Topical Review

Interface high-temperature superconductivity

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Abstract

Cuprate high-temperature superconductors consist of two quasi-two-dimensional (2D) substructures: CuO\textsubscript{2} superconducting layers and charge reservoir layers. The superconductivity is realized by charge transfer from the charge reservoir layers into the superconducting layers without chemical dopants and defects being introduced into the latter, similar to modulation-doping in the semiconductor superlattices of AlGaAs/GaAs. Inspired by this scheme, we have been searching for high-temperature superconductivity in ultra-thin films of superconductors epitaxially grown on semiconductor/oxide substrates since 2008. We have observed interface-enhanced superconductivity in both conventional and unconventional superconducting films, including single atomic layer films of Pb and In on Si substrates and single unit cell (UC) films of FeSe on SrTiO\textsubscript{3} (STO) substrates. The discovery of high-temperature superconductivity with a superconducting gap of \(\sim20\) meV in 1UC-FeSe/STO has stimulated tremendous interest in the superconductivity community, for it opens a new avenue for both raising superconducting transition temperature and understanding the pairing mechanism of unconventional high-temperature superconductivity. Here, we review mainly the experimental progress on interface-enhanced superconductivity in the three systems mentioned above with emphasis on 1UC-FeSe/STO, studied by scanning tunneling microscopy/spectroscopy, angle-resolved photoemission spectroscopy and transport experiments. We discuss the roles of interfaces and a possible pairing mechanism inferred from these studies.

Keywords: interface-enhanced superconductivity, charge transfer, electron–phonon coupling, molecular beam epitaxy, scanning tunneling microscopy, angle-resolved photoemission spectroscopy

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

1. Introduction

Superconductivity, as a remarkable macroscopic quantum phenomenon, was discovered a century ago [1]. It is characterized by the disappearance of electric resistance and complete expulsion of magnetic field below a critical temperature \(T_C\), and has been observed in many materials. More than five decades ago, Bardeen, Cooper and Schrieffer constructed the microscopic theory of superconductivity, known as BCS theory [2]. According to the BCS theory, electrons in a superconductor collectively bind into ‘Cooper’ pairs and simultaneously condense in much the same way as bosons condense into a superfluid state. The binding interaction is the coupling between electrons and vibration of the lattice (phonons). The scheme normally leads to an isotropic s-wave pairing of electrons with opposite momenta near the Fermi
According to the modified BCS theory [2],

\[ T_C = 1.14 \theta_D \exp\left[-1/N(E_F)\right] V, \]

where \( \theta_D \) is the Debye temperature, \( N(E_F) \) the electron density of states at \( E_F \), and \( V \) the attractive electron–electron interaction. In principle, \( T_C \) can be quite high by increasing one or more of the three parameters. Unfortunately, not all three parameters are completely independent. Ceramic materials have a large \( \theta_D \) but very small \( N(E_F) \), thus insulating unless heavily doped. Most elementary metals superconduct, but the \( T_C \) is low because of small \( \theta_D \). \( T_C \) had reached 25 K in Nb₃Ge in 1973 [5], and then 39 K in MgB₂ in 2001 [6], which is probably the upper limit of \( T_C \) for a typical conventional superconductor [7]. It had been generally accepted by the community that there is not much room for further raising \( T_C \).

A new era of superconductivity research was ushered in with the discovery of \( T_C \) well above 30 K in La-based cuprate in 1986 [8] and its rapid raising to a temperature well above the boiling point (77 K) of liquid nitrogen in Y-based and Hg-based cuprates in the early 1990s [9, 10]. Historically, cuprates have been coined as unconventional superconductors, since their high \( T_C \) cannot be explained directly by the BCS theory for conventional superconductors, although it is not fully proved. The second upsurge in unconventional superconductivity was the discovery of iron-based superconductors in 2008; \( T_C \) of 26 K was first reported in LaFeAsO₁₋ₓFx [11], and the record of 55 K for bulk iron-based superconductors in SmO₁₋ₓFₓFeAs in the same year [12]. The term ‘unconventional’ is notably justified by the primary \( d_{x^2–r^2} \) pairing symmetry (with nodes in the superconducting gap) [13, 14] and the pseudogap state [15–17] for cuprates, possible \( s_\pm \) pairing symmetry for iron-based superconductors [18, 19], and a BCS ratio \( (2\Delta/k_BT_C) \) much larger than 3.53, which goes beyond the conventional wisdom in the framework of BCS theory. Although the contribution of \( e–ph \) coupling to high-temperature superconductivity has been revealed by various experimental studies such as angle-resolved photoemission spectroscopy (ARPES) [20], inelastic scattering [21] and tunneling spectroscopy [22], the dominant view of the community is that the \( e–ph \) interaction is, at most, peripherally relevant to high-temperature superconductivity. Most researchers believe that the high-temperature superconductivity in cuprates and iron-based pnictides and chalcogenides originates from strong electron–electron correlation and that the pairing is mediated by spin fluctuation or short-range magnetic exchange interaction [14–16, 18]. However, the exact picture remains elusive and one of the foremost open problems in condensed matter physics.

In spite of diverse microscopic electronic structures and phenomena, all the compounds with \( T_C \) higher than ~30 K, including both unconventional superconductors and conventional BCS superconductors such as MgB₂, have a layered structure. Cuprates and iron-based superconductors consist of two types of quasi-2D substructures: the superconducting layer (CuO₂-layer or FeAs/FeSe-layer) and charge reservoir layers, for example, the La(Sr)O and LaO(F) layer, as schematically shown in figures 1(a) and (b), respectively [14, 23]. The superconductivity occurs within the superconducting layer with its charge transferred from the reservoir layers, similar to the modulation-doping in semiconductor hetrostructures [24]. In MgB₂, the characteristic graphite-like 2D boron layers sandwich the triangular Mg layers, forming a structure similar to the intercalated graphite [25], and the superconductivity can be viewed as having resulted from charge transfer from Mg to graphite-like boron layers as well.

Learning from LaFeAsO₁₋ₓFx with \( T_C = 26 \) K [11], we realized that the interface between the superconducting layer and the charge reservoir layer may play a crucial role in high-temperature superconductivity. In mid-2008, we took the plunge and commenced our study on interface superconductivity. The theoretical prototype of interface superconductivity can be traced to ‘surface superconductivity’ proposed by Ginzburg in 1964 [26]. However, the experimental research in this direction has just got into its stride with the development of advanced thin-film deposition techniques that has allowed atomic layer precision in the last two decades. We recommend the excellent review by Pereiro et al [27] and Gariglio et al [28] on the superconductivity of...
various heterostructures including PbTe/PbS, LaAlO₃/SrTiO₃, La₂₋₅Sr₂CuO₄/La₂CuO₄, etc. We began with searching for high $T_C$ in the heterostructure of ultra-thin metal/oxide films epitaxially grown on semiconductor/oxide substrates. We anticipated the achievement of significantly enhanced $T_C$ by taking advantage of the high $N(E_F)$ of metal or 2D carrier gas/liquid formed at the interface, high Debye temperature of semiconductor/oxide, and probably strong electron-electron coupling at the interface. That is, we simultaneously maximize the three parameters in formula (1) by utilizing the interface effect (figure 1(c)).

