

SURPRISES ON THE WAY FROM 1D TO 2D QUANTUM MAGNETS: THE NOVEL LADDER MATERIALS.

Elbio DAGOTTO¹ and T. M. RICE²

¹*Dept. of Physics and National High Magnetic Field Lab,
Florida State University, Tallahassee, FL 32306, USA*

²*AT&T Bell Laboratories, Murray Hill, NJ 07974, USA, and
Theoretische Physik, Eidgenössische Technische Hochschule,
8093 Zürich, Switzerland*

Abstract

One way of making the transition between the quasi-long range order in a chain of $S=1/2$ spins coupled antiferromagnetically and the true long range order that occurs in a plane, is by assembling chains to make ladders of increasing width. Surprisingly this crossover between one and two dimensions is not at all smooth. Ladders with an even number of legs have purely short range magnetic order and a finite energy gap to all magnetic excitations. Predictions of this novel groundstate have now been verified experimentally. Holes doped into these ladders are predicted to pair, and possibly superconduct.

I. Introduction

The unexpected discovery of high temperature superconductivity[1] in lightly doped antiferromagnets has sparked renewed interest in low dimensional quantum magnets. The parent cuprate insulators are now considered the best examples of planar spin-1/2 antiferromagnets with isotropic and predominantly nearest neighbor coupling. They show simple long range antiferromagnetic (AF) order at low temperatures in agreement with theory which predicts an ordered ground state for the $S=1/2$ AF Heisenberg model on a two dimensional (2D) square lattice.[2] The one dimensional (1D) AF Heisenberg chain is also

well understood. A famous exact solution found by Bethe many years ago[3] showed that quantum fluctuations prevent true long range AF order giving instead a slow decay of the spin correlations essentially inversely with separation between the spins. Therefore it came as a great surprise when numerical calculations found that the crossover from chains to square lattices, obtained by assembling chains one next to the other to form “ladders” of increasing width, was far from smooth. Although there is no apparent source of frustration, quantum effects lead to a dramatic dependence on the width of the ladder (i.e. the number of coupled chains).

Ladders made from an *even* number of legs have spin liquid ground states so called because of their purely short range spin correlation. An exponential decay of the spin-spin correlation is produced by a finite *spin – gap*, namely a finite energy gap to the lowest S=1 excitation in the infinite ladder. These even ladders may therefore be regarded as realizations of the unique coherent singlet ground state proposed some years ago by Anderson in the context of the two dimensional S=1/2 AF Heisenberg systems (the so-called Resonance Valence Bond (RVB) state).[4]

Ladders with *odd* number of legs behave quite differently and display properties similar to single chains at low energies i.e. gapless spin excitations and a power-law falloff of the spin-spin correlations, apart from logarithmic corrections. This dramatic difference between even and odd ladders predicted by theory has now been confirmed experimentally in a variety of systems.

2-leg S=1/2 ladders are found in vanadyl pyrophosphate $(VO)_2P_2O_7$ and in some cuprates e.g. $SrCu_2O_3$ (Fig.1) (here we use the convention that an “m-leg ladder” denotes m coupled spin-1/2 chains). Measurements of the spin susceptibility show that it vanishes exponentially at low temperature, a clear sign of a spin-gap. Neutron scattering and μSR measurements are consistent with short range spin order in the 2-leg ladders although, as we stressed before, they are unfrustrated spin systems which classically should order without a spin-gap. Further NMR measurements have confirmed the large spin-gap in the excitation spectrum.

3-leg ladders (e.g. $Sr_2Cu_3O_5$) by contrast show longer range spin correlations and even true long-range order at low temperature due to weak interladder forces. There is excellent

agreement between theory and experiment confirming that there is a dramatic difference between even and odd $S=1/2$ Heisenberg AF ladders.

Doped chains have long fascinated theorists because they form unusual quantum liquids, so-called Luttinger liquids with many unique properties.[5] Although doping experiments in ladder compounds are just starting, extensive theoretical studies have been made of doped ladders. Again a clear difference between even and odd ladders is predicted. Even ladders are specially interesting because hole *pairing* in a relative “d-wave” state is found using a variety of techniques which places them in a different universality class of one-dimensional systems than the Luttinger liquids found in single chains and odd ladders.

The next section reviews the theory of the $S=1/2$ AF Heisenberg model on ladders. In Section III cuprates and other compounds that are realizations of $S=1/2$ AF Heisenberg ladders are discussed together with recent magnetic measurements. Hole doping of ladders is the topic of Section IV with emphasis on theoretical studies. Finally some concluding remarks are made in Section V.

II. $S=1/2$ Heisenberg model on ladders: theoretical aspects.

The properties of $S=1/2$ Heisenberg AF models defined on 1D chains or on 2D square lattices are well-known. The model is defined by the Hamiltonian

$$H = J \sum_{\langle \mathbf{i}, \mathbf{j} \rangle} \mathbf{S}_{\mathbf{i}} \cdot \mathbf{S}_{\mathbf{j}}, \quad (1)$$

where \mathbf{i} is a vector labelling lattice sites where spin-1/2 operators $\mathbf{S}_{\mathbf{i}}$ are located. $\langle \mathbf{i}, \mathbf{j} \rangle$ denote nearest neighbors (n.n.) sites. $J(> 0)$ is the antiferromagnetic exchange coupling that provides the energy scale in the problem. This scale is material dependent and it ranges from a few meVs to about 0.1eV in the case of the high temperature superconductors. As explained above, on 2D square lattices, the Heisenberg model has a ground state with long range antiferromagnetic order, while in 1D chains the spin-spin correlation decays slowly to zero as a power-law. Both systems are spin *gapless* i.e. there is no cost in energy to create an excitation with $S=1$.

