Symmetry breaking via orbital-dependent reconstruction of electronic structure in detwinned NaFeAs

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The superconductivity discovered in iron pnictides is intimately related to a nematic ground state, where the $C_4$ rotational symmetry is broken via the structural and magnetic transitions. We here study the nematicity in NaFeAs with polarization-dependent angle-resolved photoemission spectroscopy. A uniaxial pressure was applied on the sample to overcome the twinning effect in the low temperature $C_2$-symmetric state and obtain a much simpler electronic structure than that of a twinned sample. We found the electronic structure undergoes an orbital-dependent-reconstruction in the nematic state, primarily involving the $d_{x^2}$- and $d_{y^2}$-dominated bands. These bands strongly hybridize with each other, inducing a band splitting, while the $d_{z^2}$-dominated bands only exhibit an energy shift without any reconstruction. These findings suggest that the spin fluctuations at high temperatures and their coupling with the orbital degree of freedom could be the dominant force to drive the nematicity, while the ferro-orbital ordering between $d_{z^2}$ and $d_{y^2}$ orbitals can only play a minor role here.

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I. INTRODUCTION

The parent compounds of iron pnictides share a common spin density wave or collinear antiferromagnetic (CAF) state, which is characterized by a ferromagnetic (FM) spin alignment along one direction in the two-dimensional rectangular lattice formed by iron sites and an antiferromagnetic (AFM) spin alignment along the other direction. Superconductivity in iron pnictides emerges when the magnetic order is suppressed through chemical substitution or physical pressure. The development of the CAF order is always accompanied by a lattice structure distortion, more specifically, a tetragonal-to-orthorhombic structural distortion. The transition temperature ($T_N$) at which the lattice distortion takes place either precedes or coincides with the CAF Neel transition temperature ($T_N$). The coincidence between the two transitions can be understood from a simple symmetry analysis. Besides breaking magnetic symmetry, the CAF state also breaks $C_4$ lattice symmetry of the tetragonal lattice. This lattice symmetry breaking due to the magnetic configuration can be attributed solely to the development of a nematic electronic order that only accounts for the rotational symmetry breaking but not to the broken magnetic symmetry. Based on the textbook Ginzburg-Laudau approach, the orthorhombic lattice distortion breaks the same $C_4$ lattice symmetry. Therefore, the nematic electronic order and the orthorhombic lattice distortion are allowed by symmetry to be coupled with each other. Since the observed lattice distortion in all families of iron pnictides is rather small, the electronic nematicity could be dominant.

So far, there are many experimental evidences supporting the dominance of the electronic nematic behavior in iron pnictides. Transport and neutron scattering measurements found a strong in-plane anisotropy of the resistivity and magnetic excitations. Angle-resolved photoemission spectroscopy (ARPES), scanning tunneling microscopy, and quantum oscillation studies further revealed a complex electronic structure with $C_2$ rotational symmetry. Moreover, the Fermi surface was suggested to be orbital polarized with almost pure $d_{x^2}$ orbital character in the CAF state. The energy splitting of the $d_{x^2}$- and $d_{y^2}$-dominated bands was further observed far above $T_N$. The observation of such a ferro-orbital ordering has led to a strong debate on the origin of the nematicity: whether the electronic nematicity stems from the magnetic or orbital degree of freedom. Theoretically, some suggest that the nematicity is originated from spin fluctuations, while others suggest the CAF state comes from a ferro-orbital ordering. Although following the symmetry argument that the electronic nematicities from both origins must be coupled to each other, direct experimental evidence is needed to answer how strongly such a coupling is, and whether one is dominant over the other.

