Unusual Temperature Evolution of Quasiparticle Band Dispersion in Electron-Doped FeSe Films

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Abstract: The discovery of high-temperature (high-$T_c$) superconductivity in one-monolayer FeSe on SrTiO$_3$ has attracted tremendous attention. Subsequent studies suggested the importance of cooperation between intra-FeSe-layer and interfacial interactions to enhance $T_c$. However, the nature of intra-FeSe-layer interactions, which would play a primary role in determining the pairing symmetry, remains unclear. Here we have performed high-resolution angle-resolved photoemission spectroscopy of one-monolayer and alkaline-metal-deposited multilayer FeSe films on SrTiO$_3$, and determined the evolution of quasiparticle band dispersion across $T_c$. We found that the band dispersion in the superconducting state deviates from the Bogoliubov-quasiparticle dispersion expected from the normal-state band dispersion with a constant gap size. This suggests highly anisotropy pairing originating from small momentum transfer and/or mass renormalization due to electron–boson coupling. This band anomaly is interpreted in terms of the electronic interactions within the FeSe layers that may be related to the high-$T_c$ superconductivity in electron-doped FeSe.

Keywords: iron-based superconductors; thin films; ARPES; electronic structure

1. Introduction

Iron selenide (FeSe) is structurally the simplest iron-based superconductor with the superconducting-transition temperature ($T_c$) of ~9 K [1]. Intriguingly, one-monolayer (1 ML) film of FeSe grown on SrTiO$_3$ substrate exhibits exceptionally high $T_c$ [2]. The $T_c$ value reported by transport measurements reaches 40 K [2,3], which is about five times higher than the bulk counterpart. In addition, Cooper pairing at a higher temperature of 65 K, which exceeds the highest $T_c$ (56 K) ever achieved in iron-based superconductors, has been suggested from a gap-closing temperature by angle-resolved photoemission spectroscopy (ARPES) [4–7] and Meissner effect by mutual conductance measurements [8]. These observations triggered fierce debates on the origin of the $T_c$ enhancement in 1 ML-FeSe film. One key ingredient is a novel cross-interface electron–phonon coupling. The strong coupling between electrons in the FeSe layer and optical phonons of SrTiO$_3$ has been verified via the observation of replica bands by ARPES and theoretically proposed to enhance $T_c$ in most of possible pairing symmetries [9,10]. Later, the close link between strong electron–phonon coupling and $T_c$ enhancement has been supported experimentally, e.g., by isotope effects [11,12]. With these findings as a guiding principle, the search for high $T_c$ in atomically thin films of other iron-based superconductors interfaced with SrTiO$_3$ [13–15] has been accelerated. Another key ingredient for $T_c$ enhancement is a charge transfer from SrTiO$_3$. Heavy electron doping to the FeSe layer leads to unique electronic structure...
consisting only of electron-like Fermi surfaces [4,5,16], in contrast to the semimetallic nature of bulk FeSe [17,18]. The electron doping is essential for the high-$T_c$ superconductivity, as established by the observation of high $T_c$ above 40 K even in multilayer and bulk FeSe by doping electron carriers [19–21]. Therefore, there is accumulated evidence that the FeSe layer has the capability of inducing 40-K superconductivity through electron doping and the interfacial electron–phonon coupling will assist further $T_c$ or pairing enhancement. However, little is known about why electron doping leads to the high $T_c$’s above 40 K. In particular, interactions within the FeSe layer, which would primarily determine the pairing symmetry, remain unclear.

In this study, we performed a comparative ARPES experiment on the surface of 1 ML- and Cs-deposited 20 ML-FeSe films on SrTiO$_3$, where the interfacial effects were present and absent, respectively. We demonstrated anomaly in the quasiparticle-band dispersions in the superconducting state, which is not expected from the Bogoliubov-quasiparticle (BQP) dispersion induced by a simple $s$-wave-gap opening. We discuss implications of our observation in relation to intra-FeSe-layer interactions.

2. Materials and Methods

The molecular beam epitaxy method was used to obtain 1 ML- and 20 ML-FeSe films; the films were grown on a TiO$_2$-terminated Nb(0.05 wt%)-SrTiO$_3$ substrate (SHINKOSHA) by simultaneously evaporating Fe and Se atoms while keeping a substrate temperature at 430 °C with a deposition rate of 0.01 ML/s [19]. Electron doping to 20 ML-FeSe was realized by evaporating Cs atoms onto the film surface at room temperature using a Cs dispenser (SAES Getters) [22]. After the growth, the film was transferred to the ARPES-measurement chamber without exposure to air. ARPES measurements were performed with a SES2002 spectrometer (Scienta Omicron) with the He-I$\alpha$ resonance line ($h\nu = 21.218$ eV) at Tohoku University. The film was kept under an ultrahigh vacuum of $5 \times 10^{-11}$ Torr during the ARPES measurement, and no remarkable surface degradation was observed for a typical measurement time of 1 day. The energy and angular resolutions were set to be 7–12 meV and 0.2°, respectively. A gold film which made electrical contact with the film was referenced to calibrate the Fermi level ($E_F$).

