# What DO we know about high $T_c$ ?

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From twenty years of experimental research, we can draw several conclusions that do not depend on any particular model of high-temperature superconductivity. With these in mind, it's time to assess the theoretical landscape, and wonder whether we have been asking the right questions.

> espite the impression that those outside the field (and perhaps even experimentalists working within it) may have, I believe that over the past twenty years the theory of high-temperature superconductivity in the copper oxides has made some progress. In particular, although we are still very far from having a generally agreed microscopic model that is in any way comparable to the Bardeen, Cooper and Schrieffer (BCS) model of 'classical' superconductivity, there are certain conclusions that I believe we can reasonably draw independently of such a model - here is my own list. There is space, unfortunately, only for the tersest possible indication of the reasons I believe the propositions listed (and no space at all to discuss possible objections!).

1. Superconductivity in the copper oxides is a result of the formation of Cooper pairs (pairs of electronic excitations with charge 2e that all have the same ground-state wavefunction), or more technically, of the onset of off-diagonal long-range order in the two-particle density matrix. Evidence for Cooper pairing includes the observation of flux quantization (the magnetic flux being h/2e instead of h/e) and the Josephson effect (the supercurrent is carried by two electrons from one superconducting electrode to another through an insulating barrier).

In the early days this was not at all obvious, and all sorts of exotic mechanisms were proposed that did not obviously have to do with Cooper pairing; however, the most promising of these has been shown to be simply a disguised version of that phenomenon. Even in the absence of a detailed knowledge of the nature of the pairs — such as what the pairing 'glue' happens to be — their existence enables us, under quite broad conditions, to set up a macroscopic Ginzburg–Landau theory and use it to calculate the observable electromagnetic properties; this is one area in which few would deny substantial progress has been achieved.

2. The principal locus of superconductivity is the copper oxide planes. The qualitative universality of normal and superconducting state properties, despite the disparate nature (or complete absence in the non-superconducting 'infinite-layer' (Ca,Sr)CuO<sub>2</sub>) of a 'charge-reservoir' group, supports this statement.

3. To a zeroth approximation, pairs form independently in the different copper oxide multilayers. To hop from one layer to another, the 'inter-multilayer' hopping energy as inferred from c-axis resistivity measurements is generally small compared with the characteristic (minimum) energy scale associated with superconductivity,  $k_{\rm B}T_{\rm c}$ . These latter two conclusions justify the use of a Lawrence-Doniach model (treating the multilayers as discrete and weakly coupled) even when a three-dimensional Ginzburg-Landau picture (treating the multilayers as 'continuous' through the use of a gradient term) becomes internally inconsistent (S. Doniach and W. E. Lawrence in Proc. 12th Int. Conf. Low Temp. Phys. (ed. E. Kanda) 361 (Keigaku, Tokyo, 1971)). Again, this has been very successful.

4. The dominant mechanism of Cooper-pair formation in the copper oxide materials does not involve a net saving of ionic kinetic energy as in BCS superconductors (from the small or zero isotope effect in higher-temperature copper oxides), and therefore must either be all electronic in nature or, conceivably, involve the phonons in a way much more sophisticated than in BCS theory.

Taken together, conclusions **2**, **3** and **4** suggest that an appropriate 'zeroth-order' starting point for the consideration of superconductivity in singleplane copper oxides (such as  $Tl_2Ba_2CuO_{6+\delta}$ ) is a sum of single-plane hamiltonians that contain (a) the in-plane conduction-electron kinetic energy, (b) the lattice potential energy and (c) the inter-electron Coulomb interaction (suitably screened by the ionic cores) — and nothing else! Any model invoking anyons, spin fluctuations, excitons, and so on, should be prepared, at least in principle, to derive these entities from the above generic hamiltonian.

Apart from 1, the above conclusions constrain the input of a viable theory of high-temperature superconductivity. What can we say about the output — that is, what do we know about the nature of the Cooper pairs?