Previous ARPES studies mainly focus on the so-called “122” series of iron pnictides, such as BaFe$_2$As$_2$, CaFe$_2$As$_2$, and BaFe$_2$As$_2$. Although the mechanically detwinning methods were used to avoid the complexity due to the twinning effect in the CAF state, the observed electronic structures are still very complicated. It is thus difficult to obtain a comprehensive understanding of the complex electronic structure and its orbital character in the nematic state. In this paper, we report a systematic ARPES study on detwinned NaFeAs. The observed electronic structure is much simpler than those of the 122 series. Because of the polarization-sensitivity of orbitals in photoemission experiment, we could selectively probe the bands with different orbital characters. We found that the $C_4$
rotational symmetry of the electronic structure is broken via band reconstruction and the band reconstruction itself is also strongly orbital-dependent: the $d_{xz}$- and $d_{yz}$-dominated bands open a large hybridization gap at all the band crossings, while the $d_{xz}$-dominated bands only exhibit an energy shift without any reconstruction. Moreover, the observed total occupation of $d_{xz}$ is almost invariant. These results suggest that the ferro-orbital ordering between $d_{xz}$ and $d_{xy}$ can only play a very limited role in driving the electronic nematicity. On the other hand, the orbital weight redistribution involving the $d_{xz}$ and $d_{xy}$ orbitals could be explained through an orbital-dependent spin polarization far above the Neel transition temperature, which suggests that the nematic spin fluctuations at high temperatures and their coupling with orbital degree of freedom could be the driving force for the nematic electronic state in iron pnictides.

II. EXPERIMENTAL

NaFeAs single crystals were synthesized by a self-flux method.24 Similar to other parent compounds of iron pnictides, the high temperature phase ($T > T_S$) is in a tetragonal paramagnetic (PM) state, where each unit cell has two irons due to the inequivalent positions of arsenic ions in the FeAs layer [Fig. 1(a)]. Through the structural transition ($a_o > b_o$), there is an orthorhombic PM state ($T_S > T > T_N$) with $C_2$ rotational symmetry. The CAF magnetic order shows up when entering the orthorhombic CAF state ($T < T_N$).1-3 The $x$ or $k_x$ axes and $y$ or $k_y$ axes are defined along the AFM and FM directions, respectively. Previous ARPES study on NaFeAs observed a complex electronic structure in the orthorhombic CAF state due to twinned domains of the sample.24 Taking the advantage of the mechanically detwinning method,8,25 single domain with sufficiently large size was achieved in our experiment by applying uniaxial pressure along Fe-Fe direction [Figs. 1(a) and 1(b)]. As a result, we could investigate the electronic structure of the orthorhombic CAF state without the complexity of twinning.

The temperature-dependent resistivity of unstressed sample is consistent with the neutron scattering and transport measurements.3,26 There are two inflexion points at 54 and 43 K, corresponding to the structural transition temperature ($T_S$) and the Neel transition temperature ($T_N$), respectively [Fig. 1(c)]. Superconductivity shows up at 20 K, which might be due to the doping effect of the deficient or excess sodium ions. When a uniaxial pressure is applied, the in-plane anisotropy of the resistivity ($\rho_{bo} > \rho_{ao}$) emerges at about 75 K, much higher than $T_S$ and $T_N$ in the unstressed sample [Fig. 1(d)]. Similar behavior has been reported in 122 series.8-10 Recent neutron and x-ray scattering experiments have pointed out that the in-plane anisotropy of the resistivity in detwinned BaFe$_2$As$_2$ could be related to the lattice distortion.27,28 The applied uniaxial pressure or strain could smear the structural transition and increase the onset temperature of the lattice distortion. Therefore, we attribute the 75 K observed here to be the structural transition temperature ($T'_S$) in uniaxially stressed samples.

ARPES studies were performed at the SIS beamline of the Swiss Light Source (SLS). All the data were taken with a Scienta R4000 electron analyzer, the overall energy resolution was 15 meV and the angular resolution was 0.3 degree. The samples were cleaved in situ and measured under ultra-high vacuum of $5 \times 10^{-11}$ torr. The polarization-sensitivity of orbitals in ARPES is a powerful tool to identify the orbital characters.
FIG. 2. (Color online) The experimental setup and matrix element calculations for three 3d orbitals. The mirror planes are defined by the analyzer slit and the sample surface normal. The two-dimensional plot of the Brillouin zone of NaFeAs is illustrated by red solid squares. Note that the photoemission cross-sections are amplified by a factor shown at the upper-left corner of the panels. Thus, all the panels could be shown in the same color scales. There is a minor asymmetry in certain distributions caused by the out-of-plane component of the polarization.

