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Replace 'van Hove singularity' by 'negative-U singularity' and proceed: a comment on the analysis of HTSC Seebeck data by McIntosh and Kaiser—and related matters concerning the mechanism

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Abstract. It is shown how recent simple modelling and interpretation of the transport data from high temperature superconducting (HTSC) systems can be recast in terms of a negative-U model rather than through a standard van Hove singularity. Since it seems that U_{eff} should be ~ 0 for a negative-U circumstance to be optimally effective in raising T_c, the ideas of Khodel *et al* concerning fermion condensation are worked into the presentation.

In the paper referred to [1] McIntosh and Kaiser have presented a very impressive fitting of their large bank of Seebeck data, employing four diverse HTSC systems to illustrate the universal character of the modelling, namely Bi-2201–La; Tl-1201–Pb, La; Hg-1201–O_δ and Hg-1223–O_δ. Here we have single and multilayer systems, orthorhombic and tetragonal systems, quite ionic and more covalent systems. The work of Obertelli *et al* [2] was the first to demonstrate explicitly the high degree of universality apparent in such Seebeck data, once suitably scaled. That same ready scaling of the data from the HTSC systems runs too to the resistivity and Hall results [3]. This behaviour is a marker of the 'rudimentary' nature of the responsible agent; i.e. not dependent upon the details of structural and band structural form that the various HTSC cuprates present. Actually once chain-bearing YBCO 123 and 124 are set aside the structures of the HTSC materials do constitute a manifestly restricted and homogeneous group. The nearly 2D form of the $d_{x^2-y^2}$ band associated with the chessboard geometry of the fundamental CuO_2 arrays has led many people to look to the topologically required van Hove singularity within such a band as source of the unusual transport anomalies [1, 4]. It is evident that the resulting Fermi surface nesting of the saddle-point features indeed does play a significant role during the evolution of magnetic behaviour found in HTSC systems under hole 'doping' from a Mott localized antiferromagnet towards spin density wave formation [5]. However, the degree of nesting present in these systems has been overemphasized, and the detail of the incommensurate form to the LSCO neutron scattering data are better understood in terms of mixed-valence carrier segregation [6]. ARPES data on these quite highly correlated metals have been repeatedly misinterpreted too as implying a giant form to the inevitable band structural van Hove singularity—and this pinned in very close proximity to E_F. Recent ARPES work on Sr_2RuO_4 [7] has signalled once more the mistaken conclusions being made concerning BSCCO, etc. A wide variety of modern band structure calculations consistently have indicated that the LDA-derived van

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Hove singularity (vHS) does not of itself acquire peculiarly sharp or dominant form [8]. Likewise, there is evident little explicit register of such a standard DOS feature in static susceptibility or thermodynamic data [9]. What is more, standard calculation would suggest that, under doping, δ, the van Hove singularity, should adjust its energy always to fall a little way below E_F [10]. The present [1] (and at first sight successful) 'two-carrier' modelling from McIntosh and Kaiser of the normal state HTSC Seebeck data by contrast requires to see an exceedingly sharp peak brought right to E_F. This they incorporate within a narrow subband to yield a contribution to $\sigma(E)$ of p-type character almost equal in weight to the weight of the underlying 2D band. Note here the latter still makes a contribution of n-type sign—so conveying this as in some way a two-subsystem arrangement.

A two-element aspect to the body of HTSC transport behaviour was recognized early, though treated in very different fashion, after comparison first was made between the Hall and resistivity data [11, 12]. One such early attempt to reconcile all the data in terms of a very strong mobility difference between k-points near to and far from the van Hove singularities now has been abandoned [12]. However there still continues what appears to the present author [6] to be the mistaken pursuit of interpreting the transport data in terms of two quite distinct mobility behaviours relating to transverse and longitudinal motion within an (H, E) environment; this now embraces the anomalous magneto-resistance results [13]. A mobility separation was introduced originally by Anderson [11] in concert with his 'holon–spinon' separation, proceeding by analogy with a 1D Luttinger liquid. In an attempt formally to justify some such divergence in mobility behaviour, Kottliar *et al* [14] and Coleman *et al* [15] recently have elaborated a very ingenious scenario. Both papers involve questions of charge conjugation parity breaking and the chiral aspect this would bring to carrier scattering in an H-field—i.e. to the Hall and magnetoresistance data. However, as Coleman and co-authors note, there is a real danger here of introducing an even higher degree of complexity and arbitrariness than is exhibited in the original problem: indeed the skew scattering is now required to take on the mysterious $1/T$ dependence of the observed $R_H(T)$ data.

