Topical Review

High-temperature superconductivity in one-unit-cell FeSe films

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Abstract

Since the dramatic enhancement of the superconducting transition temperature (Tc) was reported in a one-unit-cell FeSe film grown on a SrTiO3 substrate (1-UC FeSe/STO) by molecular beam epitaxy (MBE), related research on this system has become a new frontier in condensed matter physics. In this paper, we present a brief review on this rapidly developing field, mainly focusing on the superconducting properties of 1-UC FeSe/STO. Experimental evidence for high-temperature superconductivity in 1-UC FeSe/STO, including direct evidence revealed by transport and diamagnetic measurements, as well as other evidence from scanning tunneling microscopy (STM) and angle-resolved photoemission spectroscopy (ARPES), are overviewed. The potential mechanisms of the enhanced superconductivity are also discussed. There are accumulating arguments to suggest that the strengthened Cooper pairing in 1-UC FeSe/STO originates from the interface effects, specifically the charge transfer and coupling to phonon modes in the TiO2 plane. The study of superconductivity in 1-UC FeSe/STO not only sheds new light on the mechanism of high-temperature superconductors with layered structures, but also provides an insight into the exploration of new superconductors by interface engineering.

Keywords: one-unit-cell, high-temperature superconductivity, interface, SrTiO3 substrate, transport and diamagnetic measurement, FeSe film

(Some figures may appear in colour only in the online journal)

1. Introduction

The discovery of superconductivity in fluorine-doped LaFeAsO with a superconducting transition temperature (Tc) of 26 K triggered a flurry of research activity on iron-based superconductors [1–6]. A maximum Tc was soon reported up to ~55 K [4, 5] for bulk iron-based superconducting materials. The high Tc, beyond the McMillan limit of conventional superconductors, as well as the distinct properties of their electronic structure and pairing mechanism [7], make iron-based superconductors the second class of high-Tc superconductors behind those of the cuprate variety [8]. All iron-based superconductors contain an iron pnictide (FeAs) [1–6, 9] or iron chalcogenide (FeSe, FeS,Se1−x or FeTe,Se1−x) [10–48] layer, which is generally considered to be responsible for the superconducting properties, and plays a similar role to the CuO2 plane in cuprate superconductors. Although high-temperature superconductivity was discovered over 30 years ago, its mechanism is still unknown [49–51]. The discovery of iron-based superconductors provides a new arena for the understanding of high-temperature superconductivity.

FeSe is a model system for investigating the superconducting properties of iron-based materials due to its simple crystal structure, clean superconducting phase and low
toxicity. Although the superconducting transition temperature for bulk FeSe is relatively low ($T_c \sim 8\,\text{K}$) [24, 25], it can be significantly enhanced by applying high pressure [26–28], intercalating alkali metal atoms like potassium (K) [31–39], electric field tuning [46–48], and most amazingly, by growing ultrathin FeSe films on STO substrates by MBE [10–23]. Representative work devoted to the enhancement of $T_c$ in FeSe and its related systems is summarized in table 1.

Superconducting films, especially those in the 2D limit, have gained great attention in both basic and applied research. It was long believed that superconductivity in a 2D system was impossible, since fluctuations would destroy long-range order even at very low temperatures; however, that was until Thouless and Kosterlitz revealed the superfluid or superconducting topological phase transition in 2D systems [52]. Some representative discoveries regarding 2D superconductivity emerge in the following aspects. (1) 2D superconductivity may occur at the interface between two non-superconducting materials, e.g. the LaAlO$_3$/SrTiO$_3$ interface [53]. (2) Fluctuations and disorder play an important role in low dimensional systems [56], some new phenomena beyond the classical mean field theory have emerged in superconducting films, such as the quantum phase transition [57] and quantum Griffith singularity [58]. (4) Disordered superconducting thin films close to the quantum phase transition provide a platform to detect the Higgs mode [59]. (5) Film thickness becomes an important parameter for tuning the superconducting properties in ultrathin films, even with a single atomic layer change; for instance, $T_c$ oscillates with the film thickness in Pb films on Si substrates [60].

Recently, a state-of-the-art film-growth technique in an ultrahigh vacuum chamber has made the fabrication of atomically flat crystalline thin films possible. For example, high-quality samples with accurate stoichiometry and few defects can be realized in an MBE chamber. Furthermore, the MBE chamber can be linked to the STM or the ARPES chamber in an ultrahigh vacuum environment, which enables the study of pristine surfaces without exposing the sample to the atmosphere.

Superconducting FeSe crystalline thin films can be prepared via layer-by-layer epitaxial growth on double-layer graphene along the (001) crystal direction (figure 1) [61, 62]. Graphene substrates are acquired by the heat-treatment of a SiC (0001) substrate to evaporate the silicon atoms at the surface [63, 64]. An FeSe unit-cell consists of three atomic layers with a central Fe layer sandwiched between two adjacent Se layers (see inset of figure 1(a)). The FeSe film and graphene substrate are coupled by weak van der Waals type interaction. Thus, the lattice constant of the FeSe film on graphene is very close to that of the bulk material (~3.77 Å). In addition, the FeSe films grown on graphene show a lower $T_c$ and are less thick. A superconducting coherence peak can be clearly observed in the differential tunneling conductance ($dI/dV$) spectra by scanning tunneling spectroscopy (STS). Analysis of the normalized $dI/dV$ spectra at various temperatures gives $T_c \sim 7.8\,\text{K}$ for 8-UC FeSe films (figure 1(b)) and $T_c \sim 3.7\,\text{K}$ for 2-UC films (figure 1(c)).

Superconductivity cannot be observed in 1-UC FeSe films down to 2.2 K. The relation between the superconducting transition temperature $T_c$ and film thickness $d$, as shown in figure 1(d), exhibits a good agreement with the empirical formula $T_c(d) = T_{c0}(1 - d/d_0)$ [65], which is adoptable in both conventional superconducting films, like Pb films on Si (1 1 1) substrates [66], and unconventional ones like YBa$_2$Cu$_3$O$_{x}$ films on STO substrates [67]. The extrapolated $T_{c0} \sim 9.3\,\text{K}$ is consistent with the $T_c$ of bulk FeSe, suggesting that the graphene substrate has little effect on the superconducting properties of FeSe films.

However, 1-UC FeSe films grown on STO substrates by MBE exhibit extremely enhanced superconductivity (figure 2) [10, 68] compared with bulk FeSe and FeSe films on graphene. After the epitaxial growth of 1-UC FeSe on Nb-doped (001)-orientated single crystal STO with a proper annealing process, the sample was transferred into the STM chamber without breaking the ultrahigh vacuum condition. An unexpectedly large superconducting-like gap of 20.1 meV was identified from the $U$-shaped conductance spectrum of the 1-UC FeSe/Nb: STO film (figure 2(c)). This gap size is almost one order of magnitude larger than 2.2 meV for bulk FeSe, and it would correspond to $T_c \sim 80\,\text{K}$ [10] if the ratio between the superconducting gap and $T_c$ ($2\Delta/k_B T_c$) was assumed to be similar to that of bulk FeSe. Surprisingly, the tunneling spectra taken from the 2-UC or thicker FeSe films do not show any sign of superconductivity (figure 2(d)). This feature is in sharp contrast to that observed in FeSe/graphene, where $T_c$ decreases upon reducing the film thickness (figure 1(d)), and superconductivity disappears in the 1-UC films. This difference indicates that the STO substrate and interface play an essential role in the enhanced superconductivity of 1-UC FeSe/STO.