of band structure. The photoemission intensity is proportional to the matrix element of the photoemission process \( |M_{\beta,i}| \), which can be described by \( |M_{\beta,i}| \propto |\langle \psi_{\beta} | \hat{\epsilon} \cdot r | \psi_{i} \rangle|^2 \), where \( \hat{\epsilon} \) is the unit vector of the electric field of the light, and \( \psi_{\beta} \) is the initial-state wave function. For high kinetic-energy photoelectrons, the final-state wave function \( \psi_{\beta} \) can be approximated by a plane-wave state \( e^{i k \cdot r} \) with \( k \) in the mirror plane. By rotating the polarization of incoming photons or the azimuth angle of the sample, four experimental geometries could be achieved named as \( P, P_R, S, S_R \), respectively (Fig. 2). The subscript “R” stands for the rotation of the sample. Since the low-lying electronic structures at the Fermi energy \( (E_F) \) of iron pnictides are commonly believed to be mostly composed of the \( dxz, dyz, \) and \( d_{xy} \) orbitals, the matrix element distributions for these three orbitals were calculated in four geometries. As shown in Fig. 2, the matrix element distributions of the \( dxz, dyz, \) and \( d_{xy} \) orbitals exhibit a strong polarization dependence throughout the first Brillouin zone, reflecting the opposite symmetry of these two orbitals. The matrix element distribution of \( d_{xy} \) is suppressed along the direction parallel to the in-plane component of the polarization, due to its odd spatial symmetry with respect to the mirror planes.

III. THE ELECTRONIC STRUCTURE IN THE TETRAGONAL PARAMAGNETIC STATE

Based on previous studies, Fig. 3(a) sketches the Fermi surface of NaFeAs, which consists of two hole pockets (\( \alpha \) and \( \beta \)) around the zone center (\( \Gamma \)) and two electron pockets (\( \eta \) and \( \delta \)) around the zone corner (\( M \)) in the tetragonal PM state. The orbital characters are dominated by the \( dxz, dyz, \) and \( d_{xy} \) orbitals. The distribution of \( dxz \) is equivalent to that of the \( dyz \) orbital after rotating 90 degrees, which is protected by the \( C_4 \) rotational symmetry in the tetragonal PM state. For simplicity, the continuous orbital evolution on the Fermi surface and the orbital mixing on the bands are not illustrated here, which should be finite due to the hybridization of the bands with different orbitals. According to the calculation in Sec. II, we could predict the observable Fermi surfaces in different geometries as shown in Fig. 3(b). The
polarization dependence of the Fermi surface is only sensitive to the in-plane component of the polarization. The low-lying electronic structure of NaFeAs is illustrated in Fig. 3(c). The bands dispersed along \( k_x \) and \( k_y \) directions are distinguished with the “\( \alpha \)” and “\( \gamma \)” subscripts here and throughout the paper. As shown by the band calculation, the \( \beta_x \) and \( \gamma_x \) bands should connect with the band bottoms of \( \eta_x \) and \( \delta_x \) at the \( M \) point, respectively, which is guaranteed by the continuous evolution of bands in the unfolded Brillouin zone for one iron ion per unit cell. Note that, since the electronic structure of NaFeAs shows little \( \eta \) dependence in both the tetragonal PM and CAF states, we only consider a two-dimensional electronic structure hereafter.

To further confirm the orbital character of the electronic structure, the photoemission intensity maps obtained in the \( p \) and \( s \) geometries are compared in Fig. 4(a). Around \( \Gamma \), the photoemission intensity distribution rotates 90 degrees from \( p \) to \( s \) geometry, which is consistent with the predicted distribution of the \( d_{xz} \) and \( d_{yz} \) orbitals on the \( \alpha \) and \( \beta \) hole pockets. The electron pockets are observed either around \( M_y \) in the \( p \) geometry or around \( M_x \) in the \( s \) geometry, which correspond to the \( \delta \) electron pockets dominated by the \( d_{xy} \) orbital. We did not observe the \( \eta \) electron pocket here, which could be due to particular photon energy or experimental setup, since the outer \( \eta \) electron pockets could be observed in previous studies or with 118 eV photons as shown later in Fig. 5.

