

Direct Imaging of Electron Transfer and Its Influence on Superconducting Pairing at FeSe/SrTiO₃ Interface

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The interplay among various degrees of freedom at interfaces of a complex system could lead to an exotic and emergent state of matter. Recent experiments on 1 unit-cell (UC) thick FeSe films grown on SrTiO₃ (STO) substrate suggest that interface effects can be used as a means to enhance superconducting critical temperature T_c of the material to ~ 77 K [1], or even to above 100K when grown on doped STO [2]. This is ten times the T_c of bulk FeSe of 9.4K [3]. Nevertheless, the exact mechanism responsible for the significant T_c enhancement of the monolayer FeSe on STO over that of bulk FeSe remains a mystery.

To understand the T_c enhancement mechanism we carried out complementary electrical transport and low-temperature atomic imaging and EELS measurements on the same FeSe films of different thicknesses (1UC, 2UC, 8UC and 14 UC) on STO (Fig.1). HAADF reveals an additional Se layer residing between the FeSe film and the TiO_x-terminated STO substrate (Fig.1A,B). Atomically resolved EELS mapping across the FeSe/STO interface (Fig.1 C,D) both in core-loss and low-loss modes at room temperature (Fig. 1C, D) and at 10K (Fig. 2 A-F) shows clear and direct evidence of electrons transferred from STO to the first two atomic layers of FeSe films below T_c . Due to the interfacial band bending effect (Fig.2H-J), the transferred electrons are accumulated within the first two UC of FeSe away from the FeSe-STO interface, irrespective of the total thickness of the FeSe film. Quantitative EELS measurements also reveal a blue shift in Fe $L_{3,2}$ peaks. Green's function based FEFF simulations indicate the blue shift is not originated from the local chemical environment of FeSe as no shift was observed, even for the Fe ions at the 1st UC (Fig.1G). The strain effect can also be excluded since FeSe is experiencing tensile stress near STO, which would result in a red shift instead. Our analysis suggests that the screening potential in first two FeSe layers near the interface, induced by transferred electrons from the STO substrate to the FeSe layers, is responsible for the blue shift.

Our transport measurements confirm that the T_c of the 1UC films can be enhanced by thermal annealing which introduces electrons into the film and also by gating from the back surface of STO with a positive potential (Fig.1E). We also found backgating is particularly effective in enhancing the T_c without changing significantly the carrier density of the films. This lends support to the model of electron-phonon coupling across interface as the mechanism for high T_c since the positive potential tends to 'pull' the interfacial electrons closer to the high Debye temperature STO phonon bath. We note that the electron-electron interaction effect should also be strengthened by gating in such an interfacial two-dimensional electron gas (2DEG) as compared to bulk carriers due to the lowering of its dimension [4]. Our new technique of atomically resolved EELS mapping across the FeSe/SrTiO₃ interface near liquid helium temperature can be a powerful tool for the study of other emergent quantum states near interfaces and 2D systems [5].

References:

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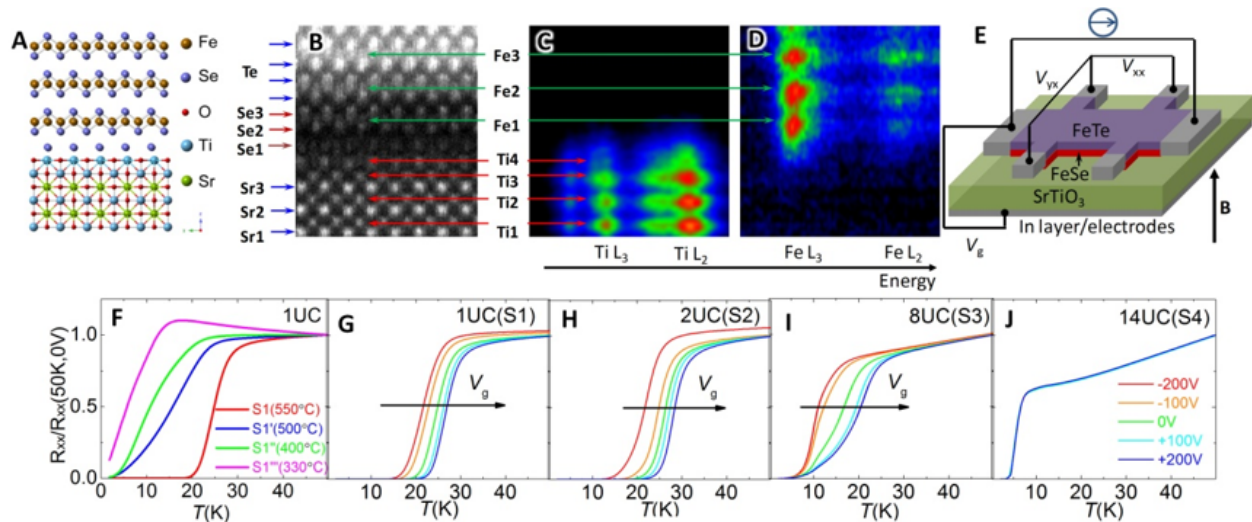


