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The energy-resolved STM results, their apparent inhomogeneity and the true bulk behaviour of the HTSC cuprates, from a negative-U perspective

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Abstract

An attempt is made to reconcile the results of the prime experiments on the high temperature superconducting cuprates from Loram and coworkers (2006 *Preprint* cond-mat/0609305) and Davis and co-workers (2005 *Phys. Rev. Lett.* **94** 197005; 2005 *Science* **309** 1048), relating respectively to electronic specific heat and to energy-resolved scanning tunnelling microscopy (STM), over the question of electronic inhomogeneity. I see the latter, *at the appropriate level*, as being essential to the evolution of HTSC in these materials, and try to bring the above key works into alignment, not only with each other but with a wealth of related work. This is undertaken around a negative-U scenario that I have long advanced, a reassessment being advocated of the way in which the STM and angle resolved photoemission spectroscopy (ARPES) results are currently being interpreted in regard to the 'coherence peaks'.

1. The perceived incompatibility of specific heat data with recent STM 'gap mappings'

Loram and Tallon have very recently restated the claim, looking at the matter from the perspective of their refined electronic specific heat analysis [1], that the energy-resolved scanning tunnelling microscopy (STM) results from the HTSC cuprates, as presented by Davis and co-workers [2] and several other groups, would seem to relate to surface effects. Their claim is that these results, the famous 'gap maps', cannot reflect what is current with regard to HTSC once away from the surface and sensed by a thermodynamic probe like specific heat measurement. The relative sharpness of the peak in the electronic specific heat found across T_c, even for the strongly 2D environment of Bi-2212, Tl-2201 and Hg-1201 and after accounting for the strongly fluctuating conditions about T_c (which signal in $C_v(T)$ as in $\rho(T)$ data [3] the presence of preformed pairs), cannot, Loram and Tallon claim, be at all reconciled with the very significant spread in bulk 'gapping' generally imputed from the STM work. Furthermore

each gap map, because it is frozen in time, appears not to relate either (directly at least) to the fluctuating stripe phenomenon registered by bulk x-ray and neutron diffraction work [4].

While there exist bulk-sensitive measurements which would support a different view to the one expressed by Loram and Tallon regarding the degree and form of inhomogeneity active in the HTSC cuprates, these Cu and O nuclear quadrupole resonance (NQR) and nuclear magnetic resonance (NMR) results usually come from LSCO [5], a material displaying special structural complications. The yttrium site NMR from YBCO appealed to now, as previously, by Loram *et al* [6] would nonetheless still support a not insignificant degree of local inhomogeneity (at NMR timescales). Muon spin relaxation (μSR) [7], unlike NMR, is drawn specifically to sample, on a faster timescale, around negatively charged 'imperfections', and in the HTSC cuprates these are legion, being deliberately inserted to transform the CuO_2 layers there to metallicity and superconductivity. It was μSR work in fact which first endorsed the view I had previously illustrated in figure 4 of [8] that this highly local inhomogeneity should inevitably play a key role in the character of the superconductivity of these 'doped' Mott insulators, in particular in their underdoped regime. Use of the 'Swiss cheese' terminology as *à propros* to the HTSC condition indeed first appeared in the μSR work of Nachumi *et al* [9] dealing with the low level substitution into the cuprates of isovalent but energetically dissimilar Ni and Zn for Cu. The doping of Sr for La in LSCO, the extra oxygen in LCO, YBCO, HBCO or BSCCO and the counter-substitution of Tl by Cu in TBCO all of course introduce *charged* lattice defects. Ultimately these 'defects' become frozen in, and randomly so when samples are rapidly cooled from their high temperature annealing conditions. Where exactly though the charge appertaining to these 'dopant' centres subsequently organizes itself is a somewhat different matter. Apart from at $p = \frac{1}{8}$, the charge self-organization is clearly dynamic, not static—see [10] for my understanding of 'stripe' formation and of the role played by the Jahn–Teller effect in this dynamic charge organization and the detailed development of HTSC. Where the Madelung potential is most strongly perturbed by random doping there it is possible for the charge stripes to become permanently pinned.