The discovery of potential high-temperature superconductivity in 1-UC FeSe/STO has opened up a new frontier in this field [55]. A high $T_c$ with a simple structure, the enhanced superconductivity from interface engineering [18, 69] and its possible topological nature [70–74] make this system a highlight in condensed matter physics and material science. In this paper, we present a brief review of this rapidly developing direction. We will illustrate the high-temperature superconductivity of 1-UC FeSe/STO in section 2. The direct evidence revealed by transport and diamagnetic measurements, as well as other evidence from STM and ARPES, are overviewed therein. In section 3, the potential mechanisms of the $T_c$ enhancement are discussed. Finally, we end with a conclusion and perspective in section 4.

2. Evidence for high-temperature superconductivity in one-unit-cell FeSe films on SrTiO$_3$ substrates

2.1. Transport and diamagnetic measurements

While a superconducting-like gap as large as 20 meV has been revealed in 1-UC FeSe/STO, direct evidence from electrical transport and magnetic measurements was still needed to confirm the observed superconductivity. The initial transport measurements on a 5-UC FeSe film covered by a 20 nm
amorphous Si protection layer show a superconducting transition with a decrease in resistance starting at ~53 K [10], but evidence of exact zero resistance is absent. Therefore, a systematic study of the electrical transport and magnetic measurements of 1-UC FeSe/STO with clear evidence of zero resistance and diamagnetism is important [11, 75, 76].

For \textit{ex situ} electrical transport measurements, an insulating single crystal STO (0 0 1) was chosen as the substrate. STO substrates were pretreated by chemical etching with 10% HCl solution followed by thermal annealing in an oxygen atmosphere to obtain the TiO$_2$-terminated surface before it was transferred into an ultrahigh vacuum MBE chamber.

### Table 1. Superconductivity in FeSe and its related systems. The representative work devoted to the enhancement of the superconducting transition temperature $T_c$ in FeSe-related systems, and the methods for characterizing the $T_c$ are summarized in this table.

| Systems | $T_c$ | Methods for characterizing $T_c$ |
|---------|-------|---------------------------------|
| Bulk PbO-type $\beta$-FeSe | $T_c \sim 8$ K | Electrical transport, diamagnetic and specific heat measurements [24, 25] |
| Bulk FeSe at high pressure | $T_{c,\text{onset}} \sim 36.7$ K, $T_{c,\text{zero}} \sim 20$ K at 8.9 GPa | Electrical transport measurements [26] |
| Bulk FeTe$_x$Se$_{1-x}$ | $T_{c,\text{onset}} \sim 14$ K at $x = 0.6$ | Electrical transport and diamagnetic measurements [29, 30] |
| A$_x$Fe$_{2-x}$Se$_2$ (A = Li, Na, K, Rb, Cs, Ca, Sr, Ba...) | $T_{c,\text{onset}} \sim 30.1$ K, $T_{c,\text{zero}} \sim 27.2$ K for K$_{0.8}$Fe$_2$Se$_2$ | Electrical transport and diamagnetic measurements [31–34] |
| A$_x$Fe$_{2-x}$Se$_2$ at high pressure (Li$_{0.8}$Fe$_{0.2}$)OHFeSe | $T_{c,\text{onset}} \sim 48$ K at 12.4 GPa for Th$_0.4$Rb$_0.6$Fe$_1.67$Se$_2$ | Electrical transport and diamagnetic measurements [36] |
| 1-UC FeSe films on Nb-doped STO (0 0 1) (for the \textit{ex situ} transport and magnetic measurements, the samples were covered by a 10-UC FeTe layer.) | $T_c \sim 25$ K | Diamagnetic measurements (SQUID) [19] |
| 1-UC FeSe films on insulating STO (0 0 1) | $T_{c,\text{onset}} > 40$ K, $T_{c,\text{zero}} \sim 23.5$ K | Ex \textit{sit}u electrical transport measurements [11] |
| 1-UC FeSe films on STO (1 1 0) | $T_c \sim 21$ K | Diamagnetic measurements (two-coil mutual inductance measurements) [11] |
| 1-UC FeTe$_x$Se$_{1-x}$ films on STO (0 0 1) | $T_{c,\text{onset}} > 40$ K, $T_{c,\text{zero}} > 21$ K | Diamagnetic measurements (two-coil mutual inductance measurements) [11] |
| 1-UC FeSe films on Nb: STO/KTaO$_3$ | $T_c \sim 70$ K | Diamagnetic measurements (SQUID) STM/STS [15] |
| K-coated multilayer FeSe films on STO | $T_c \sim 48$ K for 3-UC films | STM/STS [22] |
| K-coated multilayer FeSe films on graphitized SiC (0001) | Superconducting gap of ~13.1 meV for 3-UC films | ARPES [40] |
| K-dosed bulk FeSe/FeSe$_{0.95}$S$_{0.07}$ | Superconducting gap of ~9 meV | ST/STS [41, 43] |
| Electric field tuning of thin FeSe films on STO | $T_{c,\text{onset}} \sim 40$ K | ARPES [44, 45] |

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The FeSe films were grown by the coevaporation of Fe and Se with a flux ratio of ~1:10 on STO substrates. After a proper annealing process, the 1-UC FeSe films were covered by a 10-UC FeTe layer and a 30 nm amorphous Si layer to protect the sample from deterioration in air [11]. The major transport properties of the FeSe films were measured by a standard four-electrode method (see inset of figure 3(a)) in a commercial physical property measurement system (Quantum Design PPMS-16). It should be mentioned that the indium electrodes used here can easily penetrate the protection layers and connect to the nethermost FeSe layer on the STO substrate.

The temperature dependence of resistance for 1-UC FeSe/STO, as shown in figure 3(a), exhibits an obvious superconducting transition with $T_{c\text{onset}} \approx 40.2$ K and $T_{c\text{zero}} \approx 23.5$ K. Here, $T_{c\text{onset}}$ is acquired by extrapolating the normal resistance curve and the sharp resistance drop, and $T_{c\text{zero}}$ is defined as the temperature where resistance drops below the instrumental resolution. $T_{c\text{onset}} \approx 40.2$ K is almost five times that of $T_c \approx 8$ K for bulk FeSe, demonstrating significant interface-enhanced superconductivity. Gradual deviation from normal resistance starts at ~54.5 K, suggesting that the superconductivity may survive up to this high temperature. For comparison, the $R(T)$ curves for the 10-UC FeTe capping layer on the STO substrate, with or without an additional amorphous Si layer, exhibit insulation-like behavior. In addition, STS suggests that the proximity effect cannot turn the STO substrate or FeTe protection layer into a superconductor [11]. These facts indicate that the high-temperature superconductivity in an FeSe/STO system is limited to a single-unit-cell FeSe layer with a thickness of just 0.55 nm.