The new field of ladder systems started when Dagotto, Riera and Scalapino[6] (see also Hirsch[7] and Dagotto and Moreo[8]) found evidence that 2-leg ladders have a finite

spin-gap i.e. a finite energy is needed to create a $S=1$ excitation. They started with the simple limit obtained by generalizing Eq.(1) so that the exchange coupling along the rungs of a 2-leg ladder (denoted by J') is much larger than the coupling J along the chains, $J' \gg J$. This idealization has the advantage that rungs interact only weakly with each other, and the dominant configuration in the ground state is the product state with the spins on each rung forming a spin *singlet*. The energy in this limit is approximately $E_{\text{gs}} = -\frac{3}{4}J'N$, where N is the number of rungs and $-\frac{3}{4}J'$ the energy of each rung singlet state $|\psi\rangle_{\text{S}} = (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)/\sqrt{2}$. The ground state has total $S=0$, since each rung is in a spin singlet. To produce a $S=1$ excitation a rung singlet must be promoted to a $S=1$ triplet $|\psi\rangle_{\text{T}} = \{|\uparrow\uparrow\rangle, (|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)/\sqrt{2}, |\downarrow\downarrow\rangle\}$. An isolated rung-triplet has an energy J' above the rung singlet. The coupling along the chains creates a band of $S=1$ magnons with a dispersion law, $\omega(\mathbf{k}) = J' + J\cos(\mathbf{k})$ in the limit $J' \gg J$. The spin-gap is the minimum excitation energy $\Delta_{\text{spin}} = \omega(\pi) (\sim J' - J)$ which remains large in this limit.[9] Concurrently, the spins are mostly uncorrelated between rungs since the spin correlations decay exponentially with distance along the chains leading to the *spin liquid* nature of this state. Note, however, that the spins are not disordered but in a unique isolated quantum-coherent groundstate.

In the other extreme, $J'/J = 0$, the two chains decouple but isolated spin-1/2 Heisenberg *chains* do not have a spin-gap and excitations with $S=1$ and wavevector $\mathbf{k} = \pi$ are degenerate with the ground state in the bulk limit. To reconcile the different behavior in the limits $J'/J \gg 1$ and $J'/J = 0$, it was conjectured[6] that the spin-gap should smoothly decrease as J'/J is reduced, reaching $\Delta_{\text{spin}} = 0$ at some critical value of the coupling. Later, Barnes, Dagotto, Riera and Swanson[9] observed that the power law decay of the spin correlation in an isolated chain implies that a chain is in a critical state and thus small perturbations can qualitatively alter its properties. They predicted that the spin-gap would vanish *only* at $J'/J = 0$, so that $\Delta_{\text{spin}} > 0$ at all $J'/J > 0$ including the values of experimental interest, $J'/J \sim 1$. The ladder spin system would always be in a spin liquid state in contrast to the more familiar cases of the 1D and 2D Heisenberg models which are *gapless*.

Physical realizations of ladders like SrCu_2O_3 or $(\text{VO})_2\text{P}_2\text{O}_7$ correspond to $J' \approx J$. However, at $J' = J$ there is no small parameter to guide a perturbative calculation nor is an

exact solution known. *Numerical* techniques can handle the region $J' \approx J$ and Exact Diagonalization of small clusters and Quantum Monte Carlo techniques were used in Ref.[6,9] to study Δ_{spin} as a function of J'/J . The techniques used are not essential to the discussion. We refer the reader elsewhere for details,[10] and concentrate on the results.

In Fig.2a, Δ_{spin} calculated numerically by Barnes et al.[9] shows that indeed $\Delta_{\text{spin}} > 0$ for all $J'/J \neq 0$. At the realistic coupling $J' = J$, the gap is $\Delta_{\text{spin}} \approx 0.5J$. More recently, White, Noack and Scalapino,[11] using a novel renormalization group (RG) technique suitable for static properties of 1D systems reported a value $\Delta_{\text{spin}} = 0.504J$ at $J' = J$, in excellent agreement with Refs.[6,9]. Note, there are AF spin correlations at short distances along the chains and across the rungs, but even at $J' = J$, the latter are somewhat stronger[11] showing that the rough picture of a ground state dominated by rung-singlets[6] is robust. Finally the closely related one-band Hubbard model at half-filling also shows a spin-gap for all interaction strengths.[12,13]

By now it is clear that the presence of a spin-gap in the 2-leg ladder has been well established using a variety of techniques. A useful intuitive approximation is to visualize the groundstate as mostly rung singlets supplemented by weak AF correlations along the chains. Gopalan, Rice and Sigrist[14] suggested that a good variational description of the ground state of the 2-leg ladder could be obtained using the short-range Resonance Valence Bond (RVB) state proposed by Anderson and Kivelson et al.[4,15] with mostly adjacent rung singlets, but including resonance between 2 adjacent rung singlets into 2 n.n. singlets along the chains.[12]

