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Article

$FeTe_{1-x}Se_x$ monolayer films: towards the realization of high-temperature connate topological superconductivity

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ABSTRACT

We performed angle-resolved photoemission spectroscopy studies on a series of $FeTe_{1-x}Se_x$ monolayer films grown on $SrTiO_3$. The superconductivity of the films is robust and rather insensitive to the variations of the band position and effective mass caused by the substitution of Se by Te. However, the band gap between the electron- and hole-like bands at the Brillouin zone center decreases towards band inversion and parity exchange, which drive the system to a nontrivial topological state predicted by theoretical calculations. Our results provide a clear experimental indication that the $FeTe_{1-x}Se_x$ monolayer materials are high-temperature connate topological superconductors in which band topology and superconductivity are integrated intrinsically.

1. Introduction

High-temperature superconductors, hosts to a non-conventional electron pairing mechanism, and topological electronic materials, for which exotic metallic surface states are protected by symmetries, are two intensively studied areas. Plenty of efforts have been made to search for topological superconductors [1,2]. The simplest model deals with an unconventional chiral superconductor, but there is still no convincing experimental evidence [3,4]. The second type of popular proposals is based on the proximity effect in heterostructures made of topological insulators and *s*-wave superconductors. However, this needs relatively large coherence length and the superconducting transition temperature (T_c) is usually low [5,6].

Interestingly, there is a third type of proposals for realizing topological superconductors in superconductors with topologically nontrivial bands. Here we refer to this type of realization as connate topological superconductivity due to the intrinsic integration of band topological properties and superconductivity. Some iron-based superconductors have been proposed to fulfill this type of realization. For example, the monolayers of FeSe grown on $SrTiO_3$ (STO) substrates, which have a maximum T_c higher than 50 K [7–13], may have a nontrivial topology due to band inversion at the Brillouin zone boundary (M point) [14,15]. However, this band inversion is always far below the Fermi surface as these systems are electron doped. Another candidate is the $FeTe_{1-x}Se_x$ system, in which a topologically nontrivial band inversion takes place near Fermi surfaces around the Z point [16]. The shortcoming of these systems is that the maximum T_c is only about 14 K [17,18].

In this paper, we report that $FeTe_{1-x}Se_x$/STO(001) monolayers have a high T_c similar to that of FeSe/STO monolayers and simultaneously can carry topologically nontrivial band inversion as the bulk $FeTe_{1-x}Se_x$. Thus, this system is a possible candidate to realize high-temperature connate topological superconductivity. We substitute Se by Te in FeSe/STO monolayer films and carry out a systematic angle-resolved photoemission spectroscopy (ARPES) investigation on a series of films with different Se concentrations. The Fermi surface (FS) results suggest comparable high charge transfer in all samples. By tracing the temperature dependence of the superconducting gap, we show that superconductivity persists upon Te substitution of Se, in spite of the dramatic variations of the electronic structure. The electron- and hole-like bands at the Brillouin zone (BZ) center Γ shift towards each other and give rise to a rapid decrease of the band gap, resulting in an evolution towards a

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band inversion and nontrivial Z_2 topological invariant. Our results suggest a highly possible topological phase transition occurring at a low Se concentration in $FeTe_{1-x}Se_x$ monolayer films. We propose several ways to search for topological superconductivity in this unique system by taking advantage of its high flexibility and fine tunability.

2. Materials and methods

Monolayer films of $FeTe_{1-x}Se_x$ were epitaxially grown on 0.7 wt % Nb-doped STO(001) substrates by the same process as FeSe/STO (001) described in our previous work [12]. We have grown samples with different Se concentrations by tuning the flux ratio between Se (99.999%) and Te (99.99%). The Te/Se ratios of the films were estimated based on the Vegard's law and the linear evolution of the band positions at Γ, which will be discussed below, to give the nominal Se concentrations. This can be deviated from true value but will not affect our conclusion. Then the films were transferred *in situ* into the ARPES chamber after annealing. ARPES measurements were carried out using a R4000 analyzer and a helium discharge lamp under ultrahigh vacuum better than 3×10^{-11} torr. The data were recorded with He Iα photons ($h\nu$ = 21.218 eV). The energy resolution was set to ~ 5 meV for gap measurements and ~10 meV for the band structure and FS mapping. The angular resolution was set to 0.2°.

3. Results

The FeSe/STO monolayer films possess a simple FS topology characterized by large electron-like pockets at the M point and the absence of hole-like pockets at Γ [8,9,12], which differs from that of FeSe bulk materials and most of the iron-pnictide superconductors [19]. We show in Fig. 1a the evolution of the FS of $FeTe_{1-x}Se_x$/STO monolayer as a function of the Se content. The electron pockets at M can be observed in all samples. The size of the pockets is rather independent of the substitution, and leads to a carrier concentration of about 0.16 electrons per unit cell according to the Luttinger theorem. This indicates that the charge transfer in this heterostructure is robust, regardless of the Se concentration. We note that the coherence of the spectrum degrades upon Te substitution, which may be due to enhanced electron correlations [20,21] or antiferromagnetic fluctuations [22]. Strong intensity appears at the BZ center as the Se decreases. We will explain below that this originates from an electron band in the unoccupied side, similar to the case of K deposited FeSe/STO monolayer [12] and K deposited bulk $FeTe_{1-x}Se_x$ [23].

