Te 5p orbitals bring three-dimensional electronic structure to two-dimensional
Ir$_{0.95}$Pt$_{0.05}$Te$_2$

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We have studied the nature of the three-dimensional multi-band electronic structure in the two-dimensional triangular lattice Ir$_{1-x}$Pt$_x$Te$_2$ ($x=0.05$) superconductor using angle-resolved photoemission spectroscopy (ARPES), x-ray photoemission spectroscopy (XPS) and band structure calculation. ARPES results clearly show a cylindrical (almost two-dimensional) Fermi surface around the zone center. Near the zone boundary, the cylindrical Fermi surface is truncated into several pieces in a complicated manner with strong three-dimensionality. The XPS result and the band structure calculation indicate that the strong Te 5p-Te 5p hybridization between the IrTe$_2$ triangular lattice surfaces is responsible for the three-dimensionality of the Fermi surfaces and the intervening of the Fermi surfaces observed by ARPES.

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3d, 4d, and 5d transition-metal compounds, and 5f actinide compounds with layered crystal structures often exhibit three-dimensional multi-band Fermi surfaces, which can induce complicated spin-charge-orbital instabilities due to interplay between the two-dimensional electronic structure of the layer and the interaction between neighboring layers. The layered compounds tend to have flat cleavage surfaces which are suitable for angle-resolved photoemission spectroscopy (ARPES) measurements, and the three-dimensional electronic structures can be observed by sweeping photon energy for ARPES. For example, the ARPES study on the classical 1T-TaSe$_2$ system has shown that the large Se-Se interaction between the layers and the strong covalency between the Ta 5d and Se 4p orbitals provide three-dimensional Fermi surfaces which play important roles in the charge density wave formation [1]. In the case of Fe-based superconductors, the origin of the spin and orbital order in the parent materials is still controversial and the three-dimensional multi-band Fermi surfaces observed by ARPES are key ingredients to understand the spin and orbital instabilities as well as the nodeless and nodal superconducting states [2]. In addition, the recent ARPES study on URu$_2$Si$_2$ (which has the ThCr$_2$Si$_2$ structure) has revealed that the nesting vectors in the three-dimensional momentum space are associated with the "hidden" order [3].

Very recently, the layered 5d transition-metal chalcogenide IrTe$_2$ has been attracting great interest due to the discovery of superconductivity in doped or intercalated IrTe$_2$ by Pyon et al. [4] and by Yang et al. [5]. IrTe$_2$ undergoes a structural phase transition at $\sim 270$ K from the trigonal (P3m-1) to the monoclinic (C2/m) structure [6]. Since the Ir 5d-to-Te 5p charge-transfer energy is found to be small [7], the strong Ir 5d-Te 5p hybridization with the IrTe$_2$ layer and the Te 5p-Te 5p hybridization between the IrTe$_2$ layers can play significant roles in the structural phase transition as proposed by Fang et al. and Oh et al. [8, 9]. In addition, band-structure calculations predict that the multi-band Fermi surfaces of undistorted and distorted IrTe$_2$ are three-dimensional in spite of its layered structure [5, 8]. Although the multi-band electronic structure of the Ir 5d and Te 5p orbitals in IrTe$_2$ has been studied using ARPES [10], the three dimensionality of the Fermi surfaces has not been observed in IrTe$_2$ and its relatives. Here, fundamental questions to be addressed are (i) whether the triangular lattice Ir$_{1-x}$Pt$_x$Te$_2$ has the three dimensional Fermi surfaces as predicted by the band-structure calculation and (ii) what are the characteristics of the observed band structures in the triangular lattice Ir$_{1-x}$Pt$_x$Te$_2$. In the present study, we have performed ARPES measurements of Ir$_{0.95}$Pt$_{0.05}$Te$_2$ at various photon energies in order to study the three dimensional Fermi surfaces of the undistorted triangular lattice superconductor Ir$_{1-x}$Pt$_x$Te$_2$.

