Physics

VIEWPOINT

A Tale of Two Domes

Iron selenide films peppered with potassium atoms exhibit a high-temperature superconducting phase that emerges separately from a low-temperature superconducting phase.

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Superconductivity describes the spectacular ability of electrons in some materials at low temperatures to form Cooper pairs and coherently carry charge without resistance. Creating superconductors that operate at room temperature has long been an unrealized dream. Of particular interest are copper- and iron-based superconductors, discovered in 1986 and 2008, respectively, that possess higher transition temperatures (T_c up to 135 K under ambient pressure) than most conventional superconductors. These "high-T_c" superconductors often exhibit a peak or "dome" behavior in the transition temperature: when doping or pressure is increased, T_c rises until it reaches a maximum, and then it falls off. A new study of superconducting iron selenide (FeSe) films has revealed a double-dome behavior as the doping of electrons is increased [1]. Can-Li Song and collaborators at Tsinghua University, China, argue that the two domes arise from distinct mechanisms for binding electrons together into Cooper pairs. The unexpected discovery strengthens recent suggestions that the conventional mechanism of phonon binding, which has, for three decades, been overshadowed by more exotic mechanisms, may yet have an important role to play in further enhancing T_c.

This is not the first surprise to come from FeSe. Until recently, FeSe was a superconductor, with a modest T_c of around 8 K. In 2012, however, researchers found that a single unit cell layer ("monolayer") of FeSe grown on strontium titanate ($SrTiO_3$ or STO) exhibits high-temperature superconductivity [2], with T_c skyrocketing to 109 K [3]. This boost appears localized near the interface, as, strangely, a second layer of FeSe deposited on top exhibits semiconductor rather than superconductor behavior.

In their new study, Song *et al.* wanted to understand the giant enhancement of superconductivity in monolayer FeSe/STO. However, STO likely introduces multiple effects: strain from lattice mismatch, electron doping, and cross-interface coupling to phonons [4]. Furthermore, STO is prone to oxygen deficiency and numerous surface reconstructions. Using a reductionist approach, Song *et al.* examined the role of electron doping in isolation. First, they eliminated the effect of strain by growing thin films of FeSe on a graphitized silicon carbide (SiC) substrate, which interacts only weakly via van der Waals forces with the FeSe film. Second, they reproduced the charge transfer from STO by depositing potassium (K) atoms on top of the films. Like STO, potassium has the effect of electron doping—with the advantage that the number of K atoms donating electrons can be controlled and precisely counted.

Using scanning tunneling microscopy, Song *et al.* observed the gap in the density of states, whose width is a

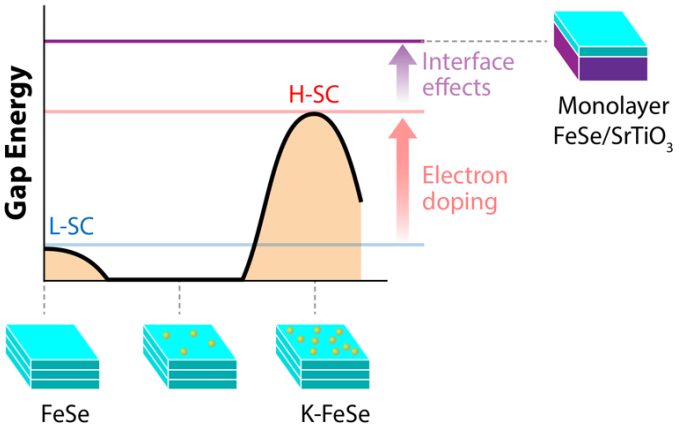

Figure 1: Schematic diagram illustrating two domes of superconductivity in FeSe. By adding potassium (K) atoms to thin films of FeSe, researchers were able to distinguish two separate superconducting phases, labeled L-SC and H-SC. The gap energy, which serves as a proxy for the superconducting transition temperature, is an order of magnitude higher in K-doped H-SC vs undoped L-SC. However, a monolayer of FeSe on $SrTiO_3$ (STO) has a gap energy even higher than H-SC. This suggests that electron doping plays a primary role in boosting superconductivity in FeSe/STO, but that additional interface effects, such as phonon coupling, further enhance the superconductivity. (APS/Alan Stonebraker)

