Classification
Physics Abstracts
74.20 — 75.10

Magnetism and high T_c superconductors

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(Received 27 July 1992, accepted in final form 1 September 1992)

Abstract. — The magnetic properties of the copper oxide planes in hight T_c superconductors are represented by a planar 2-d antiferromagnet on a square lattice. We model the magnetic effect of the addition of holes through doping by the replacement of antiferromagnetic with ferromagnetic plaquettes thereby facilitating the creation of vortices/antivortices. These magnetic arrangements lead to pair formation of the holes (Cooper pairs) at low temperatures. We explore some of the consequences of these results for high T_c superconductors.

Introduction.

There are structural and magnetic characteristics common to all high T_c superconductors. The main structural feature of these superconductors is the well defined copper oxide (CuO_2) planes separated from each other by intervening non-magnetic layers. The copper atoms in the CuO_2 planes are approximately arranged at the corners of a square lattice with a separation of 3.8 Å. Between each pair of copper atoms in the plane is located an oxygen atom [1]. Magnetic features common to all high T_c superconductors also exist. A simple valence argument based on closed shells and the use of Pauli's exclusion principle suggest an antiferromagnetic insulator with a hole of spin 1/2 localised on each copper ion [2, 3]. This magnetic feature is indeed common to all the undoped material from which ceramic high T_c superconductors are formed.

The CuO_2 planes in e.g. La_2CuO_4 thus consist of localized spin = 1/2 holes arranged in a 2-d square lattice with antiferromagnetic interactions. These spins are experimentally observed by neutron scattering [4] to be essentially confined to the plane. There appears to be strong consensus that the magnetic degrees of freedom of the undoped La_2CuO_4 are well described by the spin 1/2 antiferromagnet Heisenberg model [5]. La_2CuO_4 transforms from an antiferromagnetic insulator into the $La_{2-x}Sr_xCuO_4$ superconductor when doped with Sr ions. Strontium donates 2 electrons instead of 3 and an additional hole is formed in the appropriate unit cell. This hole is associated with the O^{-2} ions in the CuO_2 planes since there is a strong Coulomb repulsion associated with a double occupancy of the Cu (3d) states. Experimental evidence from electron energy loss spectroscopy [6] which examines 1s-2p transitions supports the presence of holes on the oxygen since this transition is only observed when doping occurs indicating the presence of O^-.

It has not yet been experimentally determined if this oxygen hole forms on the p_σ orbital extending towards the neighbouring copper atoms or on the p_π orbital extending perpendicular to this axis. Band theory calculations [7] favour occupation of the p_σ rather than the p_π orbital because the p_σ states have a better overlap with the Cu $d_{x^2-y^2}$ orbitals and thus a lower kinetic energy. This overlap produces a strong antiferromagnetic coupling, due to Pauli's exclusion principle, between the hole associated with the oxygen ion and the two neighbouring holes on the copper ions. Summing over the spin variable associated with the hole on the oxygen ion gives an effective ferromagnetic coupling between the adjacent Cu atoms. Thus the effect of doping on the magnetism of the CuO_2 planes is to produce a ferromagnetic bond between 2 adjacent copper atoms.

In our model the ferromagnetic interactions are extended to include all the bonds in a plaquette (FP) and this leads to a dramatic increase in the probability to form vortex/antivortex excitations. These excitations persist down to $T = 0$ but only if the FP's are nearest-neighbour pairs. This is a natural mechanism for the formation of real-space paired holes, bound together by the magnetic attraction between the V and its AV. Note that pair-formation of the charge-carriers is a necessary condition for superconductivity but we have not, as yet supplied all the sufficient conditions.

The model.

We now describe our model which includes these structural and magnetic characteristics for high T_c cuprate superconductors. The model, derived from a model proposed by Aharony *et al.* [8, 9], consists of a 2-d square lattice with unit vector spins free to rotate only in the plane. These spins represent the magnetic moments of the holes on each Cu atom and these holes remain localised with subsequent doping. Since the magnetic coupling between CuO_2 layers is very weak compared to the intraplane interactions, a 2-d square lattice is considered with antiferromagnetic nearest neighbour interactions [10].