Magnetic susceptibility measurements for the same sample by the commercial magnetic property measurement system (MPMS-SQUID-VSM) show the Meissner effect and demonstrate the superconductivity in the FeSe/STO system (figure 3(b)). A sharp drop in the $M(T)$ curve at ~25 K is clear evidence for diamagnetism corresponding to $T_{c\text{zero}}$ obtained by the transport measurements, and the typical magnetic hysteresis loop measured at 2 K also exhibits superconducting characteristics [11]. The diamagnetism of 1-UC FeSe films on insulating STO substrates was further confirmed by two-coil mutual inductance measurements for another 1-UC FeSe/STO sample. Magnetic susceptibility measurements were also carried out on 2-UC to 4-UC FeSe/STO films, giving clear evidence of diamagnetism in multilayer FeSe/STO [77].

Besides a high $T_c$, the 1-UC FeSe/STO also exhibits a high critical magnetic field ($B_C$) and a large critical current density ($J_C$) [11]. To determine the upper critical field $B_{C2}$, magnetoresistance at various temperatures was measured by employing a
pulsed magnetic field up to 52 T in perpendicular (figure 4(a)) and parallel (figure 4(b)) orientations. In the perpendicular field, the sample resistance at 1.4 K remains at zero until the field increases above 30 T and is still much smaller than the normal state resistance, even at 52 T. When a magnetic field is applied parallel to the film, the $B_c$ is even higher than that in the perpendicular field, reflecting the 2D nature of superconductivity in 1-UC FeSe/STO. Moreover, 1-UC FeSe film exhibits an unexpectedly large critical current density ($J_c$). The critical current at 2 K reaches 13.3 mA, corresponding to $J_c \approx 1.7 \times 10^6$ A cm$^{-2}$, which is two orders of magnitude higher than that of bulk FeSe ($J_c \approx 2.2 \times 10^4$ A cm$^{-2}$) [78].

The 2D superconducting property is further supported by the observation of a Berezinski–Kosterlitz–Thouless (BKT)-like transition (figure 5) [11], which is a topological phase transition. The temperature dependence of the power-law exponent $\alpha$ ($V \propto I^\alpha$) indicates a BKT-like transition temperature $T_{BKT} \approx 23.1$ K. This transition temperature can also be determined by using the Halperin–Nelson equation $R(T) = R_0 \exp[-h(T - T_{BKT})^{1/2}]$ [79], yielding $T_{BKT} \approx 23.0$ K, which is consistent with the result from the $V(I)$ curves.

The resistive transition of high-$T_c$ superconductors in the presence of a magnetic field is an active research topic [19, 80, 81]. The $R_{\text{eq}}(T)$ curves and corresponding $\ln[R_{\text{eq}}] - (1/T)$ curves for 1-UC FeSe/STO at various magnetic fields are shown in figures 6(a) and (b). The resistance transition in the low temperature region can be well described by the theory of thermally activated flux flow using the Arrhenius relation $R(T, H) = R(H) \exp[-U_0(H)/T]$, where $U_0$ is the activation energy. The slopes obtained from the linear fittings (pink lines in figure 6(b)) give the activation energy $U_0(H)$ at various magnetic fields (figure 6(c)). Extrapolation of these fitting lines has a common intersection point at $T_m \approx 38$ K, which is close to $T_c^{\text{mid}}$ (defined as the temperature where resistance drops to half the normal resistance) from the $R_{\text{eq}}(T)$ curves. The power-law fittings $U_0(H) \sim H^{-\gamma}$ are performed on the $U_0(H)$ data, showing a crossover from $\gamma \approx 0.14$ for $\mu_0H < 3.4$ T to $\gamma \approx 0.60$ for $\mu_0H > 3.4$ T. The noteworthy change in $\gamma$ marks a crossover from a single-vortex pinning dominated regime to a collective flux creep regime [19]. Similarly, a recent work analyzed the activation energy $U_0(J)$ by measuring the $R_{\text{eq}}(T)$ curves at various current densities $J$ [80]. $U_0(J)$ saturates to a finite value when $J \to 0$, suggesting that long-range vortex lattice correlations may be absent in 1-UC FeSe/STO, possibly due to the strong thermal fluctuations in 2D materials [80].

Hall measurements were carried out on 1-UC FeSe films to determine the carrier type and carrier density [19]. The Hall resistance $R_{xy}(H)$ curves at various temperatures exhibit a good linear relation (figure 7(a)), where the influence of the FeTe protection layers has been subtracted. The Hall coefficient and carrier density derived from the $R_{xy}(H)$ curves (figure 7(b)) indicate that the dominant carriers transform...
from the hole-type at high temperatures to the electron-type at low temperatures. The electron carrier density in the low-temperature regime is on the order of $10^{15}$ cm$^{-2}$. The high carrier density is generally believed to be a necessary factor for achieving superconductivity. Similar dominant carrier transformation from hole-type at high temperatures to electron-type at low temperatures also happens in 2-UC FeSe/STO [19].

The transport measurements of multilayer FeSe/STO with different film thicknesses and annealing conditions have been studied [19, 82]. As mentioned before, indium electrodes can penetrate to the nethermost FeSe layer on an STO substrate. Therefore, the observation of superconductivity in multilayer FeSe film by transport measurement is not contradictory to the fact that the superconducting gap is absent on the top surface of the multilayer film by STM or ARPES measurements [10, 14, 83], if the nethermost FeSe layer superconducts in a multilayer FeSe film. The FeSe/STO system exhibits unique superconducting properties, i.e. the onset $T_c$ decreases with an increasing film thickness (figures 8(a) and (b)) [82], which is the opposite to that observed in traditional superconducting films, including FeSe films grown on graphene [61]. Besides this, a proper annealing process is crucial for the observation of superconductivity in an FeSe/STO system. The as-grown 5-UC FeSe film is insulating, and annealing at 500 °C for 36 h makes it a superconductor with $T_c^{\text{onset}} \sim 39$ K (figure 8(c)); excessive annealing, however, would make the $T_c$ lower, possibly due to over-doping or the gradual evaporation of the FeSe at a high annealing temperature [83]. The annealing process is accompanied by the transformation of a dominant charge carrier from the hole to the electron at low temperatures (figure 8(d)), indicating effective electron doping from the STO substrate during the annealing process.

