PHYSICAL REVIEW B VOLUME 56, NUMBER 10 1 SEPTEMBER 1997-II

Phase diagram of the $S=\frac{1}{2}$ frustrated coupled ladder system

B. Normand
Theoretische Physik, ETH-Hönggerberg, CH-8093 Zürich, Switzerland

K. Penc
Max Planck Institut für Physik komplexer Systeme, Bayreuther Strasse 40, D-01187 Dresden, Germany

M. Albrecht and F. Mila
Laboratoire de Physique Quantique, Université Paul Sabatier, 118 Route de Narbonne, 31062 Toulouse Cedex, France

(Received 25 March 1997)

We present a theoretical study of the magnetic phase diagram of the frustrated coupled ladder structure realized recently in several materials. This system displays a nondegenerate spin-gap state in the dimer limit and an infinitely degenerate spin-gap state in the regime of weakly coupled zigzag chains. Between these we demonstrate the existence of gapless, magnetically ordered regions whose order is antiferromagnetic close to the honeycomb lattice limit, and incommensurate along the chains when all three magnetic interactions compete. [S0163-1829(97)50634-5]

Spurred by developments in the field of high-temperature superconductivity, rapid progress is now being made in the preparation of materials with similar attributes, and there is potential for fascinating new physics. These are inorganic, low-dimensional quantum magnets, where the relative strengths of the magnetic interactions, which in most cases give antiferromagnetic coupling between ions with spin $S=\frac{1}{2}$, result in systems such as chains and ladders, which are effectively one-dimensional (1D), or 2D coupled ladders and depleted planes. One structure of particular interest is the frustrated coupled ladder, shown in Fig. 1. This conformation is realized in the cases of the prototypical ''ladder'' compound[1] $SrCu_2O_3$, where the frustrating interaction J_1 is ferromagnetic (FM) and small, and in the depleted planar compounds CaV_2O_5 (Ref. 2) and MgV_2O_5.[3] In each of the latter systems, all three interactions are thought to be of similar strength, but the physical properties of the materials differ dramatically.

The phase diagram obtained on alteration of the parameters J_1, J_2, and J_2' is particularly rich. The two-chain ladder obtained at $J_1=0$ has a singlet ground state of resonating dimers with a gap to all spin excitations, while the zigzag chain at $J_2'=0$ shows for $J_2>J_{2c}$ a doubly degenerate ground state of alternating dimers which also has a gap. What appears between these limits remains poorly understood, and is one subject of the current analysis, but we may gain an initial indication from the honeycomb lattice obtained when $J_2=0$, where the gapped J_2' dimer phase is replaced by a magnetically ordered state at a critical value of increasing J_1. We investigate the system using a variety of analytical and numerical methods, proceeding (a) from the limit of ladder rung dimers, (b) from that of weakly coupled zigzag chains, and (c) by considering the ordered phases which are found to occur between these limits.

(a) Dimer limit. In the limit of large J_2', the system has a nondegenerate ground state, whose wave function is a product of rung dimer singlets, with an energy gap to spin excitations. This state is well suited to examination by the bond-operator technique,[4] which is based on transforming the four spin states on each rung to one singlet and three triplets. The gap between these gives the stability of dimer order. The method has also been shown[5] to be applicable to spin ladders (vanishing J_1) for values of J_2 up to J_2', and can be expected to have similar validity in the case of the anisotropic honeycomb lattice (vanishing J_2).

Following the treatment of Ref. 6 for a unit cell containing two dimers, and retaining in the spin-gap regime terms only to quadratic order in the triplet spin excitations $t_{i\alpha}^{\dagger}$, we obtain a system of threefold degenerate magnons with energy spectrum

$$\omega_{\mathbf{k}}^{\pm}=(\tfrac{1}{4}J_2'-\mu)\sqrt{1+d a_k^{\pm}}, \tag{1}$$

where $d=2J_2'\overline{s^2}/(\frac{1}{4}J_2'-\mu)$,

$$a_{\mathbf{k}}^{\pm}=\lambda\cos k_z\pm\lambda'\cos\tfrac{1}{2}k_x\cos\tfrac{1}{2}k_z; \tag{2}$$