For the low-lying electronic structure, the bands dominated by \( d_{xz}, d_{yz}, \) and \( d_{xy} \) could be selectively probed in different geometries [Fig. 4(b)]. The \( \alpha_x \) band appears in the \( p \) geometry, while the \( \beta_x \) band could be observed in the \( s \) geometry. The \( \gamma_x \) and \( \delta_x \) bands show weak photoemission intensities in the \( s \) geometry due to the small matrix element of the \( d_{xy} \) orbital. These two bands could be observed more clearly in Figs. 5 and 6. We further investigated the photoemission intensity maps at 50 meV below \( E_F \) [Fig. 4(c)]. Two ellipse-like features observed in the \( p \) and \( s \) geometries originate from the \( d_{xz} \) and \( d_{yz} \) orbitals, respectively. They intertwine with each other forming the cross-sections of the \( \alpha \) and \( \beta \) hole pockets. The outmost feature is the \( \gamma \) band cross-section, which is suppressed along the \( k_x \) direction in the \( p \) geometry, consistent with the matrix element distribution of the \( d_{xy} \) orbital. When the sample is cooled down to 20 K, the symmetry breaking of electronic structure is notable on the cross-sections of the \( \alpha \) and \( \beta \) hole pockets [Fig. 4(d)]. Both pockets extend along the AFM direction. The \( d_{xz} \) portions show more anisotropy than the \( d_{yz} \) portions, indicating an orbital-dependent reconstruction of the electronic structure.

![Diagram](image_url)

FIG. 3. (Color online) The sketch of the low-lying electronic structure of NaFeAs in the tetragonal PM state. (a) The sketch of the Fermi surface and its orbital character in the folded Brillouin zone for two iron ions per unit cell (solid lines) in the tetragonal PM state. (b) The observable Fermi surfaces in different experimental geometries. (c) The electronic structure of NaFeAs near \( E_F \) in the tetragonal PM state.

IV. THE ELECTRONIC STRUCTURE OF THE NEMATIC STATE

As shown in Sec. III, although the simple matrix element calculation is not quantitative, there is a good qualitative agreement between the prediction and our experimental results. Therefore, NaFeAs provides an ideal opportunity for us to study the temperature dependence of \( d_{xz}, d_{yz}, \) and \( d_{xy} \) separately along both AFM and FM directions. We will focus on the temperature dependence of the bands that make the electron pockets first.

A. The temperature dependence of the \( \eta \) and \( \delta \) bands

There are two electron pockets \( \eta \) and \( \delta \) around the zone corner. The \( \eta_x \) bands could be clearly identified crossing...
Fig. 4. (Color online) The polarization sensitivity of bands with different orbitals. (a) The photoemission intensity maps integrated over a 10-meV energy window at \( E_F \) taken at 80 K in the \( p \) and \( s \) geometries. (b) The photoemission intensity \( I(k, \omega) \) taken at 80 K in the \( p \) and \( s \) geometries along \( \Gamma - M_\alpha \) direction. The band dispersions are overlayed on the photoemission intensities with the solid lines. (c) The left and middle panels are the photoemission intensity maps integrated over a 10-meV energy window at 50 meV below \( E_F \) taken at 80 K in the \( p \) and \( s \) geometries, respectively. The observed cross-sections of bands are summarized in the right panel. (d) Same as described for panel (c) but taken at 20 K. All the data were taken with 107-eV photons.

\( E_F \) along the \( k_x \) direction in the \( p \) geometry [Fig. 5(a)]. Their Fermi crossings move outward continuously as the temperature decreases [Fig. 5(b)]. The momentum shift for \( \eta_y \) is about 0.02 \( \pi/a \) from 60 to 20 K. For the \( \delta \) electron pocket, we took the data along the momentum cut around \( M_y \) in the \( p \) geometry in order to enhance its photoemission intensity. Note that, the band dispersions along the \( k_x \) direction should be equivalent for \( M_x \) and \( M_y \), which is protected by the translational symmetry. As shown in Figs. 5(c)–5(e), when the temperature decreases, the MDCs peak intensities of \( \delta_x \) suppress near \( E_F \) promptly, indicating an energy gap opening for the \( \delta_x \) band along the \( k_x \) direction.

