Angle-Resolved Photoemission Spectroscopy of the Iron-Chalcogenide Superconductor $Fe_{1.03}Te_{0.7}Se_{0.3}$: Strong Coupling Behavior and the Universality of Interband Scattering

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We have performed angle-resolved photoemission spectroscopy of the iron-chalcogenide superconductor $Fe_{1.03}Te_{0.7}Se_{0.3}$ to investigate the electronic structure relevant to superconductivity. We observed a holelike Fermi surface (FS) and an electronlike FS at the Brillouin zone center and corner, respectively, which are nearly nested by the $Q \sim (\pi, \pi)$ wave vector. We do not find evidence for the nesting instability with $Q \sim (\pi + \delta, 0)$ reminiscent of the antiferromagnetic order in the parent compound $Fe_{1+y}Te$. We have observed an isotropic superconducting (SC) gap along the holelike FS with the gap size Δ of $\sim$4 meV ($2\Delta/k_BT_c \sim 7$), demonstrating the strong-coupling superconductivity. The observed similarity of low-energy electronic excitations between iron-chalcogenides and iron-arsenides strongly suggests that common interactions which involve $Q \sim (\pi, \pi)$ scattering are responsible for the SC pairing.

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The discovery of iron-based superconductors [1] has triggered fierce debates on the superconducting (SC) mechanism, since it is hard to explain high-T_c value of $\sim$55 K [2] within the conventional phonon-mediated SC framework [3]. Experimentally, both hole- and electron-doped $BaFe_2As_2$ superconductors (122 system [4]) exhibit an anomalously strong pairing behavior on small Fermi surfaces (FSs) which are connected by the $Q_1 \sim (\pi, \pi)$ wave vector, suggesting the importance of interband interactions for the occurrence of high-T_c superconductivity [5]. Since the parent compounds of the 122 system commonly show an antiferromagnetic (AF) long-range order with the Q_1 vector [6], remnant AF spin fluctuations with a similar wave vector may be responsible for the pairing interactions in doped compounds [7]. However, the universality of the interband scattering via Q_1 in all the iron-based superconductors as well as the role of magnetism for the pairing are still unclear. In particular, the FS topology of the iron-chalcogenide superconductor $Fe_{1+y}Te_{1-x}Se_x$ (11 system [8]), as well as its SC order parameter, is highly controversial. Although the density functional calculations for stoichiometric FeTe(Se) have predicted that the FS shape is very similar to that of FeAs compounds [9], a remarkably different FS may emerge in the actual case due to an electron doping through the introduction of excess Fe atoms [10] which partially occupy the interstitial sites of (Te,Se) layers [11]. This controversy is due to the lack of experimental data on the FS of the SC samples, because the previous angle-resolved photoemission spectroscopy (ARPES) experiments have been performed only for the AF phase of the parent $Fe_{1+y}Te$ [12]. The appearance of AF order with $Q_2 = (\pi + \delta, 0)$ in the $Fe_{1+y}Te$ [11,13], corresponding to a 45°-rotated AF order as compared to that of the 122 system, raises a major question concerning the relationship between magnetism and superconductivity. The presence of multiple spin excitations in the Se-substituted SC samples makes this issue even more complicated [14]. As for the symmetry and magnitude of the SC gap, previous experimental results, mostly from indirect measurements of the excitation gap, range from single or multiple isotropic nodeless gap(s) [15–19] to highly anisotropic nodeless or nodal gap(s) [15,19–21], with the pairing strength varying from the nearly weak coupling limit [22,23] to the strong-coupling regime [15,17–19,24]. It is thus definitely important to clarify the low-energy electronic excitations in $Fe_{1+y}Te_{1-x}Se_x$ and compare their relationship with the magnetic excitations in order to gain an insight into the high-T_c mechanism and the interplay between the superconductivity and the magnetism.