What happens if we increase the number of “legs” in the ladder? This is not a purely academic question since Rice, Gopalan, and Sigrist[16] have shown that materials like $\text{Sr}_{n-1}\text{Cu}_{n+1}\text{O}_{2n}$ contain ladder structures with a number of legs that depends on the value of n . The large J'/J limit allows us again to make predictions for the behavior of the m -leg ladder. Let us begin with the *even*-leg ladder. At $J'/J \gg 1$, the rungs decouple and at each level one has 2^m states instead of the four states of the 2-leg ladder, apparently complicating the problem. However, the ground state of the m -spins rung is also a $S=0$ singlet separated by a finite gap from the first excited state. Thus, as in the case of the 2-leg ladder, the even-leg ladder at $J' \gg J$ has a finite spin-gap $\propto J'$. Taking the analogy with the 2-leg ladder further it is again plausible to assume that a spin-gap exists in the even-leg ladder

for any $J'/J \neq 0$. Early numerical calculations on 4-leg ladders are in agreement with this picture.[8] Poilblanc et al.[17] evaluating exactly the 4×6 and 4×8 clusters with periodic boundary conditions, and extrapolating the results to the bulk limit using an exponential form obtain a spin-gap $\Delta_{\text{spin}} = 0.245J$, about half the size of the gap for the 2-leg ladder. Hatano and Nishiyama[18] found $\Delta_{\text{spin}} = 0.27J$ using a similar analysis. A reduction in the size of the gap is natural since as the width of the ladders grows, the 2D square lattice limit is approached and $\Delta_{\text{spin}} \rightarrow 0$. White et al.[11] using a RG technique on larger $4 \times N$ clusters but with open boundary conditions, which amplify the finite size effects, reported $\Delta_{\text{spin}} = 0.190J$ extrapolated to $N \rightarrow \infty$, with a spin correlation length $\xi_{AF} \sim 5-6$. Finally, a mean field approach[14] predicted $\Delta_{\text{spin}} = 0.12J$. The presence of a finite spin-gap in the 4-leg ladder seems by now well established theoretically, but some discrepancies on its value remain to be clarified.

Rice et al.[16] and Gopalan et al.[14] quoting arguments by Hirsch and Tsunetsugu made the interesting observation that ladders with an odd number of legs should behave quite differently from even-leg ladders and display properties similar to single chains at low energies i.e. *gapless* spin excitations and a power-law falloff of the spin-spin correlations. The simplest way to visualize this difference is again by analyzing the large J'/J limit, as remarked by Reigrotzki et al.[19] Let us consider for example the 3-leg ladders. At large J'/J , each rung can be diagonalized exactly leading to a *doublet* ground state, and a doublet and quadruplet excited states. The rung doublet of lowest energy will be the dominant configuration in the ground state at small temperature which thus consists now of $S=1/2$ states (doublets) in each rung. The inter-rung coupling J provides with an effective interaction between these $S=1/2$ rung states which by rotational invariance must be of the Heisenberg form with an effective coupling J_{eff} as energy scale. Thus, the ground state properties of the 3-leg ladder at large J'/J should be those of the spin-1/2 Heisenberg chain, with a coupling J_{eff} instead of J , and thus with a vanishing spin-gap. The argument can be trivially generalized to all odd-leg ladders. Since for the odd-leg case both at $J'/J \gg 1$ and $J'/J = 0$ there is no spin-gap, it is reasonable that at intermediate values of J'/J the gap always vanishes in contrast to even-leg ladders. A recent numerical RG calculation[11] verified these intuitive ideas.

Khveshchenko[20] explained the qualitative difference between even and odd ladders based on an argument used by Haldane for the 2D square lattice.[21] For odd ladders a topological term governing the dynamics at long-wavelengths appears in the effective action, while for even ladders it exactly cancels. This topological term is similar to the one that causes the well-known difference between the finite spin-gap of integer Heisenberg spin chains and the absence of a spin-gap for half-integer spin chains. The direct analogy with the Haldane state of the S=1 chain is realized in ladders with a *ferromagnetic* coupling, $J' < 0$, on the rungs. In this case in the $|J'/J| \gg 1$ limit the rungs become spin triplets rather than singlets, and the relation with the S=1 chain is obvious.[22] However, more work is needed to clarify the relationship of the Haldane state of the integer spin chains and the spin liquid of the AF spin-1/2 even-leg ladders.[23]

The single magnon spectrum $\omega(\mathbf{k})$ of the 2-leg ladder evolves from a simple cosine dispersion at $J' \gg J$, dominated by S=1 rung states,[9] to a more linear dispersion around the minimum at $\omega(\pi) \approx 0.5J$ at the isotropic coupling value, $J' = J$ (Fig.2b).[9,14] This change can be traced to a spreading of the two parallel spins in the triplet over more than one rung as J'/J is reduced, which in turn modifies the dispersion relation through longer range transfer processes. As shown by Barnes and Riera [24], the magnons near $\mathbf{k} = \pi$ remain as well defined modes separated from the 2-magnon continuum which starts at energy $\approx J$ near $\mathbf{k} = 0$. [25,26] The magnon dispersion should in principle be directly measurable through inelastic neutron scattering experiments on single crystals but as discussed below only powder spectra are available at present.

Recently, thermodynamic properties of S=1/2 ladders have also been studied by several groups. Troyer, Tsunetsugu and Würtz[27] using a quantum transfer-matrix method on 2-leg ladders obtained reliable results down to temperature $T \approx 0.2J$. The correlation length ξ_{AF} of the short range AF order values $\xi_{AF} \approx 3 - 4$ (in units of the lattice spacing), in agreement with calculations at zero temperature[11] that reported $\xi_{AF} = 3.19$. The magnetic susceptibility $\chi(T)$ [24,27,28] crosses over from a Curie-Weiss form $\chi(T) = C/(T + \theta)$ at high temperature, to an exponential falloff $\chi(T) \sim \exp(-\Delta_{\text{spin}}/T)/\sqrt{T}$ as $T \rightarrow 0$ reflecting the finite spin-gap.[29] Recently, Frischmuth, Troyer and Würtz[30], using an improved algorithm, extended their results to lower temperatures and to ladders up to 6-legs in width.

