

Soliton binding and low-lying singlets in frustrated odd-legged $S=\frac{1}{2}$ spin tubes

Andreas Lüscher,¹ Reinhard M. Noack,² Grégoire Misguich,³ Valeri N. Kotov,⁴ and Frédéric Mila⁴

¹*School of Physics, University of New South Wales, Sydney 2052, Australia*

²*Fachbereich Physik, Philipps-Universität Marburg, D-35032 Marburg, Germany*

³*Service de Physique Théorique, CEA Saclay, 91191 Gif-sur-Yvette Cedex, France*

⁴*Institute of Theoretical Physics, École Polytechnique Fédérale de Lausanne, BSP, CH-1015 Lausanne, Switzerland*

(Received 7 May 2004; published 25 August 2004)

Motivated by the intriguing properties of the vanadium spin tube $\text{Na}_2\text{V}_3\text{O}_7$, we show that an effective spin-chirality model similar to that of standard Heisenberg odd-legged $S=\frac{1}{2}$ spin tubes can be derived for frustrated inter-ring couplings, but with a spin-chirality coupling constant α that can be arbitrarily small. Using density matrix renormalization group and analytical arguments, we show that, while spontaneous dimerization is always present, solitons become *bound* into low-lying singlets as α is reduced. Experimental implications for strongly frustrated tubes are discussed.

DOI: 10.1103/PhysRevB.70.060405

PACS number(s): 75.10.Jm, 75.10.Pq, 75.50.Ee

Spin ladders, systems which consist of a finite number of coupled chains, have attracted considerable attention recently.¹ Spin- $\frac{1}{2}$ ladders with an even number of legs are expected to have a spin gap, while ladders with an odd number of legs behave like spin chains at low energy. Both predictions have been largely confirmed experimentally. Here we have taken the ladders to have open boundary conditions in the rung direction, which is the most natural definition. In comparison, spin ladders with periodic boundary conditions in the rung direction (see Fig. 1), often referred to as spin tubes, have received much less attention, mostly because the prospect for experimental realizations was remote. Nonetheless, it was noticed early on that spin tubes with an odd number of legs are not expected to behave in the same way as their ladder counterparts. The crucial observation is that the ground state of a ring with an odd number of sites is not just twofold degenerate, as for a rung in an odd-leg ladder, but is fourfold degenerate: In addition to the Kramers degeneracy of the spin- $\frac{1}{2}$ ground state, there is a degeneracy due to the two possible signs of the ground-state momentum, leading to an extra degree of freedom on top of the total spin, often called the chirality by extension of the case of a triangle. As a consequence, a standard L -leg spin tube [Fig. 1(a)], defined by the Hamiltonian

$$H = J \sum_{r,l=1}^{N,L} \boldsymbol{\sigma}_{r,l} \cdot \boldsymbol{\sigma}_{r,l+1} + J' \sum_{r,l=1}^{N-1,L} \boldsymbol{\sigma}_{r,l} \cdot \boldsymbol{\sigma}_{r+1,l},$$

where $\boldsymbol{\sigma}_{r,l}$ is a spin- $\frac{1}{2}$ operator on ring r and leg l , can be described in the strong-ring limit ($J' \ll J$) by an effective model, valid to first order in the inter-ring coupling J' ,²⁻⁵ defined by the Hamiltonian

$$H_{\text{eff}} = K \sum_{r=1}^{N-1} \mathbf{S}_r \cdot \mathbf{S}_{r+1} [1 + \alpha(\tau_r^+ \tau_{r+1}^- + \text{H.c.})]. \quad (1)$$

Here \mathbf{S}_r are the usual spin- $\frac{1}{2}$ operators which describe the total spin of ring r , while τ_r are pseudo-spin- $\frac{1}{2}$ operators acting on the chirality. The parameters of the model are an overall coupling constant $K=J'/L$ and a parameter α that

measures the strength of the coupling between spin and chirality. For ordinary spin tubes, this coupling is always strong: α is equal to 4 for three-leg spin tubes and increases with the number of legs.^{4,5} Using bosonization arguments,³ Schulz predicted that the ground state should be spontaneously dimerized and the spectrum gapped in all sectors, a prediction supported by further numerical and analytical work.⁴⁻⁶ In addition, the excitations have been argued to be unbound solitons and the gap is always a significant fraction of J' . All of this remarkable physics still awaits an experimental realization.