Aharony *et al.* [8] proposed a model where the hole associated with the oxygen ion is represented by one ferromagnetic bond. However experimental evidence [11] seems to indicate that the hole is delocalised over a slightly larger area. For this reason, we choose to model the hole by making an entire plaquette of 4 bonds ferromagnetic. These bonds are equal in magnitude to the antiferromagnetic ones. Another possibility is that 4 bonds extending radially from one Cu atom are ferromagnetic. This would produce no magnetic frustration as a few unique arrangements of spins could easily satisfy all the bonds of the lattice, and following Aharony *et al.*, we wish to retain the element of frustration, which we consider to be essential. A possible scenario leading to such a spread of the hole is a partial occupancy of the p_σ and p_π states where occupancy of the p_π leads to overlap with the p_π states of the other interstitial oxygen ions on the plaquette and occupancy of the p_σ leads to overlap with the spins on the neighbouring copper ions and hence the effective ferromagnetic interactions. We assume that these elements are also retained in the conducting phase.

Our model therefore consists of a 2-d antiferromagnet of planar spins with a dilute concentration of ferromagnetic plaquettes (FP) which are the magnetic consequences of the holes introduced by doping

$$H = -\sum_{ij} J_{ij}\, \bar{s}_i \cdot \bar{s}_j \tag{1}$$

where $J_{ij} = J$ for ferromagnetic bonds and $= -J$ for antiferromagnetic bonds.

Monte Carlo method.

Monte Carlo simulations of this model were done according to a technique outlined in detail in reference [12]. We summarise the essential elements : 1. A site, i is visited at random and the resultant vector, $R_i = \sum_j J_{ij} \bar{s}_j$ is determined from summing the spins of its neighbours. 2. The interval between 0-4 is discretised in intervals of 0.02 and the magnitude of R is rounded to the nearest multiple of 0.02. 3. A lookup table of angles is precomputed for each of the discretised value of R and temperature where the frequency of an angle in the table is proportional to its Boltzmann factor. The standard features associated with a Monte Carlo simulation are retained [13]. The simulations were done on a 30 × 30 lattice for approximately 200000 MC steps at each temperature. Sampling was done after every 5 MC steps.

Magnetic consequences of the ferromagnetic plaquettes (FPs).

To model the magnetic consequences of doping La_2CuO_4 with Sr^{+2}, we insert FPs in the lattice and measure the number of vortices/site. A vortex (V) is a configuration of spins where the sum of the differences in spins angles around a plaquette is 2π or -2π for an antivortex (AV). From the Berezinskii-Kosterlitz-Thouless (BKT) theory [10], in a system without frustration, Vs and AVs form bound pairs below a transition temperature measured at 0.9 J/k, while above this temperature they are unpaired and form a vortex gas. The density of vortices, N_v increases with temperature and figure 1 shows a plot of ln N_v (the average number of V/AV pairs per MC step per site) *versus* $1/T$ for no holes (planar model), 1 hole and for 2 holes on

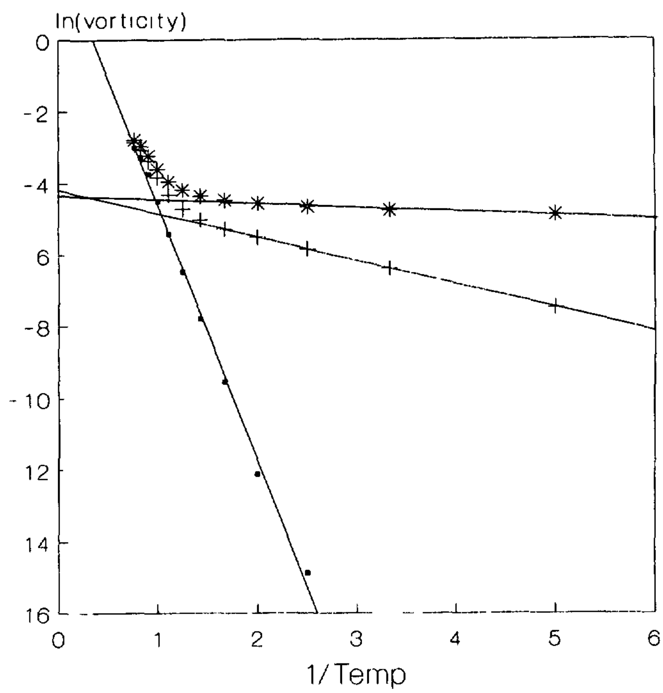

Fig. 1. — ln of N_v (number of V/AV pairs) *vs.* $1/T$ for a 30 × 30 system with 0 holes (0), 1 hole (+) and 2 adjacent holes (*). The slope is the energy required to create a V/AV pair. See text for details.