To conclude this section, the observation of zero resistance and diamagnetism unambiguously demonstrates superconductivity in 1-UC FeSe/STO. However, in most cases, the $T_c$ by ex situ transport or magnetic measurement is not as high as the expectation from the STM or ARPES measurements (see table 1). This result can be understood in the following way. First, superconductivity in a 1-UC FeSe film is dependent on the level of electron doping from the STO substrates [13, 15, 16, 83]. For the ex situ transport measurements of multilayer FeSe films, or 1-UC FeSe films with FeTe capping, carriers from STO substrates may spread over the FeSe layers in multilayer FeSe films or spread to FeTe protection layers in 1-UC FeSe films with FeTe capping, making the doping level of the interfacial FeSe layer lower than the situation with only a 1-UC FeSe layer on the STO [83], which is the situation for the STM and ARPES studies. Second, the non-superconducting FeTe layer has a different magnetic structure from the FeSe layer [84–88], and this difference may have a negative effect on the superconductivity of 1-UC FeSe/STO. Third, the doping level in 1-UC FeSe/STO depends on the annealing temperature and lasting time [13, 15]. STS suggests that a high annealing temperature (500 °C or above) is helpful in the formation of a large superconducting gap [15]. However, the FeSe would evaporate gradually during the annealing process at such a high temperature [83]. Since keeping the FeSe film well connected is necessary for ex situ transport measurements, the high annealing temperature may decrease the $T_c$ obtained by the transport measurements. Last but not least, exposure to air is damaging to ultrathin FeSe films, even though they are covered by a protection layer.

It is worth mentioning that a special in situ four-tip electrical transport measurement technique was developed (figure 9(a)) to investigate the superconductivity in 1-UC FeSe/Nb: STO films without exposing the sample to air [20, 89]. As shown in figure 9(b), zero resistance disappears at a very high temperature of ~109 K, which is even beyond the expectation of STM and ARPES measurements. Although this result is very exciting, the transport measurement is not a standard four-electrode method and the substrate does conduct; thus, further confirmation from other groups and techniques is necessary. The indication of a $T_c$ above liquid nitrogen temperature was also revealed by the ex situ diamagnetic measurements of 1-UC FeSe/Nb: STO films [19, 55]. The magnetic susceptibility $M(T)$ curves of 1-UC FeSe film after subtracting the influence of the STO substrate and FeTe capping layer exhibit a gradual
Figure 4. The high critical magnetic field and large critical current density in 1-UC FeSe/STO. (a) and (b) The magnetoresistance measured by employing a pulsed magnetic field up to 52T in a perpendicular (a) and parallel (b) orientation. (c) The $V(I)$ curves measured at temperatures ranging from 2 K–50 K at a magnetic field of $B = 0$. (d) The dependence of the critical current density ($J_c$) on temperature and the perpendicular magnetic field. Reproduced from [11]. © 2014 Chinese Physical Society and IOP Publishing Ltd. All rights reserved.

Figure 5. The BKT-like transition of 1-UC FeSe/STO. (a) The $V(I)$ curves at various temperatures plotted on a logarithmic scale; the two dashed lines correspond to $V \propto I$ and $V \propto I^3$. (b) The temperature dependence of the power-law exponent $\alpha$ ($V \propto I^\alpha$) indicates a BKT-like transition temperature $T_{BKT} \approx 23.1$ K. (c) $\ln(\ln R)/\ln T$ plotted as a function of temperature. The dashed line depicts the expected BKT-like transition with $T_{BKT} \approx 23.0$ K. Reproduced from [11]. © 2014 Chinese Physical Society and IOP Publishing Ltd. All rights reserved.
decrease in magnetization starting from \( \sim 85 \text{ K} \) [19]. Besides this, some two-coil mutual inductance measurements of the 1-UC FeSe film with a capping layer of 2-UC (Fe\(_{0.96}\)Co\(_{0.04}\)) Se/2-UC FeSe/Se also showed the formation of diamagnetic screening up to \( \sim 65 \text{ K} \) [21].

### 2.2. STM probing

Scanning tunneling microscopy/spectroscopy (STM/STS) is a powerful technique for the characterization of a superconducting

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**Figure 6.** Analysis of the thermally activated flux flow (TAFF) in 1-UC FeSe/STO. (a) The \( R(T) \) curves measured in perpendicular magnetic fields. (b) \( \ln(R_{sq}) \) versus \( 1/T \) at various magnetic fields; the data at low temperatures is fitted by the Arrhenius relation \( R(T, H) = R(H) \exp[-U_0(H)/T] \) (solid lines). (c) The field dependence of the activated energy \( U_0(H) \); the solid lines show the power-law fittings using \( U_0(H) \sim H^{-\gamma} \). Reprinted by permission from Macmillan Publishers Ltd: Scientific Reports [19], copyright (2014).

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**Figure 7.** The Hall measurements of 1-UC FeSe/STO. (a) \( R_{xy}(H) \) curves at various temperatures after subtracting the influence of FeTe layers. (b) The Hall coefficient and carrier density calculated from the data in panel (a). Reprinted by permission from Macmillan Publishers Ltd: Scientific Reports [19], copyright (2014).
the FeSe film becomes smoother, and transforms from semi-conducting to metallic and finally to the superconducting state. Figure 10(a) shows the evolution of the superconducting gap with an increasing annealing temperature at 450, 480, 500, 510 and 530 °C, respectively. Each annealing stage lasts for 2 h. The superconducting gap gradually becomes larger with an increasing annealing temperature, and reaches ~15.4 meV after annealing at 530 °C (figure 10(b)). Figure 10(c) displays a series of normalized dI/dV spectra taken at various temperatures on the FeSe surface after annealing at 530 °C. The coherence peaks are gradually suppressed and the zero bias conductance (ZBC) continuously increases with increasing temperature (figure 10(d)). The ZBC shows a linear dependence on temperature, suggesting an extrapolated \( T^c \sim 68 \) K. Although grown on insulating STO substrates, the \( T^c \) value is close to that estimated for 1-UC FeSe/Nb: STO. We know the pre-annealing process of an STO substrate in an ultrahigh vacuum environment induces 2D electron gas (2DEG) at the surface \([15, 100]\), which becomes a carrier reservoir and transfers electrons to the FeSe layer in the post-annealing process, as illustrated by the transport \([11, 82]\) and ARPES \([13, 16]\) measurements. Surprisingly, the formation of highly metallic 2DEG at the surface of the STO is independent of the bulk carrier densities over a wide range from less than \( 10^{13} \) cm\(^{-3}\) (insulating STO) to \( 10^{20} \) cm\(^{-3}\) (Nb: STO) \([101]\). This explains why the \( T^c \) for 1-UC FeSe films on insulating and conducting STO could be close.