We prepared single atomic layer films of superconductors on semiconductor/oxide substrates by state-of-the-art molecular beam epitaxy (MBE) and studied their superconductivity by combined scanning tunneling microscopy/spectroscopy (STM/STS) and transport measurements. We observed superconductivity in single atomic layer films of both conventional superconductors (Pb and In on Si) and LaAlO₃/Ga on GaN) and high-temperature superconductor in 1UC-FeSe films on SrTiO₃ (STO) (referred to as 1UC-FeSe/STO hereafter) [31]. In this short review, we summarize the interface-enhanced superconductivity in Pb/Si, In/Si and FeSe/STO systems, with focus on the FeSe/STO system, which exhibits the highest superconducting transition temperature $T_C$ among all the heterostructure systems discovered so far [32–36].

2. Interface-enhanced superconductivity in Pb/Si and In/Si systems

Bulk Pb and In are conventional superconductors with $T_C \sim 7.2$ K and $T_C \sim 3.4$ K, respectively [1]. When considering size and dimensional effects, the normal trend is that superconductivity is suppressed when the superconductor is thinner than the size of the electron pairs that form the superconducting state. For example, $T_C$ of ultra-thin crystalline Pb films is continuously reduced with decreasing film thickness from nine to three atomic layers [37]. In the 2D limit of single atomic layer, where electrons are mobile only in the planar direction, thermal and/or quantum fluctuations may disturb the coherent motion of the electron pairs and break the superconductivity. Thus, whether superconductivity exists in single atomic layer has been questioned.

It turns out that a superconducting gap appears on both single atomic layer Pb and In on Si (111) substrates [29]. Figures 2(a) and (b) display the schematic structure and atomically resolved STM topograph of the striped incomensurate (SIC) phase Pb on Si (111), respectively. Figure 2(c) shows the tunneling spectra as a function of temperature. A well-defined U-gap with zero conductance region and two symmetric sharp coherence peaks centered at $E_F$ can be seen at temperatures below 1.82 K. The temperature evolution of the superconducting gap is shown in figure 2(d). Fitting the data using the BCS gap function yields $\Delta = 0.35$ meV, $T_C = 1.83$ K and the BCS ratio $2\Delta/k_B T_C = 4.4$. Despite the $T_C$ being much lower than the bulk value of 7.2 K [1], the BCS ratio is very close to the value of 4.3 in bulk Pb, suggesting that SIC-Pb/Si is a strongly coupled BCS superconductor.

Figure 2. (a) and (e) Schematic structure models, (b) and (f) high-resolution STM images ($V = 100$ mV, $I = 50$ pA) of SIC-Pb/Si (111) and $\sqrt{7} \times \sqrt{7}$ In/Si (111), respectively. (c) and (g) the $dI/dV$ spectra measured with a Nb tip as a function of temperature ($V = 10$ mV, $I = 200$ pA), (d) and (h) the superconducting gap as a function of temperature for SIC-Pb/Si (111) and $\sqrt{7} \times \sqrt{7}$ In/Si (111), respectively. Adapted from Zhang et al 2010 Nat. Phys. 6 104 [29].

Single atomic layer In grown on Si (111) with $\sqrt{7} \times \sqrt{7}$ reconstruction is also superconducting, as shown in figures 2(e)-(h). The values of $\Delta$, $T_C$ and $2\Delta/k_B T_C$ extracted from the BCS fit are 0.57 meV, 3.18 K and 4.16, respectively. Unlike SIC-Pb/Si, where $T_C$ is strongly suppressed compared with the bulk value, the $\sqrt{7} \times \sqrt{7}$ – In phase has a surprisingly high $T_C$ close to the bulk value of 3.4 K. More significantly, $\sqrt{7} \times \sqrt{7}$ – In/Si has an enhanced BCS ratio of 4.16 in comparison to the bulk value of 3.6, implying that it is
transformed into a strongly coupled superconductor as SIC-Pb/Si.

The superconductivity observed above can be interpreted as having resulted from the interface effect demonstrated in figure 1(c): the carriers in the metal overlayer carry the supercurrent and the $e$-$ph$ interactions that glue the electrons to form pairs are provided by both the metallic layer and the interface. This interpretation is supported by ARPES studies, which demonstrate well-developed 2D free-electron-like interface. This interpretation is supported by ARPES studies, to form pairs are provided by both the metallic layer and the FeSe therein. In 3.1, we introduce the growth and structure of superconductors it is structurally the simplest and the MBE growth recipe of single-crystalline stochiometric compounds had been well established in our group [29]. Later on, two independent in situ transport studies indicate that single atomic layer In (Pb) indeed superconducts at 2.8 K (1.1 K) [39, 40]. Meanwhile, our interpretation of the correlation of superconductivity with interfacial $e$-$ph$ coupling is also supported by first-principles calculation [41]. The 1UC-FeSe films on Nb-STO and on insulating STO substrates exhibit similar superconducting property, which will be discussed in the next section. Our idea of interface-enhanced superconductivity, as demonstrated in figure 1(c), therefore, is verified elementarily.

3. Interface-enhanced superconductivity in the FeSe/STO system

We then moved to investigate ultra-thin films of unconventional superconductors to achieve higher $T_C$ in 2010. We chose FeSe, because among all the unconventional superconductors it is structurally the simplest and the MBE growth recipe of single-crystalline stoichiometric compounds had been well established in our group [42, 43]. In this section, we discuss the interface-enhanced superconductivity in the FeSe/STO system and conclude that interface charge transfer and $e$-$ph$ coupling are essential to the high $T_C$ superconductivity therein. In 3.1, we introduce the growth and structure of FeSe/STO(001). In 3.2, we demonstrate its superconducting property, including superconducting gap $\Delta$ and superconducting transition temperature $T_C$. We discuss the interface effects, such as strain, charge transfer and $e$-$ph$ coupling, and their contribution to superconductivity in 3.3. We show interface-enhanced superconductivity in other related FeSe/STO systems in 3.4.