adjacent plaquettes. We see a straight line behaviour over a wide temperature range which indicates that

$$N_v \sim \exp(-E/kT)\,. \tag{2}$$

The slope is a measure of the energy required to create a spontaneous V/AV pair. This energy, E is $E_0 = 7.1$ J with no FP (planar model), $E_1 = 0.7$ J for 1 hole and $E_2 \leqslant 0.1$ J for 2 adjacent FP. The energy to create a vortex pair drops more than 70 fold when a pair of holes is inserted because of frustration caused by the FP. The density of V/AV pairs at low temperatures is therefore greatly increased by the existence of FPs. The enhancement of V/AV by a hole implies a binding energy $E_1 - E_0 = 6.4$ J between a V and a hole. Moreover, although vortices disappear at $T = 0$ if no hole or just one hole is present, V/AV pairs persist down to zero temperature for two adjacent holes. The presence of V/AV pairs at $T = 0$ indicates that these are not thermal excitations but a distinctive ground state.

Diffusion of single holes.

Next we address the diffusion of the doped holes in the background formed by the antiferromagnetic CuO_2 plane. To study this point we randomly place the FPs on the lattice and each FP (hole) can diffuse with probability of $p = 0.5$ to a neighbouring V or AV. Following reference [14] we consider the V/AV as a double well potential for the hole, which can occupy either the AV or V with the same energy. The hole has a finite probability to move from the vortex (say) to its paired antivortex. There may be an effective barrier that restricts the movement of the hole but we have no means of reliably estimating its height within our model. Such a barrier reduces the mobility of the hole which has been calculated [14]. This reduction would simply rescale the results obtained here. The exact value of p is not critical as we perform 20 Monte Carlo steps for equilibration before each attempt at diffusion. The results of the previous section points to an attraction between a hole and a vortex, and therefore if a vortex is not on the plaquette with the hole but adjacent to it, the hole is permitted to move to the plaquette containing the vortex thereby effectively lowering its potential energy. We shall denote by H the state of a hole « dressed » with a vortex and S to denote a thermally excited vortex located on a plaquette without a hole.

This mechanism allows the hole to move through the lattice provided that vortices are present in its immediate vicinity. At very low temperatures, the density of S excitations drops to 0 and diffusion is impossible by this mechanism for the solitary hole. As the temperature increases, diffusion increases as S excitations become more common and the hole performs a random walk. At low temperatures V/AV pairs are only created in the vicinity of the hole. We find in our simulations that for temperatures up to 0.5 k/J there is complete correlation between the FP and a plaquette with either a V or AV, namely, in 100 % of the observations a V is found within 2 lattice spacings of the FP. This can also be seen in figure 1. At even higher temperatures, the diffusion of the hole is dominated by thermal energy and the dope-induced magnetic influences become negligible.

When 2 separate holes or FPs are arbitrarily placed in the lattice and allowed to move, they diffuse at moderate temperatures until their positions are adjacent. When this occurs they pair into a single H-H excitation. The holes are no longer able to diffuse away from each other and are bound. We consider the Coulomb repulsion between the holes to be strong enough to prevent two holes from occupying the same plaquette. It is possible during the course of the simulation for the vortices to drift away from the holes, and when this occurs each hole creates its own S-H excitation. Such a separation occurs more readily if we take into account the coulombic repulsion. This process leads to two independent S-Hs, each with a single hole.