In this context, the recent synthesis of $\text{Na}_2\text{V}_3\text{O}_7$,⁷ whose structure may be regarded as a spin- $\frac{1}{2}$ nine-leg spin tube, has opened up new perspectives. However, the properties reported so far⁸ do not match the properties predicted for standard odd-legged spin tubes. In particular, no spin gap could be detected in zero external field. This might not be too surprising, however: although the overall topology of

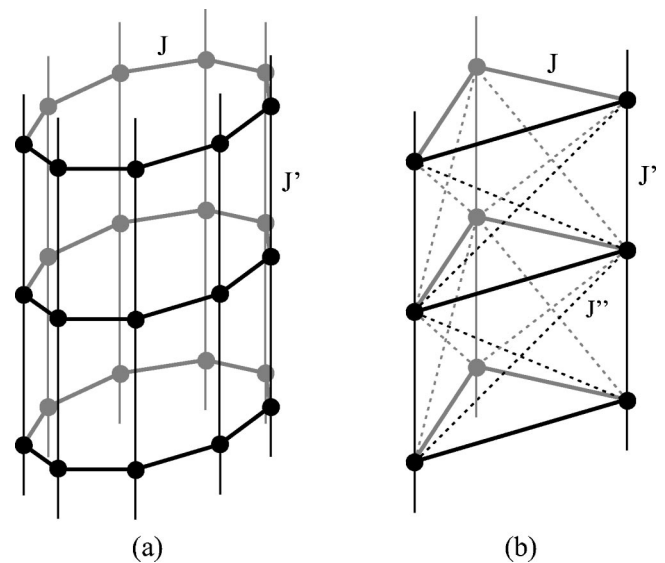


FIG. 1. Two models for $\text{Na}_2\text{V}_3\text{O}_7$. (a) Nonfrustrated, nine-leg spin tube. (b) Frustrated, three-leg spin tube.

$\text{Na}_2\text{V}_3\text{O}_7$ is indeed that of a nine-leg spin tube, the actual geometry is quite different from that of Fig. 1(a). Although ab-initio calculations have not yet reached a consensus,^{9,10} it is likely that the inter-ring coupling exhibits some kind of frustration. Since the tubes in $\text{Na}_2\text{V}_3\text{O}_7$ only have a C_3 -axis, a frustrated model of the type of Fig. 1(b) might be more appropriate.

In this Communication, we use extensive density matrix renormalization group (DMRG) simulations¹¹ supported by several analytical arguments to show that inter-ring frustration can have dramatic consequences for the properties of odd-legged spin tubes. In particular, we show that it can reduce the spin gap and bind solitons in low-lying singlets. This picture might resolve some of the puzzles of $\text{Na}_2\text{V}_3\text{O}_7$.

Our starting point is to notice that, as long as the inter-ring coupling does not break the rotational symmetry of the tube, the effective Hamiltonian is still given by Eq. (1), but with a parameter α that can take on arbitrarily small values if frustration is allowed. For instance, for the three-leg spin tube of Fig. 1(b), $K=(J'+2J'')/3$ and $\alpha=4|J'-J''|/(J'+2J'')$, leading to $\alpha=2$ if each site is coupled to two neighbors (i.e., $J'=0$),¹² and to $\alpha=0$ if each site is coupled to all sites of neighboring rings ($J'=J''$).

Therefore, we concentrate on the model of Eq. (1) with $K=1$ and consider all values of $\alpha \geq 0$ in the following. In a previous DMRG study of the effective model (1), Kawano and Takahashi⁴ reported the finite-size scaling of the spin gap for the triangular tube with $\alpha=4$. Using White's DMRG algorithm¹¹ as well, we extend their numerical analysis to the range $0 \leq \alpha \leq 20$. For this purpose, we classify the lowest-lying excitations according to the quantum numbers $[S^z, \tau^z]$ and study the sectors $[0,0]$, $[0,1]$, $[1,0]$, and $[1,1]$ for open chains with up to $N=200$ sites. From an analysis of the truncation dependence of the gaps, we find that convergence is reached by keeping 250 states and thus perform the calculations up to that limit within six finite-system sweeps. The sum of the discarded density-matrix eigenvalues is smaller than 10^{-5} in all cases.

Above $\alpha \approx 1.4$, a range that includes all nonfrustrated spin tubes, we found that a gap is indeed present, in agreement with Kawano and Takahashi's analysis of the case $\alpha=4$, but interestingly enough, we find that the first excitation appears in all sectors. In other words, spin and chirality gaps are equal above $\alpha \approx 1.4$.