Their results for $\chi(T)$ are shown in Fig.3. The difference between odd and even ladders is very clear at low temperatures as is the smaller spin-gap in the 4- and 6-leg ladders.

III. Experimental results on ladder compounds.

At present two types of ladder compounds are known. The first to be identified was vanadyl pyrophosphate $(VO)_2P_2O_7$ whose structure was shown in Fig.1. The V-ions are in an oxidation state V^{4+} i.e. $3d^1$ with the single electron occupying a non bonding t_{2g} -orbital. The superexchange interaction occurs through the $dp\pi$ -overlap of V 3d- and O 2p-orbitals. The magnetic susceptibility $\chi(T)$ shown in Fig.4a was measured by Johnston et al.[31] who found an activated behavior at temperatures $T < 100K$ crossing over to Curie-Weiss form at higher temperature. Barnes and Riera [24] by fitting $\chi(T)$, found almost equal values $J = 7.76$ meV and $J' = 7.8$ meV for the exchange along the chains and rungs, respectively. Recently Eccleston et al.[32] used a powder time-of-flight neutron scattering technique to obtain the inelastic spectrum. The powder average of the dynamic magnetic structure factor (Fig.5) shows clear evidence of a spin-gap, $\Delta_{\text{spin}} \approx 3.7 \pm 0.2$ meV at a wavevector π , a value which agrees well with the theoretical prediction of $\Delta_{\text{spin}} = 0.5J = 3.9\text{meV}$. [6,9] The data do not allow a unique determination of the magnon dispersion relation but are consistent with the form illustrated in Fig.2b.

The second type of ladder compounds are cuprates but with modified copper-oxygen planes and other structures. The key point here is the configuration of the CuO_4 squares. The high- T_c cuprate families all are based on CuO_2 -planes with all corner sharing CuO_4 -squares. This leads to 180° $\text{Cu} - \text{O} - \text{Cu}$ bonds. Since the Cu^{2+} -ion has a $3d^9$ configuration with the single hole occupying an antibonding e_g -orbital, there is an exceptionally strong superexchange interaction ($J \approx 0.13\text{eV}$) through $dp\sigma$ overlap with the O2p-orbital common to both CuO_4 -squares. In the ideal CuO_2 -plane, the O-ions form a square lattice and the Cu-ions occupy the centers of exactly one half of the O_4 -squares, also forming a square lattice. If a line defect is introduced in the Cu-occupation so that left and right different sets of O_4 -squares are occupied, then along this line the coordination of the CuO_4 -squares is edge-sharing (Fig.1b). But the superexchange path for two CuO_4 squares sharing an edge is very different and involves primarily an intermediate state with 2 holes on orthogonal

orbitals on the same O-ion. Hund's Rule then favors parallel spin alignment and as a result the Kanamori-Goodenough rules give a weak ferromagnetic (F) coupling between Cu^{2+} -ions which are edge-sharing.

Hiroi, Azuma, Takano, and Bando,[33] were the first to synthesize the family of layer compounds $\text{Sr}_{n-1}\text{Cu}_{n+1}\text{O}_{2n}$ which have arrays of parallel line defects. The copper oxide planes in the first two members were shown in Fig.1b. Rice et al.[16] pointed out that nearly ideal ladder compounds should result, since the pattern of strong AF 180° Cu – O – Cu bonds make a ladder, and the interladder coupling is very weak both because of weak F 90° Cu – O – Cu bonds and the resulting frustration. The first member ($n=3$ or SrCu_2O_3) has 2-leg ladders, the second ($n=5$ or $\text{Sr}_2\text{Cu}_3\text{O}_5$) has 3-leg ladders and so on.

Recently Azuma et al.[34] reported magnetic susceptibility measurements for the 2- and 3-leg ladder compounds (see Fig.4b,c). The difference between the two compounds is striking. The spin-gap is clearly visible in the precipitous drop in $\chi(T)$ for $T < 300\text{K}$ in the 2-leg compound, and by fitting to the low temperature form $\chi(T) \sim T^{-1/2}\exp(-\Delta_{\text{spin}}/T)$, they obtained a value $\Delta_{\text{spin}} = 420\text{K}$. This compound should have exchange constants close to the isotropic limit $J = J' \approx 1300\text{K}$ so that theory predicts a larger value for an isolated ladder $\Delta_{\text{spin}}^{\text{theory}} \sim 650\text{K}$. However, in SrCu_2O_3 there is substantial exchange coupling along the c-axis, J_c . This should lower Δ_{spin} and may account for most of the discrepancy. But, Azuma et al.[34] (see also Ishida et al.[35]) also reported NMR investigations. In particular, they observed activated behavior in the relaxation rate ($1/T_1$) at $T < 300\text{K}$ as expected but the activation energy (680K) was substantially larger than the value deduced from $\chi(T)$. At present the origin of the discrepancy is unclear.

Azuma et al.[34] measured $\chi(T)$ also for the 3-leg ladder compound, $\text{Sr}_2\text{Cu}_3\text{O}_5$, and found $\chi(T) \rightarrow \text{const.}$ as $T \rightarrow 0$ as expected for the 1D AF Heisenberg chain. Further μSR measurements by Kojima et al.[36] found evidence of a long range ordered state with $T_N = 52\text{K}$. This ordering we attribute to the interlayer coupling J_c along the c-axis. Note that no sign of long-range ordering was observed in the 2-leg compound. These results confirm explicitly the drastic difference between ladders with even and odd number of legs.