It is clear, as developed in [6], that a much simpler interpretation of the conundrums set by the transport data is to accept at face value that $R_H \propto 1/T$ might directly signal the active hole count to be slowly increasing with temperature. This does not mean transferring attention entirely from the carrier mobility to the carrier content, since, remember, there still arises the universal anomalous issue that μ varies as T^{-2} (to high T and with a very high prefactor—see the discussion in [6]). To take the carrier content as temperature dependent is a course of action which Wong and coworkers [16] have adopted in generating their recent remarkable simultaneous fits to the group's earlier ρ, R_H and S data. They have invoked in this, without realistic justification, a standard two-band, small gap formalism. The levels of fitting obtained are very comparable to those achieved by McIntosh and Kaiser in [1]; figure 3 of Wong *et al* discloses the detailed values of the fitting parameters used. Note the smallness of the extracted λ, equivalent to only a 25–50 meV excitation. The interpretive discussion given by McIntosh and Kaiser [1] is somewhat closer to that which follows.

It long has been appreciated, since the early work of Forro *et al* [17] and Moshchalkov *et al* [18], that such fitting parameters always emerge as remarkably small. One interpretation introduced by these workers drew attention to the parallel between the present HTSC situation and that occurring in heavy-fermion systems, where the f-electron ground state sits very close to E_F as established in a broad and interacting d band (see Freimuth *et al* [19]). Such a circumstance has led several to contemplate a parallel with the Kondo situation as existing in the HTSC systems [20]. However it is the long-held opinion of the

present author, bearing in mind all observed properties of the HTSC materials (not just the magnetic and transport data), and in particular the manner in which HTSC is confined to one very special niche in the periodic table, that a negative-U viewpoint is to be preferred [5, 6]. The negative-U centre plays then the role of the state embedded at E_F. The latter's origin, it is postulated, lies with the effects of $d^{10}(p^6)$ shell closure under double-occupancy fluctuations into those cuprate centres possessing a quasi-trivalent local charge count. It has to be recognized that the mixed-valence cuprate materials present a condition of structural inhomogeneity which transfers itself automatically to the electronic state.

The electronic circumstance as presented by McIntosh and Kaiser [1] within their analysis involves a standard but very narrow DOS saddle-point singularity, itself lodged within a narrow host band that emerges as no more than 0.1 eV in width. The feature as reported, what is more, does not have quite the expected characteristics. Besides its extreme narrowness it is required to sit upon the underlying broad band in such a way that E_F is always slightly above the saddle energy, and by only 1 or 2 meV at optimal doping. This arrangement would then have a negative Seebeck contribution from the broad band, while a comparable positive contribution arises from the narrow band. As the temperature rises and the 'Fermi window' increases in width the positive contribution is rapidly reduced and duly there occurs transfer to a net negative Seebeck coefficient. For 'optimal' doping early it was noted by Obertelli *et al* [2] that this crossover in Seebeck sign typically takes place somewhere around room temperature. The width of the above narrow band controls the height of the positive maximum in $S(T, x')$. From the parameters tabulated by McIntosh and Kaiser the DOS peak would appear across most of the HTSC range exceedingly closely pinned to E_F, as the latter changes in energy with doping. Ultimately at high doping E_F does move just below the DOS peak, so there then should arise a clearly pronounced lack of symmetry in the data toward the occurrence of positive and negative peaks in the Seebeck data. DOS peaks as sharp as the above are not, furthermore, really in keeping with the carrier masses, these being not particularly high ($m^* < 2\ m_e$) especially in the overdoped regime [9]. No allowance is made, what is more, in the McIntosh–Kaiser analysis for any broadening of the invoked narrow band with doping (i.e. screening). Lastly there is no evidence with systems possessing more than one CuO_2 layer per unit cell of the bonding and antibonding interactions between these leading to any multi-peak behaviour. Indeed there is no reason within a straightforward LDA band structural framework for E_F pinning to the degree imputed.

