Supplementary Materials: Superconductivity of the $FeSe/SrTiO_3$ Interface in View of BCS-BEC Crossover

Shuyuan Zhang[1,2], Guangyao Miao[1,2], Jiaqi Guan[1,2], Xiaofeng Xu[1,2], Bing Liu[1,2], Fang Yang[1], Weihua Wang[1], Xuetao Zhu[1,2,3*], Jiandong Guo[1,2,3,4*]

[1] *Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

[2] *School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100049, China*

[3] *Songshan Lake Materials Laboratory, Dongguan, Guangdong 523808, China*

[4] *Beijing Academy of Quantum Information Sciences, West Bld.#3, No.10 Xibeiwang East Rd., Haidian District, Beijing 100193, China*

Corresponding author: xtzhu@iphy.ac.cn; jdguo@iphy.ac.cn

The following Supplementary Materials include the descriptions of the basic concept and parameters of BCS-BEC crossover, the sample preparations and characterizations, as well as the BCS-BEC parameters of different superconducting systems.

I. Parameters of the BCS-BEC phase diagram:

The phase diagram of interacting Fermi system as shown in Fig. 1 is scaled by a dimensionless parameter $1/(k_F a)$, where a is the scattering length charactering the pairing interaction [1]. In the BCS limit, the weak attraction has a negative scattering length, and $1/(k_F a) \rightarrow -\infty$. In the BEC limit, $1/(k_F a) \rightarrow +\infty$. The unitary point is defined by $1/(k_F a) = 0$ with the scattering length divergence. The BCS-BEC crossover is the regime within $|1/(k_F a)| \lesssim 1$. And the BCS-regime and BEC-regime are characterized by $1/(k_F a) \lesssim -1$ and $1/(k_F a) \gtrsim 1$, respectively [2].

In ultra-cold atom experiments, the parameter a can be tuned directly by the Feshbach resonance [3,4]. However, in condensed matter, the interaction is determined intrinsically and hard to be tuned. Thus in Fig. 1, we plot the phase diagram scaled by $k_F\xi_{pair}$ which is more apparent for condensed matter experiments. The relationship between $k_F\xi_{pair}$ [the horizontal axis in Fig.1(a)] and $1/(k_F a)$ was shown in Ref. [2]. For instance in the BCS limit, $k_F \xi_{pair} \sim e^{\pi/(2k_F|a|)}$ [5]. Clearly when $1/(k_F a) = 0$, $k_F\xi_{pair} = 1$.

The value of T_C/T_F and Δ/E_F are the functions of $k_F\xi_{pair}$ as schematically plotted in Fig. 1. As for the BCS-BEC crossover analyses of superconductors shown in Fig. 2, we use Δ/E_F, instead of $k_F\xi_{pair}$ or $1/(k_F a)$, due to the lack of accurate parameter $k_F\xi_{pair}$ or $1/(k_F a)$ in the published experimental data. In the BCS limit, $\Delta = \frac{8}{e^2} E_F e^{\frac{\pi}{2ak_F}}$; in the BEC limit, $\Delta = 4E_F / \sqrt{3\pi a k_F}$ [6]. There is a monotonic one-to-one mapping between the values of Δ/E_F and $1/(k_F a)$. In the BCS limit, $k_B T_C = \frac{8E_F e^{\gamma}}{\pi e^2} e^{\frac{\pi}{2ak_F}}$, where k_B is Boltzmann constant, γ is the Euler constant with $\frac{e^{\gamma}}{\pi} = 0.567$ [2]. Then we have $\frac{2\Delta}{k_B T_C} = 3.5$ in the BCS limit which corresponds to the BCS theory [7].

II. Sample Preparations and Characterizations

All the $SrTiO_3$ substrates used in this study were 0.5% Nb-doped $SrTiO_3$(001). The monolayer FeSe films were grown by the molecular beam epitaxy (MBE) method and were characterized by scanning tunneling microscopy. The 1uc-FeSe/STO samples were post-annealed at 470 °C for 6 hours in ultra-high vacuum to make the monolayer FeSe superconducting. The TCNQ molecules were evaporated onto the 1uc-FeSe/STO sample from a low-temperature evaporator at 390 K as described in Ref. [8].

The STM topographic images were acquired in constant-current mode with the bias voltage applied to the sample with respect to the tip. The STS were measured at 4.9 K with a bias modulation of 1 mV at 987.5 Hz. All the *dI/dV* spectra in the manuscript were normalized by a background defined by a polynomial function as shown in Fig. S1.