Ladder structures occur also in other cuprates. For example, Batlogg et al.[37] reported the magnetic susceptibility for the family of compounds $\text{La}_{4+4n}\text{Cu}_{8+2n}\text{O}_{14+8n}$ which, as special cases, contain 4- and 5-leg ladder elements. These are complex structures which

contain other Cu-sites only weakly coupled to each other. These latter spins dominate $\chi(T)$ below room temperature. However, by examining the difference in $\chi(T)$ between the two compounds, Batlogg et al.[37] could identify a substantial spin-gap in a 4-leg compound ($\Delta_{\text{spin}} \approx 300\text{K}$). Note that in these compounds only weak inter-ladder coupling is expected and a value of $\Delta_{\text{spin}} \approx J/4 (\approx 325\text{K})$ is predicted theoretically, which agrees quite well with the experiment. Very recently, Hiroi and Takano [38] have found a new ladder compound $\text{LaCuO}_{2.5}$ with 2-leg ladders which are weakly connected in a three dimensional structure.

IV. Hole doping of spin-1/2 ladders

Generally it is difficult to dope transition metal oxides and produce a highly conducting state but the cuprates are exceptional in this regard. Early reports of doped cuprate ladder materials are starting to appear.[38,39] Apart from the possibility of realizing doped ladders, their behavior is of great interest to theorists because they are examples of unusual Fermi liquids that can be carefully analyzed. Hole doping of a cuprate introduces effective Cu^{3+} -sites. This oxidation state also favors square planar O-coordination similar to Cu^{2+} -ions and in this coordination a low spin $S=0$ Cu^{3+} -ion is formed which corresponds to a bound state of a $S=1/2$ Cu^{2+} -ion and a hole residing mainly on the four surrounding O 2p-orbitals (Zhang-Rice singlet).[40] Transfer of electrons between n.n. sites allows a $S=1/2$ Cu^{2+} and $S=0$ Cu^{3+} -ion to exchange positions. The canonical model describing the motion of the effective $S=0$ Cu^{3+} -ions in a background of Heisenberg coupled $S=1/2$ Cu^{2+} -ions is known as the $t - J$ model.[41]

The properties of a hole doped single chain have been much studied. It is an example of a Luttinger liquid - so called to distinguish it from the Landau Fermi liquid state that is ubiquitous for interacting fermions at low temperatures in higher dimensions. The simple alternating AF spin pattern of the parent insulator changes its period to an incommensurate value which depends on the doping. The exponent of the power-law decay increases but magnetic correlations still are the dominant ones. The most striking feature of Luttinger liquids is spin-charge separation whereby the charge and spin parts of an added hole move at different velocities and become spatially separated from each other.[5] All these properties are fascinating but do not give a sign of impending superconductivity.

The 2-leg ladder starts from a very different parent state characterized by a spin-gap and exponentially decaying spin correlations. A key question is how these features evolve with doping. Mean-field studies by Sigrist, Rice and Zhang[42] found an increase in the gap upon doping but numerical studies of finite length ladders by Dagotto et al.[6], Poilblanc et al.[17,43] and Noack et al.[12] found a decrease. Detailed studies by Tsunetsugu et al. [44] showed that it was necessary to distinguish two different types of magnetic excitations. Again the limit $J' \gg J$ is useful to gain intuition. As remarked by Dagotto et al.[6], in this limit holes *pair* on the same rung in a $S=0$ and zero momentum state to reduce the cost in magnetic interactions. One type of magnetic excitation is to promote a singlet pair of spins spatially separated from the hole pairs to form a $S=1$ triplet and this excitation evolves smoothly from the magnon we discussed earlier in the undoped case. However, a new type of spin excitation is now possible.[44] This involves separating the hole pair into a state with the holes on two spatially separated rungs, each of which now contains an unpaired spin. This new excitation still requires a finite energy so the spin-gap and the exponential decay of the spin-spin correlations remain, but its appearance at a new and lower energy than the magnon mode leads to a discontinuity in the spin-gap upon doping. Note, since these excitations require holes their number vanishes as the undoped insulator is approached.

The early calculations of Dagotto et al.[6] supported a continuous evolution of the doped system from the anisotropic limit $J' \gg J$ where strong pairing correlations signaling superconductivity were observed, down to the isotropic case $J' = J$. This is similar to the smooth connection observed in the undoped Heisenberg models. The mean field calculations of Sigrist et al.[42] were in agreement and further lead to the observation that holes were paired in a state of approximate d-wave symmetry although the lack of rotational invariance of the lattice here prevents an exact symmetry classification. Calculations by Tsunetsugu et al.[45] confirmed that this ‘d-wave’ paired state for holes persisted down to the limit $J' = J$ and realistic (for cuprates) values of $J/t \sim 0.3$. [46,47] The size of the hole pair is now larger than a single rung but they are spread only over a few lattice spacings. The excitation spectrum of the doped 2-leg ladder contrasts with the Luttinger liquid in that the ladder low energy sector contains only the collective sound mode of the bosonic liquid of hole pairs and a finite energy is needed to make a triplet excitation. These features are

similar to the case of *attractive* fermions in a single chain as analyzed by Luther and Emery [48] rather than the repulsive case which is the Luttinger liquid discussed earlier. Another interesting aspect of lightly doped 2-leg ladders is the way in which they combine features of lightly doped insulators with those of metals with large Fermi surfaces. The former behavior dominates in the energy dependence of the spectral function to add electrons but metallic behavior appears in the momentum dependence of added quasiparticles.[26,45,49]