As the $FeTe_{1-x}Se_x$/STO monolayer films share similar FS topology, which may be a key ingredient for the high superconducting transition temperature in FeSe monolayers [12], we check the evolution of superconductivity in this system. Fig. 1b displays the temperature dependence of the symmetrized energy distribution curves (EDCs) at the Fermi momentum (k_F) point of the electron FS around M. Although the introduction of Te weakens the superconducting coherence peak, a gap feature can be clearly observed at the lowest temperature for each sample. Both the gap sizes and the closing temperatures have similar magnitude. A closer inspection shows that the gap is a little larger for the samples in which additional intensity of FS appears at Γ. These results are consistent with the enhancement of superconductivity accompanying a Lifshitz transition in electron-doped FeSe monolayer [12].

We then address the electronic band structure in detail. The electron-like bands observed around the M point are displayed in Fig. 2a. The spectra are divided by the Fermi-Dirac (FD) function convoluted by a Gaussian resolution function. We fit the band dispersions using parabolic curves and investigate the evolution in $FeTe_{1-x}Se_x$/STO. The band bottom (Fig. 2b) moves towards the Fermi level (E_F) upon decreasing the Se concentration, while the effective mass (Fig. 2c) increases from FeSe/STO to FeTe/STO, which is easy to understand considering the strong electronic correlations in the FeTe bulk material [24]. As a result, the k_F value or FS size does not change much, as shown in Fig. 2d.

Fig. 3 displays the band evolution around Γ of $FeTe_{1-x}Se_x$/STO. We observe dramatic variations compared to that at the M point. The curvature plots in Fig. 3b, which can highlight the band dispersions, show two hole-like bands carrying a d_{xz}/d_{yz} orbital character [25,26]. We now examine the inner one, which is also closer to E_F. The band top moves up from FeSe to FeTe, similarly to the electron band bottom at M. In contrast, the effective mass evolves in the opposite way (Fig. 3f). We show in Fig. 3a the spectra after division by the FD function in order to check the unoccupied band. A down-shifting electron-like band is clearly observed when the Se content is smaller than 60%, similar to the case of $FeTe_{1-x}Se_x$ single crystals [23]. This electron-like band, which has a p_z orbital character hybridized with d_{xy} in the monolayer FeSe/STO, according to LDA calculations [25,26]. The electron- and hole-like bands move towards each other and the band gap between them decreases rapidly. Eventually the bands touch each other at a Se concentration of approximately 33%, which is further revealed in the plots of the constant energy contours and momentum distribution curves (MDCs), as shown in Fig. 3c and d. Since the bands deviate

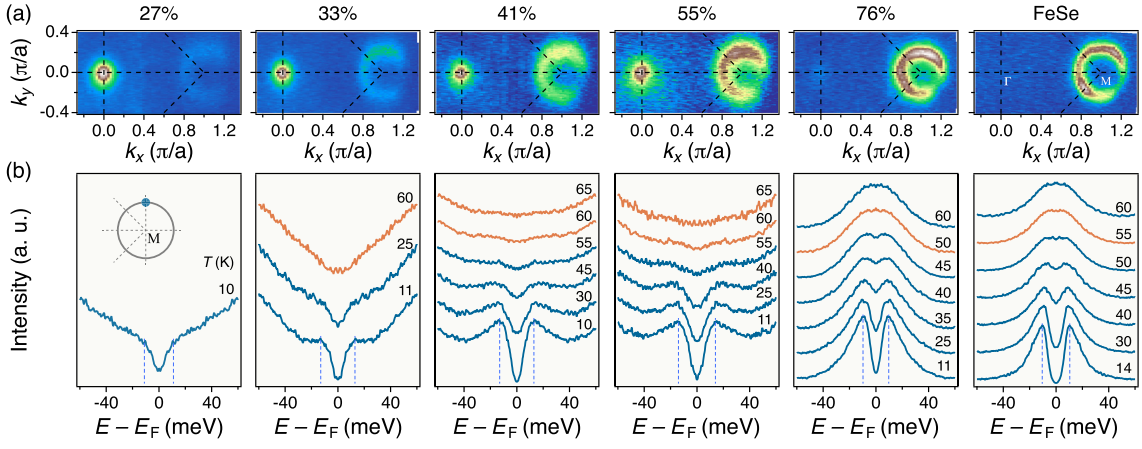

Fig. 1. (Color online) (a) ARPES FS maps of a series of $FeTe_{1-x}Se_x$ monolayers grown on STO, with the nominal concentration of Se indicated above each panel. (b) Temperature evolution of the symmetrized EDCs at the k_F point of the electron FS around M for each sample. The orange curves correspond nearly to the gap closing temperature.

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