Unidirectional Electronic Structure in the Parent State of Iron-Chalcogenide Superconductor

Fe$_{1+\delta}$Te

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We use scanning tunnelling microscopy and spectroscopy to explore the electronic structure of Fe$_{1+\delta}$Te which is the parent compound of the iron-chalcogenide superconductors. A unidirectional electronic structure with a period of $a_0$ (where $a_0$ is the lattice constant) along the $a$-axis is observed. The unidirectional pattern is visible within an energy range from approximately -200 to +130 meV. Since the direction of the unidirectional electronic structure coincides with those of the underlying antiferromagnetic and the predicted orbital orders, it is presumable that the observed unidirectional structure is closely tied to these orders and is peculiar to the parent state in iron-based superconductors.

KEYWORDS: Scanning tunneling microscope and spectroscopy, Iron-based superconductors, Iron-chalcogenide

1. Introduction

Strong electronic correlation yields several orders, which attract much attention in solid state physics since the complex entanglement of these orders creates many interesting and unique phenomena. One of the most intriguing examples of such phenomena is the high temperature superconductivity in the cuprates with antiferromagnetic (AFM) order and several types of electronic orders.\cite{1-10} Another example is the colossal magnetoresistance in the manganites with magnetic, orbital, and charge order.\cite{11-15} The recently discovered iron-based superconductors (SCs)\cite{16-24} also have AFM\cite{25-28} and orbital orders\cite{29-35} in their parent compounds. The chemical doping of the parent compounds, inducing the fluctuation of these orders, causes the high temperature superconductivity in iron-based SCs: it has been theoretically argued that the electron pairing is mediated by the spin fluctuation\cite{36-39} or the orbital fluctuation.\cite{40-43} Thus, in iron-based materials, the elucidation of the effect of these orders on the electronic structure is necessary to understand the pairing mechanism.

Recently, scanning tunnelling spectroscopy (STS) experiments based on scanning tunnelling microscope (STM) have revealed unidirectional ($C_2$ symmetric or nematic) electronic structures in the parent compounds CaFe$_{1.94}$Co$_{0.06}$As$_2$ (122 system)\cite{43} and LaOFeAs (1111 system).\cite{44} Even though the physical picture regarding such unidirectional structures is still under debate,\cite{45-47} the unidirectional electronic structure appears to be strongly tied to the underlying AFM and/or orbital orders, because the direction of the unidirectional features coincides with those of these orders. To verify whether the electronic unidirectionality is really related to the underlying orders, it is quite important to investigate the electronic structures in the parent materials containing AFM and orbital orders with different spin and orbital configurations.

One of the candidates containing a different configuration is Fe$_{1+\delta}$Te, which is the parent compound of the iron-chalcogenide SCs. This material exhibits the structural tran-

![Fig. 1. (Color online) (a) and (b) Schematic illustrations of the crystal structure of Fe$_{1+\delta}$Te. Red and blue arrows indicate the spin configurations at Fe sites in FeTe layer. The direction of AFM order coincides with $a$-axis.](attachment:Fe1+_delta_Te_crystal_structure.png)
3. Results

3.1 Typical surface structure and identification of crystal a- and b-axes

In a typical STM image of the sample surface, a square lattice structure is observed, as shown in Fig. 2(a). The lattice constants in the figure are approximately 3.8 Å which is in good agreement with the inter-chalcogen distance. In addition to this particle-hole asymmetry, kink modulation as shown in Fig. 3(c) and 3(d). Figure 3(a) shows Fourier transform image of (a). Four solid arrows (two red and two blue arrows) represent the spots corresponding to the atomic lattice of the exposed top Te atoms. (b) Line cuts of the FT image along $a_1$ (red circles) and $a_2$ (blue triangles) directions. The peak positions in these line cuts are indicated by red and blue arrows and named as $q_1$ and $q_2$, respectively. (c) Tunneling spectrum obtained by averaging the spectra on several excess Fe sites (red) and the spatially averaged spectrum all region except excess Fe sites (black).

Unidirectionality coincides with those of the underlying orders, as in the 122 and 1111 systems. Therefore, the electronic unidirectionality seems to be related to the underlying orders and to be one of the inherent features in the parent materials of iron-based SCs.

