Electronic structure reconstruction by orbital symmetry breaking in IrTe$_2$

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We report an angle-resolved photoemission spectroscopy (ARPES) study on IrTe$_2$ which exhibits an interesting lattice distortion below 270 K and becomes triangular lattice superconductors by suppressing the distortion via chemical substitution or intercalation. ARPES results at 300 K show multi-band Fermi surfaces with six-fold symmetry which are basically consistent with band structure calculations. At 20 K in the distorted phase, topology of the inner Fermi surfaces is strongly modified by the lattice distortion. The Fermi surface reconstruction by the distortion depends on the orbital character of the Fermi surfaces, suggesting importance of Ir 5$d$ and/or Te 5$p$ orbital symmetry breaking.

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Transition-metal compounds with multi-band Fermi surfaces often exhibit rich and interesting physical properties such as spin-charge-orbital order and superconductivity which originate from the topology of their multi-band Fermi surfaces. For example, the multi-orbital electronic structures of transition-metal oxides and chalcogenides including CuIr$_2$S$_4$ and Ca$_2$−xSr$_x$RuO$_4$ provide various metal-insulator transitions with spin-charge-orbital ordering. Also the multi-band structure of the Fe 3$d$ orbitals play important roles in superconductivity and magnetism of Fe pnictides and chalcogenides such as LaFeAsO$_{1−x}$F$_x$. Recently, Pyon et al. and Yang et al. have discovered interesting interplay between lattice distortion and superconductivity in triangular lattice IrTe$_2$ in which multi-band Fermi surfaces are expected to play significant roles. Since the large spin-orbit interaction of Ir 5$d$ electrons is expected to entangle the spin and orbital degrees of freedom in IrTe$_2$ and the derived superconductors, Yang et al. pointed out that the IrTe$_2$ system provides a new playground to explore and/or realize topological quantum states, which are currently attracting great interest in physics community.

IrTe$_2$ exhibits a structural phase transition at ~270 K from the trigonal (P3m-1) to the monoclinic (C2/m) structure accompanied by anomalies of electrical resistivity and magnetic susceptibility. When the lattice distortion is suppressed by chemical substitution of Pt or Pd for Ir or intercalation of Pd, IrTe$_2$ becomes superconductors. An electron diffraction study by Yang et al. observed the superlattice peaks with wave vector of $q = (1/5, 0, -1/5)$ below the structural transition temperature. Such superstructure can be explained by charge density wave (CDW) driven by perfect or partial nesting of multi-band Fermi surfaces. In multi-band Fermi surfaces derived from Ir 5$d$ and Te 5$p$ orbitals, the nesting character can be enhanced by orbitally-induced Peierls mechanism. In addition, charge modulation of Ir 5$d$ electrons is indicated by an Ir 4$f$ x-ray photoemission study. On the other hand, a recent optical study by Fang et al. on single crystal samples shows that there is no gap opening expected for CDW and, instead, band structure is reconstructed over a broad energy scale up to ~2 eV. Fang et al. conclude that the structural transition of IrTe$_2$ is not of CDW type but of a novel type driven by Te 5$p$ holes.

In this context, it is very interesting and important to study the geometry of multi-band Fermi surfaces of IrTe$_2$ using angle-resolved photoemission spectroscopy (ARPES). In the present ARPES study, above the transition temperature, the flower-shaped outer Fermi surface and the inner Fermi surfaces like six connected beads, which are predicted by band structure calculations, are partly identified. Across the structural transition, the topology of the inner Fermi surfaces is modified more strongly than that of the outer Fermi surface. Below the transition temperature, the inner Fermi surfaces consist of two straight portions, suggesting Fermi surface nesting. However, clear gap opening expected for CDW is not observed in the ARPES spectra, consistent with the optical study. Instead, spectral weight is partially suppressed at specific points of the straight Fermi surfaces.

Single crystal samples of IrTe$_2$ were prepared using a self-flux method. The ARPES measurements were carried out at beamline 9A, Hiroshima Synchrotron Radiation Center using a SCIENTA R4000 analyzer with circularly polarized light of photon energy $h\nu = 23$ eV.
FIG. 1: (color online) (a) Fermi surface map and (b) its second derivative map of IrTe$_2$ for $h\nu = 23$ eV taken at 300 K. (c) Fermi surface map and (d) its second derivative map of IrTe$_2$ for $h\nu = 23$ eV taken at 20 K. The integration energy window of $\pm 5$ meV at the Fermi level ($E_F$). The center of the hexagon roughly corresponds to the A point for $h\nu = 23$ eV. For 300 K, the flower-shaped outer Fermi surface and the inner Fermi surfaces with six-fold symmetry are schematically shown by the dashed and dotted curves, respectively. (e) Schematic drawings for the Ir triangular lattice and the hexagonal Brillouin zone at $k_z = 0$ and $k_z = \pi/c$. The Te ions indicated by solid (dotted) circles are located above (below) the Ir plane. The thin solid curves indicate possible Brillouin zone boundaries for possible three domains considering the superstructure reported in ref. 7.

FIG. 2: (color online) Broad-range band dispersions along the A-H direction of IrTe$_2$ for $h\nu = 23$ eV taken at 300 K (a) and at 20 K (b). Broad-range energy distribution curves along the A-H direction of IrTe$_2$ for $h\nu = 23$ eV taken at 300 K (c) and at 20 K (d).

The data were collected at 300 K and 20 K with an angular resolution of $\sim 0.3^\circ$ and energy resolution of 18 meV for excitation energy of $h\nu = 23$ eV. The incident beam is 50$^\circ$ off the sample surface. The base pressure of the spectrometer was in the $10^{-9}$ Pa range. The samples were cleaved at 300 K under the ultrahigh vacuum and cooled across the structural transition, and then warmed to 300 K to check the reproducibility at 300 K. The samples were oriented by ex situ Laue measurements. The spectra were acquired within 8 hours after the cleavage. Binding energies were calibrated using the Fermi edge of gold reference samples.