2. A résumé of negative-U modelling of HTSC towards a discussion of the above problem

It is in fact at certain such stochastically favoured positions that I envisage electron pairing as being most strongly effected. This is because I have assumed from the beginning [11, 8] that in HTSC one is encountering a negative-U phenomenon based upon the fluctuational conversion of the state locally approximating to $^{8}Cu^{0}_{III}$ over into its $^{10}Cu^{2-}_{III}$ double-loading condition. This is admissible by virtue of the latter's p^6d^{10} shell-closure form which entails the termination of all p/d bonding–antibonding interaction between the Cu and O atoms within the pair–recipient coordination unit. There the ensuing rearrangement of the latter's now full p and d states, specifically their energy inversion, along with the accompanying local lattice relaxation, procures stability for the $^{10}Cu^{2-}_{III}$ condition, at least on a fluctuational basis. I believe this negative-U state to have been sensed in pump–probe and further specialized optical work [12], as set out at length in [13]. Pairing obtained at the negative-U centres proceeds in the resonant view embraced in [14] (wherein the above double-loading state sits near-degenerate with the chemical potential) to induce pairing over most of the Fermi surface, and with a single T_c value. The negative-U centres, despite being inhomogeneous in location and to a certain degree therefore in energy, cooperate to bring about a condensed state which is global and phase-locked, via the intimate intersite tunnelling of the correlated pairs. In standard superconductivity the pairing envisaged is of retardedly coupled $+\mathbf{k}/-\mathbf{k}$ quasiparticles. In locally instigated negative-U pairing the pairing is direct, the pair potential is no longer the classic delta function of the BCS formalism [15], and the coherence length,

ξ, of the resulting superconductivity is, in keeping with the high T_c, high Δ and high H_{c2}, extraordinarily small. In optimally doped HTSC material the value of $\xi^{\perp c}$, as assessed from $H_{c2}^{\parallel c}$, is only ~15 Å, or about half the inter-stripe spacing [16], with $\xi^{\parallel c}$ even less. The superconductivity emerging in the p-type cuprates was early on deemed essentially d-wave ($d_{x^2-y^2}$) in form [17], the maximum gapping being at the saddles of the Fermi surface (i.e. in the basal axial directions of the crystal structure—the Cu–O bond directions there) and the gap nodes oriented in the 45° directions [18]. The gapping is clearly of strong-coupling form, the observed ratio $2\Delta(T = 0)/kT_c$ being about 5.5 for all the various HTSC cuprate families over a wide range of doping about optimal [19]. Such a value is consistently extractable from C_v, NMR, Raman, neutron and Andreev reflection data, and is to be compared with 4.3 for weak-coupling mean-field behaviour within a d-wave symmetry setting.

It is very apparent from C_v [20] as from other data that much higher ratios than 5.5 are to be gained from underdoped material if one persists in relating the outer 'pseudo' gapping often in evidence there [21] through to the value of the superconducting onset temperature, $T_c(p)$. The problem is that, upon retreating back towards the Mott-insulating condition of $d_{x^2-y^2}$ state half filling, DOS pseudogapping in underdoped material actually develops from more than one source. There is that associated with RVB spin coupling [22, 10c], as well as the residual Mott charge gapping emanating from half filling. Then, too, there is the pseudogapping local to the negative-U centres whenever or wherever these prove insufficient in number or are of an energy too far removed from resonant alignment with the chemical potential to uphold the superconductivity. It should also be recalled here that, beyond the enhanced bulk superconducting fluctuational behaviour exhibited by all these materials [23, 3], there emerges clear evidence for the presence of local pairs to several tens of degrees above T_c upon engaging appropriately sensitive probes such as the Nernst effect [24] or monitoring the electrical noise [25]. Even very precise measurements of the basal lattice parameters sense incipient susceptibility to the HTSC condition from far above T_c [26].

All this multi-sourced pre-superconductive activity and gapping help to create a pronounced minimum in the effective density of states in the centre of a band, that, if uncorrelated, would sit close to a saddle-point maximum. The outcome is that right up to $p = 0.3$ the Seebeck coefficient is positive at low temperatures from a negative sign to $(d\sigma/dE)_{E_F}$ extending through to such doping [27]. Correspondingly the strong temperature dependence of the Hall coefficient [28] comes about because of the changing balance within the $d_{x^2-y^2}$ set of conduction electrons between coherent p-type band quasiparticles and non-band-like negative carriers, around the zone diagonals and saddles, respectively. The latter become more dominant at very high temperatures, and then again at low temperatures upon approaching superconductivity. Throughout, severe and very anisotropic scattering is experienced over the Fermi surface [29]. At high temperatures this becomes sufficient to bring incoherence and even weak localization to the carriers on the saddles [30], whilst at low temperature those same carriers begin to be translated into local bosonic pairs. The saddles provide the scattering 'hot spots' for negative-U pair formation [13b], just as for potential CDW/SDW nesting [31]. It is most noteworthy that right across the underdoped regime, n_s, the number of superconducting pairs, assessed from μSR [7, 9] and other penetration depth [32] measurements, relates more closely not to n, the total number of electrons present in the $d_{x^2-y^2}$ band, but to p, the 'doping' content there below half filling. The local character to events here remains pre-eminent. The electrons instigating the HTSC phenomenon are not particularly 'good' quasiparticles, and neither indeed are those throughout the saddle region that primarily respond. This extensive impairment to standard Fermi liquid behaviour is very much in evidence in the angle resolved photoemission spectroscopy (ARPES) normal state spectra once one has left the diagonal 'nodal' segments of the Fermi surface [33].