By measuring the local response of superconductivity to impurities, and through quasi-particle interference (QPI) patterns, STM/STS has provided evidence of plain \( s \)-wave pairing symmetry in a 1-UC FeSe/STO system \([91]\). The QPI patterns (figure 11), acquired by the Fourier transform of the real space dI/dV mapping, reflect the scattering between and within the electron pockets. The annealed 1-UC FeSe/STO has four ellipse-like electron pockets at the \( M \)-points. The possible scattering channels \((q_1, q_2, q_3)\) between and within these electron pockets (figure 11(a)) correspond to three scattering rings in the QPI pattern (figure 11(c)). The integrated intensities over the different scattering rings have similar energy dependences near the gap edge (figure 11(d)), suggesting that none of these scattering channels is significantly different, and that the signs of the superconducting gap \( \Delta_k \) on different \( M \)-points are similar. This observation is consistent with the plain \( s \)-wave or \( s^\pm \)-wave pairing (\( \pm \) refers to the sign change of the superconducting order parameters between the electron and hole Fermi surfaces) but are incompatible with the \( d \)-wave pairing. The same conclusion can also be deduced from the magnetic field dependence of the QPI (figures 11(e) and (f)). Probing the response of superconductivity to local impurities (figure 12) is another common way of determining the pairing symmetry. While magnetic impurities (Cr and Mn) suppress the superconductivity and induce in-gap states, the superconducting gap is basically undisturbed by non-magnetic impurities (Zn, Ag and K). Since the \( d \) or \( s^\pm \) pairing is sensitive to non-magnetic impurities \([102, 103]\), the plain \( s \)-wave pairing is the most likely pairing symmetry in 1-UC FeSe/STO.
Figure 9. In situ special four-tip electrical transport measurement of 1-UC FeSe/Nb: STO. (a) A schematic for the in situ transport measurement setup. The resistance is measured by contacting four tips with the sample surface at an inclined angle of 20°. (b) The temperature dependence of the resistance obtained from the $\chi(I,V)$ curves. Above 79 K, the sample was cooled by liquid N$_2$. Inset: the temperature dependence of resistance taken on a bare STO surface. Reprinted by permission from Macmillan Publishers Ltd: Nature Materials [20], copyright (2014).

2.3. ARPES measurements

Angle-resolved photoemission spectroscopy (ARPES) provides another approach for examining the superconductivity in an FeSe/STO system [12–14, 16–18, 104]. Reminiscent of the STM study, the superconducting transition temperature $T_c$ can be extracted from the photoemission spectra (the energy distribution curves, EDCs) by measuring the gap-opening temperature and the temperature dependence of the gap. A superconducting signature with $T_c \sim 65$ K [13, 14] was revealed by ARPES in 1-UC FeSe/STO after the appropriate post-annealing procedures. Over other techniques, ARPES provides direct information on the band structures, and the superconducting gap can be measured at specific $k$-points in the momentum space, which sheds light on the investigation of the pairing symmetry and pairing mechanisms of the superconductivity.

Figure 13 shows the Fermi surface and band structure of 1-UC FeSe/STO. The Fermi surface only consists of electron pockets around the $M$-points (figure 13(a)). In particular, the hole-like Fermi surface at the Brillouin zone center ($\Gamma$), which exists in most iron-based superconductors, including iron pnictides [105] and bulk FeSe [106], is absent in 1-UC FeSe/STO. The lack of a hole pocket implies that the 1-UC FeSe/STO film is electron-doped. In fact, a similar feature has been observed in electron-doped iron chalcogenides, such as K$_x$Fe$_2$-$\gamma$Se$_2$ [35, 107]. The absence of a hole Fermi surface means that an $s_\pm$ pairing scenario [108] may not be the proper description of the superconductivity in 1-UC FeSe/STO. The band structures along cut 1 and cut 2 are shown in figure 13(b), exhibiting the hole and electron band, respectively. The hole-like band at the $\Gamma$-point sinks ~80 meV below the Fermi level, explaining the absence of the hole Fermi surface. High-resolution Fermi surface mapping presents the detailed structure of the electron Fermi surface [109]. Two overlapping ellipse-like electron pockets ($\delta_1, \delta_2$) are resolved at the $M$-point, though the photoemission intensity of the $\delta_2$ pocket is larger than that of the $\delta_1$ pockets (figures 13(c) and (d)).

The superconducting transition temperature $T_c$ was acquired by analyzing the temperature dependence of the symmetrized photoemission spectra along the $\gamma$ Fermi surface (figure 14(a)). The superconducting-like gap shrinks with an increasing temperature and disappears at ~65 K for well-annealed 1-UC FeSe/STO samples [13]. The evolution of the superconducting gap with temperature exhibits good agreement with the BCS fitting (figure 14(b)), giving $T_c$ ~ 65 K and a gap size of ~19 meV, which is consistent with the STS results [10]. As for the gap distribution in the momentum space, an anisotropic but nodeless superconducting gap (figure 14(c)) was revealed by a recent high-resolution ARPES experiment [109]. The gap maxima ~12 meV is located along the major axis of the ellipse, while the gap minima ~8 meV is located at the intersection of two ellipse-like electron pockets. Besides this, the pronounced anisotropy of the gap size with over 50% variation (ranging from 8.5–17.2 meV) was observed in the 1-UC FeSe films on Nb: STO/ KTaO$_3$, with a larger lattice constant due to the tensile strain from the substrate [18].

2.4. Evidence from other techniques

Some other techniques have also resulted in new insights into the detection of superconductivity in a 1-UC FeSe/STO system. For example, a superconducting transition with $T_c$ ~ 68 K and a superconducting gap of ~20.2 meV were observed in a single layer FeSe/STO system by using ultrafast spectroscopy [110]. In particular, the electron–phonon coupling strength $\lambda$ ~ 0.48 can be obtained by measuring the lifetime of the non-equilibrium quasiparticle pumped by femtosecond laser. Such information is inaccessible by most other methods and of importance in understanding high-$T_c$ superconductivity in the FeSe/STO system.

In addition, superconductivity at 62 K was reported in FeSe/STO by transverse field muon spin rotation and relaxation (TF-$\mu$SR) spectra [111]. Measurements of the field distribution in the vortex state give an effective penetration depth of $\lambda$, and hence a superfluid density of $n_s \propto \lambda^{-2}$. The density of the paired electrons is estimated to be $n_s \approx 6.08 \times 10^{21}$ cm$^{-3}$, which is consistent with the doping level revealed by ARPES (~0.12 electron per Fe corresponds to $n_s \approx 6 \times 10^{21}$ cm$^{-3}$) [13] and
transport measurements ($n^{2D} \approx 10^{15} \text{ cm}^{-2}$, or $n_s \approx 10^{22} \text{ cm}^{-3}$) [19]. The temperature dependence of $n_s$ is in good agreement with the BCS fitting, suggesting an $s$-wave superconducting state with a gap size of $\approx 10.2$ meV. Besides this, the polarized muon probe does not detect the indication of magnetism, thus supporting the absence of SDW order in single-layer FeSe.

3. Discussion on the mechanism of interface-enhanced superconductivity in 1-UC FeSe/STO

Experimental discoveries of high-temperature superconductivity in 1-UC FeSe/STO have triggered great interest in the community of condensed matter physics to figure out the
Figure 12. The local response of superconductivity to magnetic (Cr and Mn) and non-magnetic (Zn, Ag and K) impurities. (a)–(e) Topographic images of single adatoms on FeSe/STO films. (f)–(j) The differential tunneling conductance (dI/dV) spectra taken along the arrows shown in (a)–(e). The distance from the measuring points to the center of the atom is marked on the left. The in-gap states are induced by the magnetic impurities, Cr and Mn, but absent in the non-magnetic impurities Zn, Ag and K. Reprinted by permission from Macmillan Publishers Ltd: Nature Physics [91], copyright (2015).

