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Experimental Observation of Incoherent-Coherent Crossover and Orbital Dependent Band Renormalization in Iron Chalcogenide Superconductors

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The level of electronic correlation has been one of the key questions in understanding the nature of superconductivity. Among the iron-based superconductors, the iron chalcogenide family exhibits the strongest electron correlations. To gauge the correlation strength, we performed systematic angle-resolved photoemission spectroscopy study on the iron chalcogenide series Fe\textsubscript{1+y}Se\textsubscript{x}Te\textsubscript{1−x} (0<x<0.59), a model system with the simplest structure. Our measurement reveals an incoherent to coherent crossover in the electronic structure as the selenium ratio increases and the system evolves from the weakly localized to more itinerant state. Furthermore, we found that the effective mass of bands dominated by the d\textsubscript{xy} orbital character significantly decreases with increasing selenium ratio, as compared to the d\textsubscript{xz}/d\textsubscript{yz} orbital-dominated bands. The orbital dependent change in the correlation level agrees with theoretical calculations on the band structure renormalization, and may help to understand the onset of superconductivity in Fe\textsubscript{1+y}Se\textsubscript{x}Te\textsubscript{1−x}.

I. INTRODUCTION

The nature and role of many-body interaction has been a crucial yet unsettled question in the recently discovered iron-based superconductivity\textsuperscript{1−3}. Among all the iron-based superconductors, the correlation level in the iron chalcogenide Fe\textsubscript{1+y}Se\textsubscript{x}Te\textsubscript{1−x} (11 system) has been predicted to be one of the strongest\textsuperscript{3−5}, which is confirmed by transport\textsuperscript{6,7}, neutron scattering\textsuperscript{8}, optical spectroscopy\textsuperscript{9}, and photoemission spectroscopy\textsuperscript{10−12} experiments. The mechanism for strong correlation in the parent compound of the 11 system, Fe\textsubscript{1.02}Te was addressed in our previous angle-resolved photoemission spectroscopy (ARPES) study: the electronic structure in the antiferromagnetic (AFM) phase is featured by the characteristic “peak-dip-hump” features and quasiparticle dispersion with huge band renormalization (∼90), which we attribute to coherent polarons formed by the interplay of large magnetic moment and electron-phonon coupling\textsuperscript{13}. The coherent polaronic behavior naturally explains the metallicity in the AFM state of Fe\textsubscript{1.02}Te. However, there is up to date no systematic study on the evolution of the correlation strength with the change of selenium ratio from the metallic AFM phase (x<0.1) to the weakly localized phase (0.1<x<0.28) and finally the superconducting/metallic phase (x>0.28)\textsuperscript{8} (Fig. 1(a)), where the correlation level is described by large band renormalization, reported to be 6~20 for the optimally doped FeSe\textsubscript{0.45}Te\textsubscript{0.55} (T\textsubscript{c}=14.5 K)\textsuperscript{10}.

In this work, we present a systematic study of the electron correlation effect using ARPES on a series of Fe\textsubscript{1+y}Se\textsubscript{x}Te\textsubscript{1−x} samples with increasing selenium ratios (y<0.02, x=0, 0.11, 0.2, 0.25, 0.28, 0.35, 0.44, 0.59). Our results show that the electronic structure of x=0.11 sample in the weakly localized phase is similar to that of Fe\textsubscript{1.02}Te (x=0 sample) above the AFM transition temperature (T\textsubscript{N}). With higher selenium ratio, the spectral weight of the coherent quasiparticles becomes increasingly pronounced, indicating an incoherent to coherent crossover in the electronic structure. Furthermore, we find that the effective mass renormalization of the bands dominated by the d\textsubscript{xy} orbital character decreases with selenium substitution, while those with the d\textsubscript{xz}/d\textsubscript{yz} character do not show much change. Our results reveal an orbital dependent decrease of electronic correlations as superconductivity emerges in the iron chalcogenide Fe\textsubscript{1+y}Se\textsubscript{x}Te\textsubscript{1−x}. Such evolution of orbital dependent electronic correlation effect is observed so far only in iron chalcogenides, making them a unique family to study the interplay between strong correlations, multi-orbital physics and superconductivity in iron-based superconductors.
II. EXPERIMENTAL RESULTS

