The annealed positions of ferromagnetic bonds
doped into a 2D antiferromagnet

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Abstract

We determine where a given concentration of ferromagnetic (FM) bonds doped into a square lattice antiferromagnet must go to minimize the system’s total magnetic energy. We find (i) an infinite degeneracy of ground–state arrangements of FM bonds that correspond to completely unfrustrated configurations for classical spins, and (ii) this degeneracy is lifted when quantum fluctuations are included, and phase separated ground states, such as periodic arrays of stripes of FM bonds, are found. A discussion of the application of these ideas to doped cuprate high $T_c$ superconductors with annealed disorder is presented.
Carriers in the high $T_c$ compounds can be introduced into the ubiquitous CuO$_2$ planes of the insulating antiferromagnetically (AFM) ordered parent compounds either by doping with immobile dopants, or by altering the oxygen concentration. If, for example, one is concerned with the interpretation of experimental studies of the Bednorz–Müller high $T_c$ compound La$_{2-x}$Sr$_x$CuO$_{4+\delta}$, the carriers in the planes will be affected by the *quenched* disorder that is present as a result of the immobile Sr dopant ions. In contrast to this situation, excess oxygen ions ($\delta > 0$) in the $x = 0$ material are mobile at sufficiently high temperatures, and thus the disorder introduced by them during the crystal growth process can be *annealed*, effectively restoring translational periodicity of a perfect crystal [1]. The absence of dopant disorder leads to a simplified system, one that should be amenable to comprehensive theories of the holes in the CuO$_2$ planes.

In this report we concern ourselves with the possibility of charge inhomogeneities of the holes in the CuO$_2$ planes in the absence of dopant disorder. Such structures are potentially related to the frustrated phase separation phenomenology of Emery and Kivelson [2] in the form of striped regions rich in the holes, and many theoretical studies consistent with such charge distributions have been found via Hartree–Fock studies [3]. However, the mechanism associated with this microsegregation of holes remains unclear, and thus in this report we eliminate the kinetic energy of the holes and consider localized oxygen holes, thus focussing entirely on the magnetic interactions present in the doped system. Due to the annealing away of disorder that is possible for the super–oxygenated compounds, we imagine that the localized holes can find the annealed positions that they would assume to minimize the doped plane’s energy. Very similar physics is found in superoxygenated La$_2$NiO$_{4+\delta}$, although the holes are believed to exist as partially localized Zhang–Rice singlets and not as completely localized oxygen holes [4]. For our simplified model we explain how magnetic interactions alone can produce striped ground states — whether or not this is an important driving force associated with such charge inhomogeneities remains an open question.

The magnetic interactions in a system with completely localized oxygen holes is as follows: A localized oxygen hole neighbouring a Cu 3d$^9$ ion leads to a Kondo–like coupling between the $S = \frac{1}{2}$ Cu ion and the $S = \frac{1}{2}$ oxygen ion. Regardless of the sign of this interaction, the
coupling of the oxygen ion to both Cu sites leads to a net ferromagnetic (FM) interaction between the pair of Cu sites surrounding the oxygen hole, an idea first put forward by Aharony, et al. Since the undoped system corresponds to neighbouring pairs of Cu ions that interact antiferromagnetically, the simplest model of this system is just that of the so-called Aharony model, viz. a distribution of ferromagnetic bonds of a fixed concentration doped into a square lattice $S = \frac{1}{2}$ AFM. The Hamiltonian representing this situation is

$$H = J \sum_{<ij>} S_i \cdot S_j - (K + J) \sum_{<ij>}' S_i \cdot S_j$$

where $J > 0$ is the AFM exchange constant of the undoped background, $i$ and $j$ label the lattice sites of an infinite 2D square lattice, $<ij>$ denotes near neighbours, $K > 0$ is the ferromagnetic exchange constant for a bond containing an oxygen hole, and the primed summation indicates that only those bonds containing an oxygen hole are summed over. For a random distribution of these FM bonds the background AFM state becomes frustrated, leading to a complicated phase diagram vs. doping.

The fact that we are allowing the oxygen holes to move around and anneal their positions in the CuO$_2$ planes means that the ferromagnetic bonds can move in an attempt to lower the system’s energy. Thus, another way of phrasing the question at hand is: What is the spatial distribution of some fixed concentration of FM bonds doped into square lattice quantum AFM that minimizes the system’s energy? The answer that we have found involves the relieving of the frustration induced by the FM bonds by phase separation. To be specific, for classical spins a variety of configurations of FM bonds allows for completely unfrustrated spin textures. Then, when quantum fluctuations are included, something we have managed to do using Colpa’s para–diagonalization scheme for the bilinear, many–boson problem, one finds that, e.g., for a small FM exchange energy, stripe configurations are the preferred low energy state.