The analysis by Davidchack *et al* [16] proceeds on a quite different basis: namely increase in carrier count under thermal excitation from VB to CB (controlled by parameter λ). However λ emerges as so small that it is clearly not related to any sort of standard interband excitation of the type suggested by those authors. As discussed at length in [6], it is my understanding that the thermal production of free carriers arises from overcoming weak localization. The latter charge gap is induced by mixed-valence disorder, along with valence segregation and a tendency to disordered RVB pairing. RVB pairing is seen also as being responsible for the crucial low-temperature spin gap formation. The sign of all the quasi-particle carriers at $T = 0$ would appear to be p-type, not because of the detailed curvature of the bands as within a standard Fermi liquid, but because of the above proximity to localization, i.e. $\mathrm{d}(\sigma(E))/\mathrm{d}E$ is negative. Throughout the HTSC range the effective (hole) carrier count is very substantially below the actual $d_{x^2-y^2}$ band *electron* content, and as is well known is much more directly related to the 'dopant' count from the Mott-insulating, band half-filling, d^9 count. If ever, then, significant numbers of mobile *negative*-sign carriers were to be established located in the vicinity of E_F, particularly in underdoped HTSC systems, they would have to arise from a very distinct and unusual cause. Davidchack *et al*

in fact find from their simple model that it is necessary to suppress the way such carriers figure in their analysis, by assigning to them a low mobility, which is required to become ever smaller with underdoping. (N.B. There is a danger throughout their work which relates to $YBCO_x$ of getting caught up in the chain–plane question close to $x = 7$.)

Hence both these empirical papers [1, 16], despite their apparent success in curve fitting very substantial bodies of data, are on extremely dubious grounds when it comes to the actual physics involved. They are far too simple and conventionally based.

What these papers have not addressed in particular is the matter of advanced correlation, plus the related matter of electronic inhomogeneity, both essential to covering the HTSC circumstance [5b, 6]. Only when one goes beyond the LDA approach and indeed beyond mean-field approaches in general can an analysis become appropriate. What especially has promoted works of the latter type is the spin-fluctuation magnetic phenomena regarding the microscopic mechanism [21] and the ARPES results as regards the order parameter problem [22].

The experimental and theoretical work of Gofron *et al* [23], Dagotto *et al* [24], and Beenen and Edwards [25] has demonstrated how within the framework of a generalized Hubbard model (but still excluding any double occupancy) spin-fluctuation phenomena may become expressly associated with a very considerable sharpening up of the LDA van Hove singularities. Such sharpening is what, it has been claimed, is manifest in the 'giant anomalous extension' of the vHs features detected in angle-resolved photo-emission data above T_c [26]. However the recent ARPES observations in Sr_2RuO_4 [7] (a d^4 superconductor, isostructural with La_2CuO_4) ought to temper this view. It in fact has long been noted how photo-emission is severely modified in all sorts of families seated close to localization (see Cox *et al* [27] for the case of the ruthenates).

To come a little closer to an understanding of the formation and narrowing of the saddle points, Liechtenstein and coworkers [28] have attempted to parametrize their Hubbard treatment via reference to a full LDA band structure, including nearest- and next-nearest-neighbour interactions, etc. One feature they have found essential to include in their fitting is the second e_g band, based on $s/d_{z^2}/p_z$ interactions: it governs the apical Cu–O bond length, known to be a significant feature in the HTSC problem, associated with strong Jahn–Teller distortion and a coupled contraction of the CuO_2 planes. This widening of the modelling marks a degree of acceptance that there is more to HTSC in the cuprates than is contained within the $d_{x^2-y^2}$ band taken in isolation. An unstressed marker of this fact within the Liechtenstein paper is that their fitting proceeds with a (uniform) Hubbard on-site U reduced from the customary 7–8 eV down to 3.2 eV (or somewhat less than the bandwidth). There is also the statement made that the general results are not very sensitive to the 'real' U-value. In addition to moving to incorporate still more of the d states into the problem, there clearly is a need to improve much further the treatment of correlations, especially as regards fluctuations and spatial inhomogeneity.

A more highly generalized work has recently been undertaken by Onufrieva and Rossat-Mignod [29], using proper Hubbard operators, in an attempt primarily to understand the magnetic scattering results obtained by neutron experiments, particularly above T_c. This work becomes more clouded once one turns to the spin-flip scattering below T_c [6, 30]. Of course the magnetic spin-gap phenomena and HTSC are complexly intertwined problems, as photo-emission once again has recently emphasized in detecting the persistence of the DOS gapping to temperatures way beyond superconducting T_c for underdoped samples [31]. In these highly local systems it is inevitable that SDW, CDW and superconductive states are directly competitive and mutually dependent expressions of the complex underlying correlations.

