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Short Communication

Geometrical correlations and the origin of x values at the maximum and intersects of $T_c(x)$ in $La_{2-x}(Sr)_xCuO_4$

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Abstract. — Many observations in doped $La_{2-x}(Sr)_xCuO_4$, including the $T_c(x)$ curve, are explained by a simple model of the holes distribution in the CuO planes. The excitation cloud surrounding a single hole is argued to spread on no more than about one plaquette. The Cooper pairs are proposed to be in a combination of at least two possible states, whose energy difference is smaller than the barrier between them. The experimental value of highest T_c doping concentration $x_0 = 0.15$ emerges as the limit when the pairs are most densely packed. Further doping above x_0 is proposed to destroy superconductivity by generating normal islands, leading to a percolation-type structure. The model explains the observed disappearance of superconductivity above $x \approx 0.3$. It also accounts for observations of nonuniform x-dependent diamagnetic phase separation. Assuming low mobility of single hole excitations, this picture applies to $x < x_0$ as well, in agreement with recent heat capacity measurements, and explains the observed onset of superconductivity at $x \approx 0.075$.

It is well established that to understand the phenomenon of superconductivity in the new high temperature superconductors one should concentrate on the action in the CuO_2 planes [1]. These planes in the pure undoped system (e.g., La_2CuO_4) can be envisaged as consisting of spins located on the Cu^{2+} atoms, with antiferromagnetic nearest neighbour interactions. The experimental evidence [2] suggests that doping by Sr to form $La_{2-x}(Sr)_xCuO_4$, induces holes onto the planar oxygen atoms (i.e., in the CuO_2 plane), which are believed to form the pairs that carry the supercurrent. The magnetic effect of these holes, that lie roughly between such nearest neighbour Cu atoms, is to introduce an effective ferromagnetic interaction between the Cu spins [1, 3-5]. Such an interaction creates magnetically frustrated plaquettes, namely, plaquettes on which the four surrounding spins cannot be in a state that energetically satisfies all 'bonds' simultaneously.

It was recently proposed in both classical [6] and quantum [7] contexts that the pairing of two oxygen holes is mediated by vortex-like spin excitations. In this model the hole may hop between neighbouring planar oxygens and around plaquettes, thus fitting with the belief that such a hole is shared by more than one oxygen [1, 2]. The rotation around a plaquette introduces frustration, which greatly enhances the probability of exciting a vortex configuration in the spin system. In the following we will denote such an excitation around a hole by H. At temperatures below the Kosterlitz-Thouless transition temperature such a vortex excites in turn a neighbouring spontaneous antivortex S, which is not attached to any hole [8], and a SH pair of vortex/antivortex excitation is formed. This excitation can move in the plane, *via* self-bootstrapping [9] and it was found that if two such SH excitations come close enough, the lowest state for this complex is of one vortex/antivortex (bivortex) excitation, HH, where each hole pivots a vortex-like configuration. Thus there arises an effective pairing between the holes, mediated by the attraction between the vortices. The typical lifetime of such a SH excitation before pairing has been calculated at low carrier densities [9]. This model also led to a recent prediction of a new resonance in the low doping pre-pairing regime.

Experimental evidence [10] suggests that the superconducting compound is not homogeneous and two types of spatial domains may be observed depending on the value of x, the doping concentration. One type of domain entertains Meissner diamagnetism, while the other does not. In $La_{2-x}(Sr)_xCuO_4$ for $x = x_0 \approx 0.15$, where T_c is observed in numerous experiments to be maximal, only the first phase is seen, while the other shows up with increasing x, until superconductivity disappears altogether for $x > 0.32$ [11]. This has long been known to be a problem in measurements and calls were made to incorporate this phenomenon into theories of these compounds [12].