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measure of the binding energy between Cooper pairs. The gap energy typically scales with the transition temperature, so it acts as a proxy for T_c. As the researchers increased the electron doping by adding more K atoms to the surface, they observed two widely separated domes in the gap energy. The first dome was associated with the superconducting phase of undoped FeSe (called "L-SC" by the authors), which was rapidly suppressed by a small amount of electron doping. For intermediate doping, the film's superconductivity was lost, only to be regained upon further doping, where a new superconducting phase (called "H-SC") emerged (see Fig. 1).

The maximum observed gap energy in the H-SC phase is 14 meV, 7 times larger than that of the L-SC phase (2 meV), and just shy of the value seen in monolayer FeSe/STO (up to 20 meV [2]). This comparison suggests that the T_c enhancement in monolayer FeSe/STO is due primarily to electron doping, which is likely driven by oxygen vacancies in STO. The remaining portion of T_c enhancement could arise from interface effects (see Fig. 1), such as coupling to a high-energy phonon mode in STO [4]. These results complement recent photoemission experiments demonstrating similar T_c enhancements induced by K deposition, in FeSe/STO films [5] and bulk FeSe crystals [6].

For H-SC, Song *et al.* additionally observed that the Cooper-pair binding energy was spatially homogeneous, despite a disordered distribution of K atoms on the surface. The implications here are crucial. Anderson's theorem states that a conventional superconductor, where electrons are bound together by phonons, should be robust against the disorder of nonmagnetic impurities like K. Not only is this property useful in allowing superconductivity to survive in dirty materials, it can also be turned around into a litmus test for the mechanism of superconductivity. Using the converse of Anderson's theorem, Song *et al.* claimed that H-SC's insensitivity to disorder implies conventional, phononic pairing.

Song *et al.*'s implication that FeSe films attain high T_c through the conventional phononic interactions places FeSe in a special category and challenges the prevailing belief that high T_c *requires* an unconventional mechanism [7]. Unconventional mechanisms such as magnetic fluctuations have found favor for several reasons. First, magnetic order often persists to high temperatures, implying a large underlying energy scale. Moreover, magnetic fluctuations flip the sign of the electron wave function, allowing larger separation between the paired electrons, which reduces the deleterious effects of Coulomb repulsion. In fact, all copper-based and most other iron-based superconductors exhibit unconventionality. Even the material KFe_2As_2, which was also observed to have two distinct superconducting phases [8], exhibits an unconventional electron pairing mechanism in both phases.

Despite the excitement of phononic high-T_c superconductivity in FeSe, some caveats remain. First, beyond an optimal electron carrier concentration, the H-SC gap begins to diminish (this trend is also seen in Refs. [5, 6]). This dome evolution has been a hallmark of unconventional superconductors and is harder to explain with a conventional phonon scenario. Second, Song *et al.*'s use of the converse of Anderson's theorem is not logically fool-proof. The H-SC insensitivity to K atom disorder could be attributed simply to the fact that the K atoms sit atop instead of within the FeSe, which reduces their ability to break electron pairs of any type. Finally, an even more exotic unconventional mechanism has been proposed—incipient sign-changing $s_{\pm}$ gap symmetry—where bands with no Fermi surface still participate in pairing and perhaps provide robustness against disorder in violation of Anderson's theorem [9]. In the latter proposal, phononic and magnetic interactions work cooperatively to boost T_c in FeSe in a "best-of-both-worlds" scenario.

Beyond these caveats, Song's argument for phononic pairing in K-doped FeSe fits well with recent work suggesting the same for monolayer FeSe/STO [10]. Given these observations in FeSe, along with the recent discovery of phononic superconductivity up to 203 K in pressurized sulfur hydride (H_3S) [11], is it time to revisit our ideas of what it will take to reach room-temperature superconductivity? More surprises may be in store from FeSe, and lessons learned from this material may shape the future of superconductor searches more broadly.

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