$\overline{s}$ denotes the magnitude of the singlet condensate, μ is the global chemical potential, and we define $\lambda=J_2/J_2'$ and $\lambda'=J_1/J_2'$. Solution of the mean-field equations at $T=0$ gives the most important property characterizing the nondegenerate ground state, the spin gap

$$\Delta=(\tfrac{1}{4}J_2'-\mu)\sqrt{1-d\ f(\lambda,\lambda')}. \tag{3}$$

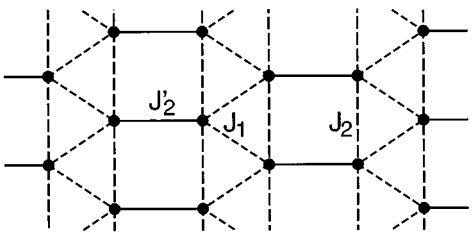

FIG. 1. Schematic representation of the frustrated coupled ladder system.

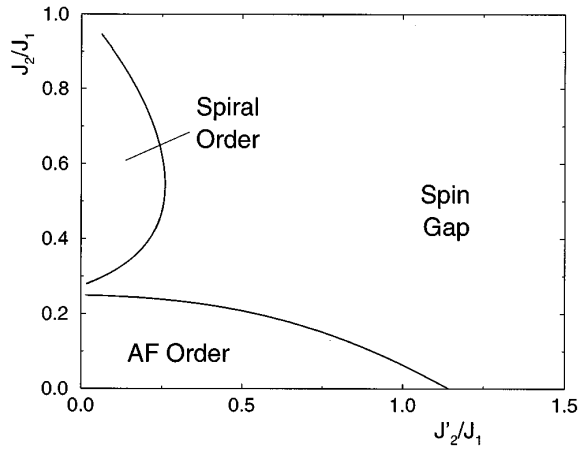

FIG. 2. Phase diagram of the coupled dimer system, shown in the plane of J_2'/J_1 and J_2/J_1, as deduced from the bond-operator method.

For $J_1>4J_2$, $f=\lambda'-\lambda=a^{-}_{k,\min}$ at the commensurate wave vector $k_{\mathrm{M}}=(0,0)$, while for $J_2>\frac{1}{4}J_1$ one has $f=\lambda+\lambda'^2/8\lambda$ occurring at an incommensurate $k_{\mathrm{M}}=(0,2\cos^{-1}(\lambda'/4\lambda))$. In the spin-gap regime, the maximum of the static structure factor $S(k)$ coincides with the minimum of the gap, and can be shown to move to incommensurate wave vectors with increasing J_2 beyond $\frac{1}{4}J_1$. The boundaries of the spin-gap phase are found where the $\Delta\to 0$, and are shown in Fig. 2. When $J_1>4J_2$, from k_{M} one may deduce a FM arrangement of rung spin pairs ($|\uparrow\downarrow\rangle$), which corresponds to antiferromagnetic (AF) order along the J_1 chains. For $J_2>\frac{1}{4}J_1$, k_{M} describes spiral order along the chains with exactly the pitch known from the classical solution for the J_1-J_2 chain. The phase boundaries in Fig. 2 appear well outside the regime of validity of the bond-operator technique, which overestimates the stability of the dimer state. However, they remain qualitatively correct, particularly in showing the instability of the dimer liquid to the different ordered phases, and furthermore provide a useful indication of where to apply alternative approaches.

(b) Frustrated chain limit. For large J_1 or J_2, the system consists of weakly coupled chains, whose properties are well known from the Bethe Ansatz solution. However, how these are affected by the weak couplings, which may or may not be frustrating, is an extremely subtle issue to which we shall return below. Here we consider the limit of small J_2', where the problem is that of the J_1-J_2 chain. This is a classic example of frustration in 1D, and we review briefly its properties (see Ref. 7, and Refs. 12–18 therein). At the Majumdar-Ghosh (MG) point $J_2=\frac{1}{2}J_1$, there is an exact dimer wave function solution[8] with a twofold degenerate ground state and gapped excitation spectrum.[9] The wave function is composed of frozen singlets on alternating J_1 bonds, and has two possible realizations differing by a translation of one unit along the chain. The gap decreases exponentially with J_2-J_{2c} (Ref. 10) on approach to the conformal point $J_{2c}/J_1=0.2412$, below which the frustrated chain retains the gapless spectrum of the nearest-neighbor spin chain. For $J_2>\frac{1}{2}J_1$ the gap increases to a maximum at $0.6<J_2/J_1<0.7$, then tends towards an exponential decay with $-J_2/J_1$ for large J_2. The static spin-spin correlation function $S(k)$ (Ref. 11) has a maximum at $k=\pi$ for $J_2\leqslant\frac{1}{2}J_1$, and for $J_2>\frac{1}{2}J_1$ is maximal between $k=\frac{1}{2}\pi$ and π, indicating predominant spiral correlations.