Along the \( k_y \) direction, the photoemission intensity of \( \eta_y \) is too weak to be distinguished [Fig. 5(f)]. As will be discussed later, this is because the \( \eta_y \) bands shift up above \( E_F \). The tiny MDCs peaks observed in Fig. 5(g) might be due to the
FIG. 5. (Color online) The temperature-dependent photoemission data of the η and δ bands around the zone corner. (a) The $I(k,\omega)$ taken at 60 and 20 K in the $p$ geometry along Cut 1. (b) The temperature dependence of the momentum distribution curves (MDCs) at $E_F$ along Cut 1. The momentum position of Cut 1 is shown by a black arrow in the inset of panel (b). (c) The $I(k,\omega)$ and its second derivative image with respect to energy $[\partial^2 I(k,\omega)/\partial \omega^2]$ taken at 60 and 20 K in the $p$ geometry along Cut 2. (d) The temperature dependence of the MDCs at $E_F$ along Cut 2. The momentum position of Cut 2 is shown by a black arrow in the inset of panel (d). (e) The MDCs selected from $I(k,\omega)$ geometry along Cut 3. (f), (g) Same as described for panels (c) and (d) but taken in the $p_R$ geometry along Cut 3. (h), (i) Same as described for panels (c) and (d) but taken in the $p_g$ geometry along Cut 4. (j) The sketch of the electron pockets in the tetragonal PM and orthorhombic PM states. All the data were taken with 118-eV photons. The color scales of $I(k,\omega)$ and $[\partial^2 I(k,\omega)/\partial \omega^2]$ are shown in the bottom-right inset. These color scales and notations are used throughout the paper. The band dispersions are overlayed on the photoemission intensities with the solid lines, which are determined with the help of the second derivative images and MDCs.

minor twinned domains of the sample. The $\delta_x$ portions of the inner electron pocket show weak temperature dependence [Figs. 5(h) and 5(i)]. The Fermi crossings of $\delta_y$ slightly shrink with a momentum shift of about 0.02 $\pi/a$ from 60 to 20 K.

The observed in-plane anisotropy of the electron pockets preserves at the temperature as high as 60 K, which is far above the structural and magnetic transition temperatures in unstressed sample. This suggests that the reconstruction of the electronic structure might be closely related to the stress and thus lattice distortion, whose onset temperature ($T_\alpha'$) increases due to the uniaxial pressure (More evidences in Fig. 8). Therefore, under the structural transition, the electron pockets reconstruct significantly as shown in Fig. 5(j). Two portions vanish in the orthorhombic PM state and the remaining electron pocket extends along the $k_x$ direction.

B. The temperature dependence of the $\alpha$, $\beta$, and $\gamma$ bands

To investigate the $\beta$ and $\gamma$ bands, temperature-dependent data taken in the $s$ geometry along the $k_x$ direction are shown in Fig. 6(a). Remarkable band reconstruction was observed. Near $\Gamma$, the $\beta_x$ and $\gamma_x$ bands open a large hybridization gap at 20 K. As a result, the Fermi crossings of $\beta_x$ are pushed outward from $\Gamma$, and the band top of $\gamma$ shifts downward to about 40 meV below $E_F$ [Figs. 6(a), 6(b), and 6(d)]. The $\beta_x$ band disperses toward $E_F$ around $M$ forming a hole-like band. It hybridizes with the $\delta_x$ electron pocket and opens an energy gap at $E_F$ upon lowering the temperature. This is consistent with the gap opening of $\delta_x$ observed in Figs. 5(c)–5(e). Moreover, the band shift and hybridization push the $\beta_x$ band above $E_F$ around $M$. Considering the fact that the $\beta_x$ band should connect with the band bottom of $\eta_x$ [Fig. 3(c)], we could further confirm the observation in Fig. 5(f) that the $\eta_x$ portions of the outer electron pocket shift up above $E_F$ in the orthorhombic PM state. Note that the long-range magnetic order could halve the Brillouin zone by folding the bands between $\Gamma$ and $M$. We thus attributed $\eta_x$ near $\Gamma$ together with $\beta_x$ and $\alpha_x$ around $M$ to be the folded bands in the magnetic state.
In contrast to the complex behavior along \( k_x \), the \( \beta \) band does not reconstruct along the \( k_x \) direction [Fig. 6(e)]. The Fermi crossings of \( \beta \) are almost temperature independent as shown in Fig. 6(f). Around the \( M \) point, the \( \beta \) band shifts downward to about \(-50\) meV at the lowest temperature. Since \( \beta \) should connect with the band bottom of \( \eta \), [Fig. 3(d)], such an energy shift of \( \beta \) is consistent with the enlargement of \( \eta \) observed in Fig. 5(a). We summarized the reconstruction of \( \beta \) in Fig. 6(g). The \( \beta \) hole pocket extends along the AFM direction in the orthorhombic PM state.