Figure 13. The Fermi surface and band structure of 1-UC FeSe/STO (0 0 1). (a) The Fermi surface mapping measured at 20 K consists only of electron pockets at the M-points. (b) The band structures along cut 1 (left panel) and cut 2 (right panel) at 20 K. The pink dashed lines denoted as α, β and the purple dashed line denoted as γ are guides to the eye, indicative of the hole- and electron-like bands, respectively. (c) and (d) The high-resolution Fermi surface mapping and corresponding second derivative image obtained in (c) circular (CR) polarization and (d) linear horizontal (LH) polarization at ~120 K. Two overlapping ellipse-like electron pockets (δ₁, δ₂) are resolved at the M-points. (a) and (b) Reprinted by permission from Macmillan Publishers Ltd: Nature Communications [12], copyright (2012). (c) and (d) Reprinted with permission from [109]. Copyright (2016) by the American Physical Society.
underlying mechanism. Now it is widely accepted that the dramatic enhancement in $T_c$ originates from the FeSe/STO interface. Several factors regarding the interface have been considered: for instance, the tensile strain due to the in-plane lattice constant difference between the FeSe film and the STO substrate, the electron doping from the STO substrate, and the electron–phonon coupling between the electrons in FeSe and the high-frequency phonons in STO, etc.

The role of tensile strain on the enhancement of $T_c$ was studied by growing 1-UC FeSe films on different substrates with various in-plane lattice constants [18, 69, 112]. For comparison, the in-plane lattice constant, the tensile strain and the corresponding $T_c$ are listed in table 2. Although $T_c$ seems to have a positive correlation with the tensile strain in a 1-UC FeSe film, the effect of tensile strain is not large enough to explain the giant enhancement of $T_c$. In particular, the $T_c$

| Substrates                           | In-plane lattice constant (Å) | Tensile strain (relative to bulk value 3.765 Å) (%) | $T_c$ (K)   |
|--------------------------------------|------------------------------|---------------------------------------------------|------------|
| Nb: BaTiO$_3$/KTaO$_3$ (rotated lattice) | 3.78                         | 0.4                                               | ~70 [69]   |
| 3-UC Nb: STO/LaAlO$_3$               | 3.79                         | 0.7                                               | ~55 [112]  |
| 5-UC Nb: STO/LaAlO$_3$               | 3.81                         | 1.2                                               | ~62 [112]  |
| Nb: STO                              | 3.91                         | 3.9                                               | ~65 [13, 14] |
| Nb: STO/KTaO$_3$                     | 3.99                         | 6.0                                               | ~70 [18]   |
| Nb: BaTiO$_3$/KTaO$_3$ (Unrotated lattice) | 3.99                         | 6.0                                               | ~75 [69]   |

Figure 14. The temperature and momentum dependence of the superconducting gap in 1-UC FeSe/STO (001). (a) The symmetrized photoemission spectra along the $\gamma$ Fermi surface measured at different temperatures. (b) The temperature dependence of the superconducting gap; the green line shows the BCS fitting with a gap size of ~19 meV and $T_c \sim 65$ K. (c) The superconducting gap distribution in momentum space along the ellipse-like electron pocket $\delta_2$ exhibits distinct anisotropy. (a) and (b) Reprinted by permission from Macmillan Publishers Ltd: Nature Materials [13], copyright (2013). (c) Reprinted with permission from [109]. Copyright (2016) by the American Physical Society.
or liquid-gated FeSe films, indicating that a conductivity with \( c \sim 40 \) K has been reported in a double-layer transistor (EDLT). A gate voltage \( V_G \) is applied through the ionic liquid. The EDLT can accomplish the function of electrochemical etching and electrostatic carrier doping separately. (c) and (d) The temperature dependence of the normalized sheet resistance \( R_s/G \) of 3.7 nm thick sample with a different gate voltage. The red (purple) solid line represents the fully charged situation at \( V_G = 5 \) V (fully discharged at \( V_G = 0 \); \( T_c^\text{on} \) is labeled with triangles. Reprinted by permission from Macmillan Publishers Ltd: Nature Physics [46], copyright (2015).

is still as high as \( \sim 70 \) K in 1-UC FeSe/Nb: BaTiO\(_3\)/KTaO\(_3\) (rotated domain), although the tensile strain can be negligible. Moreover, recent STM measurements reveal that nearly strain-free FeSe films on anatase TiO\(_2\) (0 0 1) exhibit a large superconducting gap of similar size to 1-UC FeSe/STO [113]. DFT calculations show that the magnetic interaction is sensitive to the lattice constant, e.g. the next-nearest-neighbor coupling parameter \( J_2 \) increases by about 40\% when the lattice constant increases just a few percent [114]. Since in reality the tensile strain only has a small effect (if it does have any) on the \( T_c \), the magnetic-interaction-mediated pairing mechanism alone cannot unveil the secret of a high \( T_c \) in the FeSe/STO system.

There are accumulating arguments suggesting electron doping and electron–phonon coupling are responsible for raising the \( T_c \) in a 1-UC FeSe/STO system. High-temperature superconductivity with \( T_c \sim 40 \) K has been reported in K-coated [40] or liquid-gated [46–48] FeSe films, indicating that a high \( T_c \) can be achieved in an FeSe layer once it is optimally doped. Observation of the replica bands by ARPES [17, 122, 123] reveals the crucial role of interfacial coupling between the FeSe electrons and phonon modes in STO. These findings imply that the FeSe/STO system is likely to be understood in a conventional BCS model [115].

### 3.1. The role of electron doping

Carrier density is a primary factor for superconductivity. In the BCS model, the \( T_c \) is roughly estimated as \( T_c \sim \theta_D e^{-1/N(0)V} \) in the weak-coupling limit [116], where \( \theta_D \), \( N(0) \) and \( V \) represent the Debye temperature, the density of state (DOS) at the Fermi energy and the phonon-mediated attractive interaction, respectively. Clearly, in this conventional scenario, superconductivity is not favorable in band insulators with a low carrier density, since \( T_c \rightarrow 0 \) in the \( N(0) \rightarrow 0 \) limit. However, specific band insulators, like STO [117] and MoS\(_2\) [118], can be tuned to a superconducting state by increasing the carrier density. Some common carrier density tuning methods include chemical doping (e.g. Nb-doped STO [117], alkali-metal-dopant-intercalated FeSe [31–39] or MoS\(_2\) [118], and K-coated FeSe [40–45]), electric field tuning (e.g. liquid-gated FeSe [46–48] or MoS\(_2\) [118]), thermal treatment (e.g. the STO turns from insulating to conductive after heating above 800 °C in an oxygen-free environment [101, 117]), and charge transfer at the interface (e.g. 1-UC FeSe/STO [10–23]). The dependence of \( T_c \) on the dopant concentration \( x \) in both cuprates [119] and iron pnictides [9] exhibits a dome-like shape. The doping of a Mott insulator is generally viewed as a starting point for understanding the physics of high-temperature superconductivity in cuprates [119], though the exact pairing mechanism is still being debated.