3.1. Materials and structure

The bulk $\beta$-phase FeSe is a superconductor with $T_c \sim 8$ K at ambient pressure [44]. One UC $\beta$-phase FeSe consists of two Se layers sandwiching an Fe layer with an in-plane lattice constant of 3.78 Å and an out-of-plane lattice constant of 5.50 Å (figure 3(a)). Epitaxial FeSe films with a Se-terminated (001) surface can be obtained by co-evaporating Fe and Se under Se-rich condition (typical flux ratio of $\sim$1:10) on both graphene [43] and STO [31] substrates. When FeSe is grown on graphene, it forms nearly free-standing islands due to very weak interaction and, hence, the intrinsic properties of bulk FeSe are observed, including the in-plane lattice constant of 3.8 Å, V-shaped gap of $\sim$2.2 meV and $T_c$ of $\sim$8 K [43, 45, 46]. For one UC FeSe film epitaxially grown on STO (001) substrates, its in-plane structure follows that of STO (001) with a lattice constant of $\sim$3.90 Å [31, 47], 3% expanded compared to bulk FeSe [44]. Correspondingly, it is compressed along the c-axis with a reduced Se height $h_{Se}$ (above the Fe layer) of 1.31 Å, $\sim$9% smaller than the bulk value of 1.45 Å, as resolved by atomically resolved scanning transmission electron microscopy (STEM) [48]. The Se–Fe–Se bond angle $\alpha$ is enlarged to 111.4° $\pm$ 0.9°, very close to that of a tetrahedron ($\sim$109.47°).

Macroscopic uniform single UC FeSe films can be obtained by MBE, which allows transport measurement down to single UC precision. As shown in figure 3(b), the morphology of single UC FeSe films perfectly follows the step-terrace structure of STO(001) substrates. Under the Se-rich growth condition, excess Se atoms form Se dimers (figure 3(c)) and the as-grown Se-rich FeSe$_{1+x}$ films are not superconducting at 4.2 K [49, 50]. Superconductivity occurs only when the density of the Se dimer reduces to approximately 2 per 10 $\times$ 10 nm$^2$ and lower after extensive annealing (figure 3(d)). Se vacancies appear with elongated annealing (figure 3(d)), but they do not destroy superconductivity [49]. It is worth noting that neither the lattice structure nor the superconductivity is dependent on the bulk property of the STO substrates [31, 49]. Below, we mainly show STS and ARPES results on Nb-STO(001) substrates and transport on insulating STO(001), unless otherwise noted.

3.2. Superconducting property: U-shaped gap and enhanced $T_C$

It is striking that the single UC FeSe films on Nb-STO(001) exhibit an overall U-shaped gap of 20 meV (figure 3(c)), which is nearly one order of magnitude higher than that of bulk FeSe [31]. In single UC FeSe films on insulating STO (001), we observed similar superconducting-like gap $\Delta$ $\sim$15.4 meV, roughly agreeing with the gap magnitude observed on 1UC-FeSe/Nb-STO(001) within experimental uncertainties, which is still visible at 50.1 K (figure 3(g)) [49]. A gap-closing temperature $T_{gap} \sim 66.8$ K is deduced from the linear dependence of zero bias conductance on temperature (figure 3(f)). The above observation suggests that 1UC-FeSe/STO(001) is a high-temperature superconductor with $T_C$ exceeding the known record $T_C$ $\sim$ 55 K of bulk iron-based superconductors [12]. Furthermore, in terms of the gap observed on FeSe films on graphene, which is $\sim$2.2 meV in magnitude and closes at $\sim$8 K [43], and assuming the same BCS ratio, we can even anticipate a transition temperature above the boiling point (77 K) of liquid nitrogen.

The above discovery is surprising, not simply because it suggests high-temperature superconductivity with $T_C$ above 50 K, but because the U-shaped gap hints at a conventional s-wave pairing symmetry, which has stimulated tremendous
interest in the superconductivity community. Our finding was soon confirmed by several independent ARPES and STM/STS studies, which consistently demonstrate a nodeless gap with a magnitude of 14–19 meV depending on the sample quality [51–55]. For example, the Xingjiang Zhou group performed the first ARPES investigation and demonstrated a superconducting-like gap of 15–19 meV which persists to 65 K [51, 52], agreeing well with the STS results shown in figures 3(e)–(g). More significantly, they further revealed that the superconducting-like gap is nearly isotropic around the whole Fermi surface, which matches well the observed U-shaped gap shown in figure 3(e) [31]. The Donglai Feng group investigated quasi-particle interference patterns in 1UC-FeSe/Nb-STO(001) and the response to both magnetic and non-magnetic impurities. Consequently, they deduced plain s-wave pairing symmetry based on their finding that the quasi-particle interference patterns are against line nodes and the superconductivity is suppressed by magnetic impurities, but does not respond to non-magnetic impurities [55]. This s-wave pairing symmetry is reminiscent of phonon-mediated pairing under the scheme of BCS theory, which will be discussed in section 3.3.

Since superconductivity is a macroscopic quantum phenomenon, tremendous effort has been made on transport study and $T_c$ ranging from 40 K to above 80 K has been reported by ex situ measurement [32–34, 36]. The transport measurement is challenging, because the single UC FeSe films are too thin (0.55 nm) to survive in atmosphere. By using amorphous Si and crystalline FeTe as a capping layer, we performed the first ex situ transport study and found similar $T_c$ in amorphous-Si/3UC-FeSe/STO [31, 56] and FeTe/1UC-FeSe/STO [32–34]. The schematic and the result of ex situ transport measurement on 10UC-FeTe/1UC-FeSe/STO are shown in figure 4. The resistance starts to decrease at 54.5 K and drop completely to zero at 23.5 K (figure 4(c)). The onset transition temperature $T_{onset}$, defined as the point where the normal resistance and the superconducting transition curves cross, is above 40 K (figure 4(c)), increased fourfold with respect to bulk FeSe [57]. Meanwhile, two-coil mutual inductance measurement revealed the formation of diamagnetic screening at 21 K (figure 4(d)), which is consistent with $T_{zero} \sim 23.5$ K. In addition, two independent dc magnetic susceptibility measurements provide more information. The Paul Chu group measured 10UC-FeTe/6UC-FeSe/STO(001) ($n = 1, 2, 3, 4$) samples and demonstrated a Meissner state below 20 K, a mesoscopic superconducting state up to 45 K and collective excitations up to 100 K with their nature yet to be determined [33]. The Jian Wang group observed a drop crossover around 85 K in 10UC-FeTe/1UC-FeSe/Nb-STO(001) [34]. Recently, the Yayu Wang group performed ex situ two-coil mutual inductance measurement on one total 5UC thick films (2UC-FeSe/2UC-Fe$_{0.96}$Co$_{0.04}$Se/1UC-FeSe/Nb-STO(001)) capped by Se and revealed an onset of diamagnetic screening at 65 K [36], coinciding with the gap-closing temperature determined by previous ARPES [52] and STS studies [49].
transition temperature observed in the latter two cases could be due to the fact that FeSe films on Nb-STO(001) are more uniform than those on insulating STO(001) [31, 49]. Since the second UC and thicker films do not superconduct even at 4.2 K [31], we believe that all the superconducting behavior observed on 1-5 UC FeSe films originates from the first UC FeSe right above STO(001), while the additional FeSe layers serve as protection layers. Similarly, in the case of 3UC-FeSe films capped by amorphous Si, we guess amorphous Si mixes into the underlying 1-2 UC FeSe films, so that only the bottom layer remains as FeSe and the observed $T_C$ equals to that of 1UC-FeSe films capped by single-crystalline FeTe. For films thinner than 3UC capped by Si, no superconducting transition is observed, the same as in the case of Pb films capped by Si [37].