Firstly, we describe the solution of this problem for classical Heisenberg spins (it is to be noted that the following arrangements of spin directions are also the ground states for XY and Ising spin systems). Figures 1 → 3 show some of the configurations of FM bonds for which unfrustrated spin textures may be found. The simplest such configuration is that shown on the left of Fig. 1 — we shall refer to this as the “plus sign” arrangement. To see that an
unfrustrated spin texture may be found for this set of bonds note that with the introduction of the 4 FM bonds of the plus sign configuration the central sign is isolated from the AFM background. Thus, by flipping this and only this spin, relative to the undoped AFM spin arrangement, all FM and AFM bonds in the lattice are satisfied. (One may generalize the idea of this configuration to isolated core regions of arbitrary size.) For a given concentration of FM bonds one may simply place these plus sign configurations anywhere in the lattice. Then, for a set ratio of $K/J$, the energies of all such arrangements are equal, a fact that applies to all unfrustrated arrangements that we have found.

There are a variety of other allowed spin textures: The (horizontal or vertical) striped phase of width $W$ is shown on the right of Fig. 1. The unfrustrated spin state is achieved by flipping all spins on one side of the stripe, beginning with the sites at the ends of the FM bonds; these states are unfrustrated for all $W$. Figure 2 shows another non-frustrating arrangement of FM bonds, a pair of crossed stripes (here shown for $W = 1$). Note that crossed stripes lead to a completely FM region at the intersection point, and thus may be desirable at large $K/J$. Figure 3 shows something that we shall refer to as a $W = 1$ staircase configuration — this is somewhat like a $W = 1$ stripe that is rotated by $45^\circ$, although the internal structure of the staircase (a continuous line of FM bonds) is clearly different from that of a stripe.

Clearly, there is an infinite degeneracy of such structures that at $T = 0$ minimize the system’s energy for classical spins. However, since all of these structures correspond to differing local spin environments, when zero–point spin–wave fluctuations are included the energy of these configurations for quantum spins will, in general, be different. We expect that at $T = 0$, since these systems are a long way from any phase transition linear spin–wave theory should adequately represent the spin excitations. Then, since spin waves effectively dress a classical state, we begin with the above determined classical configurations, on which we then superimpose spin waves, finally determining which of these configurations has the lowest energy for a given concentration of FM bonds and for a given ratio of $K/J$ for quantum spins of length $S$.

It is easy to exclude many configurations from further consideration: The inclusion of
spin waves means that arrangements of FM bonds that are highly localized in space, such as, e.g., the plus sign arrangements, require a large number of spin waves of all wave vectors, essentially equally weighted, to represent the interaction of the surface of the region of FM bonds with the background AFM environment — since the short–wavelength spin waves are of a high energy, they will make configurations that require their inclusion to be high as well. Thus, we expect and have limited our theoretical studies to configurations that are devoid of the plus sign structure. (To confirm the legitimacy of this approximation we have checked that for all $K/J$ and for the concentration range of FM bonds that we have studied, periodic arrangements of plus signs (with square or rectangular unit cells) for $S = \frac{1}{2}$ have energies higher than the ground state configurations.) Further, in order to access and bias the lowest energy spin waves to be those dominating the interface regions at which the FM bonds impinge on the AFM background we consider periodic arrangements of the stripes, or crossed stripes, or staircases.

Once one of these periodic arrangements is selected, the relationship between the density of FM bonds to the periodic structure is easily determined. From now on we define the concentration of oxygen holes per Cu – O – Cu bond to be $c$. Then, if one is considering a periodic repetition of stripes of width $W$, the repeat distance $L$ between stripes must be taken to be $L = W/(2c)$. For this particular arrangement this means that the number of sites in the magnetic unit cell is $2 \times L$ (or possibly $2 \times 2L$). Thus, standard Holstein–Primakoff theory of spin waves of the two–dimensional quantum AFM [7] is not directly applicable. Instead, one must allow for different quantum fluctuations to exist at every inequivalent site, something that we have accomplished by using a different boson at each equivalent site (where, say, $a_{II}^{$\dagger$}$ represents the creation of a spin deviation in unit cell $I$ at basis site $i$.) Due to the large number of sites per unit cell, the resulting bilinear Hamiltonian involves a very large number of coupled bosons. We have managed to extract the spin–wave eigenfrequencies and total system energy for this problem using the para–diagonalization formalism developed by Colpa — an extensive discussion of the details of this method is contained in Ref. [8].

Our results, for a concentration $c = 0.1$ and $S = \frac{1}{2}$, are shown in Table I. The lowest energy configuration depends on the ratio of $K/J$. Since the structure of a system’s total
energy is just $E = -S(S + 1) E_{\text{classical}} + S E_{SWT}$, our results for the ordering of energies are effectively independent of $S$. Also, in the concentration range that we have studied, \emph{viz.} $0.083 \leq c \leq 0.125$, there is effectively no dependence of the ordering of the energies on concentration.