3. Results

First, we present the electronic structure of Cs-deposited 20 ML-FeSe film measured at $T = 50$ K. As shown in Figure 1a,b, there was a circular-shaped large Fermi surface at the Brillouin-zone corner (M point) which originated from $E_F$ crossing of an electron band with the bottom of the dispersion around 50 meV below $E_F$. The top of a hole-like band around the zone center ($\Gamma$ point) was about 50 meV below $E_F$, resulting in the absence of a hole-like Fermi surface in contrast to the as-grown multilayer FeSe film [4,5] or bulk FeSe [17,18]. These observations confirmed a successful electron doping by Cs deposition onto the FeSe surface. The electron carrier concentration ($n_e$) calculated from the Fermi-surface volume was $\sim 0.11 \text{ electrons/Fe}$, which corresponds to the optimal doping level with $T_c$ value of $\sim 40$ K [22]. To investigate how the band structure changes by the superconducting transition, we performed high-resolution measurements across $T_c$ (50 and 13 K) along a momentum ($k$) cut A indicated by a blue line in Figure 1a. The results displayed in Figure 1c,d show that while the electron band above $T_c$ crossed $E_F$ at the Fermi wave vector ($k_F$) of $\sim 0.17 \pi/a$, the band dispersion below $T_c$ had a local maximum below $E_F$ so as not to cross $E_F$ due to a superconducting-gap opening. It is noted that the $k$ location of the electron-band top below $T_c$ coincided with the $k_F$ point above $T_c$, consistent with the Cooper-pairing origin of the observed gap. The superconducting-gap opening is also clearly seen in energy distribution curves (EDCs) in Figure 1e, in which the peak position at $k_F$ ($k_3$ defined in Figure 1c,d) was shifted from $E_F$ to a high binding energy by $\sim 10$ meV with decreasing the temperature to form a superconducting gap. Since the nodeless $s$-wave superconductivity is realized in electron-doped multilayer FeSe [19,20], one can see the superconducting-gap opening below $T_c$ irrespective of the $k$ cut, e.g., along the $k$ cut.
crossing the M point (cut B), as shown in Figure 1f–h, where essentially the same behavior with cut A was recognized.

An important finding manifests itself when we compare the band dispersions of the normal and superconducting states. Figure 2a displays a direct comparison of the experimental band dispersions extracted from the peak position of EDCs in cut A. As mentioned above, the band dispersion below \( T_c \) exhibited an opening of the superconducting gap and resultant bending-back behavior with the top of the dispersion at \( k_F \). Such a characteristic band dispersion below \( T_c \) was qualitatively consistent with the dispersion relation of BQP\,s in the Bardeen–Cooper–Schrieffer (BCS) theory, where BQP dispersion \( (E_k) \) is expressed as \( E_k = \sqrt{\epsilon_k^2 + |\Delta|^2} \) \( (\epsilon_k \text{ and } \Delta \text{ are the normal-state band dispersion and the superconducting-gap size, respectively}) \) [24]. For a quantitative comparison, we determined \( \epsilon_k \) by performing a polynomial fitting to the ARPES data above \( T_c \) (magenta curve) and simulated \( E_k \) by assuming a \( k \)-independent superconducting-gap size of 10 meV (light blue curve). Intriguingly, the band dispersion below \( T_c \) shows a clear deviation from the simulated BQP dispersion \( E_{\nu} \); specifically, although the simulation predicted a finite downward energy shift of BQP dispersion compared with \( \epsilon_k \) even in the \( k \) region far away from \( k_F \) (at least down to \( k_y = 0.05 \pi/a \)) because of a large \( \Delta \) value with respect to the shallow electron-band bottom, the experimental dispersion below \( T_c \) became nearly identical to \( \epsilon_k \) as soon as it moved away from \( k_F \). Almost temperature-insensitive band position away from \( k_F \) was also clearly visible in the comparison of raw EDC\,s in Figure 1e (see EDC\,s at \( k_1 \) and \( k_2 \)). The same behavior was observed at different momentum, e.g., we found that the band dispersion measured below \( T_c \) along cut B deviated from the simulated BQP dispersion over a wide \( k \) region (see Figure 2b; also see a comparison of EDC\,s in Figure 1h).

