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High T_c superconductivity in Copper oxides: the condensing bosons as stripy plaquettes

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For the first time, the nature of the carriers in strongly underdoped cuprates has been mapped with spin polarized scanning tunneling spectroscopy by a group in Nanjing, revealing that these form mysterious collective entities living on plaquettes of size 4 lattice constants with intriguing internal structure.

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A long time ago, in the late 1980s, a frenzy erupted in the physics community because of the discovery of superconductivity in copper oxides with a transition temperature that reached eventually 160 K. However, since then, this was superseded by superconductors found under high pressure in metal hydrides with T_c's near room temperature that appear to follow perfectly the electron–phonon pairing rules of the 1957 Bardeen–Cooper–Schrieffer (BCS) theory under optimal circumstances. "High T_c" per se is, in 2023, hardly of intrinsic interest. But the main intrigue in the electron systems realized in the copper oxides had shifted already a long time ago in a different direction. In this intense research effort, it became gradually clear that a grand mystery is at work, associated with the fundaments of physics[1]. The electrons in the copper oxides realize a *doped Mott insulator*[2]. The Hubbard model that captures some of the essences of this physics is presently a prominent entry on the benchmarking list for the quantum simulators of the near future[3], quantum computers targeting problems in physics. The reason is that in doped Mott insulators, conditions are realized that hardwire in many body quantum entanglement of exponential complexity[4]: the existing equations of physics fall short of addressing the math required to understand this "ultra" quantum matter.

For this reason, this field turned, in the course of time, into a highly empirical affair; all one has is an experiment. The beneficial circumstance is that the experimental opportunities in this field leaped forward since the late 1980s. Arguably, the most powerful newcomer has been *scanning tunneling spectroscopy* (STS). This is the spectroscopic extension of scanning tunneling microscopy (STM). It measures the quantum physical probability for electrons to be added or extracted from the solid, with a sub-Angstrom resolution in real space and a Kelvin or so energy resolution.

Pioneered in this context early in this century[5], it revealed great surprises. Above all, in strong contrast with the electron systems in conventional metals, it revealed a remarkable *complexity* sprouting up in the cuprate electron systems. Although it is not completely understood, a lot is going on. STS works best at very low temperatures and in the course of time the phenomena that are observed in the well-developed superconductors were called "intertwined order"[6]: This appears as some kind of coordinated "symphony" of various patterns of symmetry breaking such as the "stripy" electron crystals, but also a "quantum liquid crystal" breaking of the rotational symmetry, in some systems incommensurate static antiferromagnetism, and there may also be a novel magnetic "loop current" order, all living side by side with the superconductivity—possibly in the form of a so-called pair density wave, referring to a superconducting order parameter that is modulated in space[7].

This is the context where the new STS results by the Nanjing group[8] interfere. The cuprate story begins with the insulating stochiometric "parent" compounds. These are the Mott insulators that are easy to understand: as a consequence of the copper-oxide chemistry, electrons strongly repel each other locally so that when there is an integer number of electrons per unit cell, an electronic traffic jam sets in. But the electron spins can still freely move and these form an antiferromagnetic quantum spin system condensing in a simple two sublattice antiferromagnet. By removing (or adding) electrons upon doping, a quantum physical "stop and go" traffic develops: the doped Mott insulator that defies a mathematical description[2,4]. However, when the doping is small, <5% or so, the system continues to be a soft insulator devoid of superconductivity, which sets in gradually at larger dopings. The Nanjing group has a first in the form of an extensive set of high-quality STS data in this low doping regime. For reasons that are not all that well understood, STS works only in the so-called Bismuth family of cuprates and these are in turn very hard to prepare at low doping levels: this group has two samples for a nominally insulating doping level $x = 0.08$ and at the borderline for superconductivity $x = 0.10$. Furthermore, they report on impressive experimental progress. Using a magnetic tip, they manage to observe directly the spin-polarized response, revealing for the first time the two sublattices antiferromagnetic spin order of the Bismuth family insulator! Excitingly in this way, they can also obtain a clear view of what happens with this antiferromagnetism in real space when the system is doped, a crucial part of the doped Mott insulator mystery affair.

When the doping percentage is very low one may expect that one can get a view of the nature of a single, isolated carrier. Much theoretical work was done in the past on this particular problem[2,4]. Some of these expectations appear to be confirmed by the Nanjing experiments. I already referred to the "stop and go traffic" that develops in the presence of carriers. In this context, one has to be acutely aware of the fact that one is dealing with *quantum* magnetism. The two-dimensional $S = 1/2$ Heisenberg antiferromagnetic is characterized by very strong quantum spin fluctuations. The ordered antiferromagnetic can be viewed as a "spin solid" subjected to very strong zero-point motions. The quantum mechanical delocalization of the carrier gives rise to low

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energy states that are easily seen with STS to appear in the Mott-gap of the insulator. However, these also literally "stir" the antiferromagnet with the effect that the carrier is expected to melt the spin system in its immediate vicinity into a blob of quantum spin liquid. This "spin-liquid polaron"(composite of carrier and quantum spin liquid blob) is then predicted to propagate as a quantum particle, but given the disorder, it is prone to localize.