We showed in the previous section that the post-annealing process is important for the superconductivity in an FeSe/STO system. In fact, both the transport and STM measurements reveal the insulating-like behavior in the as-grown FeSe films on STO. Subsequent annealing processes induce an insulator–superconductor transition, and within a certain range, the \( T_c \) increases with an increasing annealing temperature [15, 82]. The post-annealing process is accompanied by electron doping, which has been demonstrated by both ARPES and transport measurements [16, 82]. The ARPES results show that the hole-like pocket at the \( \Gamma \)-point gradually
sinks below the Fermi energy, and at the same time the electron Fermi surface at the $M$-point gradually enlarges, indicating that the electron concentration in 1-UC FeSe/STO increases continuously during the annealing process [13, 16]. Hall measurements also show that after annealing, $n$-type carriers dominate in FeSe/STO above $T_c$ [19, 82]. However, excessive annealing at a relatively high temperature would deteriorate the superconductivity; for instance, 5-UC FeSe/STO after 55 h annealing at ~500 $^\circ$C exhibits a lower $T_c$ than that annealed for 36 h (figure 8 (c)), as revealed by the transport measurements [82]. This dome-like behavior is reminiscent of the cuprate superconductors and probably results from over-doping or gradual evaporation of the FeSe at a high annealing temperature.

Recently, an electrical double-layer transistor (EDLT) technique has been used to electrostatically dope carriers to FeSe films on different substrates [46–48]. Figure 15 displays the electric-field-induced superconductivity in electrochemically etched ultrathin FeSe films on STO and MgO substrates [46].

The electric field has a significant effect on FeSe films, especially for those with a thickness below 10 UC (figures 15(c) and (d)). For a 3.7 nm thick sample, the onset $T_c$ increases with an increasing gate voltage and reaches around 40 K at $V_G = 5$ V (figure 15(e)). The $T_c$ enhancement from ~8 K (the bulk value) to ~40 K suggests that electron doping contributes to the observed high-$T_c$ superconductivity in the FeSe layer.

Another noteworthy example is the observation of superconductivity with $T_c$ ~ 48 K in $K$-coated 3-UC FeSe/STO [40]. As mentioned before, the superconductivity in an FeSe/STO system is limited in the nethermost FeSe layer [10, 11], and it is very hard to tune the upper layer to a heavily electron-doped region using substrates [83]. Thus, the high carrier density at the surface of 3-UC FeSe/STO is provided by the deposited $K$ atoms, which is similar to the situation in alkali-metal-dopant-intercalated FeSe [31–39]. All these electron-doped iron chalcogenides (1-UC FeSe/STO [12–14], $K$-coated 3-UC FeSe/STO [40], $K$-coated bulk FeSe [44, 45, 120] and $K_{x}$Fe$_{2-x}$Se$_{2}$ [35]), whose Fermi surfaces only consist of electron pockets, exhibit a much higher $T_c$ than bulk FeSe. Therefore, electron doping can at least partly account for the $T_c$ enhancement in FeSe/STO.

On the other hand, the $T_c$ for $K$-coated 3-UC FeSe/STO (~48 K), the $K$-coated bulk FeSe (~25 K) and $K_{x}$Fe$_{2-x}$Se$_{2}$ (~30 K) is not as high as the value of 1-UC FeSe/STO (~65 K.
as revealed by ARPES and STM, indicating that the STO substrate plays a distinct role in realizing high-$T_c$ superconductivity beyond merely doping electrons on the FeSe layer. By the systematic STM study of $K$-coated FeSe films on STO and graphene substrates [41–43], it is found that while the superconducting gap remains at ~9 meV for films on graphene by $K$ coating, it increases from ~9 meV to ~15 meV for films on the STO when the thickness is reduced to a few unit cells (figure 16(a)). The FeSe/STO interface enhancement of the superconducting gap decays exponentially with a characteristic length of 2.4 UC as the film thickness increases [43], which matches well with the decay behavior of the penetrating field intensity of the Fuchs–Kliewer phonon modes in STO [121]. However, there is almost no such interface effect between the FeSe and graphene [43, 61, 62]. These results point to interfacial electron–phonon coupling as the likely origin of the high-$T_c$ superconductivity in a 1-UC FeSe/STO system.

### 3.2. Interface-enhanced electron–phonon coupling

In the BCS model, electrons form Cooper pairs via phonon-mediated attractive interaction [116], where the electron–phonon coupling strength dominates in determining the superconducting transition temperature. Interface-enhanced electron–phonon coupling has been suggested as a feasible origin of the dramatic $T_c$ enhancement in the FeSe/STO system [14, 121–128].

The electron–phonon coupling strength is quantified as a frequency-dependent electron–phonon coupling constant $\lambda(\omega)$ and Eliashberg spectral function $\alpha^2 F(\omega)$, obeying $\lambda(\omega) = 2 \int_0^\infty d\nu [\alpha^2 F(\nu) \lambda(\omega)]$ [129]. The calculated $\lambda(\omega)$ and $\alpha^2 F(\omega)$ spectra (figure 17(a)) reflect the stronger electron–phonon coupling strength in FeSe/STO than bulk FeSe, signaling interface-enhanced electron–phonon coupling at the FeSe/STO interface [125]. Since both the inter-pocket and intrapocket electron–phonon coupling are attractive, and hence beneficial to the even-sign $s_+^\pm$ pairing in FeSe/STO, the $T_c$ is enhanced with $\lambda_{\text{int}} > 0$ and $\lambda_{\text{ext}} > 0$ (figure 17(b)) [124]. Strengthened pairing might originate from the interfacial interaction between FeSe electrons and ferroelectric phonons involving the relative displacement of Ti and O atoms. Interestingly, a ferroelectric transition near the gap-opening temperature was detected in 1-UC FeSe/STO by mutual inductance and Raman spectroscopy measurements [130]. The coincidence of the ferroelectric transition and the superconducting transition temperature suggests the crucial role of the ferroelectric phonon in promoting pairing in an FeSe/STO system.

The detection of replica bands in 1-UC FeSe/Nb: STO (001) by high-resolution ARPES provides an experimental indication for interfacial coupling between FeSe electrons and phonon modes in STO [17, 128]. As shown in figure 18(a), the band structures of the 1-UC FeSe along a high-symmetry cut centered at the $M$-point demonstrate an electron-like band with its bottom 60 meV below the Fermi energy. Surprisingly, two...
extra replicas (A' and B') of the main bands are observed. Except for an energy shift of ~100 meV, all the characteristics of the replica bands are basically identical to their corresponding main bands. Similar findings are reproduced by raw EDCs (figure 18(b)). The temperature evolution of the M-spectrum for a 1-UC film indicates that the replica bands are robust and persist to temperatures far above the gap-opening temperature (figure 18(c)). Moreover, the replica bands only exist in the 1-UC FeSe film and disappear in the 2-UC or thicker film (figure 18(d)). This feature excludes the possibility of quantum well states [60, 131] as the cause of the replica bands. Instead, the energy separation of ~100 meV is identified as the coupling to the phonon modes in the STO. Since the superconductivity and replica bands are both only detected within the bottom FeSe layer on the STO, the coupling between the FeSe electrons and STO phonons might be responsible for the enhanced Cooper pairing at the FeSe/STO interface.