The situation changes dramatically upon reducing α . Below $\alpha \approx 1.4$, the first excitation is no longer degenerate but appears only in the sector $[S^z=0, \tau^z=1]$. The first excitation is thus a chirality excitation, and the spin gap Δ_S and the chirality gap Δ_τ are no longer equal. To examine this point further, we have performed a systematic analysis for $\alpha < 1.4$ in all sectors $[0,0]$, $[0,1]$, $[1,0]$, and $[1,1]$, including careful finite-size scaling. The first excitation is always in the sector $[S^z=0, \tau^z=1]$ and is nondegenerate. Below $\alpha \approx 0.5$, the second excitation is also nondegenerate and appears in the sector $[0,0]$. In this parameter range, the first excitation that has a nonzero spin quantum number is the third excited state. This excitation manifests itself in all sectors. By following this excitation as α is increased, one can determine that it becomes the second excitation at $\alpha \approx 0.5$, and then the first excitation at $\alpha \approx 1.4$.

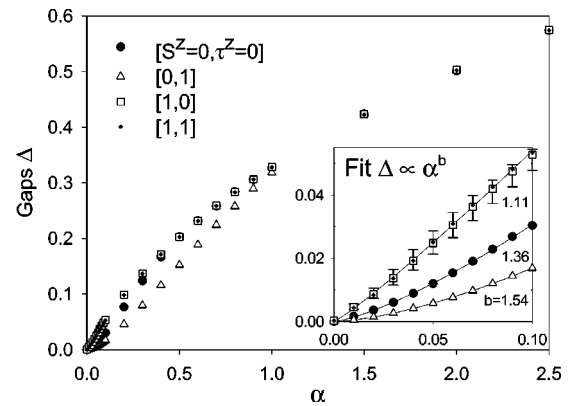


FIG. 2. Extrapolated excitation gaps in the four $[S^z, \tau^z]$ sectors as a function of α . Inset: Power-law fits of the small α results.

The gaps corresponding to these excitations are plotted in Fig. 2. In extracting the gaps, some care had to be taken regarding finite-size effects. The results were fitted with polynomials in $1/N$, where N is the length of the tube. Good fits could be obtained with third-order polynomials. The extrapolated values lie between those obtained with quadratic and quartic polynomials. The differences between these fits were used to define the error bars shown in the inset of Fig. 2.

That these gaps correspond to very different excitations is confirmed by their α dependences. As shown in the inset, the results for $\alpha \leq 0.1$ can be fitted with power laws of the form $\Delta \propto \alpha^b$ with exponents $b=1.54 \pm 0.06$, 1.36 ± 0.07 , and 1.11 ± 0.12 for the singlet excitations in the sectors $[0,1]$, $[0,0]$, and for the first spin excitation, respectively. These exponents are consistent with the simple fractions $b=3/2$, $4/3$, and 1. The large error bar and the value significantly larger than $b=1$ for the spin excitation is very probably a finite-size effect. Since the spin gap follows a linear finite-size scaling for spin tubes with up to 200 rings when α is very small, the extrapolations underestimate the gap, resulting in an exponent larger than the actual one.

The nature of the excitations can be further explored by examining the nearest-neighbor expectation values of the spin and pseudo-spin interactions $\langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} \rangle$ and $\langle \tau_i^+ \tau_{i+1}^- + \text{H.c.} \rangle$. In Fig. 3 these quantities are shown for the lowest-lying states in the important $[S^z, \tau^z]$ sectors. The spin and chirality degrees of freedom alternate synchronously. As expected, the ground state, shown in Fig. 3(a), is uniformly dimerized in both the spin and the chirality channels.

Excitations in a dimerized spin chain can be described in terms of solitons, which can be viewed as domain walls between two dimer coverings.¹³⁻¹⁵ In a chain with an even number of sites, solitons always appear in pairs, which can either be bound or unbound. The bound states can also be interpreted as excited bonds, i.e., as spin-triplet states in a background of dimers, and would therefore leave the dimer pattern unchanged. In an open chain, a bound soliton state is located with maximum probability at the center of the system, whereas its unbound counterpart tries to maximize both the distance between the solitons and the distance to the ends of the chain.