The Fermi surface mapping of IrTe$_2$ measured at 300 K above the structural transition temperature are displayed in Figure 1(a). At $h\nu = 23$ eV, the momentum perpendicular to the Ir plane approximately corresponds to $\pi/c$, where $c$ is the out-of-plane lattice constant, and the center of the hexagonal Brillouin zone is the A point. The direction from the A point to the L (H) point corresponds to the direction of Ir-Ir (Ir-Te) bond. In Figs. 1(a), several Fermi surfaces can be identified as predicted by the band structure calculations [7, 11] although the strong intensity asymmetry due to transition-matrix element effect does not allow perfect identification. In order to extract the shapes of the Fermi surfaces, the second derivative along the cut direction $d^2I(k_x, k_y)/d^2k_x$ is plotted in Fig. 1(b). The flower shape for the outer Fermi surface is more clearly seen which is schematically indicated by the dashed curve. In addition, the inner Fermi surfaces like six connected beads can be identified as indicated by the dotted curves although effect of thermal excitations at 300 K tends to obscure the relatively small Fermi pockets. The inner Fermi surfaces observed around the A point at 300 K are roughly consistent with the prediction of band-structure calculations [7, 11].

Figure 1(c) shows the Fermi surface mapping at 20 K well below the transition temperature. Across the structural transition, in the region where the outer Fermi surface is close to the inner Fermi surfaces, the outer Fermi surface at 20 K disappears due to partial gap opening or overlaps with
FIG. 3: (color online) Near-$E_F$ band dispersions along the A-H direction of IrTe$_2$ for $h\nu = 23$ eV taken at 300 K (a) and at 20 K (b). Near-$E_F$ momentum distribution curves along the A-H direction of IrTe$_2$ for $h\nu = 23$ eV taken at 300 K (c) and at 20 K (d). Near-$E_F$ energy distribution curves along the A-H direction of IrTe$_2$ for $h\nu = 23$ eV taken at 300 K (e) and at 20 K (f). The outer hole bands and the inner hole-like bands are indicated by the dashed and dotted curves, respectively.

FIG. 4: (color online) (a) Fermi surface mapping near the A point at 20 K for $h\nu = 23$ eV. (b-d) Near-$E_F$ band dispersions along the cuts parallel to the A-H direction at 20 K for $h\nu = 23$ eV. (e) Near-$E_F$ energy distribution curves at the selected Fermi surface points at 20 K.
separately from the inner one, the outer band at 20 K disappears near $E_F$. Across the transition, the outer band is shifted towards the inner one in this momentum region and is probably gapped due to the interaction with the inner band. This is consistent with the partial disappearance of the outer Fermi surface in Fig. 1(c). The inner hole-like band at 300 K is also strongly affected by the structural transition. The band located around $\sim -0.15$ eV of the A point at 300 K disappears at 20 K probably because it is shifted above $E_F$. Consequently, the hole band indicated by the dotted curve in Fig. 3(b) crosses $E_F$ at 20 K and form the straight portions of the Fermi surfaces of Fig. 1(b). Such band reconstruction cannot be explained by a simple band folding picture, indicating orbital reconstruction by Jahn-Teller-like effect.

The Fermi surface mapping around the A point at 20 K is shown in Fig. 4(a). In general, straight Fermi surfaces with nesting wave vector $q$ are expected to be gapped due to density wave formation with $q$. In IrTe$_2$, instead of gap opening, spectral weight at $E_F$ is partially suppressed at specific points of the straight Fermi surfaces. In cuts 1 and 3 along the A-H direction [Figs. 4(b) and (d)], the hole band clearly crosses $E_F$ and the spectral weight at $E_F$ is not suppressed. On the other hand, in cut 2 [Fig. 4(c)], the intensity of the hole band is suppressed near $E_F$ as seen in the EDC plot of Fig. 4(e). There are four points where the spectral weight at $E_F$ is suppressed as seen in Fig. 4(a). Such spectral weight suppression at the specific points (cold spots) would be related to the origin of the superstructure of bulk IrTe$_2$ since the wave vectors connecting the two cold spots [indicated by the arrows in Fig. 4(a)] are approximately 2/5 of the A-L distance or 1/5 of the L-L’ distance, partly consistent with its period. However, the partial spectral weight suppression would be due to surface effect or transition-matrix element effect, and no decisive conclusion can be obtained at the present stage. Here, it should be noted that the observed Fermi surfaces correspond to one of the Brillouin zone boundaries for possible domains. However, the crystal structure of low temperature phase is highly controversial (refs. 7, 11, and 13), and that, at the present stage, it is difficult to discuss relationship between the observed Fermi surfaces and the band folding due to the superstructure.

In conclusion, above the transition temperature, the observed Fermi surfaces and band dispersions are consistent with the band structure calculations. The flower-shaped outer Fermi surface (hole character) with six-fold symmetry and the inner Fermi surfaces (hole pockets) are observed. Across the structural transition, the geometry of the inner Fermi surfaces is strongly modified. In the distorted phase, the inner Fermi surfaces consist of two straight portions, suggesting that nesting character is enhanced. However, the gap opening expected for CDW is not observed in the ARPES spectra, consistent with the optical study. Also the electronic structure up to $\sim -3$ eV is reconstructed by the lattice distortion, which is also consistent with the optical study. In addition, the spectral weight at $E_F$ is suppressed at the specific points of the straight Fermi surfaces, which would be related to the origin of the superstructure.

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