In this Letter, we report the first direct observation of the FS and the SC gap and its symmetry of SC $Fe_{1.03}Te_{0.7}Se_{0.3}$ ($T_c = 13$ K) by ARPES. We show that the FS topology is essentially similar to that of the optimally-doped $BaFe_2As_2$ [5], in contrast to a recent ARPES study in the AF phase of the parent $Fe_{1+y}Te$, where an additional FS is found at the X point [12]. The observed FS topology suggests the importance of interband interactions via Q_1 for the pairing. We discuss the interplay between the AF fluctuations and the superconductivity.

The high-quality single crystals of $Fe_{1.03}Te_{0.7}Se_{0.3}$ ($T_c = 13$ K) were grown by the Bridgeman method [25]. ARPES

measurements were performed at beam line 28A in Photon Factory, KEK, Tsukuba ($h\nu = 44$ eV) and at Tohoku University ($h\nu = 21.218$ eV). The energy resolution was set at 1.7 and 12 meV for the measurements of the SC gap and the valence band dispersion, respectively. Since the sample is unstable in the atmosphere [26], almost all of the preparation procedures such as sample mounting were done in a glove box filled with Ar gas. Clean surfaces for the ARPES measurements were obtained by cleaving crystals *in situ* in a working vacuum better than 1×10^{-10} Torr.

Figure 1(a) shows the ARPES spectra measured along the Γ-M line of the Brillouin zone for the two-Fe unit cell. A holelike band is clearly observed, and appears to cross E_F around the Γ point. We also observe a less dispersive band at $\sim$100 meV around the M point and a broad feature at $\sim$300 meV around the Γ point. To see the near-E_F dispersion in more detail, we show in Figs. 1(b) and 1(c) the momentum distribution curves (MDCs) and their second derivative intensities [Figs. 1(d) and 1(e)]. As clearly seen in Figs. 1(b) and 1(d), there are two holelike bands centered at the Γ point. The outer band creates a hole FS, whereas the inner band does not cross E_F. Around the M point [Figs. 1(c) and 1(e)], we observe a shallow electronlike band crossing E_F, whose bottom of dispersion is at $\sim$50 meV. The observed outer hole and electron bands likely correspond to the α_2 and β_1/β_2 bands, respectively,

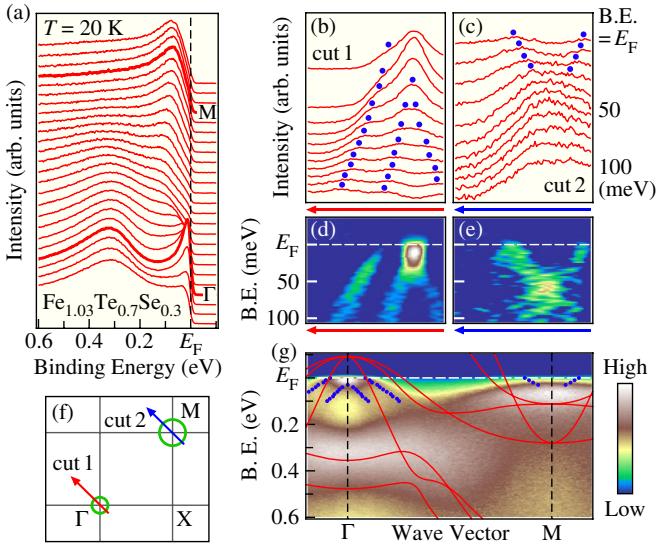

FIG. 1 (color online). (a) ARPES spectra of $Fe_{1.03}Te_{0.7}Se_{0.3}$ measured at 20 K with $h\nu = 44$ eV. (b),(c) MDCs in the vicinity of E_F measured along cut 1 and cut 2 in (f), respectively. Blue dots denote the peak positions of MDCs. (d),(e) Second derivative plot of MDCs along cut 1 and cut 2, respectively, as a function of binding energy and wave vector. (f) Schematic FS (green circles) with the locations of momentum cuts (red and blue arrows). (g) Intensity plot of (a) together with the calculated energy bands for FeTe at $k_z = 0$ (red curves) [9]. Near-E_F band dispersions extracted from the MDC peak positions are also shown by blue dots.

