Science Bulletin 68 (2023) 851–853

Contents lists available at ScienceDirect

Science Bulletin

journal homepage: www.elsevier.com/locate/scib

News & Views

Superconductivity in a strange metal

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In a gas of charged particles with the density n, mass m, and charge e, the electrical conductivity σ is given by the Drude formula: $\sigma = ne^2/m\Gamma$, where Γ is the scattering rate. Standard metals are well described by Landau's Fermi Liquid (FL) theory, in which the electric current is carried by quasi-particles, low-energy excitations of the FL that resemble electrons with some effective mass m^*. The scattering rate $\Gamma = 1/\tau = v_F/l$, where τ is the (momentum) relaxation) time, l is the mean-free path, and v_F is the Fermi velocity, can be expressed ("Matthiessen's Rule") as a sum of contributions from various scattering channels: $\Gamma = \Gamma_0 + \Gamma_{el\text{-}el} + \Gamma_{el\text{-}ph} + \ldots$, where Γ_0 describes scattering on lattice imperfections, $\Gamma_{el\text{-}el}$ the electron–electron scattering, $\Gamma_{el\text{-}ph}$ the electron–phonon scattering, etc. Of these, $\Gamma_0 = v_F/l_0$, where l_0 is the average distance between the defects, is temperature-independent. $\Gamma_{el\text{-}el}$ should scale as T^2: an electron with energy $\varepsilon \sim k_BT$ above E_F, where k_B is the Boltzmann constant and E_F is the Fermi energy, can only scatter on another electron with energy not more than ε below E_F into a state not more than ε above E_F, because of Fermi statistics and Pauli exclusion. $\Gamma_{el\text{-}ph}$ typically grows as T^5, so we expect this to overwhelm the other channels at a high enough T. But since l cannot be shorter than the distance between the atoms, the total Γ saturates at Mott-Ioffe-Regel (MIR) limit, roughly v_F/a_0, where v_F is the Fermi velocity and a_0 is the lattice constant. The resistivity should also saturate at low T, and $\rho_0 = m^*v_F/ne^2l_0$. FL theory also describes other electronic properties; e.g., it predicts that in the magnetic field B, the resistivity of the metal should increase with B^2, because $\sigma(B) = \sigma(B = 0)/(1 + (\omega_c/\Gamma)^2)$, where $\omega_c = eB/m^*$.

The discovery of high-temperature superconductivity (HTS) in cuprates caused a tectonic shock to the Condensed Matter Physics community and beyond because it seemed impossible to reconcile the HTS with the standard textbook description of superconductivity, Bardeen-Cooper-Schrieffer (BCS) theory. It was soon found that the so-called "normal" state above the critical temperature (T_c) is all but standard. As illustrated in Fig. 1a, in $La_{1.81}Sr_{0.19}CuO_4$ the $\rho(T)$ dependence is strictly linear [1]. In some cases, this extends up to 1000 K [2], and in others, down to a few tens of mK [3–6], without the expected saturation at either end, violating both FL-theory limits. Moreover, at high fields, the magnetoresistance (MR) scales linearly with B (Fig. 1b) [1].

For these reasons, cuprates have been called "strange metals", the moniker reflecting our frustration with the inability to explain this simple behavior with no energy scale other than T. In truth, many explanations were proposed — scattering on quantum-critical fluctuations of some order parameter (several candidates were put forward), Planckian dissipation, Sachdev-Yang-Kitaev model, Marginal Fermi Liquid (MFL) theory, mapping onto gravity, and more [3–6] — but so far with no clear winner. New experiments are needed to discriminate among them.

An obvious line of attack is to systematically study the doping dependence of $\rho(T, B)$ in a given family of cuprates, such as $La_{2-x}Sr_xCuO_4$ (LSCO), and then to examine other unconventional superconductors and other strange metals. The standard approach is to synthesize and study a set of samples with different chemical doping levels. A better way is to systematically vary the doping level in a single sample, e.g., by using a combinatorial-spread film synthesis, whereby all "pixels" undergo the same thermal and processing history, reducing the number of (uncontrolled) variables, and improving the (relative) accuracy of doping steps [7,8]. Quantitative characterization of the strange-metal behavior in electron-doped $La_{2-x}Ce_xCuO_4$ (LCCO) was accomplished using a combinatorial library covering the entire overdoped side [8]. The relation $\rho(T) = \rho_0 + AT$ was found to hold at temperatures right above T_c, at all doping levels. The slope A scaled with T_c^2, pointing to a link between the strange metal behavior and the superconductivity. The question is whether this holds in other unconventional superconductors, e.g., Fe-based superconductors.