Binding hole pairs gives them a bosonic character which in turn is a necessary step on the way to superconductivity. However, this alone does not suffice since a groundstate with a crystalline order of hole pairs is also possible.[6] Actually in a quasi-one-dimensional system like a ladder, true long range order will be prevented by quantum fluctuations but a power-law falloff will persist. In the doped ladder this occurs both in the channels corresponding to crystalline ordering of hole pairs and that with superfluid or Bose condensation of hole pairs. The balance between the two and the question of which dominates by means of a smaller exponent depends on the parameters of the model and more generally on residual interactions between hole pairs. This is hard to predict accurately.[45,50] The first set of experiments by Hiroi and Takano [38] on $\text{La}_{1-x}\text{Sr}_x\text{CuO}_{2.5}$, a doped 2-leg ladder system, show substantial decreases in the resistivity upon doping and evidence of metallic behavior in resistivity vs temperature at the highest value of $x = 0.2$ (see Fig.6). There are signs that the spin-gap persists upon doping at least initially but there are no signs of superconductivity. More experiments will be needed to determine if hole pairing exists and if the disorder is suppressing the superconductivity. Nonetheless conceptually the relation of the paired hole state of the doped 2-leg ladder to the superconducting state of the planar cuprates is much closer than the relation to the single chain or Luttinger liquid state.

V. Conclusions

The study of low dimensional quantum antiferromagnets has emerged as a central problem in condensed matter physics due to the discovery of high-Tc superconductivity in lightly doped cuprates with planar structures. Quantum effects are largest in a $S=1/2$ system and with isotropic Heisenberg coupling. A square lattice still has an ordered ground-state although with a substantial reduction of the sublattice magnetization due to quantum

effects. In the one dimensional analog, i.e. a Heisenberg $S=1/2$ chain, the quantum effects overwhelm the long range order but the groundstate has quasi long-range order with a decay in the spin-spin correlation function as an inverse power in the separation, apart from logarithmic corrections.

One might expect that a two leg ladder should be intermediate between a chain and a plane thus the discovery that quantum effects are much stronger in such a ladder and lead to purely short range order with an exponential decay in spin-spin correlations came as a great surprise. This result first found in numerical simulations has now been verified by a variety of techniques and more importantly has experimental confirmation in $(VO)_2P_2O_7$, $SrCu_2O_3$, and $LaCuO_{2.5}$. This difference between a single chain and two-leg ladder extends to all odd- and even-leg ladders, and the difference can be traced to the absence in even-leg ladders of the special topological term that appears in the low energy action of the single chain. This term is also absent in integer spin chains and these also display exponentially decaying spin-spin correlations and a spin-gap.

The various families of high-Tc superconductors all have a unique structural element, namely CuO_2 -planes composed of a square lattice of Cu-ions separated by O-ions. The local coordination is characterized by CuO_4 -squares which in turn are all corner sharing in the CuO_2 -planes. The ladder cuprates again have the same local CuO_4 -coordination but the pattern of the CuO_4 -squares is changed which in turn changes the pattern of magnetic exchange interactions. For example, in $Sr_{n-1}Cu_{n+1}O_{2n}$ line defects break the plane up into weakly coupled ladders. The many ways of assembling CuO_4 -squares illustrates the richness of cuprate chemistry which is only now beginning to be explored and various possibilities for novel quantum groundstates remain to be studied.

The cuprates have another unique feature among transition metal oxides, namely the possibility of hole doping without localization to realize conducting materials. The doped chain has been the paradigm of a non-Landau Fermi liquid and much attention has focussed on the unique properties of this quantum liquid, called a Luttinger liquid by Haldane, such as the complete separation of charge and spin sectors into two excitation branches at low energy. The hole doped 2-leg ladder is also essentially one dimensional but now the properties are radically different. As we discussed above, the quantum liquids in lightly doped ladders retain the spin-gap, show hole-hole pairing in approximate $d_{x^2-y^2}$ -symmetry,

and although lightly doped insulators they show features of a large Fermi surface which is metal-like. Doped ladders are a fascinating mixture of a dilute Fermi gas with strong attractions and a concentrated Fermi system with a large Fermi surface.

Returning to the high- T_c cuprates we see a paradox. The parent insulating antiferromagnets show long range order, which represents a smooth evolution or crossover from the properties of single chains but not from 2-leg ladders. Lightly doped cuprates by contrast show a spin gap and $d_{x^2-y^2}$ -superconductivity, properties we can imagine evolving smoothly from the 2-leg ladders. While much remains to be done to understand how these features fit together, it is clear that the study of ladders has given us not only surprises but valuable new insights into low dimensional quantum systems and a new impetus to broaden our horizons and explore the rich solid state chemistry of cuprates and related materials.

Acknowledgments

We thank M. Azuma, T. Barnes, H. Hiroi, A. Moreo, J. Riera, D. Scalapino, M. Takano, M. Troyer, and H. Tsunetsugu for their help in the preparation of this review. E.D. specially thanks the Office of Naval Research under grant ONR N00014-93-0495 for its support. He also thanks the NHMFL and MARTECH for additional support.

FIGURE CAPTIONS

Fig.1: (a) Ladder compound $(\text{VO})_2\text{P}_2\text{O}_7$. O- and V-ions are indicated (from D. C. Johnston et al., Phys. Rev. **B35**, 219 (1987)); (b) Schematic representation of the 2-leg compound SrCu_2O_3 and the 3-leg compound $\text{Sr}_2\text{Cu}_3\text{O}_5$ (from M. Azuma et al., Phys. Rev. Lett. **73**, 3463 (1994)). The black dots are copper, while the intersections of the solid lines are oxygen locations. The dashed lines are Cu-O bonds. The 2- and 3-leg structures are highlighted. J is the coupling along the chains, and J' along the rungs.