In the superconducting state one similarly therefore has to be very careful in one's reading both of the ARPES and the energy-resolved STM spectra, in particular where the latter relate to the saddle point states and maximal gapping. One does not have a simple, classical, mean-field, BCS-type situation here, any more than in the normal state one had a classical Fermi liquid. What is remarkable is that the classical approach of the BCS, Eliashberg–Nambu and McMillan–Dynes formalisms can be extrapolated as far into the ultra-correlated regime as appears to be the case and yet emerge with a meaningful parametrization [34]. The Uemura plot [35], namely $\log T_c$ versus $\log(n_s^{2/3})$, portrays the superconductors $NbSe_2$, MgB_2, $PbMo_6S_8$ and K_3C_{60} as bridge materials between the classical and the cuprate materials, but these still seem to be reasonably well dealt with by the traditional approach in spite of their rapidly reducing coherence lengths. This is very comparable to what is observed in the normal state, where effectively classical quasi-particle Fermi liquid behaviour extends much farther into the highly correlated regime than might have been anticipated—witness the degree of preservation of the classical Lorenz number, L_0, for $\kappa_{th}/\sigma T$ [36]. Indeed such a value is approached in very heavily overdoped $(La_{1.7}Sr_{0.3})CuO_4$, although the prefactor A exhibited on the T^2 resistivity behaviour stands there five times greater than the Kadowaki–Woods plot would support [37].

One very striking feature with the cuprates, and in particular with underdoped cuprates, is that close approach to the onset of superconductivity brings considerable apparent improvement in the quasiparticle mean free path (e.g. the empirical Lorenz number drifts down towards L_0 [38]). From thermal Hall data, Ong *et al* [39] have deduced quasiparticle mean free paths growing rapidly towards 10 nm in the run down to T_c, this despite the rapid growth in electrical noise brought about by proximity to stable pairing and bosonization of the active electronic system [25]. What the effect might be here from preformed pairs remains to be analysed.

3. Gaps and pseudogaps in the negative-U approach; questions of size and symmetry

The question now arises both with the low temperature ARPES and tunnelling results about how one should read off the superconducting gap from these spectra when there is occurring in the underdoped and even optimally doped HTSC materials so much correlated activity beyond that appertaining specifically to the global superconducting state, with its set T_c. Indeed Loram and co-workers themselves earlier expressed their C_v and NMR results in terms of a combination of superconducting gap and pseudogap [40]. As indicated above, the former gap is related to T_c by the equation $2\Delta(p, T = 0)/kT_c(p) \approx 5.5$. In the underdoped regime $T_c(p)$ drops away from T_c^{max} quadratically as $(1 - (p_o - p)^2)$ to vanish when $p \sim 0.05$ ($p_{o(pt)}$ universally being $\approx$0.16) [41]. Moreover it is observed that into the underdoped regime n_s reduces steadily from optimal doping roughly as $p_o - p$, thereby making n_s not proportional to T_c. Rather the remarkable Uemura relation $\log n_s^{2/3} \propto \log T_c$ holds [35]—endorsing in this a Bose-related behaviour. Just as significantly the superconducting condensation energy, E_c, extracted from the C_v data is proportional neither to n_s nor to T_c. Indeed E_c emerges as being a steeply augmenting function of p, growing to a sharply cusped maximum at $p = 0.185$, a little beyond where T_c is smoothly maximized [40]. Note that where T_c maximizes at $p = 0.155$, E_c is still only one-third of what it is to become by $p = 0.185$. $p = 0.185$ is actually very close to the p value at which the specific heat work and other related assessments consistently indicate the pseudogap to be rapidly vanishing [42]. It is, perhaps, this doping level that marks the high point in HTSC behaviour, as the electrons are most effectively brought into the superconducting condition. For p beyond this concentration, despite the value of n_s continuing (for a short while at least) to rise, both T_c and the condensation energy per mole already are well in decline. Niedermayer *et al* [43] record in fact for Tl-2201 that T_c, n_s and p are interrelated across the

entire range of p by the expression

$$n_s/p \propto m^* \cdot [1 + \sqrt{p} \cdot (T_c(p_o)/T_c(p))].$$

This confers upon the full T_c versus n_s plot (using p as running parameter) a characteristic, inclined and sharply lobed shape, often referred to as the 'butterfly wing' or 'boomerang effect'. It might too be recalled that in $YBCO_{7-\delta}$ as $\delta \to 0$ and virtually all the chain electrons there are finally carried through to superconductivity, this incorporation occurs without any growth in T_c [44]. I have in [10a] taken the above planar 'hole' concentration of $p = 0.185$ to afford the optimized combination of doubly occupied negative-U centre population and energy location. Namely, a sizeable population coming into resonance with the chemical potential at a level of metallicity that is high enough to uphold marginal Fermi liquid behaviour and yet not so high as to overly screen away the local Madelung potential necessary for local-pair creation at the negative-U centres.