To fine-tune the doping level, another powerful approach is the electrolyte-gating of a thin film. The principle is simple: run the current through a sample and measure the voltage. Cover the film with a suitable liquid electrolyte, add a gate electrode, and apply a gate voltage V_g (typically up to a couple of Volts). This modifies the carrier density and hence the $\rho(T, B)$ in the sample. However, the actual microscopic mechanism of doping via electrolyte gating has been a matter of some controversy. In somewhat simplistic terms, the question is whether it works like a transistor or a battery. In the field-effect transistor (FET), the electric field modifies the carrier density in the semiconductor channel and thus, the device resistance. In principle, one could gate metals as well, but given that the screening length is just 1–2 Å, the electric field penetrates only an extremely thin layer near the surface, and the overall change in film resistance may be too small to detect. Similarly,

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https://doi.org/10.1016/j.scib.2023.03.038

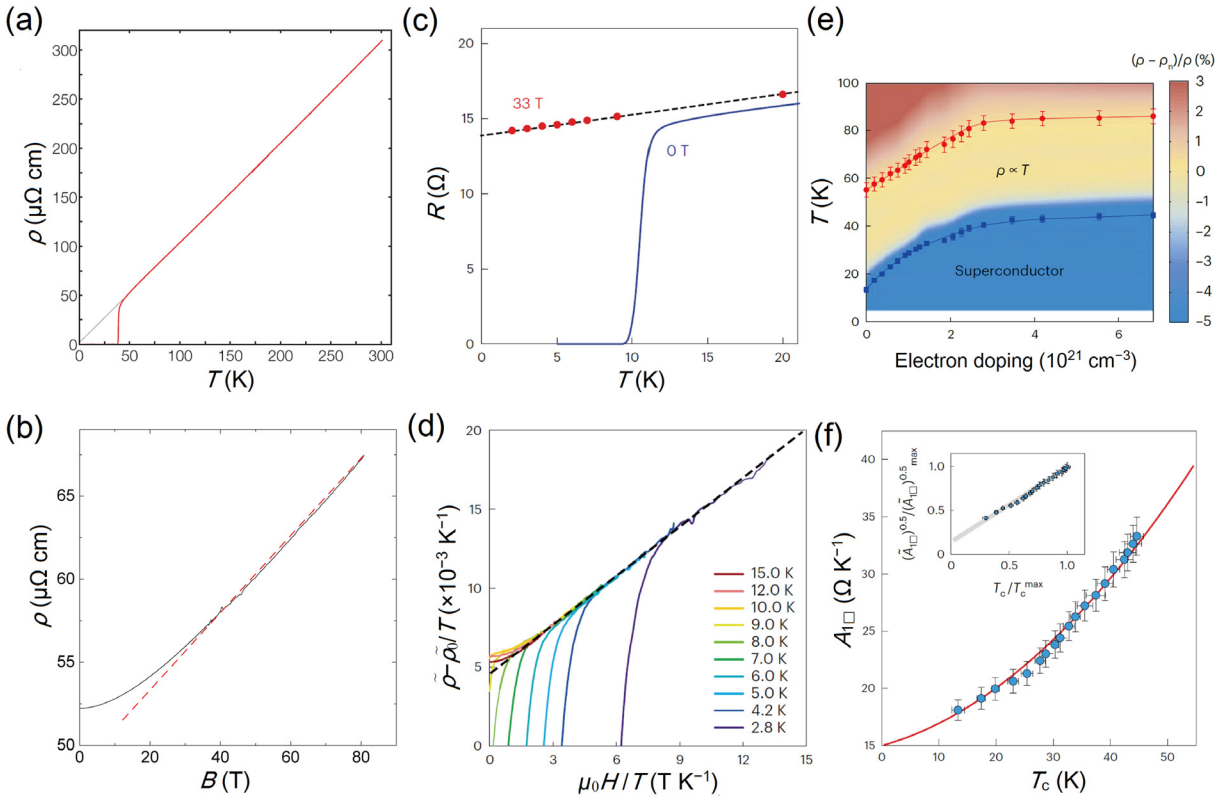

Fig. 1. (Color online) The strange metal behavior in LSCO and FeSe. (a) For LSCO (x = 0.19), the resistivity $\rho(T)$ is linear in T, and (b) the high-field magneto-resistivity $\rho(B)$ is linear in B. (c) For FeSe, after the suppression of superconductivity by the applied magnetic field, the resistivity $\rho(T)$ under 33 T field is linear in T. (d) The magneto-resistivity $\rho(B)$ of FeSe at different temperatures is proportional to $1 + \beta(\mu_0 H/T)$ where β is a constant. (e) For FeSe, the superconducting state is encompassed by the strange metal state, so superconductivity emerges out of the strange metal. (f) The slope A of the linear $\rho(T)$ shows parabolic dependence on T_c. Figures are reproduced from Refs. [1] and [13].

the changes in T_c in standard superconductors are also minute, if any.

A chemist's ingenuity worked a way around this hurdle. Instead of a solid gate dielectric, McDevitt and coworkers [9] used an ionic liquid electrolyte and were able to tune T_c in $YBa_2Cu_3O_7$ over a remarkably broad range (by 50 K). The narrative at that time was that cations (or anions) are driven toward the metal channel and crowd near the surface, attracting extra electrons (or holes) as the image charge at the metal surface. Within this "Helmholtz double layer", the local field can exceed 10^8 V/cm, inducing a surface charge density of up to 10^{14}–10^{15} electrons/cm^2. Given the screening length of ~5 Å in cuprates, this effect should be substantial only in ultrathin