Fig.2: (a) Spin gap Δ_{spin} vs. J'/J for the 2-leg ladder. The results are extrapolations to the bulk limit using numerical results obtained on finite $2 \times N$ clusters (from T. Barnes et al., Phys. Rev. **B47**, 3196 (1993)); (b) Triplet spin-wave excitation spectra for the isotropic point $J' = J$, with $J = 7.79$ meV and using the $(\text{VO})_2\text{P}_2\text{O}_7$ lattice spacing (from T. Barnes and J. Riera, Phys. Rev. **B50**, 6817 (1994)). k_r (k_c) denotes momentum along the rungs (chains).

Fig.3: Magnetic susceptibility $\chi(T)$ calculated with Monte Carlo techniques on m -leg ladders and $J' = J$ on clusters of $m \times 100$ sites. $\chi(T)$ for even-leg ladders show at low temperature the exponential suppression caused by the spin-gap, while the odd-leg ladders extrapolate to a finite number as $T \rightarrow 0$ (from B. Frischmuth, M. Troyer and D. Würtz, preprint).

Fig.4: (a) Experimental magnetic susceptibility $\chi(T)$ for $(\text{VO})_2\text{P}_2\text{O}_7$ (from D. C. Johnston et al., Phys. Rev. **B35**, 219 (1987)); same but for (b) SrCu_2O_3 and (c) $\text{Sr}_2\text{Cu}_3\text{O}_5$ (from M. Azuma et al., Phys. Rev. Lett. **73**, 3463 (1994)).

Fig.5: Neutron scattering data for $(\text{VO})_2\text{P}_2\text{O}_7$ showing the finite spin-gap E_g (from R. S. Eccleston et al., Phys. Rev. Lett. **73**, 2626 (1994)). We refer the reader to this reference for further details.

Fig.6: (a) Resistivity vs temperature parametric with the Sr-concentration x for $\text{La}_{1-x}\text{Sr}_x\text{CuO}_{2.5}$; (b) Magnetic susceptibility vs temperature for the same compound shown in (a) (from Z. Hiroi and M. Takano, Nature Vol.**377**(7), 41 (1995)).

REFERENCES

- 1 J. Bednorz and K. Müller, *Z. Phys.* **B 64**, 188 (1986).
- 2 E. Manousakis, *Rev. Mod. Phys.* **63**, 1 (1991).
- 3 H. Bethe, *Z. Phys.* **71**, 205 (1931).
- 4 P. W. Anderson, *Science* **235**, 1196 (1987).
- 5 For a recent review see H. J. Schulz, *Proceedings of Les Houches Summer School LXI: Mesoscopic Quantum Physics*, Eds. E. Akkermans, G. Montambaux, J.L Pichard, and J. Zinn-Justin, Elsevier, Amsterdam, to be published; and references therein.
- 6 E. Dagotto, J. Riera, and D. Scalapino, *Phys. Rev.* **B45**, 5744 (1992).
- 7 R. Hirsch, Diplomarbeit, Universität Köln (1988).
- 8 Dagotto and Moreo, *Phys. Rev.* **B38**, 5087 (1988).
- 9 T. Barnes, E. Dagotto, J. Riera and E. Swanson, *Phys. Rev.* **B47**, 3196 (1993).
- 10 E. Dagotto, *Rev. Mod. Phys.* **66**, 763 (1994).
- 11 S. White, R. Noack, and D. Scalapino, *Phys. Rev. Lett.* **73**, 886 (1994).
- 12 R. Noack, S. White, and D. Scalapino, *Phys. Rev. Lett.* **73**, 882 (1994).
- 13 M. Azzouz, L. Chen, and S. Moukouri, *Phys. Rev.* **B50**, 6233 (1994).
- 14 S. Gopalan, T. M. Rice and M. Sigrist, *Phys. Rev.* **B49**, 8901 (1994).
- 15 S. A. Kivelson, D. S. Rokhsar, and J. P. Sethna, *Phys. Rev.* **B 35**, 8865 (1987).
- 16 T. M. Rice, S. Gopalan, and M. Sigrist, *Europhys. Lett.* **23**, 445 (1994). See also H. J. Schulz, *Phys. Rev.* **B 34**, 6372 (1986); I. Affleck, *Phys. Rev.* **B 37**, 5186 (1988); D. S. Rokhsar and S. A. Kivelson, *Phys. Rev. Lett.* **61**, 2376 (1988); N. E. Bonesteel, *Phys. Rev.* **B 40**, 8954 (1989); and references therein.
- 17 D. Poilblanc, H. Tsunetsugu, and T. M. Rice, *Phys. Rev.* **B50**, 6511 (1994).
- 18 N. Hatano and Y. Nishiyama, to be published in *J. Phys. A: Math. Gen.*
- 19 M. Reigrotzki, H. Tsunetsugu, and T. M. Rice, *J. Phys. C: Cond. Matt.* **6**, 9325 (1994).
- 20 D. V. Khveshchenko, *Phys. Rev.* **B50**, 380 (1994) and D. G. Shelton, A. A. Nersesyan and A. M. Tsvelik, preprint.
- 21 F. D. M. Haldane, *Phys. Rev. Lett.* **61**, 1029 (1988).
- 22 For work with ferromagnetic rungs see K. Hida, *J. Phys. Soc. Jpn.* **60**, 1347 (1991), H. Watanabe, *Phys. Rev.* **B 50**, 13442 (1994), and references therein.