5. The spin state of the pairs is a singlet from NMR Knight shift and spin-lattice relaxation  $(T_1)$  measurements. 6. In copper oxides with tetragonal symmetry the orbital state is  $d_{x^{-\nu}}^{2}$ (also known as B<sub>1g</sub>), unambiguously determined by phase-sensitive experiments such as scanning SQUID microscopy. These points refer strictly to the behaviour of the order parameter under spin rotation and the symmetry group  $C_{4v}$  of the square, respectively; in particular, 6 puts no constraints on the detailed momentum dependence over the Fermi surface of the superconducting order parameter, still less of the 'energy gap' (if that concept is indeed meaningful). All of my conclusions so far are independent of any implicit assumption that the pairing process has anything in common with that described by BCS theory, in particular that the numerical relations predicted by that theory hold to within an order of magnitude.

If we are prepared to make such an assumption, we can draw two further conclusions. **7. The size of the pairs is somewhere in the range 10–30** Å — from measurements of the upper critical field, Fermi velocity and  $T_c$ . This means that the pair size is only moderately greater than the inter-conductionelectron in-plane spacing, putting us in the intermediate regime of the so-called Bose–Einstein condensate to BCS superconductor (BEC–BCS) crossover, and leading us to expect very large effects of fluctuations (these are indeed found).

8. The pairs are formed, as in BCS theory, from electrons in time-reversed states (the argument against the alternative scenario, 'inter-band' pairing between electrons from different energy bands, is technical, involving the absence of strong electromagnetic absorption just above the gap edge).

There is one other generic aspect of the copper oxides that I think would by now be fairly generally agreed: either the normal state is not, even in principle, a Fermi liquid, or if it is, the incoherent background not explicitly described by the lowenergy phenomenology is vastly more important quantitatively than in a typical 'textbook' metal (as determined from angle-resolved photoemission spectroscopy (ARPES), optical spectroscopy and transmission electron energy loss spectroscopy (EELS), to name a few). On the other hand, the superconducting state seems to behave in many ways suspiciously like a classic 'superfluid Fermi liquid' such as helium-3.

The way in which I have framed, above, the theoretical problem posed by high-temperature

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superconductivity is, I believe, accepted at least implicitly by most though not all theoretical papers in the field over the last twenty years; in particular, inputs **2**, **3** and **4**, as well as a number of other more specific assumptions, are automatically implicit in any calculation that starts from a two-dimensional Hubbard (kinetic energy plus on-site repulsive potential) or t-J model (strong correlation limit of Hubbard model).

So where to go from here? With few exceptions, theoretical papers on copper oxide superconductivity have tended to model themselves on the original BCS description of classical superconductivity, in the sense that they have assumed that the solution will be found if one can write down the appropriate effective low-energy hamiltonian and use it to calculate the experimental behaviour. Here, 'low energy' is usually implicitly taken to mean that the hamiltonian in question should describe correctly states in which, crudely speaking, the relevant one-electron energies are  $\sim k_{\rm B}T_{\rm c}$  or less. As a limited selection of such models is presented on page 138 in this issue, I will not attempt to comment on any of them individually, but will rather ask the question: is it obvious that any low-energy effective hamiltonian will be adequate?

To be sure, one would not expect that states of the N-body system with energies that are large compared with  $Nk_{\rm B}T_{\rm c}$  would play a significant role, but that is not the same criterion. And one very striking fact that has emerged from recent optical experiments is that the superconducting transition is accompanied by changes (of the order of a few per cent) in the optical properties, even at energies of the order of 100 times  $T_{\rm c}$  — which seems very strange within any model that works only with a strictly low-energy effective hamiltonian. (The 'trivial' explanation within such a model, namely that the energies of the 'initial' (lowenergy) states are shifted whereas the 'final' (highenergy) states are unaffected, appears impossible to make work quantitatively). Of course, the above considerations are not meant to imply that there is no point in working with Hubbard, t-J or other low-energy models. In particular, if such calculations should lead, as many of their practitioners hope, to the conclusion that some or all of these models

## **COMMENTARY**

inevitably imply either Cooper pairing or some more exotic kind of phase transition or both, that would in itself be an extremely valuable piece of information. However, a complete theory of high-temperature superconductivity would still have to give an account of the high-energy effects of its onset.