This is the first aim of this paper. I propose that the distribution of the paired holes in the CuO plane can be described in the context of the percolation model. Namely, that domains fully occupied with pairs form a nonuniformly distributed superconducting cluster that spans the plane. This leads to some constraints on the range of x for which superconductivity can exist. It also excludes the possibility of the single hole excitation cloud being spread on more than about one plaquette, fitting in with the above bivortices pictures. It thus implies that models for pairing mechanisms that rely on excitations of wider spreads are inconsistent with these simple considerations. It will also be shown that the observed value of x_0, at which T_c is maximal, corresponds to the densest possible packing of at least two pair states that are slightly shifted in energy. It will be further suggested that increasing the doping concentration above x_0 disrupts the local dense packing and pairing. This leads to an in-plane percolation-like quenched structure of the superconducting wave function, making it possible to deduce $x \approx 0.3$ as the *maximal* doping above which superconductivity disappears for any $T > 0$, as indeed observed [11]. Assuming low mobility at low doping, this picture may be extended to the regime $x < x_0$, leading to explanation of the observed $x_{\min} = 0.075$ as the *minimal* doping below which superconductivity disappears. In addition to the accurate explanation of $x_{\min}$, this picture is supported for $x < x_0$ by recent heat capacity measurements. To the best of this author's knowledge this is the first model that yields x_0, $x_{\min}$ and $x_{\max}$ without resorting to different mechanisms on either side of x_0.

Let us first consider the two different pair excitations in figure 1. In 1a we have a HH excitation for which the holes are located one lattice parameter, d apart ($|a\rangle$ state), while in 1b ($|b\rangle$ state) the separation is $2d$. If the entire lattice is covered by either one of these configurations we have a 'superstructure' of either of the basic building blocks in figure 1. The unit cell of this structure is $S_a = 12$ plaquettes large for $|a\rangle$ and $S_b = 15$ for $|b\rangle$. Thus had the entire lattice been covered by the $|a\rangle$ ($|b\rangle$) excitation, the doping concentration would have been $x_a = 2/12$ ($x_b = 2/15$).

If we calculate the interaction energy (measured in units of the antiferromagnetic coupling J_{AF}) at $T = 0$ for classical spins, we find that the excessive energy (above the ground pure antiferromagnetic state) per plaquette is, respectively, $E_a = 14/12$ and $E_b = 18/15$ (see Fig. 2). The energy difference between the two states is then $1/30\ J_{AF}$ per plaquette.

Carefully going over the bond interactions one can deduce that to move from configuration $|b\rangle$ to configuration $|a\rangle$, which is more favourable, the system has to cross a barrier of $1 J_{AF}$. Since this barrier is higher than both (1/30) x 12 and (1/30) x 15 (the energy difference per basic unit in either case), then at very low temperatures the pure $|b\rangle$ state is metastable and can evolve into $|a\rangle$ only by tunneling. As the temperature increases one expects hopping to become important,

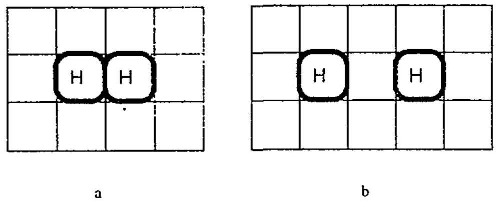

Fig. 1. — The lowest (a) and the second lowest (b) pair excitations in the plane.

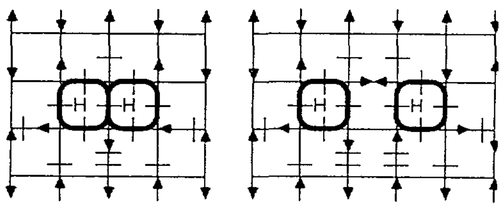

Fig. 2. — The energy of the lowest states at $T = 0$. Bonds crossed once (twice) cost $1J_{\mathrm{AF}}$ $(2J_{\mathrm{AF}})$ more than the ground antiferromagnetic state (the magnitude of the ferromagnetic coupling is irrelevant at $T = 0$). a) Lowest energy configuration : $+14J_{\mathrm{AF}}$; b) slightly higher state : $+18J_{\mathrm{AF}}$. The energy difference between the states is 18/15-14/12 = 1/30 J_{AF} per plaquette.