A. Incoherent-Coherent Crossover

The measured Fermi surfaces (FSs) and band dispersions along the high symmetry M-Γ-X directions (the high symmetry points are defined in the Brillouin zone in the reciprocal space of 2-Fe unit cell as shown in Fig. 1(b)) for samples with selenium ratios between 0 and 0.44 are plotted in Fig. 1(b) and (c), respectively. Firstly, the electronic structure of x=0 and x=0.11 at low temperatures are drastically different. \(\text{Fe}_{1+y}\text{Se}_x\text{Te}_{1-x}\) in the AFM phase is characterized by an electron pocket at \(\Gamma\), finite spectral intensity around \(X\) and almost no intensity at \(M\) (see\textsuperscript{13} for details). At the selenium ratio of 0.11 where the system is in the weakly localized state, the electron-like pocket at \(\Gamma\) becomes hole-like, producing a peanut-like shape on the FS. Bands at \(M\) start to be noticeable while the spectral weight around the \(X\) point weakens. Notably, such a doping evolution in the electronic structure is very similar to that of the temperature induced change in \(\text{Fe}_{1.02}\text{Te}\) from below to above the AFM transi-
different orbital characters. The momentum distribution curve (black curve) along \( k_x = 0 \) is plotted on the right side of each figure, together with the three-Lorentzian fitting curve (red curve) showing the actual positions of the Fermi surface crossing (\( k_F \)). (b) Fermi surface map of features around the \( \Gamma \) point from \( \text{Fe}_{1+y}\text{Se}_x\text{Te}_{1-x} \) with various \( x \) measured with 26 eV excitation energy at \( T = 10 \) K. Schematic of the Fermi surface is plotted on top of the map with different colors representing bands with different orbital characters. The momentum distribution curve (black curve) along \( k_y(k_z) = 0 \) is plotted on the right (top) side of each figure, together with the two-Lorentzian (Lorentzian+Gaussian) fitting curve (red curve) showing the actual positions of the Fermi surface crossing (\( k_F \)).

The momentum distribution curve (EDC) evolution of the \( \beta \) band around \( \Gamma \) (Fig. 1(d)(i)) and the \( \epsilon \) band around \( M \) (Fig. 1(d)(ii)); and also the Momentum Distribution Curve (MDC) evolution on \( E_F \) across the \( M \) point (Fig. 1(d)(iii)). The increase of the spectral weight (Fig. 1(d)(iv)) and decrease of the line width (Fig. 1(d)(v)(vi)) of the coherent quasiparticles are very similar to ARPES observations of the doping-dependence of other strongly correlated materials (e.g., cuprates\(^{18,19} \)); it is a direct manifestation of the incoherent to coherent crossover behavior of the electrons, concomitant with \( \text{Fe}_{1+y}\text{Se}_x\text{Te}_{1-x} \) evolving from the weakly localized phase to the metallic phase when the selenium ratio increases from 0.11 to 0.44. From 0.44 to 0.59, our measured quasiparticle spectral weights are fluctuating due to sample quality variations rather than showing the systematic trends of evolution.

More significant change occurs in the evolution of the spectral weight and line width. As the selenium ratio increases, we found that in the high symmetry cuts (Fig. 1(c)) the broad and smeared dispersions become narrower and sharper, indicating that the spectral weight of the coherent quasiparticles becomes stronger over the incoherent background, as is clearly shown by the Energy Distribution Curve (EDC). When the selenium ratio increases from 0.11 to 0.44, the changes in the electronic band structure become more gradual. From the FS mapping (Fig. 1(b)), we see that the feature at \( M \) becomes stronger in intensity with a gradual emergence of electron-like pockets, while the intensity at \( X \) fades out. Such a FS evolution may be closely related to the suppression of the \((\pi, 0)\) short range magnetic order and the enhancement of the \((\pi, \pi)\) magnetic fluctuation with selenium substitution, as reported\(^{6,16} \). At the same time, the band dispersions do not have drastic changes in energy, except for the electron-like band around \( \sim 400 \) meV below \( E_F \) at \( \Gamma \) (labeled as the \( \eta \) band in Fig. 1(c) with dominantly \( d_{z^2} \) orbital character), which shifts systematically from \(-400 \) meV to \(-300 \) meV. This band shift is well captured by the density functional calculation results\(^{17} \).

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are plotted in different panels. In each panel different colors represent samples with different x. (c) Plot of the
side for better visualization of each band. The extracted dispersion for each band is plotted on top of each image plot with
different colors representing different dominating orbital characters. (b) Stack plot of fitted band dispersions. Bands with
different orbitals are plotted in different panels. In each panel different colors represent samples with different x. (c) Plot of the
extracted effective band mass renormalization factor versus the selenium ratio. Band mass with different orbitals are plotted
in different panels.