and atomically smooth films [10]. However, experiments showed that the carrier density and T_c were modified in much thicker layers, up to 1000 Å and even more. That prompted a different model: electrochemical doping. The field-driven ions penetrate the entire volume of the cuprate channel, saturating at the density proportional to V_g. This was demonstrated by *operando* experiments, gating the samples while they were characterized by spectroscopy, structural, and transport measurements. Parkin's group [11] showed that when VO_2 is electrolyte-gated, oxygen anions move in and out, depending on the field polarity. Our group showed that H^+ (proton) cations dope WO_3 [12]. When an ionic liquid is exposed to air, it absorbs some water molecules, which ionize into $(OH)^-$ and H^+ (the ionic liquid is indeed a polar solvent!). And H^+ is just a proton, so it has no steric hindrances to penetrate any material.

The above background sets the stage for the study by Jiang et al. [13] highlighted here. Using pulsed-laser deposition, they synthesized 200 nm thick films of FeSe. Using atomic force microscopy, they found the root mean square (RMS) of surface roughness was several nm. This detail is essential since it implies that one could not expect a purely electrostatic, FET-like effect to modify or induce HTS in more than one or two top FeSe layers, and since the surface is not smooth, the thin top HTS layer would be disconnected. Jiang et al. [13] patterned these FeSe films into transport devices and measured the dependence of resistivity on T, B, and V_g. As they varied V_g, they observed dramatic changes in the film's superconducting properties; T_c increased from 10 to 45 K. Moreover, by observing the Meissner effect via the two-coil mutual-inductance technique *in situ*, they could fine-tune T_c in a controllable fashion. In this way, they collected a dense $\rho(T, B, V_g)$ data set, allowing a detailed analysis.

The fact that T_c changed, apparently homogeneously, in the 200 nm thick film indicated that electrochemical insertion and removal of some ionic species occurs throughout the entire FeSe film volume. Using the tomographic measurement of the chemical composition in the film by secondary ion mass spectroscopy (SIMS) — a new trick in this field — Jiang et al. proved that H^+ intercalated in FeSe. The main findings are illustrated in Fig. 1c and d. When the magnetic field suppresses superconductivity, the T-dependence remains linear (Fig. 1c) down to the lowest temperature accessible, 2 K. The B-dependence is also linear (Fig. 1d) in fields up to 50 T. FeSe is thus a canonical example of a strange metal.