assisting transition. However, at low temperatures one expects the pair to be in some combination of $|\mathrm{a}\rangle$ and $|\mathrm{b}\rangle$. In principle the pair may be in a combination of a few more sates with the same spatial separation between the holes but at slightly different energy levels. Nevertheless, I will show below that the modification this introduces does not affect the present qualitative and quantitative results. Therefore let us consider only the two states shown in figure 1. Since the system is in a combination of them one expects that the spread of the pair excitation would be somewhere between 12 and 15 plaquettes. More precisely, one should calculate the projection of the state on either $|\mathrm{a}\rangle$ or $|\mathrm{b}\rangle$ to calculate from it the probability to find the pair in either of these states (e.g., *via* double-well potential formalism, which is applicable here) and then weighting the area according to this probability. But in the following all we need is the realisation that this probability cannot be too different from 1/2, $p_{\mathrm{a}} = 1/2 + \delta$. It will be shown below that even a relatively large value of δ ($<1/2$) modifies the present quantitative results only negligibly and does not affect the qualitative conclusions at all. It follows that the doping concentration at such dense packing can be evaluated through the relation

$$x_0 = 2/\left[p_{\mathrm{a}}\, S_{\mathrm{a}} + (1 - p_{\mathrm{a}})\, S_{\mathrm{b}}\right] \tag{1}$$

Substituting the above values for $S_{\mathrm{a}} = 2/x_{\mathrm{a}}$, $S_{\mathrm{b}} = 2/x_{\mathrm{b}}$ and p_{a} we find $x_0 = 0.15 + \delta/30$. This value of x_0 agrees excellently with the one measured in $\mathrm{La}_{2-x}(\mathrm{Sr})_x\mathrm{CuO}_4$ for the highest T_{c}, (the correction is ca. 0.01 for δ as large as 1/3). This simple calculation implies that at x_0 the entire plane is covered by pair excitations and hence is entirely superconducting. This implication is supported by the observations of complete diamagnetism in this particular concentration [10]. As temperature increases (then in the spirit of statistical mechanics) one expects the two states to be equally probable, which reduces δ even further and solidifies this result. There is another ramification to this description : the mean distance between the basic building blocks at x_0 is of order $4d$. Therefore, in the neighbourhood of this doping concentration, and as the system goes superconducting with decreasing temperature, one would expect to see *softening* of phononic excitations that relate to this superstructure. Such softening has indeed been observed at the vicinity of x_0 [13], but no connection has been made to the superlattice of pairs. Since this structure is constructed by a random process and relaxes very slowly due to the large mass (see also discussion below) it cannot form a perfect lattice. It follows that the phonon spectrum is expected to be smeared ; as indeed seen.

With this simple picture in mind one can now analyse, in more detail, the drop in T_c with doping for $x \geq x_0$. The idea is that as the pairs-covered plane is subjected to further doping, each additional hole disrupts the local pair structure by overcrowding. Thus the effect of the surplus hole is to suppress locally the superconducting wave function and to form an 'impurity' of normal state. As x increases the two-dimensional superconducting coverage is further ruptured leading to increasingly larger islands of normal domains. The latter are composed of compressed single holes (which may or may not carry a vortex excitation, depending on the temperature) and, being normal, cannot exhibit Meissner diamagnetism. This again fits with the above experiments, where the fraction of nondiamagnetic domains is observed to increase with x above x_0.

When the superconducting fraction is sufficiently reduced, narrow 'bottle necks' start to form through which the supercurrent is forced to flow. Since the plane is compressed with excitations, both of pairs and of single holes, we can safely assume that their mobility is constrained so that the system can be considered stationary on time scales shorter than the typical lattice relaxation time. It follows that the normal domains can be regarded as randomly quenched in the plane. Thus we end up with a picture of a superconducting cluster spanning the entire plane, which contains quasi-static 'impurities' of normal domains. Such a picture has been studied extensively in the literature in the context of the percolation model [14]. As the normal domains grow the percolating superconducting cluster (i.e., the cluster connecting between the boundaries of the plane) approaches the percolation threshold where the above 'bottle necks' become extremely narrow.