2. Experiment

The samples used in this study were cut from a single crystal of Fe$_{1+y}$Te grown by a melting-growth technique. The molar ratio of the grown crystal was determined to be Fe$_{1.07}$Te by inductively coupled plasma mass spectroscopy. The AFM and structural transition temperatures $T_s$ were determined to be 64 K by SQUID magnetization and resistivity measurements. The STS measurements were performed in a helium gas environment at 4.2 K on an atomically flat surface, as shown in Fig. 2(a), prepared by cleaving the samples in-situ at 4.2 K. At this temperature, the crystal structure of the sample is monoclinic: the lattice constants along the a- and b-axes are not equivalent. We measured the $I$-$V$ characteristics and obtained the differential conductance $G(r, E = eV_b) = dI/dV$ by numerical differentiation.

3.2 Typical tunneling spectra

A tunnelling spectrum averaged over the surface except for the excess Fe sites is shown by the black curve in Fig. 2(c). A finite conductance at 0 meV ($E_F$) is consistent with the metallic state in this material below $T_s$. There is a significant particle-hole asymmetry characterized by the fact that the conductance in negative high energy is greater than that in positive. In addition to this particle-hole asymmetry, kink structures are observed at $\sim +130$ meV and $\sim \pm 50$ meV as pointed out by the arrows in Fig. 2(c). On the excess Fe sites, the conductance at $E_F$ increases and the particle-hole asymmetry becomes more pronounced as shown by the red curve in Fig. 2(c). The local enhancement of the conductance at $E_F$ near the excess Fe atoms is consistent with the previous STS experiments in superconducting Fe$_{1+y}$Te$_{1-x}$Se$_x$. 

3.3 Unidirectional LDOS modulation

To investigate the spatial variation of the local density-of-state (LDOS), we performed spectroscopic imaging measurements. Figure 3 shows an STM image and a conductance map at 0 meV [G(r, 0 meV) map] with the same FOV as in the STM image. In the conductance map, there are several bright spots at the excess Fe sites. In the background of these bright spots, a stripe like or streaky structure running along the b-axis is visible as shown in Fig. 3(b) and more clearly in the inset of Fig. 3(b): a unidirectional modulation along the a-axis exists in this conductance map. (A clear correlation between the excess Fe atoms and the unidirectional pattern cannot be observed in our measurements). FT analysis of the $G(r, 0$ meV) map also supports the existence of this unidirectional modulation as shown in Fig. 3(c). Figure 3(d) shows the line cuts of the topographic signal (red) and the simultaneously measured conductance (blue) along the white arrow in Fig. 3(a). This indicates that the conductance modulation...
3.4 Energy evolution of unidirectional LDOS modulation

Figure 4 shows the energy dependence of the conductance maps. The conductance modulations can be observed from $E \sim -200$ to $+130$ meV. To clarify the energy dependence of the electronic unidirectionality more quantitatively, we plots the FT magnitudes $G(q, E)$ of the conductance maps along the $a$- (solid circles) and $b$-axes (open circles) as a function of $q$ for different values of energy in Fig. 5. The difference between the $G(q, E)$ curves along the $a$- and $b$-axes indicates the unidirectionality of the LDOS modulations. For positive energy, the difference between the FT magnitudes at $q = 2\pi/a_0$ and $2\pi/b_0$ gradually increases from $E = 0$ to $+120$ meV, abruptly decreases at $+130$ meV and is almost undefined above $+160$ meV. On the other hand, for negative energy, the difference gradually decreases as the energy decreases from $E = 0$ to $-200$ meV, becoming invisible from $E = -200$ to $-280$ meV. Although the physical explanation of this energy asymmetry is not understood at present, this effect will likely be one of the key clues to understand the origin of the unidirectional electronic structure.

4. Discussion

4.1 Relation with underlying orders

The direction of the observed unidirectional electronic structure coincides with the spin antiparallel direction of the underlying bicollinear AFM order. Such a unidirectional electronic structure has not been observed in the superconducting Fe$_{1+\delta}$Se$_2$Te$_{1-\delta}$ ($x \sim 0.4, 0.15$) which shows no AFM order. Therefore, it is presumable that the observed unidirectional structure is closely tied to the underlying AFM order. However, it remains an open question what kind of mechanisms connects the AFM order (spin state) with the unidirectional electronic structure (electronic state).