FeTe films do protect FeSe films from oxidation in atmosphere. However, they simultaneously suppress the superconductivity through partially mixing with FeSe [48, 58] and providing an additional decay channel to the gluing bosons [59]. The atomic intermixing can be resolved from the atomically resolved STEM image shown in figure 4(b), as the top layer Se atoms look similar to the Te layer and in contrast, obviously brighter than the bottom-layer Se atoms. Thus, to pin down the $T_C$ of 1UC-FeSe/STO(001), one would need to either perform transport and magnetic measurement in situ, without exposing the film to air, or find a capping material that does not reduce $T_C$. Recently, the Jin-Feng Jia group accomplished in situ transport measurement. Remarkably, by using microscopic four-point contact probe technique [35], they found $T_C$ above 100 K [35], suggesting that 1UC-FeSe/STO(001) is the second superconductor system with $T_C$ above 77 K. This is to be confirmed by in situ magnetic measurement.

### 3.3. Interface effect

The 1UC-FeSe/STO(001) exhibits distinctive superconductivity. In contrast to the weakly interacting interface between FeSe films and graphene [43, 45], the FeSe/STO (001) interface plays a significant role in the remarkably enhanced superconductivity. Here, we summarize several distinct behaviors of this system:

1. 1UC-FeSe films on STO(001) exhibit a U-shaped gap of 20 meV (figure 3(e)) and the second UC and thicker films do not superconduct at 4.2 K [31], whereas, FeSe films only thinner than 2UC have a V-shaped gap of no more than 2.2 meV when they are grown on graphene [45].

2. With increasing film thickness, the $T_C$ (measured by ex situ transport) of multilayer FeSe films on STO(001) gradually degrades and reduces to the bulk value of 8 K at 50UC [56], which is again in sharp contrast with the case on graphene, where the $T_C$ (extracted from the linear fitting of the temperature-dependent zero bias conductance) increases and saturates to 8 K at 8 UC [45].

To elucidate the role of the FeSe/STO interface, such as strain, interface charge transfer and interface $e$–$ph$ coupling, various experimental investigations have been performed. As the superconductivity of bulk FeSe is very sensitive to pressure [60], we first consider the strain effect. The finding that 1UC-FeSe films on STO(001) undergo in-plane expansion and out-of-plane compression, as described in section 3.1, seems to be consistent with the empirical rule in bulk iron chalcogenides that the superconductivity is enhanced when the Se height $h_{Se}$ is reduced [61]. However, according to the empirical relation between $T_C$ and the anion height for bulk iron-based superconductors [61], a $T_C$ of $\sim$20 K is expected for the Se height $h_{Se}$ of 1.31 Å, which is much lower than the value for 1UC-FeSe films on STO. Motivated by such empirical phenomena, the Donglai Feng group used NbSTO/KTaO$_3$ [62] and Nb-BaTiO$_3$/KTaO$_3$ [63] as substrates to achieve FeSe films subjected to even stronger in-plane expansion. However, their ARPES study demonstrated that the gap-closing temperature is enhanced only 5 K with an additional 5.5% lattice expansion [63]. Meanwhile, we observed a similar superconducting gap of 17–20 meV in 1UC-FeSe films both on TiO$_2$ [64] and STO(110) [65, 66] substrates (see sections 3.4.3 and 3.4.4 for details, respectively), although they possess distinct in-plane structure, i.e. square lattice with the same lattice constants as bulk FeSe versus rectangular lattice with 5% anisotropy. Strain is, therefore, excluded from the critical factors for the interface high-temperature superconductivity in FeSe/STO. Such a conclusion is further supported by the observation of an identical superconducting gap across a domain boundary in 1UC-FeSe/STO(001), despite locally compressed lattice [55].
Furthermore, the lattice variation would significantly affect the antiferromagnetic superexchange interactions between Fe moments [67]. Hence, the minor role of strain refutes the notion that antiferromagnetic-interaction/spin-fluctuation plays the dominant role in mediating Cooper pairing in the FeSe/STO system.

On the other hand, it is widely believed that interface charge transfer and interface-enhanced \( e-\text{ph} \) coupling are essential to the high-temperature superconductivity in 1UC-FeSe/STO, for their contribution has been explicitly identified by various experimental investigations and theoretical calculations. Below, we successively discuss these two effects.

3.3.1. Charge transfer. Significant charge transfer at the FeSe/STO interface, resembling that from the carrier reservoir layer to superconducting layer in high-temperature superconductors (figures 1(a) and (b)), has been revealed by extensive ARPES [51–54], STS [49, 50] and transport studies [49]. For example, as shown in figure 5(a), the Fermi surface of superconducting 1UC-FeSe/STO(001) consists of only electron-like pockets centered around the Brillouin zone corners with a band bottom lying 60 meV below the Fermi level, indicative of the formation of 2D electron gas (2DEG). Compared with the Fermi surface of bulk FeSe, the lack of hole pockets in the Brillouin zone center of 1UC-FeSe/STO (001) implies that the 1UC-FeSe films are heavily electron doped. As estimated from the Fermi surface volume, the doping level in the FeSe layer is \( \sim 0.12 \) electrons per Fe atom [52], which could originate from the oxygen vacancies in the STO substrates [53, 68] and be induced by band bending at the FeSe/STO interface [64, 69]. In another example, as demonstrated by the Hall measurement shown in the inset of figure 5(c), the carriers change from p-type to n-type with decreasing temperature, indicating that the superconductivity is dominated by n-type carriers. Moreover, the superconductivity transition shifts to higher temperature with more electrons injected into the FeSe films from STO under electrical field (figure 5(c)). It is evident that the interface charge transfer from STO to FeSe films indeed plays a role in promoting high-temperature superconductivity.