Raising the temperature at a given value of x increases thermal fluctuations, which act to break the pairing. However, superconductivity is destroyed not merely due to the attack on the overall fraction of superconducting material in the plane. Had this been the case T_c would have decreased in proportion to the total superconducting area and hence, at most, linearly with x. Since the cluster is almost completely quenched the superconducting component is nonuniformly distributed, which suggests that the 'bottle necks' are the first to go normal upon increasing thermal fluctuations. The fluctuations then cut these narrowest channels and thus destroy the connectivity of the spanning cluster. The larger the fraction of normal domains, the narrower are the bottle necks and the smaller are the thermal fluctuations needed to destroy superconductivity. This model then gives an explanation of the decrease in T_c with x above $x = 0.15$, in accordance with all observations. As shown below, it also suggests a way to deduce the value of x_{max}, i.e., the maximal doping at which superconductivity still exists.

It is well known [14] that in random two-dimensional percolation model (specifically, bond-percolation on a square lattice, see also discussion below on the applicability of the assumption of quenched randomness) at the thermodynamic limit the infinite cluster breaks at the percolation threshold where the overall fraction of dilution $1 - p_c$ exceeds 1/2. The configuration where the infinite cluster is barely connected by the narrowest possible channels occurs for $p(x)$ just above this value. Thus as $T \rightarrow 0$ the value of $p(x)$ at which superconductivity disappears is determined by the most loosely connected cluster since the smallest fluctuation disconnects the cluster. Therefore at this doping, $p(x)$ must be extremely close to p_c, which yields an equation for x_{max} as follows. We write the general doping concentration x, as a function of the occupancy probability of superlattice units that belong to the superconducting cluster $p(x)$,

$$x = p(x)x_0 + [1 - p(x)]x_h \ , \tag{2}$$

where x_h represents the mean doping concentration of the gas of single holes in the compressed normal domains. The concentration within the superconducting cluster remains constant when x increases since no further in-cluster compression is possible without overcrowding the pairs. Contrarily, the value of x_h does depend on x. The value of x_{max} can be evaluated now by putting $p(x_{max}) = p_c = 1/2$. To estimate the maximal value of x_h (which increases monotonically with x) recall that at $T = 0$ a single hole is surrounded by a vortex which occupies at least one plaquette as was shown numerically in reference [6]. Therefore one expects that ideally the most compressed situation occurs when a hole occupies every other plaquette, corresponding to $\mathrm{Max}\{x_h\} = 1/2$. Substituting this value and $p_c = 1/2$ into (2) yields $x_{max} = 0.325 + \delta/60$. This result agrees exceptionally well with the observed value of x_{max} [11].

Recalling that δ is a number that lies between 1/2 and 0, we thus established the above claim that the difference in occupation probabilities between the states, $|a\rangle$ and $|b\rangle$, plays no significant role in the calculation of x_{max}. Moreover, this observation also puts to rest the aforementioned issue of more than two lowest states as follows : if other closeby excitations exist with the same spatial configuration of bivortices as $|a\rangle$ and $|b\rangle$, but with slightly different energies — E_a, E_b,..., E_n. Then the pair is in a combination of all these n states and p_a is the weighted probability to be on nearest plaquettes. The calculation of δ may be more complicated then by the above double-well-potential formalism. However, the little effect that δ has on both x_{max} and x_0 suggests that their values are robust against such modifications of the pair state.