Interestingly, these $\rho(T, B)$ data can be scaled to a simpler, single-variable dependence. Let us introduce two dimensionless variables, $u \equiv k_B T/\mu_B B$, and $F \equiv ne^2\hbar\Delta\rho/(m^* k_B T)$. Then, as shown in Fig. 1d, $F(u) = \alpha + \beta u$, where α and β are numerical constants of

the order of unity. Generally, the scaling with fewer variables implies some hidden symmetry in the data — in this case, between the roles of T and B. Given that $\hbar\Gamma = \alpha k_B T + \beta\mu_B B$, the resistivity is controlled by the sum of thermal and magnetic field energies. We note that this is different from the scaling in $BaFe_2(As_xP_{1-x})_2$ reported by Analytis and coworkers [14], $\hbar\Gamma = [\alpha(k_BT)^2 + \beta(\mu_BB)^2]^{1/2}$. A new physical picture may be needed, not yet clear but apparently challenging Landau's FL theory.

Jiang et al. [13] mapped out a detailed doping dependence of magnetoresistance, strange metal behavior, and superconductivity in FeSe (Fig. 1e). A caveat is that the carrier density was estimated roughly, in Faraday's manner, by integrating the charging current. However, T_c was measured accurately and could be used as a proxy. The critical observation is that the strange-metal behavior is seen in a large portion of the T-B-doping phase diagram that wraps around the superconducting dome. At all doping levels, HTS emerges out of the strange-metal state.

Going further, Jiang et al. [13] fit the linear part as $\rho = \rho_0 + AT$, where $A \equiv d\rho/dT$, and check how the slope evolves with doping. As shown in Fig. 1f, they find a parabolic dependence, $A = A_0 + CT_c^2$, with some offset A_0. What this means is a challenge to theory. But good experiments frequently answer one question while opening new ones; this work is not an exception. In search for answers, Jiang et al. recalled similar observations in other unconventional superconductors, including $BaFe_2(As_{1-x}P_x)_2$, hole-doped cuprates such as $La_{2-x}Sr_xCuO_4$, $Bi_{2-x}(Pb,La)_xSr_2CuO_6$, and $Tl_2Ba_2CuO_{6+x}$, n-doped cuprates such as $La_{2-x}Ce_xCuO_4$, and Bechgaard salt $(TMTFS)_2PF_6$, and speculated that these findings might be universal.

What can we infer from the above? A few conclusions seem apparent. First, the standard FL theory faces challenges in gated FeSe as in other strange metals. The fact that this occurs in a large portion of the phase diagram and a broad doping range seems incompatible with ascribing this to quantum-critical fluctuations of some order parameter emanating from an isolated quantum critical point (QCP). The doping dependence of A may be a challenge to the Planckian dissipation scenario: if the scattering rate is linear in T because it saturates at $\hbar\Gamma = k_BT$ for the highest doping ($T_c \approx 45$ K), why is it linear also at the lowest doping ($T_c \approx 10$ K) where the dissipation rate is twice lower?

As the authors argue, superconductivity and strange-metal behavior are likely related. But in some cuprates, T_c is higher than in the Bechgaard salt by more than a factor of 100. Hence, strange-metal behavior may be necessary but is not a sufficient condition for HTS. The same conclusion was reached three decades ago since all MFL properties (including T-linear resistivity) were also observed in cobalt- and ruthenium oxides that are metallic but not superconducting [15].

This paper is an important new entry into our HTS materials and strange metals knowledge base. But some fundamental questions remain open. What controls T_c in these unconventional superconductors? Why is T_c so high in cuprates and pnictides? There is still much room left for breakthroughs in our understanding — and this is good news for the next generation of condensed matter physicists.

Conflict of interest

The authors declare that they have no conflict of interest.

Acknowledgments

J.W. was supported by the National Natural Science Foundation of China (12174318) and the Foundation of Westlake University. I.B. was supported by the U.S. Department of Energy, Basic Energy Sciences, Materials Sciences and Engineering Division.

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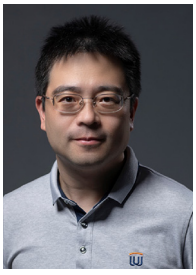

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