Regrettably most of those working in the field of HTSC have no regard for negative-U centres. The physics of HTSC is seen solely as the preserve of $d_{x^2-y^2}$ or B_{1g} symmetry [17]. Where quasiparticles of a more standard form do exist takes on B_{2g} 'nodal' symmetry [45]. An insistent emphasis has developed upon aspects of the physics holding these two symmetries, a quasi-standard Fermionic physics, as with, for example, the SDW formation purported to emanate from the saddle 'hot spots' of the Fermi surface [46]. A further case is to be found in the detailed review of the electronic Raman results very recently published by Devereaux and Hackl [47], in which all emphasis is placed on the B_{1g} and B_{2g} anomalous spectra to the detriment of the somewhat more difficult to acquire A_{1g} spectra. The latter spectra are, though, even more striking, but are largely confined in [47] to the comments in the conclusion as a 'mystery'. Note, however, that it is the A_{1g} peak which at 330 cm^{-1} (i.e. 41 meV) in $T_c^{max} \sim$ 95 K systems would correspond to $2\Delta_0 \sim 5\frac{1}{2}\, kT_c$. Furthermore, it is this A_{1g} feature that exhibits the temperature and doping dependence [48] anticipated for global superconductivity; indeed the same 2Δ behaviour as tracks the much examined (π, π) 'resonance' peak met with below T_c in inelastic neutron work [49]—the spin-flip pair excitation [50]. (Note the latter scattering feature pointedly displays no isotope effect.) By contrast the B_{1g} and B_{2g} electronic Raman peaks are not attained until 440 cm^{-1} (55 meV) or more, up in the range of the pseudogap phenomena [51]. In very similar fashion standard infrared work casts its emphasis upon a still higher energy pseudogap region, strong IR gapping setting in below around 800 cm^{-1} or 0.1 eV (i.e. $\sim J$), this quickly coming to dominate the optical self-energy and helping to mask any action at 2Δ relating directly to the superconductivity itself [52]. Cardona was the first to stress the key behaviour of the A_{1g} electronic Raman spectrum [53] and to show that the peak energy arises precisely where small sharp discontinuities in the phononic Raman spectrum mark the superconductive gap to lie [54]. A_{1g} symmetry covers not only isotropic s-wave superconducting symmetry but, more generally, extended-s-wave symmetry as well. Nodes in the latter are suitably able to assist at the Cu coordination unit centre in holding down the $+U$ repulsive component to the overall U value.

4. Misreading the superconducting gap size from STM and ARPES data

How then are we to read the underdoped gap 'bit maps' generated by Davis and co-workers from their energy-resolved STM experiments [2]? If not as extremely as Davis and colleagues seem inclined to, then certainly not either as Loram and Tallon would construe them [1]. The latter imply that if the pseudogap energy were to be significantly inhomogeneous, the superconducting gap is necessarily going to be inhomogeneous too. And if indeed inhomogeneity were to exist on anything approaching the scale of Davis and colleagues'

mapping, then the superconducting onset temperature ought itself to be much more disordered than patently is the case from behaviour of the electronic specific heat peak to be found across the superconductive onset. Their view rests, however, upon treating the superconductivity as being of rather standard form, fermion based and density of states driven. All changes in the local condition in such a case would be reflected directly in local T_c values, especially if, as the bit maps have been taken to imply, the superconductive gap value were at all local. Yet T_c from the C_v results, as with other data from good samples, does not reflect any such marked spread. Therefore it is claimed in [1] that the inhomogeneity evident in the STM map must in its entirety be a surface effect and cannot in any way extend to the circumstances of the bulk.

However, if one looks carefully at the 4 K STM results, as indeed was originally pointed out by McElroy *et al* themselves in [2a], the inner part of the gap below ∼25 meV actually *is* rather invariant across the field of view. It is only the outer wings of the STM spectrum that are strongly inhomogeneous—and it is the latter which dominate the bit map as it is customarily generated (via use of an overall $(\mathrm{d}I/\mathrm{d}V)^{\max}$). Already, however, we have observed that these increased energies relate to the pseudogap and not to the superconducting gap itself. Close in to T_c the thermodynamics is governed by the final opening up of the deeper inner gap, without any discontinuous change there to the pseudogap. The coherence length of the superconductivity remains throughout in excess of the dopant nearest neighbour separation at the levels of substitution involved in HTSC samples (namely $p > \frac{1}{18}$, leading to a local nearest neighbour spacing of very rarely more than $3a_0$). Within the random distribution of dopants the movement of quasiparticles is not prevented once above $p = 0.05$, and still less so should be the transfer of bosonic pairs under microscopic quantum tunnelling. A uniform phase angle becomes duly established right across the field and a unique T_c value results.