in the previous ARPES study on $Fe_{1+y}Te$ [12]. While an additional hole band (α_3) which crosses E_F is observed in the previous study, it is not clearly seen in the present study. On the other hand, we observed an inner hole band corresponding to the α_1 band which was missing previously. The estimated Fermi velocity is $\sim$0.4 eV Å for both the hole and the electron bands, comparable to that for optimally doped 122 compound [27], while it is smaller than the value reported for the parent $Fe_{1+y}Te$ ($\sim$ 0.7 eV Å) [12]. This difference may reflect the change of band dispersions and/or correlation strength by the Se substitution. In Fig. 1(g), we plot the ARPES intensity compared with the density functional calculations [9]. The calculated bands plotted in Fig. 1(g) are renormalized by a factor of 2 and hence the bandwidth is twice narrower as compared to the original calculation. Although the observed overall band structure appears to roughly track the renormalized band calculations, there are remarkable discrepancies especially in the near-E_F region. For instance, the observed two holelike bands at the Γ point are located at higher binding energy as compared to the calculated bands. On the other hand, the observed electron pocket at the M point is located at lower binding energy. This opposite shift between the hole and the electron pockets cannot be simply explained in terms of electron doping by the presence of excess Fe atoms. Interestingly, a similar behavior has been reported for FeP- [28] and FeAs-based [27,29] superconductors, suggesting that it is a general trend of Fe-based superconductors.

In Fig. 2, we plot the ARPES intensity at E_F. We clearly identify FSs centered at the Γ and the M points,

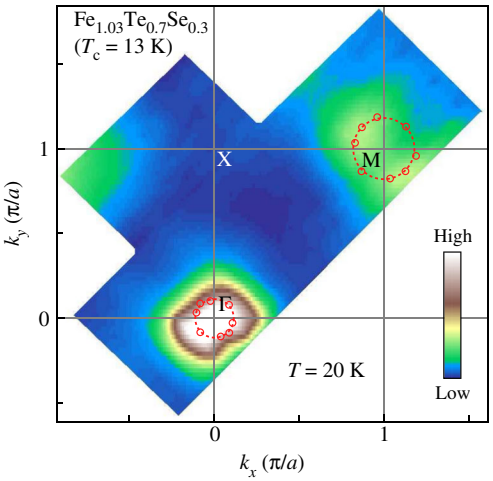

FIG. 2 (color online). ARPES intensity plot at E_F of $Fe_{1.03}Te_{0.7}Se_{0.3}$ as a function of the two-dimensional wave vector measured at 20 K with 44 eV photons. The intensity at E_F is obtained by integrating the spectra within $\pm$10 meV with respect to E_F. Solid and dashed red circles show experimentally determined k_F points and schematic FSs, respectively. There are sizable experimental uncertainties on the k_F points, mainly due to weak intensity around the M point.

corresponding to the hole and the electron pockets, respectively, essentially similar to the FS topology of the 122 superconductors [5]. No indication of a FS is found at the X point, in contrast to a recent ARPES study in the AF phase of the parent $Fe_{1+y}Te$ [12]. The disappearance of the FS around X is consistent with the disappearance of long-range AF order with the Q_2 vector in the SC sample, which would fold the FS at Γ to the X point in the parent compound.

