Temperature insensitivity of Fermi surface in electron-doped high-temperature FeSe films on SrTiO$_3$

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Abstract. We have performed high-resolution angle-resolved photoemission spectroscopy study of electron-doped multilayer FeSe films on SrTiO$_3$. We determined the evolution of the electronic structure in a wide temperature range and found temperature-insensitive nature of the Fermi surface in contrast to the strong temperature dependence observed in non-doped FeSe. Such a striking doping dependence would provide a key to understanding the origin of unconventional fermiology and physical properties in FeSe.

1. Introduction
The discovery of iron-based superconductors [1] opened a new avenue for the study of high-temperature superconductivity. An intriguing character of iron-based superconductors is their multiband nature which is believed to play an important role for antiferromagnetic and nematic orders in the non-doped parent compound and for high-temperature superconductivity in the doped compound [2]. Theoretical calculations for iron-based superconductors generally predict the formation of hole-like and electron-like Fermi surfaces at the Brillouin-zone center and corner, respectively [3-5]. While such multiband nature has been confirmed by experiments such as angle-resolved photoemission spectroscopy (ARPES) studies [6-9], there are some disagreements between the theoretical and experimental results and their origin remains unclear. One of the major issues is the pronounced temperature dependence of the Fermi surface; namely, recent ARPES studies on FeSe revealed that the size of the Fermi surface strongly changes with temperature even in the normal state [10, 11]. This unusual temperature evolution lays the foundation for the emergence of nematic order which leads to the $d_{x^2-y^2}/d_{xy}$ orbital splitting [12, 13] and also for unconventional superconductivity in the Bardeen-Cooper-Schrieffer (BCS) and Bose-Einstein condensation (BEC) crossover regime [14], suggesting the importance of further systematic study on the electronic structure over a wide temperature range.

Recently, FeSe is also attracting considerable attention due to the discovery of high-temperature superconductivity in single-layer films on SrTiO$_3$ substrate [15]. The reported superconducting
transition temperature (\(T_c\)) exceeds 65 K [16-19] which is significantly higher than \(T_c\) of \(~8\) K for bulk FeSe [20]. The importance of interfacial effects, such as the electron-phonon coupling across the interface between FeSe and SrTiO\(_3\), has been proposed as the origin of the observed drastic \(T_c\) enhancement [15, 21, 22]. In addition to the interfacial effects, the electron doping from SrTiO\(_3\) is essential for high-temperature superconductivity [16, 17]. In fact, electron doping by alkaline-metal deposition leads to high-temperature superconductivity above 40 K in multilayer FeSe films [23] and even in bulk FeSe [24], where the interfacial effects are absent. A question naturally arises as to how the electronic structure evolves with temperature in electron-doped high-temperature FeSe superconductor; is it similar to that of non-doped FeSe? Answering this question would provide a key for understanding the origin of unusual fermiology and physical properties of FeSe.

In this paper, we report high-resolution ARPES study on electron-doped multilayer FeSe films on SrTiO\(_3\). We have succeeded in determining the electronic structure in a wide temperature range. We compare the obtained ARPES data with the previous results on non-doped FeSe, and found marked difference in the temperature dependence of the band shifts.

2. Experiment

The 20-monolayer (20-ML) FeSe films were fabricated on TiO\(_2\)-terminated Nb(0.05wt\%)-doped SrTiO\(_3\) substrate (SHINKOSHA) with the molecular-beam-epitaxy (MBE) technique in a vacuum better than \(2 \times 10^{-10}\) Torr [23]. After the fabrication, the film was transferred to the ARPES chamber without exposing to the air. Electron doping necessary for the realization of high-temperature superconductivity was achieved by the Cs deposition onto the film surface at room temperature using a Cs dispenser (SAES Getters) [25]. ARPES measurements were performed in an ultrahigh vacuum better than \(5 \times 10^{-11}\) Torr using a Scienta-Omicron SES2002 electron analyzer with a He discharge lamp (\(h\nu = 21.218\) eV) at Tohoku University. The energy and angular resolutions were set to 12-30 meV and 0.2°, respectively. The Fermi level (\(E_F\)) of the film was referenced to that of a gold film evaporated onto the sample holder.

3. Results and discussion

![Figure 1](image-url)

**Figure 1.** (a) ARPES-intensity mapping at \(T = 30\) K for the Cs-deposited 20-ML FeSe film plotted as a function of a two-dimensional wave vector. The intensity is obtained by integrating the spectral intensity within \(\pm 5\) meV of \(E_F\). (b) and (c) ARPES-intensity plots near \(E_F\) as a function of binding energy (B.E.) and wave vector measured along the cuts A and B shown in (a), respectively. Black and orange dashed curves are a guide for the eyes to trace the band dispersions.
First, we show the electronic structure of Cs-deposited 20-ML FeSe films measured at $T = 30$ K (Fig. 1). Around the Brillouin-zone corner (M point), one can find a large electron-like Fermi surface originating from the $\gamma$ band with the bottom of the dispersion around 50 meV below $E_F$ [Figs. 1(a) and 1(c)]. The formation of the large electron pocket is a manifestation of heavy electron doping to the topmost FeSe layer from the Cs atoms. As a result, the hole-like bands ($\alpha$ and $\alpha'$) at the Brillouin-zone center (\Gamma point) do not cross $E_F$ as seen in Fig. 1(b). The electron carrier concentration ($n_e$) estimated from the size of the electron pocket $\gamma$ is $\sim$0.1 electrons/Fe which is close to the optimal doping where the highest $T_c$ of $\sim$ 40 K is realized [25]. It is noted that, while there is another hole-like band ($\alpha_2$) approaching $E_F$ near the $\Gamma$ point [see finite intensity near the $\Gamma$ point in Fig. 1(a) and orange curve in Fig. 1(b)], it comes from the interior FeSe layers beneath the topmost layer which remain nearly non-doped even after Cs deposition [25]. Therefore, in the following, we will focus only on the $\alpha$, $\alpha'$, and $\gamma$ bands originating from the electron-doped topmost FeSe layer to discuss the temperature-induced evolution of the band structure in the electron-doped region.