Coming at the issue from a different angle, let's consider the question: what kind of energy is saved when the system makes the transition from the normal to the superconducting state? In principle

## HOW WILL WE KNOW WHEN WE HAVE A DECENT THEORY OF HIGH-TEMPERATURE SUPERCONDUCTIVITY?

this question can be answered by experiment, but in interpreting the data one must beware of the fact that what looks like kinetic energy at one level of theoretical description may look like potential energy at another more phenomenological level, or vice versa. A spectacular example of this feature may be found in the BCS theory: a consideration of the (effective) Bardeen-Pines hamiltonian (J. Bardeen and D. Pines Phys. Rev. 99, 1140-1150; 1953) would lead to the conclusion that it is the potential energy that is saved, although a firstprinciples argument given by G. V. Chester (Phys. Rev. 103, 1693-1699; 1956) shows that actually this saving is exactly balanced by the expenditure of electronic kinetic energy, and it is the saving of the ionic kinetic energy that tips the balance. Anyway, if we work at the level of the generic in-plane hamiltonian defined above, the default option is that the saving is principally of the inter-conduction-electron Coulomb energy, and I will make this assumption for the sake of the following argument.

For a homogeneous three-dimensional system, the Coulomb energy can be expressed (up to a numerical constant) as simply the integral over wave vector **q** and frequency  $\omega$  of the loss function  $-\text{Im}(1/\varepsilon(\mathbf{q},\omega))$ , which is a quantity directly measurable, for example, by EELS; in a strongly layered system, such as a typical copper oxide, the situation is a little more complicated, but analogous relations can still be derived. So the obvious question is: where in the phase space of **q** and  $\omega$  is the energy necessary to drive the superconducting condensation principally saved? In the absence, so far, of an unambiguous experimental answer to this question, I believe it is reasonable to use what I have called elsewhere the 'Willie Sutton principle': that it makes most sense to take it from the places where there is a lot of it already. And in which regions of **q** and  $\omega$  is the energy mostly stored in the normal phase? We can at least use existing EELS and optical

data to say one thing - that for reasonably small **q** (of magnitude less than 0.5  $Å^{-1}$ , say) there is very little energy stored below around 0.1 eV, and a lot stored in the MIR (mid-infrared) peak, which for most copper oxides can be taken as extending. roughly, from around 0.1 to 1-1.5 eV. As there are independent reasons (of which I cannot go into details here) to believe that in layered systems the 'small-q' regime is at least as important for the Coulomb energy as larger values, my personal bet would be that it is indeed the MIR regime that contributes most substantially to the condensation energy of the copper oxides. Should experiments eventually confirm this (existing optical experiments probe only very small **q** and are thus inconclusive), this state of affairs would presumably require an approach quite different from most of those currently being explored. But for the moment, we are in the hands of the experimentalists ...

One other question that I believe systematic experimental study can address is: how universal are the normal and superconducting state properties of the copper oxide family, and what is the origin of the lack of universality? The most-commented-on aspect of the latter question is of course the dependence of  $T_c$  on the number of layers *n* in homologous series, but I would also give a lot to know why, for example, the transition temperatures of the single-plane Hgand Tl-based compounds are reproducibly different, despite the *prima facie* close similarity of the copper oxide planes in the two materials.

More generally, my personal belief, for what it is worth, is that the history of theoretical research over the last twenty years on these materials teaches us that, although it is obviously important to explore the implications of specific microscopic models, progress is at least as likely to come by asking questions (such as 'does the macroscopic Ginzburg–Landau description work?', 'what is the symmetry of the order parameter?' and 'where is the energy saved?') that can be posed independently of any such model and can, at least in principle, be answered unambiguously by experiment.

Finally, how will we know when we have a decent theory of high-temperature superconductivity (other than from the purely sociological evidence constituted by its universal acceptance)? I believe such a theory should either provide us with a blueprint for a robust room-temperature superconductor, or, failing that, enable us to state with confidence that no such superconductor will ever be built (at least from copper oxide or related materials). Or, failing even that, at least explain lucidly why we cannot do either of those things. Currently we seem to be some way from being able to do any of the above.