In order to elucidate the character of the SC gap, we have performed ultrahigh-resolution ($\Delta E = 1.7$ meV) ARPES measurements in the close vicinity of E_F. Figure 3(a) shows the temperature dependence of the near-E_F ARPES spectrum measured across T_c at k_F of the outer holelike band [30]. The midpoint of the leading edge at 5 K is shifted toward higher binding energy by ~0.6 meV, suggesting the opening of a SC gap, and the intersection of the ARPES spectra measured above and below T_c is also away from E_F (see inset). To eliminate the effect of the Fermi-Dirac distribution function, we have symmetrized the ARPES spectrum at each temperature as shown in the bottom of Fig. 3(c). On decreasing temperature, the spectral weight near E_F is transferred to high binding energy below T_c. The spectral shape is distinctly different between 5 K and 17 K, whereas no essential difference is seen between 17 K and 25 K, indicating that the spectral feature

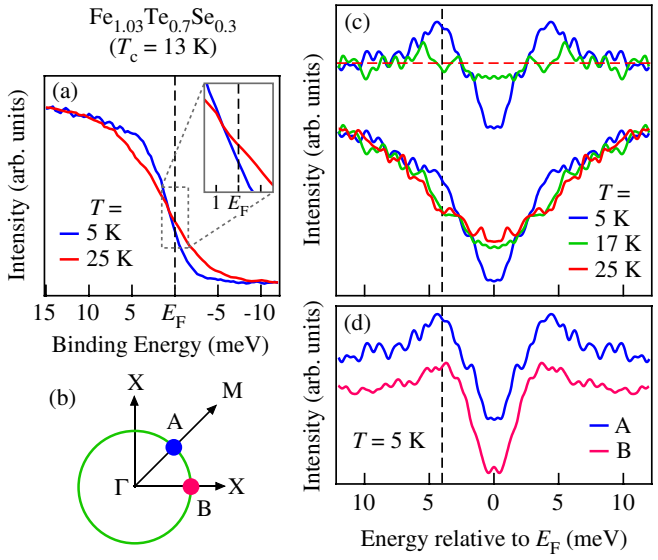

FIG. 3 (color online). (a) ARPES spectra of $Fe_{1.03}Te_{0.7}Se_{0.3}$ with the He Iα resonance line, measured at point A displayed in (b). The inset shows the expansion in the vicinity of E_F. (b) Schematic holelike FS at the Γ point with the location of the k_F points A and B. (c) Temperature dependence of symmetrized ARPES spectra (bottom) at point A, and the same but divided by the spectrum at $T = 25$ K (top). (d) Comparison of the symmetrized spectra at k_F points along Γ-M (blue curve) and Γ-X (red curve) high-symmetry lines measured at 5 K divided by the 25 K spectrum. Dashed lines at 4 meV in (c) and (d) represent the energy scale of the SC gap Δ.

varies drastically across T_c. We divided the symmetrized ARPES spectra measured at 5 and 17 K by the 25 K spectrum [top of Fig. 3(c)] to cancel out the V shaped spectral density of states due to the tail of the inner holelike band. Apparently, a coherence peak emerges at the binding energy of ~4 meV at 5 K, unambiguously demonstrating the opening of a SC gap. The estimated SC gap size ($\Delta \sim 4$ meV) corresponds to a $2\Delta/k_BT_c$ value of ~7, indicating the strong-coupling nature of superconductivity in $Fe_{1.03}Te_{0.7}Se_{0.3}$. This is in good agreement with the strong-coupling behavior suggested by some measurements [15,17–20,24]. On the other hand, a much smaller $2\Delta/k_BT_c$ value of ~3.5 has been reported by scanning tunneling spectroscopy measurements [22,23]. This discrepancy may be due to the opening of a smaller SC gap on different FS(s) as observed in the 122 system [5] or the presence of a large SC gap anisotropy [15,19–21]. To examine a possible gap anisotropy, we measured an ARPES spectrum along the Γ-X cut [point B in Fig. 3(b)]. We immediately notice that the observed energy position of the coherence peak for points A and B almost coincides, suggesting that the SC gap is likely to be isotropic on the hole FS within the present experimental accuracy. Consequently the reported small gap value is not understood in terms of the gap anisotropy on the hole FS, while we cannot rule out the possibility of opening of a small gap or a large gap anisotropy on the electron FS.