At this point it is also appropriate to put another worry to rest. The picture presented here for the disappearance of superconductivity may seem to ignore the penetration Λ of the normal wave function into the superconducting domains. It may appear that if Λ is bigger then the mean distance of about $4d$, one cannot use $p(x) = p_c$ and the calculation for x_{max} should be modified accordingly. This, however, is not the case. Close to the percolation threshold the quenched planar system is invariant under scale transformation [14]. This means that for scales larger than $\Lambda(T)$ (but smaller than the system's size, or any other upper cutoff length), the structure looks statistically self-similar under scale transformation. Then the above analysis can be carried out in the renormalized scale and the above calculation is exact as long as the scales that are considered are well above Λ.

It is interesting to note that the above arguments are qualitatively independent of the specific type of excitation that mediates the pairing, and it may apply to bipolarons and holon-holon excitations as well as to our bivortices. However, having accepted the picture proposed here, one cannot escape the conclusion that the spread of a single hole excitation cannot be much larger than ca. one plaquette. If this were not true and single excitations (e.g., polarons, holons or single vortices) could occupy a larger area, the concentration, at which the entire plane is covered by pairs, will be much smaller $x_0 < 0.15$ and so will be x_{max}, which contradicts the observations in Meissner diamagnetism measurements.

This picture may tell us something about $x < x_0$ as well. The above arguments were developed for $x \geq x_0$, since for lower doping the plane is not overcrowded, the pairs are more mobile and in principle the quenched picture may not be valid. Thus whether this discussion is extendable or not to the regime $x < x_0$, depends on the mobility of the holes in the antiferromagnetic background. If the excitations are massive enough even without overcrowding, we can still apply the picture of diluted quenched cluster-like structure (alternatively, one can discuss shorter time scales at which the system can still be considered quenched). In this case, the normal domains are only sparsely doped rather than composed of compressed single holes as for $x \geq x_0$. An indication that this is a fair description comes from the experimental data in reference [13]. When trying to determine T_c by measurements of the heat capacity it has been found that the behaviour is somewhat different on either side of x_0. When $x \geq x_0$ the value of T_c is very well defined as the temperature where the heat capacity is peaked, while for $x < x_0$ the maximum is smeared and T_c is less accurately estimated (T_c is always defined there as the temperature where the maximum occurs). Nevertheless, for $x < x_0$ the departure from the normal behaviour of the curve upon decreasing T seems to appear at a roughly x-independent temperature (about 40 K) that matches T_c at x_0. This may reflect the fact that below x_0 there are no weak links to determine the disappearance of the global superconductivity, but rather pairs are locally breaking by thermal fluctuations, independent of one another. Thus the geometrical correlations that determine T_c above x_0 are absent in this region.

There is another supporting indication in reference [13]. If the percolation model is still applicable for low doping then at $T = 0$ superconduction occurs only when x is high enough to form a spanning cluster of pairs with x_0. Assuming for simplicity that all single holes pair, so that the normal domains are empty of single holes, we can use again $p_c = 1/2$ and conclude that the value of x, below which no superconduction exists, is $x_{min} = x_0/2 \approx 0.075$. Fluctuations in this value may occur due to : i) finite size effects and ii) some mobility that can modify the picture of quenched randomness (see discussion below). The heat capacity measurements reported in reference [13] indeed show no peak for $x = 0.06$, while the sample with the closest higher doping, $x = 0.08$ already enjoys such a peak, which fits nicely with the above result.

If one allows for higher in-plane mobility of holes at low doping concentration (e.g., by a mechanism similar to that suggested in reference [9] then the structure is not ideally quenched. This modification means that the dipolar interaction between excitations, may effect clustering of the pairs and a correlation in the spatial distribution will appear. Such correlations, due to dynamic clustering, give rise to superconduction at even lower doping, which may account for observations of systems with x_{min} lower than 0.075.