A highly interesting development in this direction is that presented by Khodel and coworkers [32], which pursues a Landau quasi-particle picture into the strong-correlation regime where the Fermi sea can develop instabilities in its k-space geometry. These lead to state rearrangement and ultimately to fermion condensation. Unlike the case in a standard Fermi sea there is no discontinuity in n_p at $T = 0$ across the Pauli filling point, the quasi-particle lifetime becomes proportional to T, and the density of states or effective mass to $1/T$. A 'fermion condensate' develops on susceptible portions of the LDA Fermi surface, and one can end up with non-dispersing plateau-like sections and a two-subsystem circumstance of fermions and Cooper pairs able to support superconductivity. The latter arises when with strong correlations in play there occurs a circumstance in which U_{eff} becomes attractive. There is, of course, an enormous difference in the physical circumstances between $U = 0$, which is the situation in a fully screened free-electron gas, and a highly tight-binding case such as the HTSC materials. Then $U_{eff} = 0$ results from the cancellation of very large terms (see figure 1 of [6] or the figure on p 346 of the article by the present author [5]). The above treatment by Khodel and coworkers gives also the result that the scattering amplitude $\Gamma(q, \omega)$ is for low ω proportional to T^2. All the above characteristics are so familiar it is hard to believe this is not coming close to describing HTSC.

What now it seems is required is to make formal contact between the approach given by Khodel and the standard negative-U theory for local pair superconductivity, which has developed over the past few years following the early work of Friedberg and Lee [33] and Micnas *et al* [34]. Listed in [35] are some of the papers that have recently appeared concerning this topic. What one sees as lacking in much of this work (besides active interest in the source of the negative U) particularly as regards the HTSC materials, is that the level of mixed-valence inhomogeneity is not adequately incorporated [6]. However considerable progress is being made now in that direction by introducing CPA routines within a Bogoliubov–de Gennes approach to the superconductivity [36].

In much negative-U work it has become apparent that the maximum enhancement of T_c comes about when the value of $|U_{eff}|$ is approximately zero. I have previously expanded at length upon how I think such a value arises in the mixed-valence cuprates, this under double-occupancy fluctuations to achieve shell closure and state dehybridization at quasi-trivalent sites (see the articles by the present author [5, 6] and references therein). In describing such pair fluctuations and making distinction from the various static situations, I have employed a simple expanded 'chemical' notation that over the past 10 years I have found invaluable in labelling the complex set of configurations and settings relevant to HTSC. The energetics of all these states, as estimated from my earlier spectroscopic work on transition metal compounds [37], are to be found summarized in figure 3 of [38], or figure 3 of [39].

Using this background and notation, figure 1 is now presented as interpretation of what is to be inserted vis à vis the simple analyses of the transport data [1, 16] with which we commenced. The figure is at the same time close in spirit to the theoretical ideas injected by Khodel *et al* [32] as to how best a negative-U circumstance might be seen at the stage where $U_{eff} \sim 0$ (see figure l of [6]). In a (uniformly) trivalent cuprate (generalized as Cu_2O_3) the negative-U pair fluctuation state in question ($^{10}Cu^{2-}_{III}$) is asserted to lie somewhat above the (then) empty $d_{x^2-y^2}$ band (site condition $^{8}Cu^{0}_{III}$). However towards the divalent end of the mixed-valence system it (i.e. the double loading fluctuation $^{10}Cu^{2-}_{III}$) has become degenerate with E_F, which should rise more steeply in energy between the two extremes of valence mixing. Of course as the divalent limit is approached the number of sites closely affected by the chemical substitutions (and to which the fluctuation $^{10}Cu^{2-}_{III}$ is specifically relevant) is rapidly moving towards zero. The reason that these fluctuations remain in evidence is by virtue of the Mott-insulating character displayed at d^9, so that for light doping (from