In contrast, with hole pockets appearing in the Brillouin zone center (figure 5(b)) and becoming stronger with increasing thickness [53], 2UC and thicker FeSe films on STO(001) prepared by the same method do not exhibit any signature of superconductivity [53, 70]. Compared with 1UC-FeSe/STO(001) [52], one may speculate that the absence of superconductivity in multilayer FeSe films is due to insufficient carrier transfer from the STO substrate. Recently, it was revealed that multilayer FeSe films convert to superconducting when they are doped with sufficient electrons. This was first reported by Y. Miyata et al., as their variable-temperature ARPES study showed that 3UC-FeSe films become electron populous with potassium (K) adsorption and exhibit a pairing formation temperature of \( 48 \pm 3 \) K at optimal doping [71]. Almost during the same period, we systematically investigated the K adsorption on 1–4UC FeSe films by STS, while the Xingjiang Zhou and Donglai Feng groups performed ARPES investigation on the surface-doping effect with K adsorption.

We found that K adsorption does induce superconductivity in the topmost layer of multilayer FeSe films (>2UC). However, the gap size is thickness dependent [47]. Potassium atoms adsorb randomly on the surface of FeSe films (figure 6(a)) and form local \( 2 \times 2 \) and \( \sqrt{5} \times \sqrt{5} \) reconstructions at the K coverage of \( \sim 0.2 \) ML (figure 6(b)). As shown in figures 6(c) and (d), for all the 2–4UC FeSe films, a superconducting gap appears at the K coverage of approximately 0.1 ML, increases with further adsorption of K atoms, and reaches a maximum value at an optimal K coverage of 0.2–0.25 ML (roughly corresponding to a charge transfer of \( \sim 0.1–0.12 \) electron/Fe). Intriguingly, the gap size decreases with increasing thickness, that is, the superconductivity enhancement degrades with the superconducting layer moving away from the interface. For example, at optimal doping, 2UC, 3UC and 4UC films exhibit superconducting gaps of \( 14.5 \pm 1.0 \) meV, \( 13.1 \pm 1.4 \) meV and \( 11.9 \pm 1.4 \) meV, respectively (figure 6(d)). In contrast, FeSe films thicker than 4UC under optimal doping universally exhibit a smaller gap of 10 meV persisting up to \( 44 \pm 2 \) K [72, 73]. Given that K atoms mainly dope electrons into the topmost FeSe layer [72, 74] and the doping level can reach and even exceed the value of \( \sim 0.1–0.12 \) electron/Fe transferred from STO to
1UC-FeSe films, the finding that the superconducting gap gradually decays with increasing thickness up to 4UC and remains constant for thicker films suggests that the STO substrate contributes an additional role in promoting the superconductivity, that is, interfacial e–ph coupling that will be discussed in section 3.3.2.

We further found that the magnitude of the superconducting gap observed on K-coated 2UC-FeSe films depends on the superconducting property of the underlying 1UC-FeSe films [75]. It reaches a maximum gap of ~17 and ~11 meV depending on whether the underlying 1UC-FeSe is superconducting or not, respectively. The underlying 1UC-FeSe films can be superconducting or nonsuperconducting, which can be achieved by controlling the annealing temperature above or below 450 °C, respectively [75]. The gap size in the latter case is consistent with the value of 10 meV achieved on K-coated FeSe films thicker than 4UC [72]. If the gaps of 10–11 meV are solely induced by electron doping, the further enhancement to 17 meV when the underlying 1UC films are superconducting could be attributed to the additional role of the FeSe/STO interface. For 2UC-FeSe films, the additional enhancement factor is ~55% (from 11 to 17 meV).

In the case of 1UC-FeSe films, they undergo stronger suppression induced by thermal/quantum fluctuation in the 2D limit, since K-coated 1UC-FeSe films on graphene exhibit an exclusively small gap of 6.6 meV in comparison with ~10 meV for K-coated 2UC and thicker films on the same substrate [73, 76]. If compared with K-coated 1UC-FeSe/graphene, the gap size of 1UC-FeSe/STO(001) [31] is impressively increased threefold.

Very recently, a high $T_C$ of 35–48 K [77–79] and a superconducting gap of 9–14 meV [73, 76] were successively achieved in heavily electron-doped FeSe thin films/flakes by using a liquid-gating technique and K-coated FeSe films on graphene, respectively. Moreover, this high-temperature superconductivity occurs after the native low $T_C$ superconductivity ($T_C \sim 8$, V-shaped gap of ~2.2 meV) vanishes completely with increasing doping, showing up as a separated phase [76, 78]. The native low $T_C$ superconductivity disappears with an extra electron doping of ~0.02 e per Fe atom, and the high-temperature superconductivity occurs and reaches optimal at doping of ~0.04 and ~0.12 e per Fe, respectively [76]. This finding disclosed that the nonsuperconducting behavior of multilayer FeSe films on STO(001) is due to weaker electron doping from STO (~0.02 e per Fe atom for the second UC films) which suppresses the low $T_C$ superconductivity, but is insufficient to boost high $T_C$ superconductivity. On the other hand, 1UC-FeSe/STO(001) is directly promoted to the high $T_C$ regime with a large amount of charge transfer from STO (0.12e/Fe), which correlates to the double-TiO$_x$ layer (figure 4(b)) at the FeSe/STO interface [48, 80]. The above results clearly indicate that the charge transfer from the STO substrates to FeSe films plays a crucial role.

### 3.3.2. e–ph coupling

Compared with all the other electron-doped or alkali-metal intercalated FeSe superconductors, 1UC-FeSe/STO(001) exhibits the following unique features:

1. The 1UC-FeSe/STO(001) possesses a $T_C$ of ~20 K higher and a gap of at least 5 meV larger than the corresponding values of all the other FeSe-correlated superconductors [76, 78, 81–83].

2. Compared with alkali-metal intercalated FeSe superconductors, for example, (Tl,Rb)$_x$Fe$_{2–y}$Se$_2$ superconductor ($T_C = 32$ K, effective electron mass ~6.1m$_e$) [84], the electron correlation in 1UC-FeSe/STO(001) is weaker (effective electron mass ~2.7m$_e$) [51]. This fact, together with (1), refutes the notion that the high
$T_C$ in 1UC-FeSe/STO(001) is due to a stronger electron–electron interaction.

3) In contrast to the competition between superconductivity and nematic order observed in multilayer films (>2UC) [72, 85], neither $xz$/$yz$ band-splitting signaling the nematic order [31, 51, 54] nor any sign of strong nematic fluctuation [55] is observed in 1UC-FeSe/STO(001).