The single crystal samples of Ir$_{0.95}$Pt$_{0.05}$Te$_2$ were prepared as reported in the literature [12]. The ARPES measurements were performed at beam line 28A of Photon Factory, KEK using a SCIENTA SES-2002 electron analyzer with circularly polarized light. The total energy resolution was set to 20-30 meV for the excitation energies from $h\nu = 79$ eV to $h\nu = 54$ eV. The base pressure of the spectrometer was in the $10^{-9}$ Pa range.
single crystals of Ir$_{0.95}$Pt$_{0.05}$Te$_2$, which were oriented by 
ex situ Laue measurements, were cleaved at 20 K under the ultrahigh vacuum and the spectra were acquired within 24 hours after the cleaving. The x-ray photoemission spectroscopy (XPS) measurement was carried out using JEOL JPS9200 analyzer. Mg K$_\alpha$ (1253.6 eV) was used as x-ray source. The total energy resolution was set to $\sim$ 1.0 eV, and the binding energy was calibrated using the Au 4f core level of the gold reference sample. For the band structure calculations, we employ the code WIEN2k (13) based on the full-potential linearized augmented-plane-wave method and present the calculated results obtained in the generalized gradient approximation (GGA) for electron correlations, where we use the exchange-correlation potential of Ref. [14]. The spin-orbit interaction is taken into account for both Ir and Te ions.

Before presenting the ARPES result, let us discuss the fundamental electronic structure of Ir$_{0.95}$Pt$_{0.05}$Te$_2$, which provides the three-dimensional Fermi surfaces in the layered triangular lattice material, based on the XPS result. In Fig. 1(a), the Ir 4f core-level XPS spectrum of Ir$_{0.95}$Pt$_{0.05}$Te$_2$ is compared with that of CuIr$_2$S$_4$. While the formal valence of Ir is +4 in Ir$_{0.95}$Pt$_{0.05}$Te$_2$, the binding energy of the Ir 4f core level is much lower than that of CuIr$_2$S$_4$ with Ir$^{3.5+}$, indicating that the actual valence of Ir should be smaller than +3.5. Therefore, some of the Te 5p electrons are transferred from the Ir 5d orbitals, and the Te 5p orbitals are expected to be partially unoccupied. This picture is consistent with the band structure calculation in which the electronic states near the Fermi level ($E_F$) have substantial Te 5p character. Indeed, as shown in Fig. 1(c), the valence-band XPS spectra of Ir$_{0.95}$Pt$_{0.05}$Te$_2$ exhibit a peak around 2.5 eV below $E_F$ whereas the spectral weight near $E_F$ is rather weak. Since the photoionization cross section of Ir 5d is much larger than that of Te 5p at this photon energy, the valence-band XPS result suggests that the electronic states 2-3 eV below $E_F$ are dominated by Ir 5d while those near $E_F$ have more Te 5p character.

Figures 2(a)-(j) show the second derivative plots of ARPES spectra approximately along the Γ-K or A-H direction taken at $h\nu = 79$ eV (a), 76 eV (b), 73 eV (c), 70 eV (d), 67 eV (e), 64 eV (f), 61 eV (g), 58 eV (h), 56 eV (i), and 54 eV (j). The dots indicate the Fermi surfaces predicted by the band-structure calculation.
and \( k_z \) is given by \( h\nu - W + V_0 = \hbar^2/2m \times \sqrt{k_x^2 + k_y^2 + k_z^2} \) at photon energy of \( h\nu \). Here, the work function \( W \) and \( V_0 \) are set to 4.4 eV, and 14 eV, respectively. The momentum parallel to the surface is conserved at the \( \Gamma \) point, and \( k_z, k_y \) are electron momenta along the x-direction (the \( \Gamma \)-K or A-H direction) and the z-direction (the \( \Gamma \)-A direction), respectively.

The dispersion while the Ir \( 5d \) orbitals, and all the Ir \( 5d \) bands exhibit large three dimensionality. The photon energy dependence of the observed band dispersions displayed in Fig. 2 shows that all the Ir \( 5d \) bands near \( E_F \) strongly depend on \( k_z \), consistent with the prediction of the band structure calculation considering the spin-orbit interaction.