Figure 1. Superconducting FeSe films on SrTiO₃ (STO) substrates. (A) Super-cell of FeSe on top of STO (001), inferred from combined HAADF image and EELS data. (B) The HAADF-STEM image of the 1UC FeSe film on STO with FeTe capping layers. (C) and (D) The integrated Ti L_{3,2} and Fe L_{3,2} EELS after subtracting background in false color with increasing intensity in the black-blue-green-red sequence. (E) Schematics of the gate tuned six-terminal Hall-bar device of FeSe films on STO with FeTe capping layer. For clarity, Te film on FeTe is not shown. (F) Normalized R_{xx} vs. T for 1UC films annealed at different temperatures for 2h post MBE growth: S1(550°C), S1'(500°C), S1''(400°C) and S1'''(330°C). (G-J) Normalized R_{xx} vs. T at various backgating voltage V_g for the S1(1UC)(G), S2(2UC)(H), S3(8UC)(I) and S4(14UC)(J) films under the optimal annealing condition at 550°C.

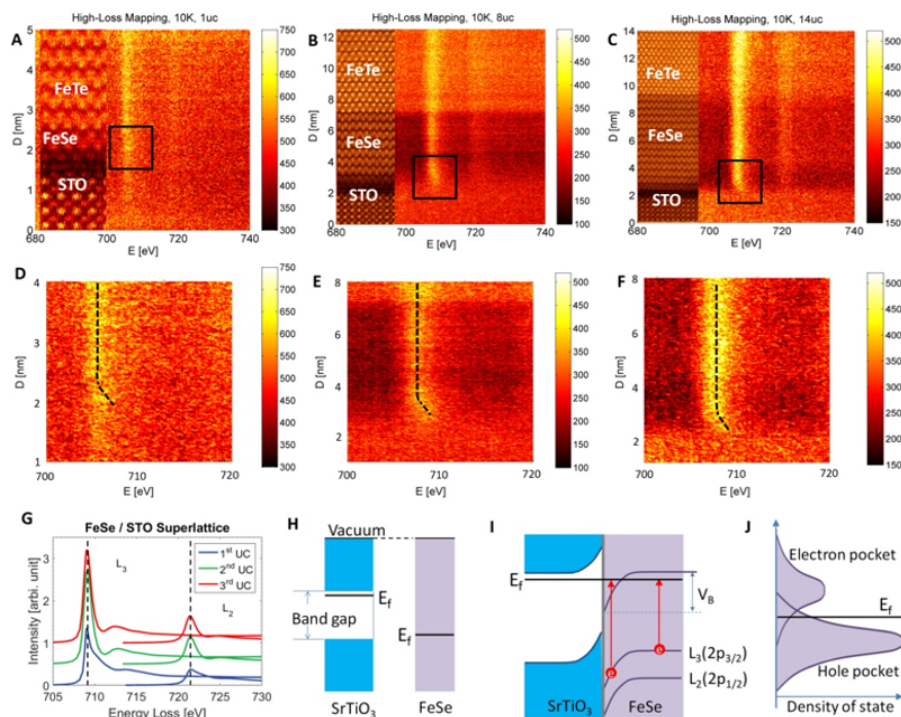


Figure 2. Atomically resolved STEM-EELS results at 10K. (A-C) Core-loss EELS mapping of Fe-L_{3,2} with an energy range between 680 and 740eV for 1UC (A), 8UC (B) and 14UC (C). (D-F) The zoom-in images at the interface region in (A-C), respectively, revealing a clear blue-shift. The dash lines are guides for eyes. (G) FEFF simulation of the core-loss EELS spectra using the supercell in Fig. 1A. (H) Schematics of work function difference between STO and FeSe. (I) Schematics of screening potential profiles in the FeSe region induced by electron transfer from the proximal STO interface. Due to the finite screening length, Fe's L₃ (2p_{3/2}) and L₂ (2p_{1/2}) levels close to the interface bend accordingly, giving a blue-shift of the electron energy loss. V_B is total potential variation. (J) At the interface, density of states of hole-pocket is higher than that of electron-pocket. It needs more electrons to fill up the hole-pocket as compared with electron-pocket for the same E_F shift.