4) The superconductivity in 1UC-FeSe/STO(001) is always continuously suppressed with K adsorption (figures 6(c) and (d)), independent of the initial state (superconducting or not) [75]. In contrast, the superconductivity can be further enhanced when electrons are injected from STO with field effect (figure 5(c)) [49].

The features (1)–(3) imply that the FeSe/STO interface contributes additional profound effects besides doping and, hence, provides additional enhancement in superconductivity. The additional role could be the interface-enhanced $e$–$ph$ coupling, as proposed [31], which has been experimentally identified by successive APRES [54], STS [47], ultrafast optical spectroscopy [59] and high-resolution electron energy loss spectroscopy (HREELS) [86] investigations and theoretically supported [87–91]. First, ARPES observation of ‘replica’ bands with energy separation of $\sim$100 meV in 1UC-FeSe/STO(001) suggests the coupling of FeSe electrons with oxygen optical phonons in STO at such frequency [54]. Furthermore, an $e$–$ph$ coupling constant $\lambda \sim 0.5$ is obtained, as estimated from the intensity ratio of the replica bands. Second, in agreement with previous ARPES [51, 52, 54] and STS [31, 49] results, $T_C = 68(\pm 5/2)$ K, $\Delta = 20.2 \pm 1.5$ meV and $\lambda = 0.48$ were revealed by ultrafast optical spectroscopy study [59]. The $e$–$ph$ coupling constant $\lambda = 0.48$ is triple the value of 0.16 measured by the same method for bulk FeSe [92]. Third, a Fuchs–Kliewer (F–K) phonon at a frequency of $\sim$92 meV was recently revealed by surface phonon investigation using HREELS, consistent with previous ARPES results [54] in terms of phonon energy. More importantly, the electric field generated by this F–K phonon decays exponentially with a decay length of 2.5UC-FeSe [86], which agrees with the observed thickness-dependent superconducting gap shown in figure 6(d). These findings point out that the oxygen longitudinal optical (LO) mode of STO with energy reaching $\sim$100 meV (partially) contributes to the additional enhancement beyond doping. This special oxygen LO phonon is demonstrated to be a special oxygen-vacancy- induced flat phonon mode, mainly composed of relative Ti and O atomic displacements along the [001] direction in the top two layers of the STO substrate [88]. Meanwhile, a quantum Monte Carlo computation showed that this interface $e$–$ph$ coupling can significantly enhance the pairing strength, irrespective of the pairing symmetry as well as its underlying electronic origin [91].

Last, but not least, the significant change in the $e$–$ph$ interaction in FeSe films owing to the coupling with STO substrates is also identified in tunneling spectra; the signature of $e$–$ph$ coupling, i.e. symmetric dip-hump features show up.

As displayed in the normalized tunneling spectra shown in figures 7(a) and (b), the emergence of the superconducting gap is accompanied by two pairs of dip-hump features, which degrade simultaneously with increasing temperature [47]. Displayed in figure 7(c) is the phonon-mode energy $\Omega$, extracted from the second derivative of tunneling conductance, as a function of the superconducting gap. The energy distribution of the phonon modes collapses basically into two distinct groups centered at 11.0 meV ($\Omega_1 = 11.0 \pm 2.1$ meV) and 21.5 meV ($\Omega_2 = 21.5 \pm 4.5$ meV), despite the superconducting gap changing significantly from 6.5 to 19 meV (depending on the film thickness and K coverage). On the other hand, for ultra-thin films of FeSe grown on and weakly
bonded to graphene, although a bosonic mode was observed, the energy is much smaller and only at a level of 2.7−4 meV [93]. The sharp contrast again proves the special role of the STO substrate in boosting the e−ph coupling and, hence, the high $T_C$ superconductivity.

The identification of the two groups of phonon modes with energy of $∼11.0$ and $∼21.5$ meV and their contribution to the high $T_C$ superconductivity in 1UC-FeSe/STO(001) is further supported by first-principles calculation [90]. According to the description in [90], one of the roles of the STO substrate is to stabilize the 1UC-FeSe films to a nearly square arrangement so as to prevent the films from undergoing a shear-type structure transition as is the case in bulk [90]. Indeed, the square structure is evidenced from the STM images shown in figure 3(d), and signal of nematic order has never been observed in 1UC-FeSe/STO [31, 51, 54], which is in contrast to bulk FeSe [94]. As a result of this substrate-bound structure, two e−ph coupling channels with phonon frequencies of 10 and 20 meV [90], in excellent agreement with experimental observation shown in figure 7(c), are opened. This excellent agreement suggests that the STO substrates indeed act as a template and further enhance the e−ph interaction from FeSe phonons. The corresponding e−ph coupling constant is calculated to be $\lambda = 1.6$ [90], ten times the value of $\lambda = 0.16$ for bulk FeSe [92].

As discussed in section 3.3, with strong electron–electron correlation being excluded as a key role, we show that the interface charge transfer and e−ph coupling cooperatively contribute to the high $T_C$ in 1UC-FeSe/STO(001) systems. We roughly estimated the respective contribution of the interface charge transfer and e−ph coupling as the former can enhance the gap to 10−14 meV and the latter takes the responsibility for additional enlargement of 6−10 meV. Our initial proposal [31] as depicted in figure 1(c) is, therefore, verified to be an efficient approach for raising $T_C$. The evidence of enhanced e−ph coupling discussed above, together with the U-shaped gap and s-wave pairing symmetry discussed in section 3.2, points out that the pairing mechanism in 1UC-FeSe/STO(001) could be rather conventional, i.e. pairing mediated by phonons. This conjecture is supported by a recent calculation based on e−ph coupling mechanism, which revealed that a $T_C$ of 77 K is obtainable for 1UC-FeSe/STO(001) when the experimentally identified LO phonon energy of 100 meV [54, 86], e−ph constant of 0.5 [54, 59] and chemical potential of 60 meV [51, 53, 54] are taken into account and the Coulomb repulsion is neglected due to the huge dielectric constant of STO [89]. Nevertheless, the detailed picture of interface e−ph coupling and some puzzles still need to be further explored. For example, exploring the isotope effect of $T_C$ will help pin down the pairing mechanism. The interaction between FeSe and double-TiO$_x$ layer terminated STO, which correlates to the significant charge transfer and e−ph coupling at the interface, deserves further study. It is also intriguing to observe the continuous suppression of the superconductivity with K adsorption on 1UC-FeSe/STO(001), feature (4) listed in section 3.3.2. Given that the interface e−ph coupling is associated with the formation of the interface electric dipole, due to the relative displacement of the Ti cations and the oxygen anions [48, 88], the intrinsic quantum paraelectric/incipient ferroelectric of STO [95−97], and the electric field generated by F−K phonon mode of STO [86], we can interpret the suppression of superconductivity in 1UC-FeSe films with electrons doped from the topside as having resulted from the counteraction of the interface electric dipoles, which, thus, weakens the screening effect and Cooper pairing strength in turn.