The theoretically suggested orbital ordering in the parent phase is one of the important candidates mediating between the spin and electronic states. In Fe$_{1+\delta}$Te, the structural distortion below $T_c$ lifts the degeneracy of $d_{xz}$ and $d_{yz}$ and shifts the energy level of $d_{xz}$ to be higher than that of $d_{yz}$. According to Turner et al., the $d_{xz}$ orbital is fully occupied by a tightly bound electron whose spin creates the bicollinear AFM order, whereas the $d_{yz}$ orbital is partially occupied by a mobile electron. Consequently, the movable electron density periodically modulates along the spin anti-parallel direction or the crystal $a$-axis. Our results indicating the unidirectional electronic structure along the $a$-axis around $E_F$ are consistent with this prediction.

4.2 Comparison with the other materials

Here, we compare the unidirectional electronic structures in Fe$_{1+\delta}$Te with those in the 122 and 1111 parent materials. The direction of the electronic unidirectionality in Fe$_{1+\delta}$Te corresponds to the spin antiparallel direction in the
underlying AFM order as seen in the 122 and 1111 systems, although the directions of the AFM order are different from those in the 122 and 1111 systems. Therefore, the electronic unidirectionality appears to be strongly tied to the underlying order and to be the inherent phenomenon in the parent phase. On the contrary, we find several different features in Fe\textsubscript{1+y}Te: (i) No signature clearly indicating that the unidirectional structure is pinned by some impurities could be observed. (ii) The period in Fe\textsubscript{1+y}Te is \(a_0\) which is considerably shorter than that in CaFe\textsubscript{1+y}Co\textsubscript{0.06}As\textsubscript{2} (which is approximately \(8a_0\)). From these differences, it might be believed that the coupling mechanism of the electronic unidirectionality to the underlying orders in Fe\textsubscript{1+y}Te is different from that in CaFe\textsubscript{1+y}Co\textsubscript{0.06}As\textsubscript{2}.

5. Summary

In summary, we have investigated the electronic structure lying in the parent state of the iron-chalcogenide superconductor Fe\textsubscript{1.07}Te by using scanning tunnelling spectroscopy. We find a unidirectional electronic structure with a period of \(a_0\) along the \(a\)-axis which coincides with the spin antiparallel direction of the underlying AFM order and the predicted orbital order. The unidirectional nature is visible within an energy range from about -200 to +130 meV. Even though the direction of the underlying orders in Fe\textsubscript{1+y}Te is different from that in other parent materials, the direction of the unidirectional structure corresponds to the spin antiparallel direction of the underlying AFM order in Ca(Fe\textsubscript{1+y}Co\textsubscript{0.06}As\textsubscript{2}.\textsuperscript{43}) and LaOFeAs.\textsuperscript{44} Therefore, it is plausible that the electronic unidirectionality is strongly tied to the underlying orders and is universal and inherent in the nature of the parent state of the iron-based SCs. The observed electronic unidirectionality provides us an expectation that an in-plane resistivity anisotropy can be found in the detwinned Fe\textsubscript{1+y}Te in the same manner for the 122 materials near the parent phase.\textsuperscript{53} On the contrary, we find several differences between the features of the unidirectional structure in Fe\textsubscript{1+y}Te and Ca(Fe\textsubscript{1-y}Co\textsubscript{y}As\textsubscript{2}.\textsuperscript{43}) (i) there is no distinct signature indicating that the unidirectional structure is pinned by some impurities, and (ii) the period is just \(a_0\) which is different from the value of approximately \(8a_0\) in CaFe\textsubscript{1+y}Co\textsubscript{0.06}As\textsubscript{2}. From these differences, the coupling mechanism of the electronic unidirectionality to the underlying orders seems to be different between these materials. The results in this study provide important hints to understand how the underlying orders couple to the electronic structure in the parent materials of the iron-based superconductors.

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