We examine the lifting of degeneracy and the closing of the singlet-triplet gap Δ by an adapted perturbational approach, which due to the degenerate nature of the ground state is in a strict sense variational. The Hamiltonian is written as $H=H_0+H'$, where

$$H_0=J_1\sum_i \hat{\mathbf{S}}_{i,l}\cdot\hat{\mathbf{S}}_{i+1,l}+J_2\sum_i \hat{\mathbf{S}}_{i,l}\cdot\hat{\mathbf{S}}_{i+2,l}, \tag{4}$$

$$H'=J_2'\sum_{i,l\ \mathrm{even}} \hat{\mathbf{S}}_{i,l}\cdot\hat{\mathbf{S}}_{i,l+1}+J_2'\sum_{i,l\ \mathrm{odd}} \hat{\mathbf{S}}_{i,l}\cdot\hat{\mathbf{S}}_{i,l+1}, \tag{5}$$

and in H' both i and l take either even or odd values only. We construct the variational wave function

$$|\Psi_j\rangle=|S\rangle_0\otimes\cdots\otimes|S\rangle_{j-1}\otimes|T\rangle_j\otimes|S\rangle_{j+1}\cdots, \tag{6}$$

in which $|S\rangle_i=\eta|S_0\rangle+\eta'|S_\pi\rangle$ is a linear combination of the two degenerate ground states on the ith chain. $|T\rangle_j=\Sigma_\alpha(\xi_{k,\alpha}|T_{k,\alpha}\rangle+\xi_{k+\pi,\alpha}|T_{k+\pi,\alpha}\rangle)$, where $|T_{k,\alpha}\rangle$ is the eigenstate with momentum k of the isolated zigzag chain, denotes a triplet excitation of the jth chain. In the thermodynamic limit these form a continuum above the gap, and their delocalization by H' gives a kinetic energy gain which reduces Δ.

In the regime $J_2<\frac{1}{2}J_1$ where the spin correlations are predominantly antiferromagnetic, the lowest-lying triplet excitations appear at $k=0$ and $k=\pi$. Here η or $\eta'=0$, and the ''gap equation'' for J_2' takes the simplified form

$$\frac{1}{J_2'}=\max\sum_\alpha\left\{\frac{|\langle T_{\pi,\alpha}|\hat{S}^z_\pi|S_0\rangle|^2}{\epsilon_\alpha-\epsilon_S-\Delta},\ \frac{|\langle T_{0,\alpha}|\hat{S}^z_\pi|S_\pi\rangle|^2}{\epsilon_\alpha-\epsilon_S-\Delta}\right\}, \tag{7}$$

where ϵ_S and ϵ_α are the ground and excited state energies, and may be evaluated in terms of the spin-spin correlation function

$$\mathrm{Im}\,S^{zz}_q(\omega,k)=\sum_\alpha|\langle T_{k+q,\alpha}|\hat{S}^z_k|S_q\rangle|^2\delta(\omega-\omega_{k+q}), \tag{8}$$

in which $\omega_{k+q}=\epsilon_{k+q,\alpha}-\epsilon_S$. These expressions are calculated using a Lánczos algorithm for chain lengths up to $N=22$ sites. Setting $\Delta=0$ yields the value J_{2c}' where the ordered phases appear. J_{2c}' scales with the unperturbed gap magnitude Δ_0, and vanishes at $J_2=J_{2c}$. For $J_2>\frac{1}{2}J_1$, the situation is more complicated because the lowest triplet excitations appear at intermediate momenta, corresponding to predominant incommensurate spin correlations.[11] There is no decoupling as above, necessitating solution of the full J_2' equation and calculation of the energy by minimizing with respect to η and η'. $\Delta\to 0$ for a value of J_2' whose dependence on Δ_0 varies because of the incommensuration, as shown in the inset of Fig. 3. That gap closure occurs at a wave vector away from π we take as an indication of spiral magnetic order. The method of solution is most accurate where the gap of the dimerized phase is large, and we display in Fig. 3 the phase diagram for the parameter regime