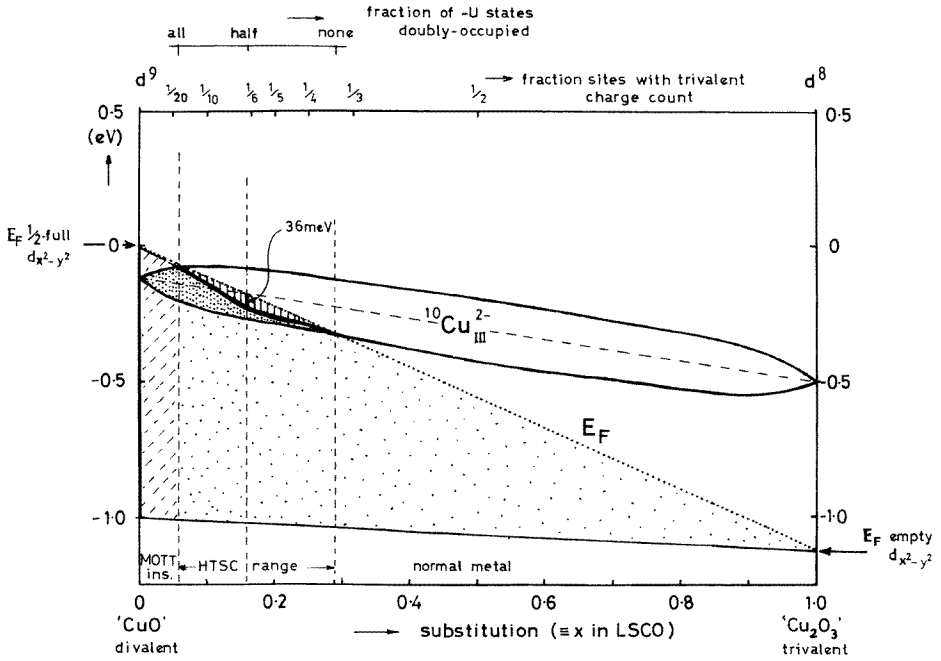

Figure 1. The scale diagram indicates the *suggested* (see text) relative energy location across the mixed di-/tri-valent HTSC cuprate regime of the Fermi energy in the $d_{x^2-y^2}$ band as opposed to the negative-U pair fluctuation state $^{10}Cu^{2-}_{III}$. The band is empty at d^8 and half-filled at d^9, and at both these extremes is Mott insulating. Superconductivity is supported through the overlap region, the situation being optimized in the vicinity of $x = \frac{1}{6}$ where the pair fluctuation state is exactly degenerate with E_F—see the text.

At $x = \frac{1}{6}$

half-width of negative-U state $= 72$ meV

$\frac{1}{2}$ half-width, doubly occupied $= 36$ meV

so stabilization per electron $(\frac{1}{6}\times) = 6$ meV

implying

gap excitation energy $2\Delta(0) \quad \sim 12$ meV.

Mott insulation and appearance of local moments prevent the HTSC range extending below 6% substitution.

the latter reference point) there is little metallic screening and the dopant centre is local and well defined electrostatically. I discussed in some detail in section 4 of my article [5] how the degree of definition secured for this state acts as control over the value of T_c attainable within and between particular HTSC cuprate families. This control is seen as being achieved by the particular selection of the counter-ion involved (in addition to applied pressure): too ionic (as with LBCO) and the system becomes too magnetic or undergoes charge segregation and superlattice formation (see appendix B of [6]): too covalent and the state is broadened so much through charge delocalization that the pair fluctuation can no longer become effective in seeding superconductivity (as with certain Bi-2201). Hg-1223 currently marks the optimum in this regard.

The figure, as presented, would have a fermion condensation possible over the observed HTSC range. Here the upper limit falls just short of 30% substitution, optimization occurs around 16% (when state centre and E_F are degenerate), and a lower limit of around 6% is perceived as set by Mott localization, uncontrolled spin fluctuations, and a rapidly decreasing

population of negative-U centres. The portrayed effective breadth acquired by the negative-U state follows from the degrees of inhomogeneity versus delocalization. Inhomogeneity is of more import towards the Mott insulating ends of the mixed-valence system (remember d^8 $LaCuO_3$ and $KCuO_2$ are each Mott insulators), whilst delocalization is more significant toward the centre of the mixing range.

If one were to push one's luck with figure 1, one might say that it also displays an estimate of the superconducting gap energy. The chemical potential at $x = 0.16$ is lowered by ~ 36 meV (due to double occupancy of the negative-U state). Per *unit* electron equivalence this becomes 6 meV, giving a $2\Delta(0)$ gap value of 12 meV, much as for LSCO. Of course the figure was constructed with this potential in mind, but the fact that it could be accomplished satisfactorily within the one modelling is of reassurance, and was not all that easy to achieve, given the energy constraints which derive from the earlier quoted figure (figure 3 of [38] and [39]).

It is hoped this paper will stimulate both experimentalists and theorists alike to cast their work on the HTSC problem in the negative-U manner outlined here and see what emerges.

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