3.4. Interface-enhanced superconductivity in related systems

It is natural to ask whether the interface-enhanced superconductivity scenario works in other systems. To test this idea, we have grown 1UC FeTe$_{1−x}$Se$_x$ films on STO(001), 1UC K$_x$Fe$_2$Se$_2$ films on STO(001), 1UC-FeSe films on STO (110) and on TiO$_2$(001) by MBE and studied the superconducting properties by in situ STS and ex situ transport. We found that all the systems exhibit remarkably enhanced superconductivity compared with the corresponding bulk materials. Below, we briefly discuss the main results in the four systems.

3.4.1. FeTe$_{1−x}$Se$_x$ films on STO(001). Similar to FeSe, FeTe$_{1−x}$Se$_x$ films are prepared by co-depositing Fe, Se and Te and form an ordered Se/Te−terminated (001) surface on STO (001) substrate. Displayed in figure 8(a) is a typical morphology of 1UC FeTe$_{1−x}$Se$_x$ films, where bigger Te atoms are imaged brighter, and smaller Se atoms darker. U-shaped gaps with vanishing conductance centered at $E_F$ are observed in all the 1UC FeTe$_{1−x}$Se$_x$ films ($x = 0.1$, 0.3, 0.5, 0.6) and the gap size varies from 12−16 meV depending on the Se composition (figure 8(b)). Compared to the superconducting gap $∼1.7$ meV of the optimally doped bulk FeTe$_{0.6}$Se$_{0.4}$ single crystal [98], the gap size of 1UC-FeTe$_{1−x}$Se$_x$/STO(001) is enlarged at least six times, indicative of interface-enhanced superconductivity. In addition, several groups of phonons with frequency of $∼10$, $∼20$ and $∼25$ meV, which are consistent with $E_g$(Te/Se), $A_{1g}$(Te/Se)/TO$_2$(STO) and $B_{1g}$(Fe) modes, respectively, are observed. However, the spin resonance mode at $∼6$ meV as observed in bulk FeTe$_{1−x}$Se$_x$ [99, 100] is not observed on this single UC film. The above results again demonstrate the deterministic role of the FeTe$_{1−x}$Se$_x$/STO interface in the enhanced superconductivity. It is worth noting that in contrast to the filamentary superconductivity reported in bulk FeTe$_{1−x}$Se$_x$ when the Se ratio is smaller than 0.29, the superconducting gap is persistent on the whole surface of the 1UC FeTe$_{0.6}$Se$_{0.4}$ films with quite weak position dependence (about 14 meV on Se sites and 12 meV on Te sites), indicating that the interface-enhancing effect is so strong that it supasses the contribution of Te substitution. The resemblance to 1UC-FeSe/STO(001), especially the enlarged U-shaped gap and evident e−ph coupling, implies that interface engineering is a rather general approach for raising superconductivity temperature. Moreover, 10UC-FeTe/1UC-FeTe$_{0.2}$Se$_{0.5}$/STO(001) and 10UC-FeTe/1UC-FeSe/STO (001) exhibit almost similar R-T behavior and equal
Figure 8. (a), (c), (e) and (g) Atomically resolved images ((a) V = 200 mV, I = 100 pA, (c) V = 100 mV, I = 50 pA, (e) V = 70 mV, I = 300 pA and (g) V = 50 mV, I = 100 pA) of 1UC films of FeFe₀.₆Se₀.₄ on Nb-STO(001), K₀.₃Fe₂Se₂ on Nb-STO(001), FeSe on Nb-STO (110) and FeSe on TiO₂(001), respectively. (b), (d), (f) and (h) The corresponding dI/dV spectra (V = 30 mV, I = 100 pA) taken at 4.6 K on the films shown in (a), (c), (e) and (g), respectively. (a) and (b) Adapted from Li et al 2015 Phys. Rev. B 91 220503 [48], (c) and (d) from Peng et al 2015 Phys. Rev. B 92 180507 [75], (e) and (f) from Zhou et al 2016 Appl. Phys. Lett. 108 202603 [65], and (g) and (h) from Ding et al 2016 Phys. Rev. Lett. 117 067001 [64].

T_{onset} \sim 40$ K and $T_{zero} \sim 21$ K [58], indicative of the substitution of Te for Se during the growth of the capping layer, consistent with the intermix of Se and Te at the FeTe/FeSe interface, as shown in figure 4(b).

3.4.2. $K_xFe_2Se_2$ films on STO(001). 1UC $K_xFe_2Se_2$ films were obtained after the K-coated 2UC-FeSe films were appropriately annealed so that the K atoms intercalated between the two FeSe layers [75]. In morphology, 1UC $K_xFe_2Se_2$ films are characterized by a step height of 0.7 nm [75] and $\sqrt{2} \times \sqrt{2}$ reconstruction (figure 8(c)). As expected, a spatially uniform U-shaped superconducting gap of 14.5 meV is observed (figure 8(d)), which is significantly larger than $\Delta \sim 7$ meV of bulk $K_xFe_2Se_2$ [101], $\Delta \sim 4$ meV for $K_xFe_2Se_2$ films on graphene [102], and $\Delta \sim 9$ meV for thicker $K_xFe_2Se_2$ films on STO [103]. Given that bulk $K_xFe_2Se_2$ is heavily electron doped, the enhancement observed here should be mainly due to the interface-enhanced $e-ph$ coupling effect. Meanwhile, the superconducting gap of 14.5 meV in 1UC $K_xFe_2Se_2$ films is about 3 meV smaller compared with K-coated 2UC-FeSe films at optimal doping, probably owing to the similar counteraction of the interface-enhancing effect with K atoms adsorbed on 1UC-FeSe films, as shown in figure 6(c).

3.4.3. FeSe/STO(110). Different from the STO(001) surface, STO(110) is polar and has anisotropic in-plane lattice 3.91 and 5.53 Å along the [001] and [110] direction, respectively. FeSe films form a Se-terminated (001) surface with anisotropic lattice $a = 3.93$ Å and $b = 3.78$ Å along the [100] and [1–10] directions of the STO substrate, respectively (figure 8(e)). Despite anisotropic lattice structure, the 1UC-FeSe films exhibit a gap of 14–17 meV (figure 8(f)), similar in magnitude to most reports of superconducting gaps, but a little bit smaller than the maximum value (20 meV) in 1UC-FeSe/STO(001) [31, 50]. This result excludes strain as a critical factor and is inconsistent with the fact that antiferromagnetic-interaction/spin-fluctuation plays the dominant role in mediating Cooper pairing, as discussed in section 3.3.