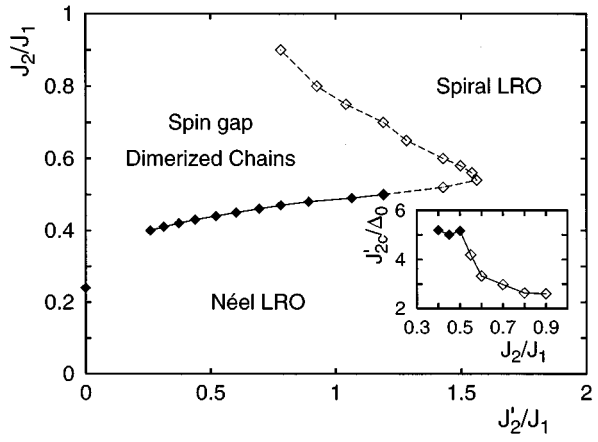

FIG. 3. Phase diagram of the coupled zigzag chain system, as deduced from the variational method. Solid symbols denote the commensurate solution ($J_2 < \frac{1}{2}J_1$) and empty symbols the incommensurate.

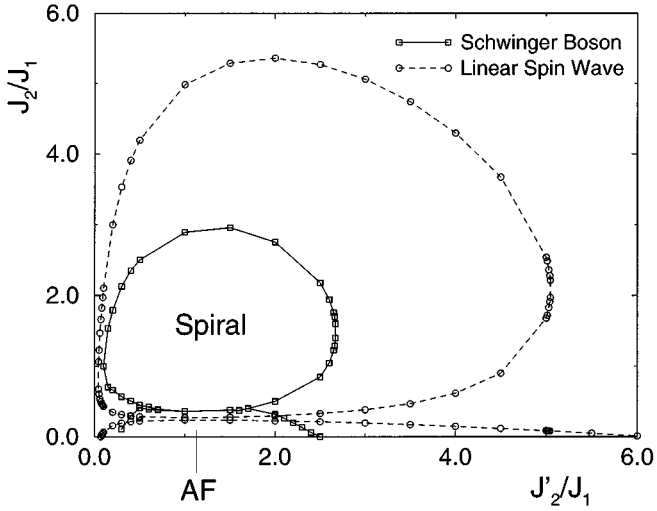

FIG. 4. Phase boundaries of the ordered regions, deduced by the Schwinger boson and linear spin-wave techniques.

$0.4 < J_2/J_1 \leqslant 1$. The nature of the magnetic order stabilized by increasing J_2' corresponds to the wave vector k_M maximizing $S(k)$.[11]

We may analyze the energy shift and splitting of the ground state by applying second-order degenerate perturbation theory. At this level one need only consider two coupled chains, which have a fourfold degenerate ground state, and the energy correction per site is a 4×4 matrix conveniently calculated using the Lánczos algorithm. The correction is found to be proportional to the unit matrix, so the degeneracy is not lifted in the thermodynamic limit to second order. This result suggests that the degeneracy of the ground state will be reflected in the presence of low-lying singlet states in the singlet-triplet gap for small J_2'. We note that this behavior contrasts with the lifting of degeneracy which occurs in a system of gapped spin chains where i and l may take all values in Eq. (5).

(c) Ordered phases. These may be treated by expanding in fluctuations around a state with fixed moments, of periodically varying orientation, on each site. We present here results from the Schwinger boson and linear spin-wave methods. In the Schwinger boson transformation,[12,13] ordered solutions are described by a Bose condensation where $\langle b \rangle \neq 0$ becomes a parameter in the system of mean-field equations, and the chemical potential μ is determined by the requirement that the excitation spectrum is gapless (Goldstone modes). We find both long-range order of Néel type, where all correlations are AF only, and spiral order where there are one condensate and five bond-order parameters in the solution. The latter number arises because all links J_1 are equivalent, as are all links J_2, i.e., no solutions can be found where there is a symmetry breaking analogous to the dimerization in the MG state, so in general only two order parameters are required for each type; J_2' links are AF only, requiring one. Disordered solutions have no Bose condensation, a gapped excitation spectrum and a six-parameter solution including μ. The phase diagram is shown in Fig. 4. It is gratifying to find that the AF and spiral ordered phases appear in the expected locations, and that the phase boundaries are in qualitative agreement with those deduced from both the dimer and zigzag chain limits. We note in particular the two features that (i) there is an upper boundary for the regime of spiral order at sufficiently large J_2 and (ii) there is a continuous transition directly from AF- to spiral-ordered regimes.