- 23 S. P. Strong and A. J. Millis, Phys. Rev. Lett. **69**, 2419 (1992); Phys. Rev. **B 50**, 9911 (1994); Y. Xian, Manchester preprint; Y. Nishiyama, N. Hatano and M. Suzuki, to appear in J. Phys. Soc. Jpn. **64**, No.6, (1995); S. R. White, preprint; H. Watanabe, preprint; D. Sénéchal, preprint.
- 24 T. Barnes and J. Riera, Phys. Rev. **B50**, 6817 (1994).
- 25 The dynamical spin structure factor $S(\mathbf{Q}, \omega)$, with $\mathbf{Q} = (\pi, \pi)$ has a sharp peak at $\omega = \Delta_{\text{spin}} \approx 0.5J$ that carries most of the weight. See Refs.[9,14].
- 26 M. Troyer, H. Tsunetsugu and T. M. Rice, ETH-preprint.
- 27 M. Troyer, H. Tsunetsugu, and D. Würtz, Phys. Rev. **B 50**, 13515 (1994).
- 28 A. W. Sandvik, E. Dagotto, and D. J. Scalapino, preprint.
- 29 In addition, the nuclear spin relaxation rate has been calculated by Troyer et al.[27] predicting $1/T_1 \sim \exp(-\Delta_{\text{spin}}/T)(a + \ln T)$ at low temperatures. Sandvik et al.[28] have also calculated $1/T_1$ using numerical techniques.
- 30 B. Frischmuth, M. Troyer and D. Würtz, preprint.
- 31 D. C. Johnston, J. W. Johnson, D. P. Goshorn and A. P. Jacobson, Phys. Rev. **B35**, 219 (1987).
- 32 R. S. Eccleston, T. Barnes, J. Brody and J. W. Johnson, Phys. Rev. Lett.**73**, 2626 (1994).
- 33 Z. Hiroi, M. Azuma, M. Takano and Y. Bando, J. Sol. State Chem. **95**, 230 (1991).
- 34 M. Azuma, Z. Hiroi, M. Takano, K. Ishida and Y. Kitaoka, Phys. Rev. Lett. **73**, 3463 (1994).
- 35 K. Ishida, Y. Kitaoka, K. Asayama, M. Azuma, Z. Hiroi and M. Takano, J. Phys. Soc. Japan **63**, 3222 (1994); K. Ishida, Y. Kitaoka, Y. Tokunaga, S. Matsumoto, K. Asayama, M. Azuma, Z. Hiroi and M. Takano, preprint.
- 36 K. Kojima, et al., Phys. Rev. Lett. **74**, 2812 (1995).
- 37 B. Batlogg et al., Bull. Am. Phys. Soc. **40**, 327 (1995).
- 38 Z. Hiroi and M. Takano, Nature Vol.**377**(7), 41 (1995).
- 39 M. Azuma, M. Takano, T. Ishida, and K. Okuda, preprint, recently presented magnetic susceptibility measurements for Zn-doped 2- and 3-leg ladders.
- 40 F. Zhang and T. M. Rice, Phys. Rev. **B 37**, 3759 (1988).

- 41 Note that in the case of $(\text{VO})_2\text{P}_2\text{O}_7$, it is not clear whether the $t - J$ model would be suitable for describing its properties upon doping. A detailed analysis based on a many orbital Hubbard model for V- and O-ions is needed.
- 42 M. Sigrist, T. M. Rice and F. C. Zhang, Phys. Rev. B **49**, 12058 (1994).
- 43 D. Poilblanc, D. J. Scalapino, and W. Hanke, Phys. Rev. B (in press).
- 44 H. Tsunetsugu, M. Troyer and T. M. Rice, Phys. Rev. B **49**, 16078 (1994).
- 45 H. Tsunetsugu, M. Troyer, and T. M. Rice, Phys. Rev. B **51**, 16456 (1995). See also J. A. Riera, Phys. Rev. B **49**, 3629 (1994).
- 46 C. A. Hayward, D. Poilblanc, R. M. Noack, D. J. Scalapino and W. Hanke, Phys. Rev. Lett. **75**, 926 (1995).
- 47 Calculations of superconducting correlations for the Hubbard model were discussed in Ref.[12] and also in Y. Asai, Phys. Rev. B **50**, 6519 (1994), and preprint; Yamaji and Y. Shimoi, Physica C **222**, 349 (1994); and R. M. Noack, S. R. White and D. J. Scalapino, Europhys. Lett. **30**, 163 (1995), and preprint.
- 48 A. Luther and V. J. Emery, Phys. Rev. Lett. **33**, 589 (1974).
- 49 Note the similarities between the results for 2-leg ladders and those corresponding to models of high- T_c superconductors where the infrared Drude weight scales as the hole concentration, while the Fermi surface is large and electron-like.[see Ref.10]
- 50 Several recent publications have addressed these issues: N. Nagaosa (Univ. of Tokyo preprint) claimed that the low energy physics of doped ladders is described in terms of bipolarons. L. Balents and M. P. A. Fisher (ITP preprint) studying the Hubbard model ladder with a controlled RG method found a phase with a finite spin-gap and a single gapless charge mode that they interpret as the analog of a 1D superconductor or a charge density wave. H. Schulz (Orsay preprint) studied two coupled Luttinger liquids reporting a spin-gap in the spectrum and either dominant charge density wave or singlet pairing correlations in the ground state.