The reconstruction of the inner \( \alpha \) hole pocket is shown in Fig. 7. Along the \( k_y \) direction, the \( \alpha \) band splits into two parts, indicating possible hybridization with other bands [Figs. 7(a) and 7(c)]. The hybridization with \( \delta_1 \) could be first excluded. According to the temperature-dependent data, while band shift of the lower part of \( \alpha \) is notable at 60 K [Fig. 7(d)], the folding of \( \delta_1 \) occurs at a much lower temperature of about 49 K [Fig. 7(b)]. Moreover, no hybridization was found for the \( \delta_1 \) band in Fig. 5(h). Therefore, the \( \alpha \) band should hybridize strongly with the \( \gamma \) band in the orthorhombic PM state, as shown in the inset of Fig. 7(c). In this case, a large proportion of the \( d_{yz} \) orbital on \( \alpha \) would be mixed into \( \gamma \) due to such hybridization. Consequently, we note that the photoemission intensity of \( \gamma \) is suppressed at 20 K around the \( \Gamma \) point in Fig. 6(e), as the \( d_{yz} \) orbital could not be observed in the \( s_{R} \) geometry. Along the \( k_x \) direction, the \( \alpha \) bands show weak temperature dependence. Their Fermi crossings slightly move outward about 0.02 \( \pi /a \) from 60 to 20 K [Figs. 7(e)–7(g)]. Therefore, similar to the \( \beta \) hole pocket, the \( \alpha \) hole pocket extends along the AFM direction in the orthorhombic PM state as shown in Fig. 7(h).

Our previous APRES study on twinned NaFeAs has found that the short range magnetic order emerges just below the structural transition temperature of 54 K, which causes a band folding.\(^\text{24}\) However, when the structural transition is enhanced to 75 K here by the uniaxial pressure, we still observe the folding of \( \delta_1 \) bands below \(-49\) K [Fig. 7(b)]. Therefore, the short-range magnetic order observed above \( T_N \) in previous study should be related to the fluctuation of the long-range magnetic order, instead of a direct correlation with the structural transition.

V. DISCUSSION

To further investigate the orbital-dependent behaviors of the reconstruction in the orthorhombic PM state, the bands dominated by the \( d_{x^2-y^2} \) and \( d_{x^2+y^2} \) orbitals are highlighted in Fig. 8(a). The most remarkable feature found here is the hybridization gap opening at almost all band crossings. As a result, the bands split into the upper and lower bands around \( \Gamma \) and open an energy gap at \( E_F \) around \( M \). Such reconstruction could redistribute the orbital weight of \( d_{yz} \) and \( d_{xy} \), inducing a
The momentum position of Cut 1 is shown by a black arrow in the inset of panel (b). (c) The MDCs selected from Cut 2. (h) The sketch of the CAF state in the tetragonal PM and orthorhombic PM states. All the data were taken with 118-eV photons.

FIG. 7. (Color online) The temperature-dependent photoemission data of the α band around the zone center. (a) The $I(k,\omega)$ and $\delta^2 I(k,\omega)/\partial \omega^2$ taken at 60 and 20 K in the $p_F$ geometry along Cut 1. (b) The temperature dependence of the MDCs at $E_F$ along Cut 1. The momentum position of Cut 1 is shown by a black arrow in the inset of panel (b). (c) The MDCs selected from $E_F$ along Cut 1. Each MDC has been individually normalized by its integrated weight to highlight weak features. (d) The temperature dependence of the MDCs at 50 meV below $E_F$ along Cut 1. (e), (f), and (g) Same as described for panels (a), (b), and (c) but taken in the $p$ geometry along Cut 2. (h) The sketch of the α hole pocket in the tetragonal PM and orthorhombic PM states. All the data were taken with 118-eV photons.