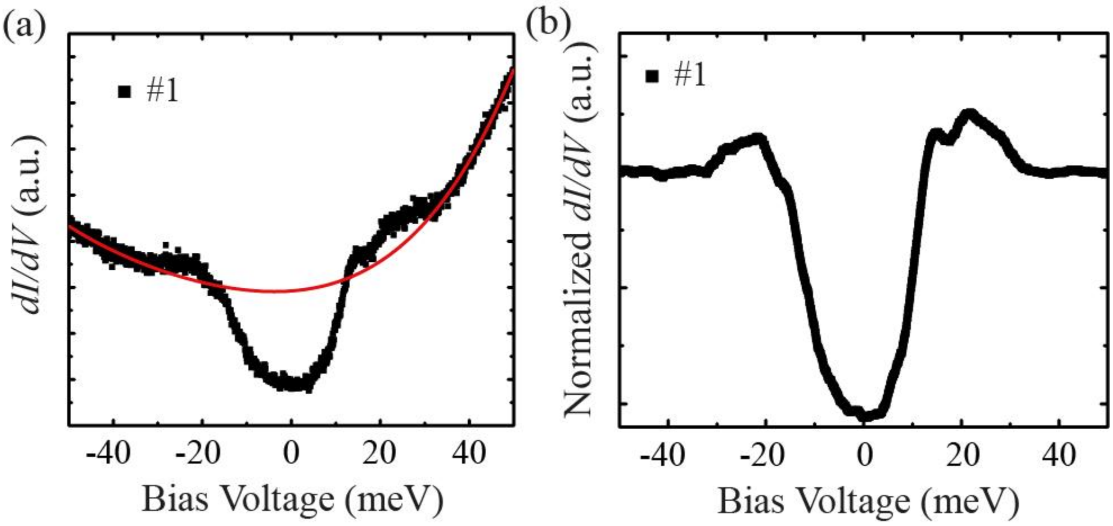

Figure S1| (a) Original *dI/dV* spectrum of 1uc-FeSe/STO at point #1 marked in Fig. 3. The background for normalization is fitted by a polynomial function and plotted in the red line. (b) Normalized *dI/dV* spectra at point #1 as shown in Fig. 3(c). The original *dI/dV* spectra are normalized by the red line in (a) and smoothed by the average of adjacent 20 points.

III. Parameters of different superconducting systems

Values of $k_F\xi_{pair}$, Δ/E_F and T_C/T_F presented in the manuscript were determined based on the existing independent experimental measurements. The pair size ξ_{pair} is roughly replaced by the phase coherent length ξ_{phase}, because in BCS regime the two length is only differed by a factor $3/\sqrt{2}$ [2]. The parameters of k_F, Δ, E_F, and T_C of 1uc-FeSe/STO were extracted from band structure measured by angle-resolved photoemission spectroscopy [9]. The parameters of other superconducting systems were extracted from references as listed in Table SI.

Table SI. Values of $k_F\xi_{pair}$, Δ/E_F and T_C/T_F for different superconducting systems.

Systems and References.	k_F (1/Å)	ξ_{pair} (Å)	$k_F\xi_{pair}$	Δ(meV)	E_F (meV)	Δ/E_F	T_C (K)	T_F (K)	T_C/T_F
FeSe bulk [10,11]	0.065	50	3.25	2.5	2 - 3	0.83 - 1	8	23 - 35	0.229-0.348
FeSe bulk [10,11]	0.153	50	7.65	3.5	10.0	0.30	8	116	0.069
(LiFe)OHFeSe [12,13]	0.280	33	9.24	10.5	43.0	0.24	40	449	0.080
$k_{0.8}FeSe$ [14]	-	-	-	10.3	60.0	0.17	32	696	0.046
1uc-FeSe/STO	0.208	20[15]	4.20	20.0	56.0	0.36	65	650	0.100
1uc FeSe/STO under-annealed [16]	-	-	-	10.0	48.3	0.21	40	561	0.071
	-	-	-	14.0	50.6	0.28	55	587	0.094
	-	-	-	16.0	55.7	0.29	60	646	0.093
	-	-	-	19.0	54.9	0.35	65	637	0.102
1uc-FeSe/BTO [17]	-	-	-	17.0	63.0	0.27	70	731	0.096
	-	-	-	19.5	63.0	0.31	75	731	0.103
1uc-FeSe/TiO_2	-	-	-	14.6	45.0	0.32	63	522	0.121

[18]									
Cuprates 123 [19-21]	-	-	-	-	-	-	54	1232	0.044
	-	-	-	-	-	-	62	1325	0.047
	-	-	-	-	-	-	69	1478	0.047
	-	-	-	-	-	-	75	1740	0.043
	-	-	-	-	-	-	90	1977	0.045
	-	-	-	-	-	-	87	2204	0.040
	-	-	-	-	-	-	79	2741	0.029
	-	-	-	-	-	-	91	2947	0.031
	-	-	-	-	-	-	91	3170	0.029
Cuprates 2223 [19-21]	-	-	-	-	-	-	110	2413	0.046
	-	-	-	-	-	-	126	3112	0.040
	-	-	-	-	-	-	108	3228	0.034
Cuprates 214 [19-21]	-	-	-	-	-	-	36	922	0.039
	-	-	-	-	-	-	30	812	0.036
	-	-	-	-	-	-	40	1561	0.026
	-	-	-	-	-	-	30	1709	0.018
	-	-	-	-	-	-	28	1709	0.016
$CeRhIn_5$ [22]	-	-	-	-	-	-	-	-	0.016
Ce_2PdIn_8 [22]	-	-	-	-	-	-	-	-	0.009
UPt_3 [22]	-	-	-	-	-	-	-	-	0.008
Ce_2CoIn_8 [22]	-	-	-	-	-	-	-	-	0.003
$NbSe_2$ [21,23]	-	-	-	-	-	-	7	802	0.009
Nb [21,23]	-	-	-	-	-	-	10	12607	$7.62E^{-04}$

Sn [21,23]	-	-	-	-	-	-	4	115638	$3.26E^{-05}$
Zn [21,23]	-	-	-	-	-	-	1	109496	$7.98E^{-06}$
Al [21,23]	-	-	-	-	-	-	1	133721	$8.56E^{-06}$

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