Now we discuss the implication of the present results in comparison with the 122 compounds. It is useful to summarize the present ARPES results: (i) we observed a hole pocket at the Γ point and an electron pocket at the M point, (ii) the hole pocket at the X point observed in $Fe_{1+y}Te$ [12] is absent in $Fe_{1.03}Te_{0.7}Se_{0.3}$, and (iii) a large and nearly-isotropic SC gap ($\Delta \sim 4$ meV) with a strong-coupling value ($2\Delta/k_BT_c \sim 7$) opens at the hole pocket. These experimental facts show that the low-energy electronic structure responsible for the superconductivity in $Fe_{1.03}Te_{0.7}Se_{0.3}$ is qualitatively similar to that of the 122 superconductors [5], indicating that the effect of electron doping by an excess of Fe is overestimated in a recent theoretical study which predicted the appearance of a large square-type FS both at the Γ and the X points due to the chemical potential shift [10]. The present results further suggest that the Q_2 AF spin fluctuations observed by the inelastic neutron scattering of the Se-substituted SC sample [11,13,14] are less important for the SC pairing in $Fe_{1.03}Te_{0.7}Se_{0.3}$, since Q_2 does not connect the observed FSs. Instead, the observed hole and electron FSs are connected by Q_1, similarly to the case of the 122 system [5]. Interestingly, recent neutron scattering measurements revealed the presence of AF spin fluctuations extending at least up to 20 meV near Q_1 even well above T_c [14,31], consistent with the interband scattering condition between hole and electron pockets observed in the present ARPES study, and those fluctuations appear only in the SC sample.

It is thus inferred that the interband scattering through Q_1 AF fluctuations plays an important role for the emergence of superconductivity. The observed large SC gap size is consistent with the scenario that the interband scattering promotes the SC pairing.

Finally, we discuss the composition dependence of physical properties in $Fe_{1+y}Te_{1-x}Se_x$. It has been reported that the SC transition is better defined (e.g., a clear specific heat jump at T_c) in crystals with a smaller amount of excess Fe (y) and a larger Se concentration (x) up to $x = 0.5$ [32]. We think that the likely competing Q_2 and the Q_1 AF spin fluctuations play different roles to the superconductivity and that this competition would be a key to understand the composition dependence of the SC character. For samples with a larger excess of Fe (y) and smaller Se values (x), the presence of Q_2 spin excitations [11,13,14] may cause a reduction of the SC volume fraction, since those spin fluctuations would not be favorable to the SC pairing as discussed above. With decreasing excess Fe and increasing Se concentration, the Q_2 spin fluctuations are suppressed, whereas the Q_1 spin fluctuations, which would assist the pairing and lead to stronger superconductivity, are enhanced, in agreement with the inelastic neutron scattering measurements [11,13,14,31]. A systematic study of the electronic structure for a wide range of excess Fe (y) and Se compositions (x) is the next step toward a full understanding of the SC mechanism in $Fe_{1+y}Te_{1-x}Se_x$.

In conclusion, we have determined the band dispersions, the FS topology, and the momentum dependence of SC gap of $Fe_{1.03}Te_{0.7}Se_{0.3}$. While the experimentally determined low-energy electronic structure is distinctly different from that of the parent AF phase, it is qualitatively similar to that of the 122 superconductors. Our results suggest that the coupling to Q_2 AF correlations is suppressed in $Fe_{1.03}Te_{0.7}Se_{0.3}$ and the SC state arises from the interband interactions between hole and electron FSs via Q_1, suggesting the importance of common AF spin fluctuations to the SC pairing of Fe-based superconductors.

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Note added.—After submission of this manuscript, we became aware of related ARPES works [33,34] on the $Fe_{1+y}Te_{1-x}Se_x$ superconductor. They studied the electronic structure in the normal state of $x = 0.34$ [33] and 0.42 [34] and revealed the presence of hole and electron FSs at Γ and M points, respectively, and the absence of FS at X point. These results on the normal state are consistent with the present study for $x = 0.3$. In this study, we further performed the low-temperature and high-resolution measurements and clarified the SC gap and its symmetry.

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