The onset temperature of such splitting could be estimated to be around 75 K, where the in-plane anisotropy of resistivity emerges ($T_N^\#1$). This result is consistent with previous study on the “122” series of iron pnictides,12 and suggests that the orbital weight redistribute significantly under the structural transition. Moreover, the smooth evolution of the electronic structure reconstruction into the CAF state indicates that both the magnetic and structural transitions share the same driving force.24

So far, there are mainly two categories of theoretical proposals for understanding the origin of the nematic electronic ordered state above $T_N$. One category of theories suggests that the nematicity could originate from spin fluctuations.17–19 The spin fluctuations could couple with the nematic electronic order and break the lattice symmetry via electron-lattice coupling.17 The other category of theories suggests that the orbital degree of freedom could be dominating. Different types of orbital ordering were proposed to be accompanied by the magnetic order.20 Especially, the ferro-orbital ordering originated from the inequivalent occupations of $d_{xz}$ and $d_{yz}$ was proposed to drive the structural transition and stabilize the long-range magnetic order.21–23 Our results provide several important quantitative tests for these theoretical proposals. We could first check the inequivalent occupation between the
FIG. 8. (Color online) The orbital-dependent reconstruction of the electronic structure. (a) The low-lying electronic structure in the tetragonal PM and orthorhombic PM state, respectively, where only the $d_{yz}$- and $d_{xy}$-dominated bands are highlighted. The band dispersions above $E_F$ are shown by dashed lines, considering the continuous dispersion of the bands with certain orbitals. (b) Cartoon shows the orbital weight distributions near $E_F$ in the tetragonal PM and orthorhombic PM states. (c) The low-lying electronic structure in the tetragonal PM and orthorhombic PM state, respectively, where only the $d_{xz}$-dominated bands are highlighted. (d) The temperature dependence of the EDCs at $k_1$ as indicated by the gray line in panel (a). (e) The temperature dependence of the EDCs at $k_2$ as indicated by the gray line in panel (c). (f) The peak positions of the $\beta_x$ and $\beta_y$ bands as functions of temperature.

d_{xz}$ and $d_{yz}$ orbitals, which is considered to be important for the formation of ferro-orbital ordering.\textsuperscript{21–23} The occupation change of the $d_{xz}$ orbital could be calculated from the size increase of the $\eta_x$ electron pocket subtracted by the size increase of the $\alpha_x$ hole pocket. The momentum shift of the $\eta_x$ Fermi crossings is about 0.02 $\pi/a$ from 60 to 20 K. Considering the band shift above 60 K [Fig. 8(f)], this momentum shift might be doubled to 0.04 $\pi/a$, corresponding to a maximum occupation change of about 0.04 electron per iron. Moreover, the Fermi momenta variations are almost the same for the enlargement of the $\eta_x$ electron pocket and the $\alpha_x$ hole pocket. Therefore, the occupation change of the $d_{xz}$ orbital should be infinitesimal, which suggests that the ferro-orbital ordering only involving $d_{xz}$ and $d_{yz}$ plays a minor role in driving the nematic transitions here.\textsuperscript{21} Note that, although the occupation of $d_{xz}$ is almost invariant, the orbital polarization could be strong only at the low energy. Since the orbital weight of $d_{xz}$ and $d_{yz}$ is partially gaped away from $E_F$, the Fermi surface could be dominated by the $d_{xz}$ orbital in the orthorhombic state.\textsuperscript{16,37} More intriguingly, instead of the pure ferro-orbital ordering between $d_{xz}$ and $d_{yz}$ orbitals, the reconstruction of the electronic structure involves mostly the $d_{yz}$ and $d_{xy}$ orbitals. These two orbitals are found numerically to contribute much larger magnetic moment than $d_{xz}$ in the magnetic ordered state.\textsuperscript{22,34–37} Therefore, the orbital weight redistribution observed here below $T'_S$ could be related to the spin polarization of the $d_{xz}$ and $d_{yz}$ orbitals. In this scenario, the strong coupling between spin fluctuations and the orbital degree of freedom could induce an orbital-dependent spin polarization far above
the Neel temperature, which further reinforces the spin fluctuations and the nematicity of electronic structure. Such a positive feedback process can break the C4 rotational symmetry and thus induce the structural transition. Consistently, the enhancement of spin fluctuations was observed below the structural transition temperature. Our results therefore suggest that the nematic spin fluctuations at high temperatures and their coupling with the orbital degree of freedom could be the dominating cause for the nematic electronic state in the iron pnictides. Note that the feedback process could be further accelerated by an uniaxial stress. It thus could explain the dramatic enhancement of $T_S$ by the rather small pressure applied in our experiment.