Applying the linear spin-wave technique, again (Fig. 4) we find regimes with both AF and spiral long-range order, and again there is a definite upper limit in J_2 on the latter. In this case there is no direct transition between the ordered phases, which are always separated by a disordered region. This can be shown in detail by studying the ground-state energy and staggered magnetization. A similar contradiction has been found previously[13] in the square lattice with frustrating next-neighbor superexchange. For this feature we have no way of distinguishing between the phase diagrams of Fig. 4 on the basis of these studies alone, and leave as open the possibility that the transition between ordered phases may be continuous along a finite line in parameter space. We may not exclude the existence of a phase which is none of the four discussed hitherto, as an intermediate between both the two ordered phases and the two (degenerate and nondegenerate) gapped phases.

All of the approximations applied have tended to overestimate the extent of the parent region, although the deduced phase boundaries are in good qualitative agreement. The quantitative aspects may be addressed by numerical techniques. When J_2 is zero, the system is an anisotropic honeycomb lattice, which has an unfrustrated ordered state for suitably large J_1 ($J_2' \neq 0$), and a spin-gap phase otherwise. Because the absence of frustration removes the sign problem, the quantum critical point on this axis can be found essentially exactly by large-system quantum Monte Carlo (QMC) studies. The result $J_2'/J_1 = 1.74$ (Ref. 17) fixes the unknown end of the phase boundary of the nondegenerate spin-gap regime. Comparison with the bond-operator (1.15) and Schwinger boson (2.65) results indicates the validity of each. We have in addition performed exact diagonalization (ED) studies on small systems, from which for the present purposes it is possible to locate crossover regions between phases by analyzing the quantum numbers of the ground states. This work remains in progress, and the results will be presented in detail elsewhere.

We conclude with a more schematic discussion of the important limits of large J_1 and large J_2. When J_1 is large, the system is one of weakly coupled chains with a small, marginally relevant next-neighbor frustration J_2. The action of the interchain coupling J_2' has been discussed extensively in recent literature for the anisotropic square lattice.[14–16] Despite the difference in behavior noted above for this geometry when $J_2 > J_{2c}$, we may, following Ref. 15, cast the interchain interaction as an effective, staggered mean field and deduce that any finite J_2' leads to magnetic order while the spectrum of the isolated chains remains gapless. Thus for any frustration $J_2 < J_{2c}$, the presence of a J_2' term can be expected by this argument to induce AF order. It remains to compute the value of J_{2c} on a frustrated chain in the presence of the effective staggered field: if this quantity does not track the phase boundary of the multiply degenerate MG-type ground state, we are presented with the existence of an intermediate phase in the region of the conformal point and the known phase boundary. This may be the state of no magnetic order and no spin gap arising in the above discussions,[14,16] which, following the analysis of the ordered states, may also be a candidate intermediate regime between AF and spiral order.

At large J_2 the problem is one of chains with competing ladder and frustrated couplings. The ladder coupling is relevant, and opens a gap Δ linear in J_2'/J_2 to the nondegenerate state. The zigzag chain coupling opens an exponentially small gap[7] $\Delta \sim \exp(-J_2/J_1)$ to the multiply-degenerate state. How each bond type affects the gapped state established by the other is unknown, as both gaps may close, leaving some intermediate phase, or there may be a crossover between the gapped states. In the former case, our results provide a strong indication against the possibility that the gapless phase is ordered, but not that of a disordered, gapless phase in 2D arising due to the competition of the couplings. In the latter case, one may postulate a crossover where the singlet-triplet gap changes smoothly, remaining finite while the low-lying singlet states of the zigzag chain limit are simply split by increasing J_2'/J_1, or may also undergo a level crossing with a higher-lying singlet. Examination of ground-state quantum numbers by ED will be particularly useful in resolving this issue.