For $K/J$ less than unity, the lowest energy state is a $W = 1$ striped phase. This is thus an example of magnetic interactions producing a striped ground state \[8\]. The driving energy per spin associated with some kind of stripe ordering is of the order of 6 K for La$_2$CuO$_{4+\delta}$, and is thus quite small — this suggests that interactions such as the Coulomb interaction \[8\] must be included to fully account for the observed charge orderings. That we find a striped ground state may be understood in a simple non–interacting approximation: \emph{ignoring the interface energies} and exactly evaluating the energies of a width $W$ stripe with open boundary conditions, and then a width $(L - W)$ AFM region, and then weighting the energies of the two regions by $W/L$ and $(L - W)/L$ to finally represent the energy of the bulk lattice, one finds that the minimum energy for small $K/J$ corresponds to $W \to 0$ (this is simply a statement that for small $K/J$ FM bonds are not helpful in lowering the system’s energy, no matter where they are put). However, since we are imposing that some fixed concentration of FM bonds must be present, $W = 1$ is the smallest allowed $W$. The $W = 1$ geometry which we find minimizes the energy for small $K/J$ is that of a periodic repetition of $W = 1$ stripes.

For large $K/J$ we expect an entirely different configuration to be the minimum energy state, \emph{viz.}, if one completely phase separates the system into a FM region of concentration $c$, and then a AFM region of concentration $1 - c$, \emph{and} is able to produce the unfrustrated interface separating the two regions, one should find the minimum energy configuration. A representation of this kind of arrangement is shown in Fig. 4. Clearly, this is simply the $W \to \infty$ staircase with a fixed concentration of FM bonds; alternatively, this phase is a maximally dense clustering of plus sign structures forming a $45^\circ$ interface. One may write down an expression for the energy per spin of this state that is exact in the bulk limit (since the interface energy will scale as $1/L$, $N = L^2$ being the number of lattice sites) and find

$$\frac{E}{N} = -2K c < S_0 \cdot S_1 >_{FM} + 2J (1 - c) < S_0 \cdot S_1 >_{AFM}$$

(2)
Evaluating the near–neighbour correlation functions for an AFM in the spin–wave approximation our results for this two–phase structure are also listed in Table I. We see that for \( K/J > 1.0 \) this completely phase separated structure has the lowest energy.

In conclusion, we have made an in depth study of the energy–minimizing configurations of FM bonds doped into a 2D square lattice AFM. The infinite degeneracy of unfrustrated classical spin ground states is lifted by zero–point spin–wave excitations which subsequently leads to striped microsegregated ground states for small \( K/J \), and macroscopically phase separated ground states for large \( K/J \).

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[8] U. Löw, et al., Phys. Rev. Lett. 72, 1918 (1994) have found striped ground states in their competing interactions model only if Coulomb-like interactions are included.
Figure 1: Two frustration–free configurations of FM bonds. The solid circles represent the transition metal ions that interact AFM with their nearest neighbours with strength $J$ in Eq. (1). The solid lines are the FM bonds of strength $K$ in Eq. (1). On the left of the figure is four FM bonds organized in the shape of a plus sign. These bonds do not frustrate the AFM background since the central spin can be flipped over from the direction it would chose in the undoped AFM state. On the right of the figure is a vertical stripe of FM bonds, stacked one on top of another. That this configuration is unfrustrated follows from these changes: take every spin–up sublattice site on the right of the stripe and make it a spin–down sublattice. Similarly, change the down–sublattice sites to be up–sublattice sites. For an infinite system, one requires that this stripe be of infinite length for the total spin texture to be unfrustrated. The stripes can be of any integral width, $W$, with the figure displaying a $W = 2$ stripe.

Figure 2: The intersection of two $W = 1$ stripes, which also leads to an unfrustrated spin arrangement. The overlapping portion corresponds to a fully FM region.

Figure 3: A staircase configuration. The solid lines are seen to create a continuous line of FM bonds, thus effectively producing a 1D FM chain embedded in a 2D AFM.

Figure 4: A $W \to \infty$ staircase configuration. This leads to a two–phase, completely phase separated region of either FM (lower right) or AFM (upper left) bonds. If the interface is taken to be along this direction, the resulting ground state spin arrangement is completely unfrustrated.
Table 1: The energy per spin, relative to $J$, for $S = 1/2$ and $c = 0.1$, of some periodic arrangements of classically unfrustrated FM configurations for $W = 1$, the $W \to \infty$ crossed stripes configuration, and the two–phase state shown in Fig. 4. The starred energies are the lowest energy configurations for a given $K/J$.

| $K/J$ | Striped | Staircase | Crossed Stripes | Crossed Stripes | Staircase |
|-------|---------|-----------|-----------------|-----------------|----------|
|       | $W = 1$ | $W = 1$   | $W = 1$         | $W \to \infty$ | $W \to \infty$ |
| 0.1   | -0.6136* | -0.6071   | -0.6133         | -0.6095         | -0.5971  |
| 0.25  | -0.6167* | -0.6117   | -0.6163         | -0.6130         | -0.6046  |
| 0.5   | -0.6243* | -0.6209   | -0.6239         | -0.6218         | -0.6171  |
| 1.0   | -0.6430  | -0.6416   | -0.6429         | -0.6430         | -0.6421  |
| 2.0   | -0.6863  | -0.6868   | -0.6866         | -0.6894         | -0.6921* |
| 4.0   | -0.7801  | -0.7821   | -0.7810         | -0.7864         | -0.7921* |
| 10.0  | -1.0740  | -1.0763   | -1.0754         | -1.0884         | -1.0921* |