In the magnetic state, the electronic structure is folded between $\Gamma$ and $M$ due to the long-range magnetic order. According to the photoemission intensity maps in the $s$ and $p$ geometries [Figs. 9(a) and 9(b)], the Fermi surface morphology and the band structure in the orthorhombic CAF state are shown in Figs. 9(c) and 9(d). The electron pocket folds from $M$ to $\Gamma$ and then hybridizes with the $\beta$ hole pocket. As a result, four small pockets are formed. There are two bright spots along AFM direction dominated by the $d_{xz}$ and $d_{yz}$ orbitals. Since they are formed by two bands with opposite symmetries [Figs. 9(c) and 9(d)], these two small pockets are intensively discussed as Dirac points in 122 series. The other two pockets are formed by the $\delta y$ and $\beta y$ bands along the FM direction [Figs. 9(c) and 9(d)].

We summarized the reconstruction of Fermi surface in Fig. 9(e). The $C_4$ rotational symmetry of the Fermi surface is broken under the structural transition. The anisotropic Fermi surface observed in the orthorhombic PM state could be responsible for the in-plane anisotropic transport behaviors above $T_N$. All the pockets show anisotropic shape with an extension along the $k_x$ direction. Especially, the transport properties contributed by the electron pockets could be highly anisotropic, since two portions of electron pockets are gaped away from $E_F$ in the orthorhombic PM state. When entering the orthorhombic CAF state, the Fermi pockets are folded...
between $\Gamma$ and $M$. The Fermi surface is thus characterized by four distinct pockets originated from the hybridization between the hole and electron pockets. These features qualitatively agree with previous studies in the 122-series,\textsuperscript{1,11,12,15,35} which highlight the unified nature of the nematic magnetic state in iron pnictides. Furthermore, the Fermi crossings of $\beta$, $\delta_x$, and $\eta_x$ are sensitive to the structures, $k_z$, and doping levels. Therefore, the topology of these four pockets could be variable in different compounds. For example, the pockets distributed along FM direction are hole-like in NaFeAs, but electron-like in 122 series.\textsuperscript{11,12,15,35} In that case, the topology change of these distinct features could be responsible for the nonmonotonic doping dependence of the in-plane resistivity anisotropy observed in 122 series.\textsuperscript{8-10,42}

VI. SUMMARY

To summarize, our polarization-dependent ARPES study on detwinned NaFeAs clearly demonstrates the details of the electronic structure reconstruction in the nematic state. We found that the rotational symmetry breaking of electronic structure is closely related to the in-plane anisotropy of the resistivity, whose onset temperature is far above the Neel transition temperature. More intriguingly, such a symmetry breaking originates from the reconstruction of the electronic structure and exhibits orbital-dependent behaviors. The $dx_{yz}$- and $d_{x^2-y^2}$-dominated bands split near $\Gamma$ and open an energy gap around $M$, while the $d_{yz}$-dominated bands only exhibit an energy shift without any reconstruction. Our observations exclude the theories based on the ferro-orbital ordering only involving the $dx_{yz}$ and $d_{x^2-y^2}$ orbitals, since the occupation of $dx_{yz}$ is almost temperature independent. Instead, the strong coupling between the nematic spin fluctuations and the orbital degree of freedom are suggested to drive the nematicity. Such a coupling causes an orbital-dependent spin polarization, and thus the development of finite magnetic moments of the $dx_{yz}$ and $d_{x^2-y^2}$ orbitals as manifested in their weight redistributions. Our findings provide the experimental foundation for a microscopic understanding of the nematic state of iron pnictides.

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