What is more problematic for the system, however, is what proceeds in the single particle spin system. The field of the spin pseudogap is certainly disjoint under the variety of J values between sites. While RVB holds in some microregions, in others spin glass behaviour shows up, especially as one drops the p value and shifts substantially towards the antiferromagnetic condition of the Mott insulator [55]. Ultimately local charge gapping becomes dominant as the $+U$ circumstances of band half-filling and non-metallicity begin to prevail. Hence the gap bit maps, as they customarily are being created and presented, are automatically transferring emphasis onto the non-superconductive reaches of this multi-stranded pseudogap and away from physics directed specifically toward events at T_c. These latter are much more homogeneously expressed by the inner gap [2a]. And, what is more, right at the surface it is inevitable that pseudogapping will be promoted by the local escape of dopant oxygen from the opened cleavage plane. Remember that O^{2-} is not a stable species when not fully embedded, and the surface O^{2-} 'excess' is accordingly open to restructuring.

5. Bosonic modes internal and external to the non-BCS condensate

A comparable complexity of behaviour is enfolded in the ARPES results, which have been so well publicized but which again would seem to have been so unfortunately interpreted [56]. The weakly defined spectral location of E_F at the saddles in the normal state condition becomes replaced in the superconducting state of Bi-2212 by a contrasting strong peaking there. This peak feature (also seen in other bilayer and trilayer HTSC cuprates) has, despite persisting to 30 K above T_c and with its energy throughout being almost temperature independent, become widely ascribed in its entirety to a superconducting 'coherence peak'. The outcome has been that the latter invariably becomes allowed to deform the outer shape of the superconducting gap, causing many ARPES papers to cite Δ values considerably in excess of the true superconducting gap. This, for example, includes the paper from Mesot *et al* [57] looking

to enumerate $\Delta(\theta)$ values around the Fermi surface—and showing indeed roughly $|d_{x^2-y^2}|$ azimuthal form. The STM papers from the Davis group embrace these same overstated gap values, a matter that I have already taken issue with in [14].

However, as is to be seen in figure 13 from the recent ARPES paper by Peets *et al* [48], it is the leading-edge maximum-gradient (LEM) point that in such spectra properly identifies the real $2\Delta(p, T = 0)/kT_c(p)$ ratio ~5.5 near p_0. The higher energy ARPES peak value in fact does not even extrapolate to zero as T_c^{opt} moves to zero under a change of HTSC system, unlike the LEM point. Nevertheless the photoemission peak binding energy plainly is diminishing as T_c^{opt} is decreased, and hence it is not registering some transfer out into the pseudogap in the underdoped regime in the way that the STM peak did. The separation between the ARPES peak and the LEM energy is found to reduce steadily between HTSC systems from 16 meV (or 128 cm^{-1}) when $T_c^{opt} \sim 95$ K to 8 meV (or 64 cm^{-1}) as $T_c^{opt} \to 0$ K [48]. This separation very conceivably might relate to the reflectivity edge in the *c*-axis far-infrared spectra to be associated with the interplanar 'Josephson' plasma resonance and its attendant energy loss function [58]. In [58b] the peak in the energy loss for LSCO $p = 0.17$ is located at 80 cm^{-1}. The ARPES peak then would betoken the latter corporate excitation of the condensed electrons being strongly coupled into the photoemission process. Accordingly, the ARPES peak needs to be viewed, as with the STM peak, as being far removed from the classical coherence peak of a standard superconductor. The above would account quite naturally for why the photoemission peaking is so prominent at the highly correlated saddles, where by every measure, ARPES included, one detects the normal state coherence being relinquished under the intense scattering active there—in my own view from local pair formation, Boson–Fermion interaction and pair destruction [14b]. In keeping with the above perception it should be noted that below T_c no peak is observed to decorate the $(0, \pi)$ edge in the corresponding ARPES spectrum from *single*-Cu-layer materials like Bi-2201 [59] and Hg-1201 [60], where it becomes much more difficult to secure any *c*-axis charge transfer. As a pertinent reminder of just how highly correlated these cuprates are, ARPES now brings us the 'waterfall effect' of complete one-electron d-band degeneration once well removed from E_F [61].

The above puts us in a position to be able to understand the more recent energy-resolved STM results just released by Lee, Davis and co-workers [62]. These are in my view being completely misinterpreted, and are highly likely to form the basis of much future confusion given that these colour maps are so compelling. As before, the new STM work permits the leading peak value effectively to control the size of the local superconducting gap $\Delta(r)$. It then proceeds through d^2I/dV^2 to identify a coupled bosonic mode of energy $\Omega(r)$, measured from this Δ across to the gradient inflection point on the well-known hump that follows. This mode will shortly be related intimately to the binding energy of the negative-*U* state below E_F, written here in cursive form as $\mathcal{U}$ to distinguish it from the Hubbard-*U* energies themselves—respectively much larger positive and negative component energies of +3 and −6 eV, yielding $U_{eff} = -3$ eV per pair [13]. While the values extracted for this coupled modal energy $\Omega(r)(\approx \mathcal{U}(r))$ in fact prove roughly correct, those for $\Delta_0(r)$ remain much overstated. Near optimal doping, respective values for Ω and $2\Delta_0$ of 55 and 40 meV are more acceptable [14b]. As emphasized in [62], the value of the modal energy Ω (properly measured from $\Delta(\mathbf{r})$) alters rather little in any given sample across the scanning field of view (typically 50 nm × 50 nm), whilst the apparent local gap energy would itself seemingly fluctuate by a factor of at least 2. The authors of [62] advance with their figure 5(c) that Δ and Ω are actively 'anticorrelated', a finding most appropriate to the present interpretation, where the Fermionic gap Δ gets larger as the negative-*U* state moves up towards resonance (i.e. $\mathcal{U}(r)$ diminishes). By contrast, in the present underdoped circumstances $\Delta(\mathbf{r})$ and $O(\mathbf{r})$ (the location of the extra oxygens on the surface) show the expected strong positive correlation. Finally $\Omega(\mathbf{r})$ and $O(\mathbf{r})$ display no