However, even if the geometrical picture is the same and the percolation model is applicable in both regimes, they still differ significantly. For example, for $x \geq x_0$ just above T_c, the single holes coexist with the remnants of the bosons cluster. The fraction of the domains occupied by both species will be roughly equal, but their concentration is different. For $x \approx x_0$ just above T_c, the number of unpaired holes is similar to the number of paired ones, while for x close to x_{max} the normal domains are much denser and one expects the ratio of paired to single holes of order $0.15/0.5 \approx 0.3$. Now two scenarios may occur : i) the single holes carry no vortex excitations and hence they are simple fermions. Then when the temperature is lowered, we may get Bose condensation in contact with a dense Fermi liquid, which was studied recently in reference [15] ; ii) the holes may keep carrying their excitation clouds, which results in the normal domains composed of a dense gas of vortex excitations. In this case, it is not obvious what is the statistics, since the carriers may form two sub-lattices, one of antivortices and the other of vortices, displaced by a lattice parameter d. It is reasonable that the second possibility is more likely to occur when T_c is low, namely, for x close to x_{max}. The first scenario is more plausible at higher values of T_c near x_0. In between there can be a mixing of bare holes with vortices-dressed ones.

For $x < x_0$ just above the critical temperature there can appear several phases : i) superconducting domains of paired carriers, ii) heavily doped normal domains of compressed holes, either with or without vortices attached to them, and iii) very dilutely doped normal domains. The latter dominate the normal phase behaviour near x_{min}, while the second possibility is likely to govern the normal domains near x_0. This difference should be accessible to experimental observations. In general, in this regime above T_c, single holes originate from both thermal breaking of pairs and overcrowding. The effect of the latter mechanism is greatly reduced for low x and hence the ratio of paired to single holes is expected to be generally larger than for the $x > x_0$ regime. Therefore the condensation with decreasing T in this regime should be more similar to the conventional Kosterlitz-Thouless picture and different from the $x \geq x_0$ regime. By this reasoning, increasing x from $x_{\min}$ to x_0 this ratio should decrease due to the increasing concentration of single holes in the normal domains.

Another factor that has to be considered is the entropy, which would act to stabilise and homogenise the planar occupation distribution when T_c is high. Finite size effects are then expected to play an increasingly important role as T_c decreases, on both sides of x_0. However, since above x_0 the holes are compressed in the plane, entropy cannot be too dominant and hence it is more likely that these effects will be more pronounced for the $x < x_0$ regime. Another way of compressing holes is by doping at high oxygen pressure, which will tend to homogenise the entire plane and may overcrowd and destroy, rather than assist, superconductivity.

To conclude, I have presented a simple geometrical picture that explains the three observed numerical values of the doping concentration : i) $x_{\max}$ that corresponds to most heavily doped possible superconducting compound, ii) x_0, where T_c is highest, and iii) $x_{\min}$ that corresponds to most lightly doped possible superconducting compound. The above analysis not only agrees numerically with the three 'magic' values of $x_{\min}$, x_0 and $x_{\max}$, but it also describes correctly the qualitative behaviour of the $T_c(x)$ curve on both sides of x_0. This suggests that the geometrical correlations discussed here play indeed a central role in determining the phase diagram of $La_{2-x}(Sr)_xCuO_4$.

The results obtained here depend slightly on the basic model. The 2d system does not fall categorically into either bond- or site-percolation ($p_c \approx 0.59$) . If the latter is assumed then $x_{\max} \approx 0.29 + \delta/50$, still in good agreement with observations, and $x_{\min} \approx 0.09$, which is slightly too high. But, as discussed above, higher mobility at low doping can lead to clustering, which reduces $x_{\min}$, so that the site-percolation is also acceptable. In both cases the basic premise is that the CuO plane is structurally homogeneous and the only disorder stems from the locations of holes. Since this is only an ideal case, disorder from other origins may affect the results.

Insight gained from this picture may also be used to understand better the transport mechanism just above T_c, where the conductivity is governed by the broken narrow channels between superconducting domains. The good numerical agreement also supports the suggestion that the excitation cloud carried by a single hole cannot spread on more than about one plaquette. This model explains the observed nonuniform x-dependent phase separation with respect to Meissner diamagnetism and can account for recent heat capacity measurements. Finally, experimental ways to confirm, or disqualify, this picture have been discussed.

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