**Figure 1.** (a) ARPES intensity map at \( E_F \) as a function of two-dimensional wave vector for Cs-deposited 20 monolayer (ML)-FeSe film obtained at \( T = 50 \text{ K} \) with \( h\nu = 21.218 \text{ eV} \). Intensity at \( E_F \) was obtained by integrating the spectral intensity within \( \pm 10 \text{ meV} \) of \( E_F \). Green circle is a guide for the eyes to trace the Fermi surface. (b) Plot of ARPES intensity along the \( \Gamma M \) cut at \( 50 \text{ K} \) as a function of binding energy and wave vector. (c,d) Near-\( E_F \) ARPES intensity along cut A in (a) at \( T = 50 \text{ and } 13 \text{ K} \), respectively, divided by the Fermi–Dirac distribution (FD) function at each temperature convoluted with the resolution function. Intensity above \( E_F \) is displayed up to \( 3k_BT \). Red and blue circles in (c,d), respectively, are the band dispersion determined by fitting the energy distribution curves (EDCs) with Bardeen–Cooper–Schrieffer (BCS) spectral function [23]. (e) Comparison of EDC\,s between \( T = 50 \text{ K} \) (red) and \( 13 \text{ K} \) (blue) taken at representative \( k_F \) points \([k_1, k_2, \text{ and } k_3 \text{ indicated by magenta lines in (c,d)}]\). Red and blue dots indicate the local maxima corresponding to the peak position. (f–h) Same as (c–f) but obtained along cut B in (a).
symmetry

To clarify whether the energy difference between the experimental and simulated BQP dispersions below \( T_c \) was an essential ingredient of electron-doped high-\( T_c \) FeSe films, we investigated the band-structure evolution in 1 ML-FeSe (Figure 3). For this purpose, we performed high-resolution measurements on slightly underdoped 1 ML-FeSe (\( n_e = 0.09 \)) with \( T_c \approx 40 \) K because a sharp spectral line shape compared with the heavily doped sample (\( T_c = 65 \) K; \( n_e = 0.12 \)) [4] is suited for accurately determining the quasiparticle band dispersion. As is well known, 1 ML-FeSe has a large electron-like Fermi surface centered at the M point. The electron band which forms the Fermi surface showed an opening of the superconducting gap (\( \Delta \approx 10 \) meV) below \( T_c \), as highlighted by the characteristic bending-back behavior with the minimum-gap locus at \( k_F \) (see Figure 3b). A direct comparison of the band dispersions above and below \( T_c \) in Figure 3c demonstrates that an energy shift due to the superconducting gap opening was limited to the \( k \) region around \( k_F \approx 0.16 \pi/a \) (compare red and blue circles), in sharp contrast to a clear downward shift of the simulated BQP dispersion for \( k_F \leq 0.1 \pi/a \) (light blue curve). Similarly, the case of Cs-deposited 20 ML-FeSe suggested that the deviation of the experimental band dispersion from the simulated BQP dispersion below \( T_c \) is a common feature of electron-doped FeSe films irrespective of film thickness.

![Figure 2](image1.png)

Figure 2. (a) Comparison of the near-\( E_F \) band dispersions in Cs-deposited 20 ML-FeSe at \( T = 50 \) K (red circles) and 13 K (blue circles) along cut A in Figure 1a. Magenta curve is the normal-state band dispersion \( \epsilon_k \) extracted from a polynomial fitting to the red circles. Light blue curve is the calculated Bogoliubov-quasiparticle (BQP) dispersion based on the BCS formula \( E_k = \sqrt{\epsilon_k^2 + |\Delta|^2} \) with a constant \( \Delta \) of 10 meV. (b) Same as (a) but for cut B in Figure 1a.

![Figure 3](image2.png)

Figure 3. (a,b) ARPES intensity divided by the FD function measured along the \( \kappa \) cut crossing the M point in 1 ML-FeSe at \( T = 50 \) K and 13 K, respectively. Red and blue circles show the band dispersion extracted from the peak position of the EDCs. (c) Comparison of the near-\( E_F \) band dispersions at \( T = 50 \) K (red circles) and 13 K (blue circles), together with \( \epsilon_k \) determined by polynomial fitting to the red circles (magenta curve) and the BQP dispersion \( E_k = \sqrt{\epsilon_k^2 + |\Delta|^2} \) simulated with a constant \( \Delta \) of 10 meV (light blue curve).
4. Discussion and Conclusions