Figure 5 summarizes the preceding analyses and discussion. The shaded regions represent those where open questions remain to be addressed. Returning to the materials which have the frustrated coupled ladder structure, the original ladder compound $SrCu_2O_3$ would appear close to the isotropic point on the axis $J_1=0$, and is well described as a liquid of resonating singlets residing primarily on the ladder rungs. That the frustrating interaction is actually FM is of little consequence here. CaV_2O_5 shows a large spin gap, in qualitative agreement with expectation for a system with $J_1 \sim J_2 \sim J_2'$,[18] as we have seen that the frustrating interaction J_1 must be significantly larger than the ladder terms in order to remove the system from the nondegenerate gapped phase. MgV_2O_5 is found to have a very small spin gap and a peak in the static susceptibility at a temperature three times smaller than that in the Ca analog, results which may indicate close proximity to the phase boundary to spiral order. Finally, the zigzag chain compounds $SrCuO_2$ (Ref. 1) and LiV_2O_5 (Ref. 19) do not have exactly the structure considered here. However, both materials appear to fall in extreme limits of J_2/J_1, and characterization of their ground states would nevertheless yield valuable information about the effects of weak interchain coupling in the regimes discussed above.

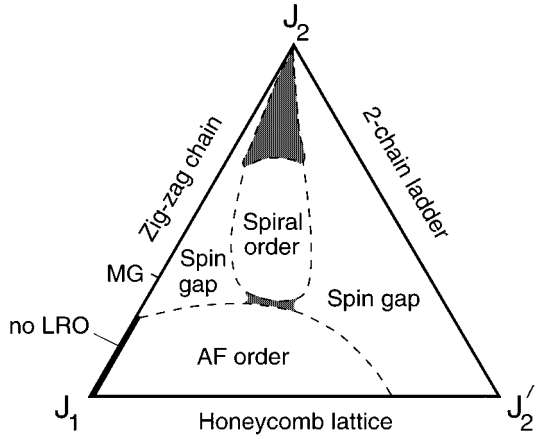

FIG. 5. Schematic phase diagram of the frustrated coupled ladder system.

In summary, the frustrated coupled ladder system represents well the wealth of interesting physics to be found in low-dimensional quantum magnets, and provides a valuable means of studying quantum phase transitions.

We are indebted to M. Troyer for the quoted QMC result, and acknowledge also useful discussions with J. Bonvoisin, P. Millet, and D. Poilblanc.

[1] Z. Hiroi, M. Azuma, M. Takano, and Y. Bando, J. Solid State Chem. **95**, 230 (1991).
[2] See J. C. Bouloux and J. Galy, J. Solid State Chem. **16**, 385 (1976), and references therein.
[3] P. Millet and J. Bonvoisin (private communication); Y. Ueda (private communication).
[4] S. Sachdev and R. Bhatt, Phys. Rev. B **41**, 9323 (1990).
[5] S. Gopalan, T. M. Rice, and M. Sigrist, Phys. Rev. B **49**, 8901 (1994).
[6] B. Normand and T. M. Rice, Phys. Rev. B **54**, 7180 (1996).
[7] S. R. White and I. Affleck, Phys. Rev. B **54**, 9862 (1996).
[8] C. K. Majumdar and D. K. Ghosh, J. Math. Phys. (N.Y.) **10**, 1388 (1969).
[9] B. S. Shastry and B. Sutherland, Phys. Rev. Lett. **47**, 964 (1981).
[10] F. D. M. Haldane, Phys. Rev. B **25**, 4925 (1982).
[11] T. Tonegawa and I. Harada, J. Phys. Soc. Jpn. **56**, 2153(1987).
[12] D. P. Arovas and A. Auerbach, Phys. Rev. B **38**, 316 (1988).
[13] F. Mila, D. Poilblanc, and C. Bruder, Phys. Rev. B **43**, 7891 (1991).
[14] I. Affleck, M. P. Gelfand, and R. R. P. Singh, J. Phys. A **27**, 7313 (1994); **28**, 1787(E) (1995); I. Affleck and B. I. Halperin, *ibid.* **29**, 2627 (1996).
[15] H. J. Schulz, Phys. Rev. Lett. **77**, 2790 (1996).
[16] Z. Wang, Phys. Rev. Lett. **78**, 126 (1997).
[17] M. Troyer (private communication).
[18] A recent analysis by S. Marini and D. I. Khomskii, (unpublished), argues that this system is significantly closer to the dimer limit (the vertex J_2' in Fig. 5), improving agreement with experiment.
[19] M. Isobe and Y. Ueda, J. Phys. Soc. Jpn. **65**, 3142 (1996).