correlation, with $\Omega(\mathbf{r})$ being effectively global whilst $O(\mathbf{r})$ is locally random. Note that $\Omega(\mathbf{r})$ in its exhibited lack of dependence upon position expressly does *not* manifest the characteristics of a local phonon mode, and yet in [62], from some errant isotope work, the authors have tried to impute a familiar phononic sourcing to the above findings. In the scheme I have outlined earlier, the above bosonic mode at 55 meV relates to the 'Anderson mode' for the local-pair condensate, this sitting at a slightly deeper binding energy than the more dispersed mode formed by local pairs remaining outside the condensate. It is this latter mode that features in figure 2 of [14b] constructed from the Fourier transformed STM scattering data secured by Hoffman *et al* [63] and McElroy *et al* [2a].

6. The matter of symmetry for negative-U modelling of the BCS–BEC crossover

Let us now return to the matter of symmetry, for that must fundamentally reflect what is happening in HTSC cuprate physics. ARPES, being involved with ψ^2 rather than ψ, does not sense the phase angle component to the superconducting order parameter, only its magnitude. With the nodal arrangement for $|d_{x^2-y^2}|$ and extended-s states co-aligned in the 'diagonal' 45° directions (i.e. bisecting the Cu–O basal bond directions), the two channels should cross-couple whenever they relate to ψ^2 rather than to ψ. But surely this is the circumstance within the current charge-based negative-U scenario, the latter as constrained by the geometry of the coordination unit and the tight-binding electron bands. From a phonon normal mode analysis for the cuprate structures, A_{1g} (or Γ_1) symmetry there covers the longitudinal breathing mode of a coordination unit—just one notes as the electronic Raman data echoes. The charge flow and the lattice respond with the same natural symmetry, as modification is incurred in the electron loading of the $d_{x^2-y^2}$ σ^*-state of the coordination unit. Electron energy loss (EELS) experiments [64] indeed have given evidence of a very lossy peak with precisely the above specified 60 meV location. Although this EELS feature appears at T_c, the subsequent dependence of intensity on temperature would imply that the true onset to activity is not in fact T_c itself but, for BSCCO-2212, some 30 K above T_c, much as is now also intimated by the Nernst data [24, 65].

What additionally commences well above T_c and adheres to this same Γ_1 or A_{1g} symmetry is found to be an extremely strong and highly anomalous basal softening of the uppermost (80 meV at the zone centre) LO phonon mode, which develops sharply from beyond around half-way to the zone boundary [66]. This lattice breathing mode is, it would seem, coupling very strongly with some short wavelength mode. Now, long ago a zone edge mode seemingly superfluous to the customary phonon normal mode analysis was detected in neutron scattering work—in $YBCO_7$ that feature indeed lay around 55 meV (or 440 cm^{-1}) [67]. The occurrence of a striking gain was observed in the line-width/coupling-strength for this A_{1g} symmetry coupled mode upon cooling across T_c. Furthermore, when examining the electronic Raman spectrum over the same energy range one can clearly see the development of some underlying, highly temperature dependent activity. Puchkov [68] was the first to emphasize that the Raman activity in this energy range, for both A_{1g} and B_{1g} symmetry, is expressly associated with the superconductivity and manifests the above (440/330) cm^{-1} or equivalently (53/40) meV, i.e. 4/3, energy relation to 2Δ. The upper feature here (which can be resonantly enhanced) is found to adjust its energy between different HTSC systems in step with T_c. In simple phonon-related Raman scattering, with a photonic probe, one is normally examining what transpires near the zone centre. However, in electronic Raman work direct excitation can arise from whichever k states hold 'addressable' Fermions, and this should be the case for the present composite bosons as well. The bosonic excitation in question here is perceived [14b, 69] as being not that of the Γ point condensate itself, but, as mentioned earlier, of the addressable