Figure 2 shows a plot of the average number of Monte Carlo steps in which the holes remain within two lattice spacings of each other while otherwise independently diffusing through the lattice. A significant feature here is that below $T = 0.15$ k/J, the lifetime of the hole pair (H-H) is practically infinite and the holes remain paired by the V/AV for the duration of the simulation. Note that this does not imply that all the V/AV pairs are always on the FPs — but only that no S excitations are created which will allow separate diffusion of the holes to take place. The temperature of $T = 0.15$ k/J may be considered as the upper bound of the superconducting transition temperature for this class of superconductors, because above it pairing of holes is not permanent.

We now consider stationary holes and ask what is the lifetime of a H-H excitation at a given temperature. Thermal fluctuations can disengage the V/AV from the adjacent holes on which they « sit ». To measure this we simulate two adjacent FPs and monitor the percentage of H-H. The results are displayed in figure 3. At very low temperatures H-H pairing is a dominant event, for example 83 % of all V/AV created are located on the adjacent holes. As the temperature increases slightly to 0.4 k/J the percentage of paired holes drops to 20 %. At this temperature well below the BKT temperature, the V/AV pairs are still firmly bound, but many of these V/AV pairs are displaced from the holes. This allows holes the potential to form H-S pairs and diffuse apart. At a temperature of 0.05 k/J almost all the V/AV are part of a H-H. We propose that this temperature constitutes a lower bound for the superconducting transition

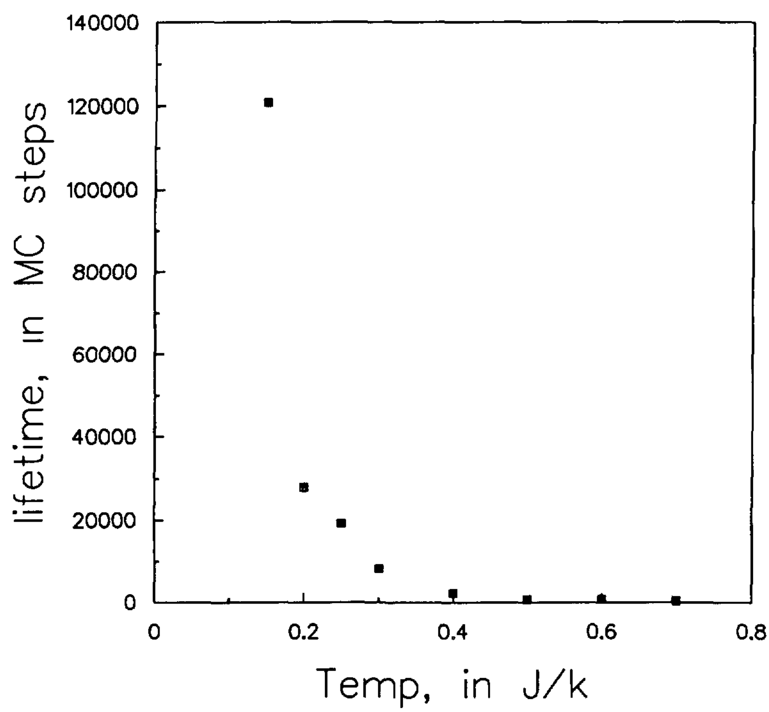

Fig. 2. — Number of MCS (Monte Carlo Steps) for which the V/AV pairs are bound to neighbouring holes (ferromagnetic plaquettes) and where the holes may diffuse but only if an « empty » V or AV is a neighbour. Below a temperature of ~ 0.15 T the magnetic V/AVs are essentially bound to two adjacent ferromagnetic plaquettes. This temperature may be considered as an upper bound to this class of superconductors.

