15 K. The samples were fractured *in situ* in an ultra-high vacuum below 1×10^{-10} Torr.

The valence-band spectra of $\text{LaFeAsO}_{1-x}\text{F}_x$ ($x = 0, 0.06$) and $\text{LaFePO}_{0.94}\text{F}_{0.06}$ measured at $h\nu = 57$ eV and $T = 15$ K are shown in Fig. 1. These spectra show the following common features: a sharp peak near E_F , a weak structure at ~ -1.5 eV, a shoulder at ~ -4 eV and a broad peak at ~ -5.5 eV. Comparison reveals good agreement between our experimental data and band-structure calculations displayed in the bottom of Fig. 1. Thus, we attributed the peaks near E_F to Fe 3d states, while the shoulder around -4 eV and the peak around -5.5 eV to As 4p/3p and O 2p states, respectively. In the inset of Fig. 1, a blowup of the valence-band spectra near E_F shows that the peaks are shifted towards higher binding energies with F doping in $\text{LaFeAsO}_{1-x}\text{F}_x$. This shift can be explained as a chemical potential shift due to the electron doping. Also, we notice that the Fe 3d band of $\text{LaFePO}_{0.94}\text{F}_{0.06}$ is broader than that of $\text{LaFeAsO}_{1-x}\text{F}_x$.

The valence-band spectra were also measured at various photon energies in the Fe 3p \rightarrow 3d core excitation region. The intensity of the near- E_F peaks as well as the -1.5 eV structure and the -4 eV shoulder show strong photon-energy dependence: They exhibit an in-

crease from $h\nu \sim 54$ eV to $h\nu \sim 60$ eV as shown in Fig. 2(a) and (b) for LaFeAsO . The $h\nu$ -dependence plotted in Fig. 2(b) is indicative of the Fe 3p \rightarrow 3d resonance, and reconfirms that the near- E_F states down to binding energies as deep as ~ -2 eV are mainly Fe 3d states and that the -4 eV shoulder representing the As 4p band is significantly hybridized with Fe 3d. These results are in good agreement with those reported in Refs. [3, 4]. To obtain the experimental Fe 3d partial density of states (PDOS), we have subtracted the off-resonance spectra (taken at $h\nu = 54$ eV) from the on-resonance spectra ($h\nu = 60$ eV) as shown in Fig. 2(a). In order to extract the Fe 3d band, the As 4p/3p band (the broad peak in the range $-(3-4)$ eV in the difference spectrum of Fig. 2(a)) has been approximated by a Gaussian and subtracted from the Fe 3d PDOS, leaving the Fe 3d-band part, shown in Fig. 2(c) and (d) for $\text{LaFeAsO}_{1-x}\text{F}_x$ and $\text{LaFePO}_{1-x}\text{F}_x$, respectively. The experimental Fe

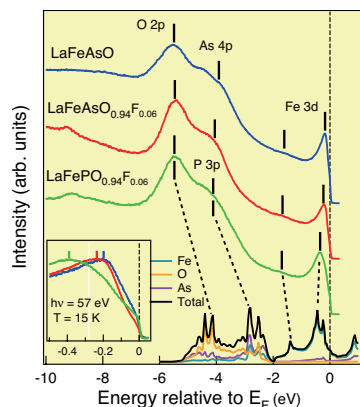


Figure 1 Valence-band photoemission spectra of $\text{LaFeAsO}_{1-x}\text{F}_x$ and $\text{LaFePO}_{1-x}\text{F}_x$ (top) compared with band-structure calculations (bottom). Vertical bars mark main features observed in these spectra and the inset represents the near- E_F spectra.

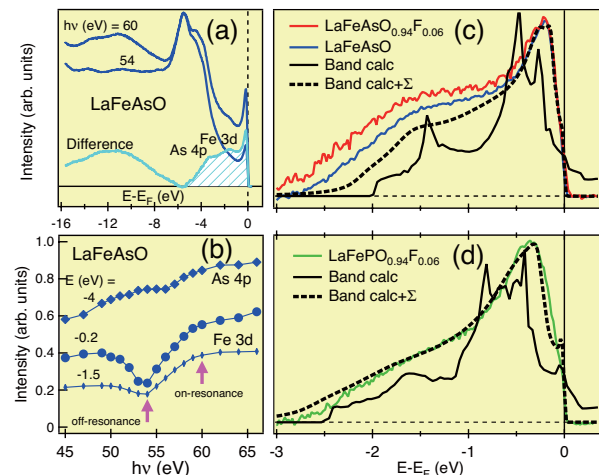


Figure 2 (a) Valence-band photoemission spectra of LaFeAsO in Fe 3p \rightarrow 3d core absorption region and their difference. (b) Plot of the photoemission intensities at $E = -0.2$ eV, -1.5 eV and -4.0 eV as functions of photon energy. (c, d) Fe 3d PDOS of $\text{LaFeAsO}_{1-x}\text{F}_x$ and $\text{LaFePO}_{0.94}\text{F}_{0.06}$. The results of calculations with and without self-energy correction are shown.

3d PDOS show small deviations from the calculated ones (black solid curves in Fig. 2(c) and (d)). These deviations are attributed to the interactions among electrons (electron correlations) which are accounted for by the self-energy Σ . Therefore, we have applied a self-energy correction to the band-calculation result to take these electron correlations into account. Here, we have taken a simple analytical form of the self energy $\Sigma(\omega) = -g\omega(\omega + i\Gamma)^2$, where g and Γ are fitting parameters. Following an empirical approach, these parameters were fitted to reproduce the experimental spectra as has been previously done for Fe chalcogenides [5]. The corrected spectra thus obtained (dotted curves in Fig. 2(c) and (d)) exhibit better agreement with experiment. The electron correlations result in a renormalization of the energy bands and quantitatively, this is given by the mass enhancement factor m^*/m_b where m_b is the bare band mass and m^* is the enhanced mass at E_F . In this case, using the formula $m^*/m_b = 1 - \partial \text{Re} \Sigma(\omega) / \partial \omega = 1 + g/\Gamma^2$, we have obtained $m^*/m_b \sim 1.8$ for $\text{LaFeAsO}_{1-x}\text{F}_x$, and $m^*/m_b \sim 1.5$ for $\text{LaFePO}_{0.94}\text{F}_{0.06}$. This indicates that the degree of electron correlations in $\text{LaFeAsO}_{1-x}\text{F}_x$ is slightly larger than that in $\text{LaFePO}_{1-x}\text{F}_x$, which may originate from the smaller Fe 3d bandwidth in $\text{LaFeAsO}_{1-x}\text{F}_x$ as compared to $\text{LaFePO}_{1-x}\text{F}_x$. Also, these values suggest that the electron correlations in iron pnictides are moderate in contrast to high- T_c cuprates which are strongly

correlated electrons systems. Recent angle-resolved photoemission spectroscopy (ARPES) results on single crystals of the same compounds [6] show good agreement with our results [2].

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