The electronic band dispersions near the Fermi level
evolution of samples with selenium ratios from x=0.28 to
0.59 (the spectral weight of the dispersions for x<0.28 is
too small, making such analysis difficult). Fig. 2 plots
the measured FSs at both the Γ (a) and M (b) points. At
the Γ point, we find the “peanut shape” in the FS is part
of a circular hole pocket with mixed orbital content of d_{xy}
and d_{xz}/d_{yz} due to band hybridization (see supplemental
material, part II). The reason we did not observe a full
circle is due to the suppression from the matrix element
(see supplemental material, part I). The intensity at ex-
act Γ comes from the band top of the inner d_{xz}/d_{yz} band.
At the M point, we observe two intersecting ellipses, with
different segments coming from different orbitals. Some
parts of the two ellipses are not visible in the data due
to the suppression from the matrix element. To deter-
mine the Fermi pocket size, we find the maximum and
minimum openings of the pockets from fitting the con-
tour plots. Schematics of the FSs can then be drawn by
considering the four-fold crystal symmetry of the tetrag-
onal state. The carrier concentration level can then be
evaluated by counting the Fermi surface volume. The
calculated net doping level is 0.003~0.012 electron/Fe,
indicating that the 11 system is almost electron-hole bal-
anced with the additional electrons may come from the
excessive interstitial Fe atoms\(^{15}\). Furthermore, we do
not observe any obvious change in either the electron or
hole pockets for samples with different selenium ratios,
confirming the isovalent nature of the selenium/tellurium
substitution.

The electronic band dispersions near the Fermi level
do not shift noticeably in position but exhibit systematic
change in curvature. We extracted the dispersions by
locating peak positions from both the energy and mo-
mentum distribution curves. Around the Γ point we
could identify three different hole bands which are domi-
nated by different orbital characters (see supplemental
material, part I). By fitting each of the dispersion to
a parabolic curve, we can extract the effective mass of
each band (Fig. 3 (a), (b)). As Fig. 3 (c) shows, the
d_{xy} band has the largest effective mass renormalization
compared with density functional calculation\(^{17}\), while the
other bands have much smaller renormalization factors
(∼3 for the d_{yz} and the d_{xz} band). Moreover, only the
renormalization factor of the d_{xy} band shows significant
doping dependence, decreasing from 22 to 14 as the sele-
nium ratio increases from 0.28 to 0.59. For the d_{xz}/d_{yz}
bands, the extracted effective masses do not change no-
ticeably over this doping range.

We applied the same analysis to the electron pockets at
M and observed similar behavior (see supplemental
material, part III). All together, we have found that bands
dominated by the d_{xy} orbital character have a much
larger renormalization comparing to bands of other or-
bitals, and the renormalization monotonically reduces as
the selenium ratio increases while the other orbitals do
not have such effect. Therefore, by using effective band
mass as the correlation level indicator, we have discov-
ered there is a reduction of (yet still strong) correlation as
tellurium replaces tellurium in Fe\(_{1+x}\)Se\(_{1−x}\), and this
correlation reduction has most significant effect in bands
with the d_{xy} orbital character.

![Figure 3](image-url)

**FIG. 3.** (a) Photoemission intensity of the cut around Γ along the Γ-M direction for Fe\(_{1+x}\)Se\(_{1−x}\) with various x. The
original data is plotted on the left side of each panel while the second derivative of the same data is plotted on the right
side for better visualization of each band. The extracted dispersion for each band is plotted on top of each image plot with
different colors representing different dominating orbital characters. (b) Stack plot of fitted band dispersions. Bands with
different orbitals are plotted in different panels. In each panel different colors represent samples with different x. (c) Plot of the
extracted effective band mass renormalization factor versus the selenium ratio. Band mass with different orbitals are plotted
in different panels.
mean-field theory, ref 11 system. In a theoretical study based on dynamical correlation and unique orbital dependent physics in the 11 system, there have been several theoretical works discussing the level of interaction is a unique feature for the 11 system. So far, there lurium atoms would modify the structural parameters to other orbitals. Given the structural sensitivity, the orbitals are more localized in iron chalcogenides compared to iron chalcogides. As a result, electrons in the in-plane d hole band is higher than the dzy orbitals which controls the crystal field splitting, is much smaller from that of an ideal tetrahedron in iron chalcogenides. As a result, electrons in the in-plane dhole band is due to the band hybridization between the d hole electron band and d hole band. (c) Calculated mass renormalization for different bands from samples with different selenium ratio x in FeSeTe1−x. The mass renormalization is calculated by dividing the effective mass from the calculated band structure with U=2.45 eV, J/U=0.25 by that from U=J=0. Different marks with color indicate the renormalization of different bands, and different colors label different orbitals. (d) Calculated phase diagram of FeSeTe1−x as a function of selenium ratio x and Coulomb interaction U. The blue dotted line denotes where U=2.45eV and the calculated mass renormalization best agrees with the actual experimental results. WCM: weakly correlated metal. SCM: strongly correlated metal. O SMP: orbital selective Mott phase. MI: Mott insulator.