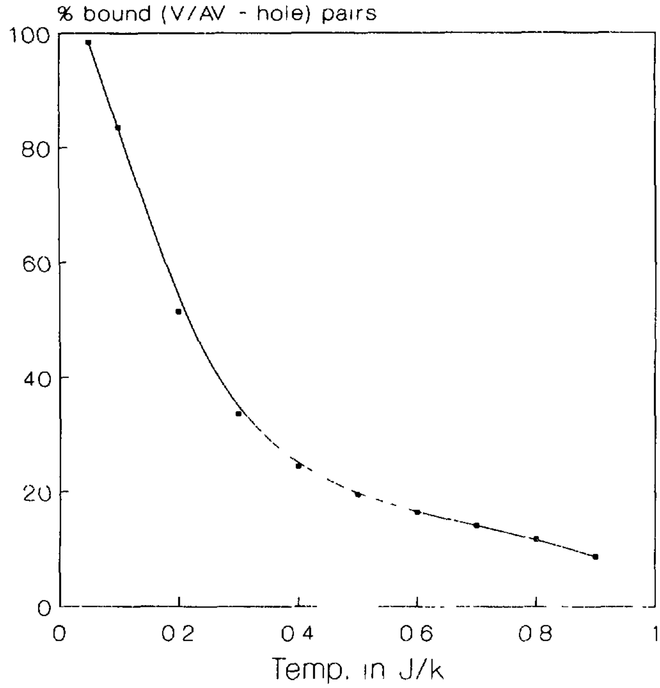

Fig. 3. — Fraction of V/AVs formed on two static adjacent ferromagnetic plaquettes (holes). This is a measure of bound pairing where diffusion between the plaquettes may occur by any means including magnetic ones. This is a lower bound to the optimal superconducting transition for a moderate concentration of holes.

temperature since only for $T < 0.05$ k/J the pair of holes is stable against *all* forms of hole diffusion not only as above where $T = 0.15$ k/J from magnetic excitations.

Figure 4 shows the energy of the ground state as we vary the distance between two FPs. We consider the undoped system as our zero-point energy. The solid point at separation = 0 is the energy of a single plaquette. When the separation between the two plaquettes exceeds 7 lattice units the energy saturates and the FPs can be considered as independent. As we move the plaquettes closer together we find that there is a sharp drop in energy reaching a minimum when the plaquettes are 1 unit spacing apart. The energy increases to 8.0 units when FPs are placed on the same cell. Coulombic repulsion may increase the distance at which the minimum energy occurs to more than 1 lattice spacing.

In most annealed magnetic models with a mixture of ferromagnetic and antiferromagnetic bonds the ground state energy is minimised by forming ferromagnetic and antiferromagnetic regions. Such a ground state would invalidate our pairing mechanism at low temperatures. To test if clustering occurs we have considered three holes in our system and allowed them to diffuse in the manner described above. However if a hole finds itself next to another hole and a V/AV pair forms on them they are considered bound and immobile. Otherwise they are allowed to wander on. This is a crude attempt to take into account coulombic repulsion and to note its effect on clustering of the ferromagnetic bonds. We find no evidence of clustering of three holes at low temperatures (0.1 k/J to 0.2 k/J). Adding further holes should inhibit the formation of domains due to the increased coulombic repulsion. There is an attractive interaction we have not considered, namely, the dipolar attraction between V/AV pairs of the