In Fig. 3, the ARPES intensity at \( E_F \) is plotted as a function of \( k_x \) and \( k_z \) for various photon energies and is compared with the calculated Fermi surfaces indicated by the dots. The observed ARPES intensity at \( E_F \) roughly follows the calculated Fermi surfaces. In the region around the \( \Gamma \) point (4.8 Å\(^{-1} < k_z < 4.4 \text{ Å}^{-1} \)), the observed Fermi surfaces are rather simple. The inner Fermi surface is almost parallel to the \( \Gamma \)-A axis, and the distance between the \( \Gamma \)-A axis and the inner Fermi surface is \( \sim 0.25 \text{ Å}^{-1} \). In the region around the A point

![FIG. 3: (color online) Fermi surface map in the \( k_x-k_z \) plane. Here, \( k_x \) and \( k_z \) are electron momenta along the x-direction (the \( \Gamma \)-K or A-H direction) and the z-direction (the \( \Gamma \)-A direction), respectively.](image)

![FIG. 4: (color online) Fermi surface maps in the \( k_x-k_y \) plane with various photon energies. The Fermi surface maps are compared with the calculated Fermi surfaces which are indicated by the solid curves.](image)
(4.4 Å⁻¹ < k_z < 4.1 Å⁻¹), the inner and outer Fermi surfaces show the complicated structure. In particular, whereas the calculation predicts that the inner Fermi surfaces are broken around k_z ∼ 4.2 Å⁻¹ in the k_y-k_z plane, substantial spectral weight is observed in this region due to the flat dispersion [see Figs. 1(g)-(j)]. The residual spectral weight at E_F due to the flat band dispersion would play important role in the nesting picture although the nesting vector suggested by Yang et al. [5] is not seen in the Fermi surface map in the k_y-k_z plane.

In Figs. 4(a)-(j), the ARPES intensities at E_F are plotted as functions of k_x and k_y, and the Fermi surface maps in the k_z-k_y plane are compared with the prediction of the band structure calculations for various photon energies. Here, the relationship between k_z and k_x (k_y) in Fig. 4(k) is considered in the calculation. In going from Fig. 4(a) to Fig. 4(e), the inner and outer Fermi surfaces are relatively insensitive to the k_z value both in the calculation and the experiment. In the calculation, the inner Fermi surface is separated from the outer one along the Γ-M line in Figs. 4(b)-(d) and finally almost touches to the outer one in Figs. 4(a) and (e). On the other hand, in the experiment, substantial spectral weight is observed already in Figs. 4(b)-(d) in the region where the calculated inner and outer Fermi surfaces touch to one another in Figs. 4(a) and (e). This subtle discrepancy between the experiment and the theory suggests that the coupling between the inner and outer Fermi surfaces is more important than the prediction of the band structure calculation. In going from Fig. 4(f) to Fig. 4(i), the inner and outer Fermi surfaces dramatically change with k_z, and the experimental result is basically consistent with the band structure calculation. However, substantial spectral weight is observed in the region between the calculated inner and outer Fermi surfaces.

The intervening between the inner and outer Fermi surfaces can be related to the substantial distribution of Ir-Ir distance observed by the recent EXAFS study on IrTe_2 [15]. The Ir-Ir bond disorder indicates out-of-plane displacement of Ir atoms which can mix the a_{1g} and e_g orbitals in addition to the spin-orbit interaction. Most probably, the Ir-Ir bond disorder with the out-of-plane displacement is driven by the Te-Te interaction between the layers [5], and therefore, the Te 5p orbitals play important roles both in the intervening between the Fermi surfaces and in the three dimensionality of the Fermi surfaces. Here, one can speculate that the orbital fluctuations (and the superconductivity) would be enhanced by the particular three-dimensional Fermi surfaces via the interlayer Te-Te interaction.

In conclusion, we have studied the three-dimensional multi-band Fermi surfaces of Ir_{0.95}Pt_{0.05}Te_2 using ARPES and band structure calculation. The strong Te 5p-Te 5p hybridization between the IrTe_2 triangular lattice layers is responsible for the strong k_z dependence of the band dispersions which is confirmed by the ARPES experiment. The observed inner and outer Fermi surfaces with strong k_z dependence are basically consistent with the band structure calculation. The strong Te-Te interaction between the layers is responsible for the three dimensionality of the Fermi surfaces. The intervening between the inner and outer Fermi surfaces is larger in the ARPES result than the calculation, suggesting possibility of orbital fluctuation which would be enhanced by the Te-Te interaction and the three-dimensional Fermi surfaces.

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