III. THEORETICAL INTERPRETATION

Comparing to the band renormalization change that happens to all the t2g bands in Co-doped BaFe2As2 system21, such orbital dependent renormalization evolution is a unique feature for the 11 system. So far, there have been several theoretical works discussing the level of correlation and unique orbital dependent physics in the 11 system. In a theoretical study based on dynamical mean-field theory, ref6 has pointed out that the correlation effects in Fe pnictides/chalcogenides come from the Hunds coupling and that the structural parameters have a strong impact on the overall correlation strength and orbital-selectivity. The longer Fe-chalcogen bond length compared to that of Fe-pnictigen would result in more localized electrons. Meanwhile, the Ch-Fe-Ch bond angle, which controls the crystal field splitting, is much smaller from that of an ideal tetrahedron in iron chalcogenides. As a result, electrons in the in-plane dxy orbitals are more localized in iron chalcogenides compared to other orbitals. Given the structural sensitivity, the substitution of smaller selenium atoms for bigger tellurium atoms would modify the structural parameters and decrease the correlation level and orbital selectivity. In another work based on a slave-spin mean-field method22,23, the authors proposed that due to the strong intra-orbital Coulomb repulsion U and Hunds coupling J, the iron chalcogenide family is in proximity to an orbital selective Mott phase (OSMP), where the dxy orbital is Mott-localized while the other orbitals remain itinerant. The physical scenario of OSMP has been previously proposed and discussed in Ca2−xSr2RuO424–26 and iron based superconductors22,23,27,28.

Both works would explain our observations on the correlation level change and orbital selectivity in FeSeTe1−x. As an illustration, we applied the model from22,23 to the FeSeTe1−x family and the calculation results are summarized in Fig. 4 (details of the calculation could be found in the supplemental material, part VI, we also note the fact that the dxy hole band is higher than the dzy orbitals in many cases). With the inclusion of moderate U and J (U=2.45 eV, J/U=0.25) in the model, the dispersion of FeSe0.5Te0.5 [Fig. 4(b)] is found to be greatly renormalized compared to the U=J=0 case [Fig. 4(a)]. The calculated mass
replacements at the same value of U and J for different x values in FeSe$_1$Te$_{1-x}$ well reproduce the change in the overall correlation strength and orbital-selectivity as observed [Fig. 4(c)]. The agreement with the experimental result shows that FeSe$_1$Te$_{1-x}$ are overall in the strongly correlated metal phase and loses correlation with increasing x [Fig. 4(d)]. The calculation further proposed that the correlated metal phase is in proximity to an orbital selective Mott phase and raising temperature is one potential path to enter such phase$^{29}$.

IV. DISCUSSIONS

It should be noted that our observation of the correlation evolution of Fe$_{1+y}$Se$_2$Te$_{1-x}$ has only extended to selenium ratio up to 0.59. Single crystals with selenium ratio higher than that have been found to be hard to stabilize$^{30}$. However, in our recent ARPES measurement, we have observed very renormalized d$_{xy}$ hole bands with renormalization factor $\sim$10 for K$_x$Fe$_{2-y}$Se$_2$$_{28-29}$. In addition, a recent ARPES report on single crystal of FeSe found the d$_{xy}$, d$_{yz}$ and d$_{xz}$ hole band renormalizations to be 9, 3 and 3.7 respectively, fully consistent with our observations of the trend$^{12}$. Therefore, the large d$_{xy}$ orbital band renormalization appears to be universal to all iron chalcogenides, making it unique among all iron-based superconductors$^{29}$.

The nature of the strong correlation may be critical to the understanding of the superconductivity in iron chalcogenides. In Fe$_{1+y}$Se$_2$Te$_{1-x}$, the level of correlation seems to be the primary tuning factor for superconductivity since the doping level and the underlying Fermi surface topology do not change. For K$_x$Fe$_{2-y}$Se$_2$, where Tc is comparable to iron pnictides, the lack of hole pockets makes a weak-coupling Fermi surface nesting picture unlikely. Hence, the superconducting pairing mechanism may stem from strong correlations that lead to strong local pairing. As the d$_{xy}$ band participates superconducting pairing and is most sensitive to the change of correlation among all Fe 3d bands, its band renormalization would serve as an accurate gauge for the correlation level and pairing strength.

As we are preparing our manuscript, a similar report$^{31}$ also addressed the evolution from incoherent to coherent electronic states in FeSe$_1$Te$_{1-x}$, consistent with our conclusion. In addition to$^{31}$, our first discovery on the orbital dependent band renormalization provides a deeper understanding of the nature of the strong correlations in multiple orbital systems such as Fe$_{1+y}$Se$_2$Te$_{1-x}$.

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