population of free pairs which remain outside the condensate. In the HTSC cuprates the latter mode is centred away from $\mathbf{K} = 0$ around each saddle point $\mathbf{K} = (0, \pi)$, etc. de Llano *et al* [70] in their theoretical treatment of BCS/BEC crossover have indicated that this bosonic excitation mode of finite-$\mathbf{K}$ pairs should exhibit linear dispersion in $\mathbf{K}$, the local-pair momentum, as against the customary quadratic variation. As has been detailed in [14b], and already alluded to here, I regard this to constitute the dispersed bosonic mode encountered in the original Fourier transform STM work [63, 2]. Gigahertz spectroscopy afforded the first direct evidence of the presence of these bosons outside the condensate [71]. The above mode has to be distinguished from that relating to the local-pair condensate itself and figuring first in the ARPES work. The latter much more weakly dispersed mode automatically takes a slightly greater binding energy of about 60 meV. It is this mode for which such a wide variety of origins have been proposed other than that advocated here—origins all invariably of B_{1g} rather than A_{1g} lineage [72]. The upper, more dispersed, pair mode is observed for k-values where it runs between E_F and $2\Delta^{max}$ [14b]. By contrast, as Belkhir and Randeria have deduced [73], the lower, condensate mode can form only at k values greater than ξ^{-1}—namely here (where $\xi \sim 4a_0$) beyond about halfway across the zone—precisely as is observed.

7. What STM 'gap' bit maps really disclose about cuprate HTSC

Many theoretical papers on the BCS/BEC crossover do not accommodate any negative-U aspect to events [74] (and the converse similarly is true [75]). It is very apparent, though, that to make decisive headway theoretically with the cuprates the inhomogeneous nature of these near-localized, mixed-valent systems must be appropriately embraced throughout. The stance taken by Loram and Tallon, for example, that the real situation is not in any way as fragmented as the STM work would suggest, appears to me to be a distinctly retrograde one, directly in conflict with chemical understanding as to where the HTSC systems stand. Loram and Tallon's earlier reported observations regarding the effect of a magnetic field upon specific heat data (see [76]) serve in fact to endorse what is currently being baulked at (see section E17 (p 1021) in [10c]). These comments are just as applicable in regard to most magnetic modelling of what arises in the HTSC cuprates [77].

Where the present confusion arises is in viewing the STM peak as representing the local superconducting gap energy as if it were the 'coherence peak' of an extrinsically perturbed classical superconductor, when it would look to me to mark the the negative-U state energy in a highly local, intrinsically inhomogeneous, non-BCS superconductor: a superconductor at the BCS/BEC crossover driven by the resonant energy location of an appropriate subset of the negative-U centres [13, 14]. At some given doping, diverse nanometre regions find themselves holding negative-U centres for which the energy is off resonance. When an optimally doped sample is replaced by an underdoped, less metallic, one, the number of subresonant centres mounts as their typical energy falls away from resonance, under the now less strongly screened trivalent Madelung potential to which they owe their existence as metastable doubly loaded entities. We are, then, talking about intrinsically different sorts of Swiss cheese, and with the cuprates it is not the type met with in $(Nb/Mo)Se_2$ or even $(Mg/Li)B_2$. Recall that we observed from the old STM work of Maggio-Aprile *et al* [78] that within field vortices established through an underdoped HTSC sample the negative-U peak section of the tunnelling response still persists, and it is only the inner, truly superconducting part of the signal which becomes suppressed.

This brings us finally to consider the intrinsic level of inhomogeneity in the inhomogeneity itself. That being picked up by Davis and colleagues [2, 62], being on the scale of 30–40 Å, definitely implies the presence of a degree of chemical dopant (oxygen excess) clustering which