Now we are going to discuss the origin of the observed anomaly in quasiparticle dispersion. To simulate BQP dispersion $E_k = \sqrt{\epsilon_k^2 + |\Delta|^2}$, we assumed that $\epsilon_k$ is the same as the dispersion above $T_c$ and $\Delta$ is $k$-independent. It would be natural to consider that one or both of these assumptions are incorrect, rather than thinking that the BQP picture was broken in the electron-doped FeSe. For simplicity, we consider in the following the two extreme cases that the deviation was induced by a change in either $\epsilon_k$ or $\Delta$. First, to examine the $k$ dependence of $\Delta$ as the origin, we put the experimental band dispersions below and above $T_c$ into $E_k$ and $\epsilon_k$, respectively, and estimated $\Delta(k)$ which reproduces the experimental band dispersion below $T_c$. The obtained $\Delta(k)$ was strongly $k$-dependent as seen from Figure 4a,b for 1 ML- and Cs-deposited 20 ML-FeSe, respectively; namely, $\Delta(k)$ is finite only in the narrow $k$ region centered at $k_F$ (within $\pm0.02 \, \pi/a$ of $k_F$), so that band dispersion only around $E_F$ was shifted toward high binding energies by the superconducting transition, consistent with our observations. An unusual Cooper pairing in the limited $k$ space near $k_F$ may be caused by pairing interactions which have small momentum transfer $q$ [25–27]. For instance, it has been proposed by Migdal–Eliashberg theory for 1 ML-FeSe that forward scattering with small $q$ phonons produces highly anisotropic superconducting gap peaked at $k_F$ and also leads to temperature-independent band structure away from $k_F$ [25], in qualitative agreement with $\Delta(k)$ in Figure 4a as well as band dispersion in Figure 3. Although this theory assumes a cross-interface coupling between small $q$ phonons of SrTiO$_3$ and electrons in 1 ML-FeSe as the key pairing interactions, our observation of anisotropic $\Delta(k)$ in 20 ML-FeSe (Figure 4b) where interfacial effects are negligible suggests that small-$q$ interactions within the FeSe layers may be also responsible for superconductivity if the $k$-dependent pairing was indeed a source of the deviation from the simulated BQP dispersion.

Next, we consider another possibility that $\epsilon_k$ is temperature-dependent whereas $k$ dependence of $\Delta(k)$ is small. To explain the observed deviation between the experimental and simulated BQP dispersions, $\epsilon_k$ below $T_c$ must be shifted toward $E_F$ compared with the normal-state band dispersion above $T_c$ while keeping the same $k_F$ position. Such an energy shift would be a consequence of mass renormalization linked to the superconducting transition, likely due to coupling with bosonic modes as reported for bulk crystals of high-$T_c$ superconductors [28–33]. Here we defined the energy difference between the experimental and simulated BQP dispersions as $\Delta E$ (see black arrow in Figure 3c) and plotted it in Figure 4c,d for 1 ML- and 20 ML-FeSe films, respectively. As seen from Figure 4c,d, $\Delta E$ showed a broad peak around 20 meV in 1 ML- and 20 ML-FeSe.
obtained $\Delta E$ value can be used as a measure of the mass enhancement similarly to the real part of self-energies. By analogy with the fact that the peak position in the real part of self-energies below $T_c$ corresponded to $\Delta + \Omega$, where $\Omega$ is the energy of bosonic modes coupled to electrons, the observed peak structure at ~20 meV suggests a coupling to low-energy modes with $\Omega \sim$10 meV (here, $\Delta \sim$10 meV). The origin of the corresponding modes is an important open question; candidates include phonons [34] and magnetic resonance [35]. Nevertheless, one important outcome from our observation is that low-energy modes intrinsic to the FeSe layer must be involved because the mass renormalization was found not only in 1 ML-FeSe, but also in 20 ML-FeSe.

In summary, we reported the evolution of low-energy band dispersion across $T_c$ in 1 ML- and Cs-deposited 20 ML-FeSe films on SrTiO$_3$. We found deviation of the band dispersion below $T_c$ from the simple BQP dispersion simulated with the temperature-independent $\epsilon_k$ and $k$-independent $\Delta$. We proposed two possible scenarios as the origin of this observation; (i) anisotropic $\Delta(k)$ peaked around $k_F$ due to the superconducting pairing by small $q$ transfer and (ii) enhancement in the effective mass in the superconducting state due to the coupling to low-energy bosonic modes. In either scenario, the observed similarity between 1 ML- and 20 ML-FeSe suggested intra-FeSe-layer nature of the interactions. Our result lays a foundation for understanding the mechanism of high-$T_c$ superconductivity in electron-doped FeSe.

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