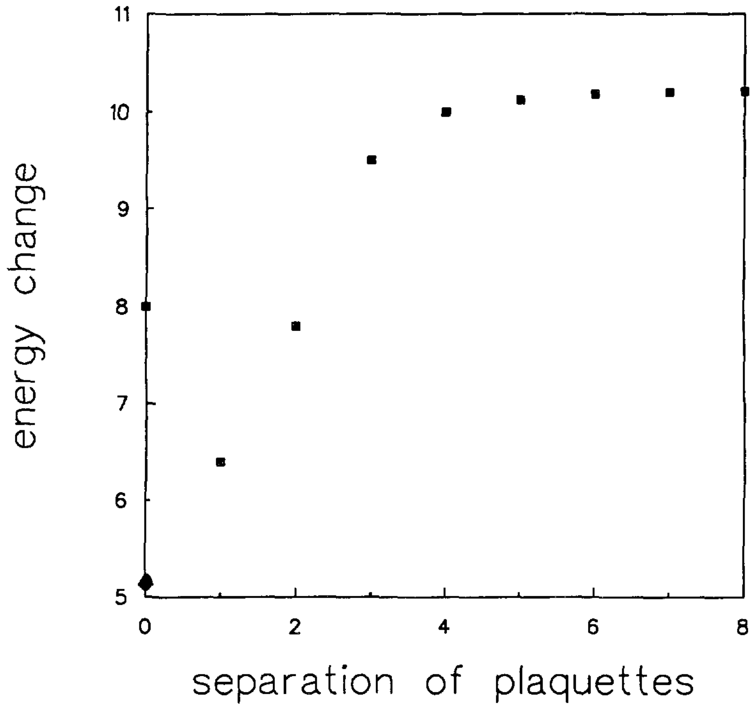

Fig. 4. — Energy change in units of J at $T = 0$ for two ferromagnetic plaquettes as a fucntion of distance. The energy is a minimum when these plaquettes are adjacent to each other. The energy of a single plaquette is shown (◆).

H-H excitations. This attraction is not strong enough to lead to clustering but may provided the necessary attraction for a Bose-Einstein condensation. Thus we conclude that provided the V/AV binding energy is greater than the coulombic repulsion, H-H pairs will form, rather than clustering of the FPs.

Consequences for superconductivity.

We now consider the implications of our results for superconductivity. If the magnetic effects of the holes introduced by doping the antiferromagnetic insulator lead to FPs as described here then these plaquettes enhance the creation of V/AV pairs by two orders of magnitude. We have found that the magnetic energy of the excitations is minimised if the FPs are adjacent, which leads to magnetically induced pairing of the holes. We have observed that an isolated hole which is permitted to oscillate from an antivortex to its neighbouring vortex diffuses until its position is adjacent to another isolated hole. When this happens pairing occurs with high probability and its lifetime depends on the temperature. We have assumed that the spins localised on the copper ions are in equilibrium, i.e. the time scale of a spin flip is shorter than the diffusing FP. We also assume that this is true even in the superconducting phase.

At high temperatures we expect to observe normal conduction and short range spin fluctuations. As the temperature is lowered correlations appear among the vortices, and at the BKT temperature, $T = 0.9$ k/J, the vortices become paired but due to the abundance of S-S excitations the density of pairs is greater than half the density of holes. There will be H-H pairs, S-H pairs and S-S pairs. As the temperature is reduced to $T < 0.15$ J/k the V/AV pairs consist almost exclusively of H-Hs. Reducing T further does not change the number of frustration induced V/AV pairs any more, which persist down to $T = 0$. As the density of holes is increased by doping it will be impossible at some point to localise the V/AV pairs on the holes even down to 0 K. Preliminary results indicate that this would occur at a density > 0.33.

If our scenario is correct then these superconductors are real-space paired holes bound together by magnetic interactions. At sufficiently low temperatures Bose-Einstein condensation to the superconducting phase occurs. In this phase there are spin fluctuations (representing the wake of the bound pairs) due to the rearrangement of the spins as the bound pairs of holes move through the system. This is expected at all temperatures in the superconducting phase and has been observed experimentally [15] but this would indeed be true for all spin-bipolaron theories. At temperatures above the superconducting transition but still below the BKT transition we should observe all three spin excitations : H-H, H-S and S-S. These excitations are identical with those of the spinon/holon theory of Laughlin and others [16], and it has been speculated that this is the classical analog of the fractional charge picture [16b]. At high densities of holes it will be impossible to achieve the pairing necessary for superconductivity because of the destabilising effect of the close proximity of the V/AV pairs to each other. At low concentration of holes the rate at which the system is cooled may determine whether there is a superconducting transition or not. It is possible e.g. for the system to be quenched rapidly from a temperature where the V/AV are mostly of the S-S and S-H type to a low temperature where these can no longer be excited, not even in the vicinity of a single hole, as our simulations show. A gentle rate of temperature decrease will allow magnetically induced diffusion and eventual pairing to take place.

The BKT transition has been used to describe the superfluid transition and superconducting transition of thin films [17]. In this approach the important pair excitations which destroy the « superfluidity » property are bound below, and are free above, the BKT transition. The unbinding of the V/AV pairs is used to describe the onset of the normal phase. The bosons of the superfluid are assumed to be present at the onset at all temperatures. Here we are proposing a new picture in that the *creation* of the bosons which eventually become superconducting is due to the attraction between magnetic vortices formed by the holes. These V/AV are paired below the BKT transition of 0.9 k/J and are tightly coupled to the holes at a lower temperature of about 0.15 k/J.