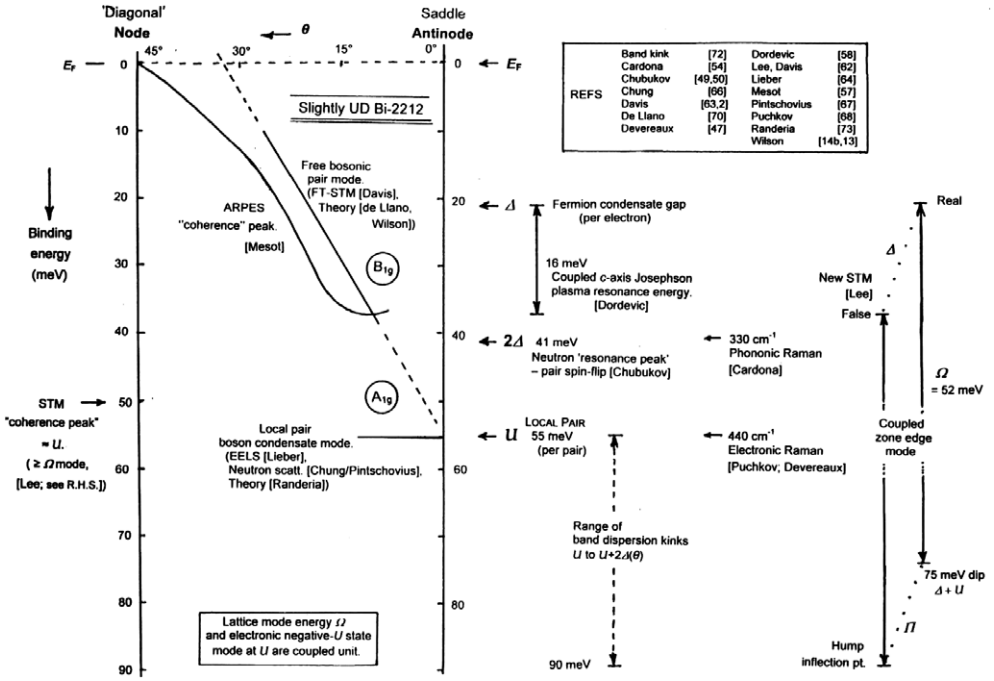

Figure 1. Representation of the composite situation adjudged to hold in slightly underdoped BSCCO, in which the negative-U pair state $^{10}Cu_{III}^{2-}$ formed from saddle states drives superconductivity on a system-wide basis by virtue of its near-resonance with the chemical potential. The local-pair state, strongly coupled to the lattice, takes on A_{1g} symmetry as a consequence of its closed shell and coordination unit breathing character. By contrast the Fermionic system acquires B_{1g} gap symmetry, in line with the $d_{x^2-y^2}$ band symmetry and the very short length scales relevant. The induced gap function is distorted towards the local-pair state, so that the measured $\Delta(\theta \to 0)$ become greater than anticipated from other measurements based on physics centred around the nodes ($\theta = 45°$). The local-pair state diminishes in binding energy, $\mathcal{U}(p)$, with doping, p, coming into resonance with the fermionic pair chemical potential at optimal doping. The local pairs are responsible for a weakly dispersed condensate mode and for a more strongly dispersed free pair mode. These have been sensed in the experiments indicated (references as in main text). The ARPES spectra are viewed as being decorated by a c-axis plasmon peak, rather than for this peak to represent the classical 'coherence' feature. The effect once again here is to contribute to an overstatement of Δ_0 as invariably extracted in ARPES papers. The information appearing on the right-hand side is first as it appears in the STM paper from Lee *et al* [62], and then after reappraisal in line with the above. Note that energy Ω is essentially energy $\mathcal{U}$, the two symbols simply emphasizing the lattice and the electronic aspects to the coupled negative-U local-pair condensate mode. This mode is supported only at short wavelengths $\lambda \leqslant \xi \sim 4a_0$.

is not simply statistical. In the latter case one would expect an inhomogeneity of somewhat less than half this length scale. Is the above experimental situation therefore an outcome of the sample annealing routine followed, or is it a surface related problem as posed earlier? The fact that Fischer and co-workers [79] have since recorded considerably less striking, presumably 'finer-grained', STM mappings would support the former conclusion. The *secondary level* of graining now being reported by Lee *et al* in figure 2 of [62] through their process of 'gap referencing' is much more on the scale of what might have been expected, and the fact that it displays too some indication of axial checker-boarding is clearly intriguing from the present point of view [14b]. How tunnelling into the negative-U centres must actually proceed is clearly intriguing, and it will be very valuable to find out how the results are modified by employing a

superconducting tip rather than the customary non-superconducting one—typically Au, W or Ir; perhaps one could use drawn, stranded Nb_3Sn wire.

8. A forward look on finalizing the negative-*U* understanding of HTSC

I have already pointed in [10a] to an appropriate cluster geometry with which one should attempt now to press forward to a formal dynamic cluster analysis [80] of the advocated inhomogeneous *negative-U* situation. The cluster suggested is quite large, but its analysis should become tractable within the near future. In order to encourage those whose *forte* this is to expend the time and money required, I would in closing like to draw attention to one final empirical result of much import to the current matter. An extensive study has very recently been circulated of a new system stated to manifest strong evidence for superconductivity at 84 K [81]. The material is not a layered perovskite but a cubic perovskite related one, and, moreover, it is not predominantly a copper-based system, but a ruthenium-based one. And yet the new material does contain copper. Indeed, from its highly oxidized stoichiometry, it once more must contain Cu(III) for most certainly it cannot contain solely Cu(II) in the doping-induced presence of Ru(VI). Accordingly with this new find, $Ba_2Y(Ru_{1-u}Cu_u)O_6$ with $u \sim 1/6$, one is again back at the very situation that originally confronted us in LSCO, etc. Namely of $^{8}Cu^{0}_{III}$ sites incorporated into a highly correlated σ^* d-band, open to fluctuational conversion to $^{10}Cu^{2-}_{III}$, with the latter state resonantly attuned to E_F as dictated by the one-electron system.

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Note added in proof. Following the recommendations of the referees the title of this work has been changed from the original used in *Preprint* cond-mat/0610887 so as better to represent the overall content of the paper. Subtitles also have been added to help to reveal the paper's structure. Finally, by way of summary, figure 1 ought to aid in unifying some of the points being made. The references on the latter are as within the main body of the text, the names of the leading authors being attached for convenience.

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