This is pretty much precisely what is seen in the Nanjing experiments: they find strongly inhomogeneous textures formed from the Mott-insulating antiferromagnet, interrupted by blobs characterized by low energy states revealing the presence of carriers where the antiferromagnetism is strongly suppressed. Although predicted a long time ago[9], it is exciting to see this basic motif of the doped Mott insulator for the first time directly confirmed in the experiment.

But what comes next is actually very different from these old theoretical expectations—when I saw this first I was struck by surprise. It is about a new principle at work in doped Mott-insulators pertaining to the connection with superconductivity. We just know for sure that the full-fledged high Tc superconductors are Bose condensates formed from "bosons" carrying a charge equal to twice the electron charge. A bound pair of fermions is a boson, just like the Cooper pairs of conventional low-temperature superconductors. But this poses a problem of principle. The Cooper mechanism behind the conventional Cooper pairs is a very special quantum physical effect: exposing a highly degenerate Fermi gas to a weak attractive interaction leads to a binding exclusively in the pairing channel. Dealing, for instance, with a classical system, attractive interactions lead to clumping, drops of cohesive liquid form being "N body bound states". In doped Mott insulators, especially at low doping, there is just nothing like a Fermi gas with a Fermi surface and so forth. What protects the pair channel here? This is a question that is, after 30+ years of research, still not answered satisfactorily. Given that the superconductivity starts to emerge at $x = 0.10$ it may be that the Nanjing experiments can shed light on this question.

They appear to do so but in a highly surprising fashion. Back to the carrier "blobs" in the inhomogeneous texture: Upon increasing the doping from $x = 0.08$ to $x = 0.10$ it appears that the system is just approaching the percolation threshold, the blobs being isolated at $x = 0.08$ to form a percolating network at $x = 0.10$ where the antiferromagnetism is strongly suppressed everywhere and weak link superconductivity sets in. Hence, one can zoom in on the isolated blobs and in the supplementary material, the authors make a compelling case that one can associate two electron charges to every blob in the $x = 0.10$ system, suggesting that these blobs reveal somehow how the "preformed pairs" look like in real space. The surprise is that the blobs have a remarkably complex internal structure, revealed by the position dependence of the low energy electronic density of states. This is perhaps best summarized by the cartoon presented in their Fig. 5b. The first surprise is that this "effective pair" is remarkably large: it involves a plaquette of the tetragonal Cu–O lattice of size 4×4 lattice constants. Next, the ways that the low energy states are distributed are very inhomogeneous and anisotropic inside this plaquette: most of it is concentrated in the line of oxygen atoms connecting the Cu atoms in the middle of the plaquette, with some extra low energy density of states appearing mostly on the oxygen atoms on the lines in the lattice above- and below this central line. That the density of states piles up on the oxygens is not surprising; among the first facts that were thoroughly established is that the cuprates are so-called charge transfer insulators where the hole states are associated with the oxygen 2*p*-states instead of the "lower Hubbard band" *d*-states. This was actually the "Zaanen–Sawatzky–Allen" notion[10] introduced in my 1986 Ph.D. thesis that helped me much in my early career.

However, it is just greatly surprising to find this highly textured affair inside the big plaquettes—I am not aware of any theoretical work pointing at such a phenomenon. On the other hand, it sheds more light on what happens at higher dopings. In the well-developed superconductors up to optimal doping, the "intertwined order" is observed as I already emphasized. A most distinctive signature is the coexistence of the superconductivity with the "stripy" or "nematic" patterns breaking the translational symmetry in the low energy density of states. By just stacking the Nanjing two-hole plaquettes in a percolating way, taking care that the translational symmetry breaking is in optimal synchrony one just obtains these intertwined order patterns of the full-fledged superconductors!

This does interfere with the way that I and many others used to think about these matters. The precise nature of the symmetry breaking is still the subject of debate. Conventionally it was believed that the density of states modulations reflect a change in electron density, showing the presence of a charge density wave. However, evidence started to accumulate that it could be even a so-called pair density wave[11] associated with spatial modulation of the superconducting order parameter[7]—perhaps the best bet is that it may be both since in a strongly coupled situation (small pairs in real space) one expects that charge density and superconducting phase always communicate. But thinking about such density waves typically departs from highly collective physics in the strongly interacting fluid, think nesting instabilities and so forth, that should be irrelevant for isolated carriers.

Now we get offered by the Nanjing group the "lego-block" perspective. Apparently, the quantum antiferromagnet together with the charge-transfer holes and whatever microscopic physics we do not quite understand conspire to produce these two-hole "boson" like states living on these 4×4 Cu–O lattice plaquettes exhibiting already the fully developed "nematic" or "stripy" like anisotropy that is a famous ingredient of the intertwined order. This represents a very interesting challenge for the theoretical community—to have a chance to get anywhere the few-hole problem is a lot easier than the high-density metallic case but apparently, although these two holes may be responsible for the most prominent anomalies of the superconducting state, they are presently plainly mysterious.

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J.Z. wrote, revised, and finalized the manuscript.

COMPETING INTERESTS

The author declares no competing interests.

ADDITIONAL INFORMATION

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