Some of the features of this model are close to those of other spin-bipolaron theories (de Jongh [18], Goddard [19], Schreifer *et al.* [20], Micnas *et al.* [21], etc.). Mott [22] e.g. assumes that the bosons exist above the superconducting transition temperature to account for the linear rise in the resistivity with increasing temperature up to 1 000 K. This phenomenon is easy to understand in our picture since 1 000 K is around the T_{BKT} for La_2CuO_4 ($J \sim 0.1$ eV) and we expect that most holes would exist as H-H or S-H. A measurement of the charge carriers in this temperature range would probably show a mixture of 2 e and e with the ratio of 2 e to e decreasing with increasing temperature. Stamp *et al.* [23] (see also Wiegmann and Dzyaloshinskii *et al.* [24]) have already considered the possibility that a BKT transition describes the onset of superconductivity in the ab planes of $YBa_2Cu_3O_{7-\delta}$. They suggest that the excitations exist in the normal state, *do not carry flux* but pair up at T_{BKT} to form quasi two dimensional superconducting condensates. There is some controversy on the nature of these excitations and whether there is indeed flux. There are two transitions implicit in our approach, the BKT transition for the pairing of the vortices (no flux) and the bound-pairing of these with holes (flux). Under certain conditions of doping these two transitions may be close to each other in temperature and we would expect an observation as reported by Stamp *et al.* In other cases where the transitions are widely separated then the normal BKT transition associated with a planar superconductor will be observed i.e. the excitations will carry flux when the transition occurs from the superconducting to the normal state [25, 26].

Conclusion.

Many authors have focused on the magnetic properties of the high T_c superconductors as essential to the understanding of superconductivity and we have referred to some of them above. In addition a few have highlighted the creation of magnetic vortices as an essential element in the pairing mechanism [27] but in a somewhat different manner to the approach we have taken here.

We have considered the magnetic properties of the CuO_2 superconductors and have modelled the CuO_2 planes as a planar antiferromagnet. We have assumed that the holes introduced by doping create ferromagnetic plaquettes. At low temperatures these plaquettes act as seeds for V/AV pairs which in turn bind the holes in pairs (H-H). We have found that the H-H pairs persist at $T = 0$ in contrast to S-S and S-H excitations that disappear at zero temperature. We expect a Bose-Einstein condensation of the hole-pairs as observed for ^{4}He at its transition to superfluidity. This occurs when the typical distance between neighbouring bosons is less than a threshold distance proportional to the de Broglie wavelength. In the same manner, we expect our bosons, the hole pairs, to undergo the transition to superconductivity when their typical distance of separation is small enough, which happens at low temperatures and a high enough density of hole-pairs. The superconducting phase diagram follows naturally from this — the number of bosons increases with the increase of holes and the transition temperature is expected to increase with the concentration of bosons (compare the temperature variation of the λ-transition as the fraction of ^{4}He bosons is increased in ^{3}He/^{4}He mixtures). A too high concentration of holes would bring about a decrease in the transition temperature as a result of overcrowding of the V/AV pairs thereby destabilising the pairing mechanism [14]. At a concentration of 0.33 at $T = 0.10$ k/J we find that the V/AV are no longer localised on the holes. We postulate that at low concentration of holes the *onset* of the superconducting phase may depend on the rate of cooling. We have proposed a specific mechanism at low concentration of holes on how the onset of the superconducting phase may depend on the rate of cooling, namely, at a slow rate the number of H-H pairs increases, leading to a higher critical temperature. Our results further suggest that the highest transition temperature should be between 0.05 k/J and 0.15 k/J. It may be possible to experimentally detect the presence of V/AV pairs [28].

Acknowledgments.

We would like to thank the following for criticisms, discussions and encouragement : Sir Neville Mott, J. M. J. van Leeuwen, H. E. Stanley, A. Aharony, A. Coniglio, L. J. de Jongh, J. Vannimenus and J. M. Wheatley. DLH and NJ are supported by grants from NSERC, Canada and RB is supported by a grant from SERC, UK.

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