By assuming both electron and hole bands couple to phonon modes with an energy of ~80 meV and optimizing the coupling strength and forward-scattering parameters, the typical replica bands acquired by a high-statistics scan at low temperature (figure 18(e)) can be well simulated (figures 18(f) and (g)). The electron–phonon coupling constant is estimated as $\lambda \approx 0.5$, which is close to the value ($\lambda \approx 0.48$) obtained by ultrafast optical spectroscopy [107]. Theoretical analysis points out that phonon-mediated attraction can effectively enhance the magnetic-interaction-mediated Cooper pairing [132]. Specifically, the $T_c$ enhancement factor $T_c(v_{\text{eff}})/T_c(0)$ is determined to be directly proportional to the ratio between the phonon-mediated attraction strength $v_{\text{eff}}$ and antiferromagnetic exchange constant $J$ (figure 18(h)). A conservative estimate gives $T_c(v_{\text{eff}})/T_c(0) \approx 1.5$. Considering that the $T_c$ for electron-doped FeSe systems without an interface effect is at the level of 30–40 K (see table 1), this enhancement factor yields fairly good agreement with the gap-opening temperature. Whether the coupling to the STO phonon modes makes a beneficial contribution to the Cooper pairing depends on the superconducting pairing symmetry. In the case of FeSe/STO with two electron pockets at $M$ and the sign-unchanging $s$-wave pairing (supported by STM [91], ARPES [12, 17, 109] and theory [17, 124]), the total contribution of electron–phonon coupling to the pairing $\lambda_{\text{ph}} > 0$, thus enhancing $T_c$. Therefore, the interface-enhanced electron–phonon coupling is experimentally substantiated as an interpretation of the dramatic $T_c$ enhancement in 1-UC FeSe/STO.

The essential role of interfacial electron–phonon coupling in raising the $T_c$ is further verified by a recent comparison experiment on several FeSe-based systems [122]. In general, FeSe-based superconductors can be categorized into three classes: intrinsic FeSe systems (such as FeSe films on graphene, bulk FeSe and FeTe$_x$Se$_{1-x}$), electron-doped FeSe systems (including K-doped multilayer FeSe/STO, K-dosed bulk FeSe and K$_x$Fe$_2$Se$_2$), and electron-doped FeSe systems with interfacial coupling (including 1-UC FeSe films on STO and TiO$_2$). Figure 19 presents the Fermi surface and the band structures at the $M$-points of several representative
FeSe systems of these three classes. The intrinsic FeSe systems exhibit hole pockets around the Γ-point [14, 106, 133] and $T_c$ $< 20$K. Electron-doped FeSe systems share similar band structures, i.e. no hole pockets at Γ [12–14, 35, 40, 44] and hole bands at the $M$-points that are ~45 meV below the electron bands (figures 19(f)–(h)). The $T_c$ for electron-doped FeSe systems without interface effects is enhanced to the level of 30–40K, which is significantly higher than intrinsic FeSe systems. A $T_c$ as high as 60K, however, can only be detected in FeSe systems with both a charge transfer and additional interfacial electron–phonon coupling, e.g. 1-UC FeSe films on STO and TiO$_2$, in which replica bands are observed (figures 19(g) and (h)). The coincidental detection of a high $T_c$ value and replica bands signifies the importance of interfacial coupling for $T_c$ enhancement. In addition, the huge differences in the lattice constant and dielectric constant between TiO$_2$ and STO (the C$_4$ symmetry is broken in 1-UC FeSe/TiO$_2$ (100) with $a = 3.53$ Å and $b = 3.95$ Å, while it is preserved in 1-UC FeSe/STO (001) and TiO$_2$ at a low temperature and has a much smaller dielectric constant [122]) suggest tensile strain, and the dielectric constant is likely to be unimportant for enhancing the $T_c$ in these systems.

To conclude this section, 1-UC FeSe films grown on various TiO$_2$-terminated substrates (STO (001) [10–21], STO (110) [22, 123], BaTiO$_3$ (001) [69], TiO$_2$ (001) [113], TiO$_2$ (100) [122]) with different in-plane lattice constants and dielectric constants exhibit similar $T_c$ values (~60 K–70 K), which are well above the value (~30 K–40K) for electron-doped FeSe systems without interfacial enhancement. This fact clearly demonstrates that interface effects—specifically the charge transfer and coupling to phonon modes in the TiO$_2$ plane—play critical roles in the high-temperature superconductivity of FeSe/STO. The $T_c$ enhancement has been shown to depend on the electron–phonon coupling strength, though a quantitative inconsistency between the experimental value and theoretical calculation still exists [126]. Further investigations are necessary to better understand the high-temperature interface superconductivity observed in FeSe/STO.

4. Summary and perspective

We briefly review the experimental progresses and the physical understandings of the high-temperature superconductivity in 1-UC FeSe/STO and related systems. Some indications show that the $T_c$ could be higher than the temperature of liquid nitrogen [19, 20]. However, strong evidence is still lacking and highly desired. Since the 1-UC FeSe/STO films need to be taken out of the ultrahigh vacuum chamber for ex situ measurements, the search for a high-quality protection layer that does not reduce the $T_c$ is urgently encouraged. With this advancement, high-$T_c$ FeSe/STO interface superconductors would have tremendous potential for superconducting electronic devices.

The interface effects—specifically the charge transfer and coupling to phonon modes in the TiO$_2$ plane—have now been identified as the most likely origins of enhanced superconductivity, although some other mechanisms cannot be completely excluded. Research on FeSe/STO not only sheds light on the mechanism of high-temperature superconductors (cuprates and iron-based superconductors) with a similarly layered structure (e.g. CuO/SrO in BSCCO and FeAs/LaO in LaOFeAs) [10, 11], but also provides insight for the exploration of new superconductors by interface engineering.

Last but not least, the potential topological nature of the FeSe/STO system has gained special attention [70–74]. Dirac cone-like structures have been revealed in the FeSe thin films by band structure calculations [70–72] as well as ARPES measurements [73]. Furthermore, a recent study has predicted the existence of 1D topological edge states at the grain boundary in 1-UC FeSe/STO, which is confirmed by STM probing [74]. This experimental evidence and theoretical support has inspired people to consider the possibility of realizing Majorana fermions in systems based on FeSe/STO or other similar structures. The realization of high-$T_c$ topological superconductors would significantly promote the development of topological quantum computation in the future.

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