Figures 2(a) and 2(b) show the band dispersions around the $\Gamma$ and M points, respectively, measured over a wide range of temperature from 13 to 250 K. At the $\Gamma$ point, spectral intensities of the hole-like $\alpha$ and $\alpha'$ bands are gradually broadened with increasing temperature but are still visible even at the highest temperature of 250 K [Fig. 2(a)]. Similarly, the electron-like $\gamma$ band is clearly observed around the M point irrespective of temperature [Fig. 2(b)]. The observed qualitative similarity of the overall band dispersions between 13 and 250 K indicates the absence of temperature-induced drastic electronic reconstruction such as the lifting of $d_{xy}/d_{xz}$ orbital degeneracy by electronic nematicity found in the low-temperature phase of non-doped FeSe [12, 13].

To investigate the temperature dependence of the band structure in more detail, we have extracted the energy distribution curve (EDC) at the $\Gamma$ point at each temperature as plotted in Fig. 3(a). One can identify two peaks at 13 K at binding energies of $\sim$30 and $\sim$60 meV, which correspond to the top of $\alpha'$ and $\alpha$ bands, respectively. This two-peaked structure is more clearly visible up to higher temperatures in the second-derivative intensity plot in Fig. 3(b), where one can recognize only a weak temperature dependence of the peak positions. The estimated temperature-dependent energy shifts of the $\alpha$ and $\alpha'$ bands are 10 meV or less [see red circles in Fig. 3(b)]. Next, to evaluate an energy shift in the $\gamma$ band,

**Figure 2.** (a) and (b) Temperature dependence of the near-$E_F$ ARPES intensity plotted as a function of binding energy and wave vector measured around the $\Gamma$ and M points, respectively. These cuts nearly trace cuts A and B in Figs. 1(b) and 1(c), respectively.
Figure 3. (a) Temperature dependence of the energy distribution curve (EDC) at the $\Gamma$ point extracted from Fig. 2(a). (b) Corresponding second-derivative intensity plotted as a function of binding energy and temperature. In (b), the two-peaked structure corresponding to the $\alpha$ and $\alpha'$-band tops is magnified. Also, the $\alpha$- and $\alpha'$-band tops estimated from the peak positions in the second-derivative intensity are shown in red circles. (c) The momentum distribution curve (MDC) at $E_F$ at various temperatures extracted from Fig. 2(b). (d) Temperature dependence of the Fermi wave vector ($k_F$) for the $\gamma$ band estimated from the numerical fittings to the MDCs in (c).

at the M point, we show the temperature dependence of the momentum distribution curve (MDC) at $E_F$ in Fig. 3(c) (note that the bottom of the $\gamma$ band is ill-defined in the EDC and hence we utilize MDC here). As clearly seen from Fig. 3(c), the peak position in the MDC which corresponds to the Fermi wave vector of the $\gamma$ band is stationary to temperature (~0.18 $\pi/a$), as confirmed by the result of numerical fittings plotted in Fig. 3(d). This result demonstrates the temperature insensitivity of the Fermi surface in electron-doped FeSe.

The observed temperature insensitivity is in sharp contrast to the pronounced temperature dependence of the Fermi-surface volume reported in non-doped FeSe [10, 11]. In the previous reports, several scenarios such as the band renormalization effect by spin fluctuations, the Pomeranchuk effect, and the chemical potential effect have been intensively discussed as the origin of the pronounced temperature dependence in non-doped FeSe [10, 11]. Since these effects should be doping dependent (e.g. the coupling between electrons and spin fluctuations may be significantly weakened by the disappearance of the hole-like Fermi surface via electron doping and hence the renormalization effect would be reduced), the observed contrasting behavior of the electron-doped versus non-doped regions would provide a key to elucidating the origin of the unusual temperature evolution in non-doped FeSe. Future quantitative analyses on the doping dependence of these effects and the comparison with the present ARPES results are of crucial importance.
4. Summary
We reported high-resolution ARPES results on Cs-deposited 20-ML FeSe films grown on SrTiO$_3$ substrate. The systematic temperature-dependence study revealed the temperature insensitivity of the Fermi surface and the absence of the electronic reconstruction, both of which are in contrast to the recent observations in non-doped FeSe. The contrasting behavior between the electron-doped and non-doped regions would be a key to elucidate the underlying interactions which control the electronic structure and the physical properties of FeSe.

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