The enhanced superconductivity in 1UC-FeSe/STO (110) can be consistently explained in the interface-enhanced $e-ph$ scenario discussed in section 3.3. Basically, STO(110) substrates have comparable 2D carrier density (due to oxygen vacancies) [104, 105] and similar O-Ti-O stretching mode with energy at $\sim 100$ meV [63, 106]. Compared with 1UC-FeSe/STO(001), 1UC-FeSe/STO(110) always exhibits a slightly smaller superconducting gap. We speculate that the dielectric constant and the spatial extension of 2D carriers may play a role. On the one hand, for the 2D superconductivity, where the electrons are confined in the plane parallel to the FeSe/STO interface and form 2DEG, in-plane dielectric constant $\varepsilon ||$ reflects the strength of electron screening or Coulomb interaction. And the interface $e-ph$ coupling constant is proportional to $\sqrt{\frac{\varepsilon ||}{\varepsilon_L}}$, where $\varepsilon ||$ and $\varepsilon_L$ are the dielectric constant parallel and perpendicular to the FeSe/STO interface, respectively [87]. The finding that FeSe/STO (110) exhibits a smaller superconducting gap agrees with the
fact that $\varepsilon_0$ ($\varepsilon_{\perp}$) of STO(110) is smaller (larger) than that of STO(001) [107]. On the other hand, the spatial extension of 2D carriers along the vertical direction is larger for the STO (110) surface than for the STO(001) surface [108]. This weaker confinement of 2D carriers in STO(110) should give rise to lower charge transfer to FeSe films, therefore, lower $T_C$ and weaker $T_C$ modulation with electrostatic gating [65]. This indicates that stronger confinement of 2D carriers promotes higher $T_C$, agreeing with the quasi-two-dimensionality in the high-temperature superconductors introduced in section 1.

3.4.4. FeSe/TiO$_2$(001). Learning that FeSe bonds to double-TiO$_2$ layer at the FeSe/STO(001) interface (figure 4(b), we grow anatase TiO$_2$ directly on STO(001) as substrate for FeSe growth. The anatase TiO$_2$ is characterized with distinct O-Ti-O triple-layered planes with in-plane lattice constant of 3.78 Å, much closer to that of bulk FeSe in comparison with STO(001). Consequently, the 1UC-FeSe films epitaxially grown on this anatase TiO$_2$ possess the same in-plane lattice constant as TiO$_2$, 3.80 ± 0.05 Å, slightly larger than the value (3.76 Å) of bulk FeSe (figure 8(g)). This result suggests nearly strain-free FeSe films formed on anatase TiO$_2$(001), resembling those on graphene. Intriguingly, the 1UC-FeSe/TiO$_2$(001) exhibits U-shaped gaps of 21 meV in magnitude (figure 8(h)), while 2UC-FeSe/TiO$_2$(001) exhibits no superconductivity signature at all, resembling those on STO (001) and STO(110). Instead of single vortex observed on 1UC-FeSe/STO(001) (due to dense domain boundaries), periodic vortex lattice forms on 1UC-FeSe films on the TiO$_2$(001) substrates [64]. The above results demonstrate unambiguously the occurrence of high-$T_C$ superconductivity in 1UC-FeSe/TiO$_2$(001) and, hence, support our conclusion that strain is not crucial and that antiferromagnetic-interaction/spin-fluctuation does not play the dominant role as discussed in section 3.3. Considering the oxygen LO phonon that couples to FeSe electrons and contributes to the interface-enhanced superconductivity in 1UC-FeSe/STO (001) is composed of relative Ti and O atomic displacements (see section 3.3), we speculate that the case of 1UC-FeSe/TiO$_2$(001) holds the same mechanism. This is verified by a recent ARPES investigation of 1UC-FeSe films on rutile TiO$_2$(001) substrates, which reveals the coexistence of replica bands with an energy separation of 90 meV and superconductivity with a $T_C$ of 63 K [109]. The above finding echoes the essential role of oxygen phonons.

4. Summary and perspective

In this brief review, we have discussed the interface-enhanced superconductivity in Pb/Si, In/Si and FeSe/STO systems. For the FeSe/STO system, the compelling evidence of an enlarged superconducting gap (14–20 meV) and enhanced $T_C$ ( > 40 K) demonstrates that interface engineering provides a feasible way for rational design and preparation of high-$T_C$ superconductors. As proposed previously [31], by fabricating sandwiched heterostructure, for example, 2–3 UC FeSe sandwiched between STO [47] or TiO$_2$ [64, 109, 110] on both sides, much higher $T_C$ may be achieved.

We further demonstrate that interface charge transfer and interface-enhanced $e$–$ph$ coupling are essential to the enhanced superconductivity. A related calculation reveals that a $T_C$ of ~ 77 K is possible by a combination of three factors: high LO phonon energy, large $e$–$ph$ coupling constant and huge dielectric constant of the STO substrate, suppressing the Coulomb repulsion [89]. Most recently, a record high $T_C$ of 203 K was reported in H$_2$S under extreme pressure [111], which is a conventional BCS superconductor. Thus, the discovery of high-temperature superconductivity in 1UC-FeSe/STO and H$_2$S strongly suggests that high $T_C$ (well beyond the McMillan limit) is achievable under the BCS scenario.

In terms of the resemblance between the FeSe/STO interface and the built-in multi-interfaces in cuprates and iron pnictide superconductors, as shown in figure 1, we conjecture that both doping resulted charge transfer [112] (modulation-doping as in semiconductor heterostructure [24]) and $e$–$ph$ coupling contribute the high-temperature superconductivity in cuprates and iron-based pnictides and chalcogenides. The essential role of oxygen phonons, which has been evidenced in both the FeSe/STO system [54, 109] and cuprate [20, 21] definitely deserves to be revisited. For searching for high $T_C$ under the scheme of doping-charge-transfer and $e$–$ph$ coupling, we can follow the idea depicted in figure 1(c), e.g. fabricating heterostructures of metal and material that have high-energy phonon modes (such as diamond, BN, Al$_2$O$_3$, TiO$_2$, etc), or heterostructures involving Mott–Hubbard insulators that can be doped effectively with band-bending effect [112]. If ultra-thin or even monolayer Mott–Hubbard insulator film can be prepared on insulating substrate, electric field effect may be employed to tune it superconducting with high $T_C$. We can further fabricate superlattice of such heterostructures to achieve higher $T_C$, as in the case of the multi CuO-layer cuprates [9, 10]. Experiments in this direction are underway in our lab.

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