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High-temperature superconductivity: a conventional conundrum

High-temperature superconductivity in ultrathin films of iron selenide deposited on strontium titanate has been attributed to various exotic mechanisms. New experiments indicate that it may be conventional, with broader implications.

Ivan Božović

In the Condensed Matter Physics community, one of the most talked-about events in the last few years was the discovery of high-temperature superconductivity (HTS) in one unit cell (1UC) thick films of iron selenide (FeSe) deposited on single-crystal strontium titanate (STO) substrates. It has been attributed to a so-far unseen mechanism, enhancement of the electron pairing in one material by their interaction with some excitations in another material, across the interface. This has been hailed as a path to HTS in many new (combinations of) materials, even opening the way to room temperature (RTS). However, the new experiments of Shiogai and colleagues¹, reported on p. xxx in this issue, point to a more mundane explanation.

In bulk FeSe samples superconductivity appears below a pretty low critical temperature, $T_c = 8$ K. However, in 1UC FeSe/STO samples, resistivity starts dropping² at about 40-50 K. This is accompanied by the second telltale signature of the true superconducting state, expulsion of the magnetic field — the so-called Meissner effect. Scanning tunneling microscopy (STM) provided the third piece of evidence, a gap in the electronic spectrum — interestingly, larger (21 meV) and persisting up to a higher temperature (about 65 K) than expected, which was taken as an indication that T_c could be even higher². Angle-resolved photoemission spectroscopy (ARPES) confirmed these findings, moreover revealing an isotropic gap around a simple, circular Fermi surface³⁻⁵. In contrast, bulk FeSe samples and films thicker than 1 UC alike feature multiple electron bands and Fermi surfaces.

One technical difficulty is that superconductivity in 1UC FeSe is spoiled as soon as the film is exposed to air, or covered by any material tested so far, even including additional layers of FeSe itself. To meet this challenge, scientists have recently built a number of advanced synthesis/characterization systems in which the sample can be shuttled between modules dedicated to Molecular Beam Epitaxy (MBE) synthesis, ARPES and/or STM, respectively, without breaking the vacuum. One system, built in the group of Jingfeng Jia in Shanghai, combines MBE growth with in-situ four-probe measurements of electrical resistance. Leveraging on this technical advance, Jia and colleagues reported $T_c = 109$ K, the record in 1UC FeSe/STO, albeit only at isolated locations on the film surface⁶.

These experimental discoveries have triggered a great interest⁷ and a race among theorists. Most proposed that HTS originates in enhancement of the pairing interaction across the interface, but there is no consensus on whether this is due to the high value of the static dielectric permittivity of STO, a strong electron-phonon interaction between the electrons in FeSe and the high-frequency phonons in STO, the presence of soft phonons in STO, or something else, e.g., spin fluctuations.

Now, enter Jun-ichi Shiogai and collaborators at Tohoku University in Japan with an unlikely experimental approach. They used a very simple synthesis technique, pulsed laser deposition, and grew thicker and somewhat inhomogeneous FeSe films on STO and MgO substrates. Then they covered the film with an ionic liquid and applied a gate voltage across it. At moderate temperatures and gate voltages, this induced charge transfer and doped the film with electrons, while at higher T and V_G the film was progressively etched away. By tuning these parameters and timing the process, Shiogai and colleagues control

the film thickness, resistance, and T_c . They have observed resistivity drops starting at $T_c = 40$ -50 K even in films up to about 20 UC thick. Moreover, they see the same T_c in FeSe films grown on MgO, a material very different from STO; e.g., in MgO there are no soft phonons, the dielectric permittivity is low, etc.

These new results cast doubt on explanations that invoke exotic interfacial interactions between FeSe and STO. Rather, they point to a simple picture in which the primary factor is the carrier density – HTS occurs in a single FeSe layer or in many layers as long as they are optimally doped.

What makes this venture so exciting? Read between the lines. For the last three decades, the most important open problem in condensed matter physics has been the origin of HTS in cuprates⁸. Indeed, at least 14 Nobel laureates, plus innumerable mortals, have put forward their theories and ‘scenarios’, mostly unconventional and exotic, and contradicting one another fiercely⁹ – inspiring the sobriquet “the Battle of (Physics) Titans”. When HTS was also discovered in iron-based compounds seven years back, every camp rejoiced, hoping that as soon as we pinpoint what they have in common with the cuprates, the scales will tip (their way). But this has not happened — so far ‘ironics’ have been befuddled with as much controversy as the cuprates.

This impasse may be broken now. Given the simple circular Fermi surface and an isotropic gap, 1UC FeSe/STO looks quite conventional – and yet reportedly capable of reaching $T_c = 109$ K. And as of this year, the HTS family got yet another, record-breaking member, H_3S , claimed to reach (under enormous pressure of 1.5 million atmospheres) $T_c = 203$ K while also being conventional¹⁰. Assuming that both claims survive the scrutiny, the current picture may appear as in Fig. 1: all known superconductors conform to one well-understood theory (originally put forward by Bardeen, Cooper, and Schrieffer, hence ‘BCS’) – except for cuprates somewhere in the middle of the T_c spectrum and some but not all of iron-based superconductors. This looks very weird, indeed. One would rather hope that a single unifying theory should be able to fit them all. [For the record, I doubt that the simple BCS model will do.]

What is ahead of us, experimentally? The paper of Shiogai *et al.*¹ certainly calls for some follow up. First, we need better (read MBE grown), atomically smooth films. Next, we should elucidate whether the actual mechanism of doping FeSe by electrolyte gating is purely electronic or (my guess) ion intercalation. Certainly some information on the electron band structure and the gap in gated FeSe samples would be most valuable, but regrettably ARPES and STM are impeded once the film surface is submersed under ionic liquid; we need some new, *operando* probes. Fortunately, magnetic (e.g., two-coil inductance) measurements are feasible and could be used to demonstrate the Meissner effect. Note that so far only one such measurement has been reported on 1UC FeSe/STO, and it showed just few per cent of the expected signal; however, this was done *ex situ* and the film was covered (with FeTe and Si). Hopefully, a larger signal could be observed *in situ*, on a pristine film surface, but this calls for some additional hardware development, since there is no such setup in operation. However, the experiment of Shiogai *et al.* opens an easier route, measuring mutual inductance *ex situ* on a gated thicker film. In fact, such *operando* inductance measurements have been already reported on electrolyte-gated cuprates. Next, while several groups have reproduced the lower $T_c \approx 50$ K, we are still awaiting independent confirmation of the higher $T_c > 100$ K and reconciliation with STM, ARPES and *in-situ* magnetic data. Last but not least, we need to find out whether *that* is exclusive to 1UC FeSe/STO — with obvious implications.

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Fig. 1. The (bizzare) current superconductivity road map. Most superconductors are believed to conform to the standard (weak-coupling, electron-phonon) BCS theory, including some materials with the $T_c > 100$ K (1 UC FeSe/STO) and even the record $T_c > 200$ K (H₃S under 150 GPa pressure) — with the exception of cuprates which are generally believed to be quite exotic. Opinions on iron pnictides are divided but the majority seems to believe that they are strongly-correlated, i.e., non-BCS (albeit different from cuprates) — and yet, FeSe/STO again looks BCS-like. Personally, I find this picture untenable.