The nearest-neighbor expectation values of excited states

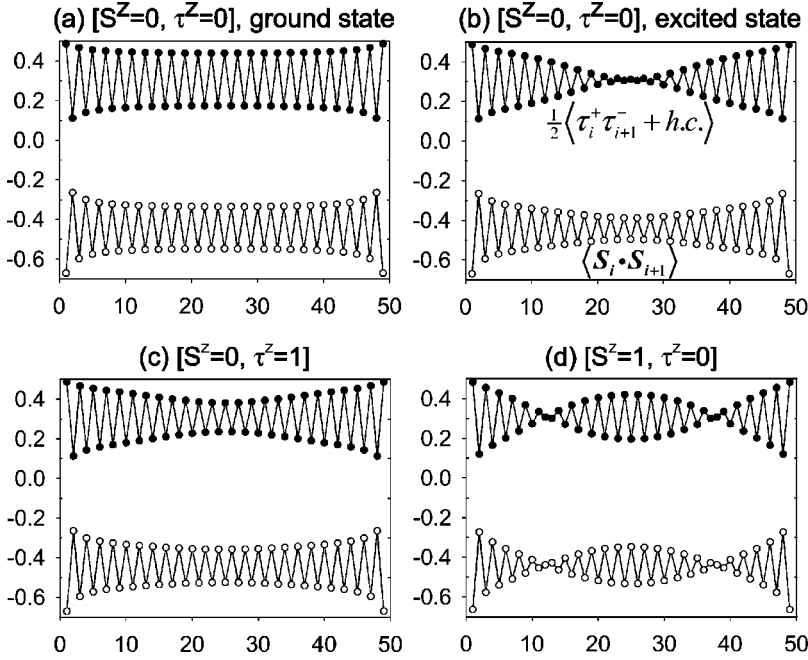


FIG. 3. Nearest-neighbor expectation values of spin and chirality along a chain with 50 sites in the sectors $[S^z=0, \tau^z=0]$, $[0, 1]$ and $[1, 0]$ for $\alpha = 0.1$. The ground state (a) is dimerized. The unaltered dimer patterns and the constrictions at the center of the chain in (b) and (c) indicate bound soliton states, whereas the two domain walls in (d) suggest the presence of two unbound solitons.

in the sectors $[S^z=0, \tau^z=0]$ and $[0, 1]$ for $\alpha=0.1$ are shown in Figs. 3(b) and 3(c). The unaltered dimer patterns and the constrictions at the center of the chain indicate the presence of bound soliton states. The very pronounced constriction in Fig. 3(b) suggests that the excitation in the ground state sector is close to unbinding, which we find to occur in the parameter range $0.5 < \alpha < 0.6$. For larger values of α , all expectation values have a structure similar to that shown in Fig. 3(d), representing the lowest-lying spin excitation. Here one can clearly identify two domain walls at around $\frac{1}{3}$ and $\frac{2}{3}$ of the chain length, suggesting the presence of two unbound solitons.

We have checked that these features are independent of the chain length by studying systems of up to 200 sites. From a complete analysis of the expectation values throughout the whole range of α , we conclude that lowest-lying bound states only exist in the sectors $[S^z=0, \tau^z=0]$ and $[0, 1]$, for which the unbinding can be observed at $0.5 < \alpha < 0.6$ and $1.3 < \alpha < 1.4$, respectively. The lowest-lying pure spin and combined spin-chirality excitations are always unbound and are therefore degenerate in the thermodynamic limit.

Interestingly, the same hierarchy of states for the bound and unbound soliton excitations as a function of α can be obtained analytically by the variational approach of Wang⁶ for the model that includes a next-nearest-neighbor coupling along the legs with relative strength $\beta=0.5$.¹⁶ Since it was shown by Kawano and Takahashi⁴ that Wang's model remains in the same phase as the additional interaction β is turned off for $\alpha=4$, we conjecture that the two models are in the same phase over the whole range of α . In the same spirit, we note that the effective Hamiltonian of Eq. (1) can also be seen as a special case of the recently investigated spin-orbital models,^{17–20} with great similarities in the roles of chirality and orbital degrees of freedom.

Next, we show that the scaling of the chirality gap $\Delta_\tau \propto \alpha^{3/2}$ can be recovered by a simple mean-field decoupling of the interaction terms $\mathbf{S}_r \cdot \mathbf{S}_{r+1} \tau_r^\pm \tau_{r+1}^\mp$ into

$$\langle \mathbf{S}_r \cdot \mathbf{S}_{r+1} \rangle \tau_r^\pm \tau_{r+1}^\mp + \mathbf{S}_r \cdot \mathbf{S}_{r+1} \langle \tau_r^\pm \tau_{r+1}^\mp \rangle - \langle \mathbf{S}_r \cdot \mathbf{S}_{r+1} \rangle \langle \tau_r^\pm \tau_{r+1}^\mp \rangle.$$

Since, according to our numerical results, the ground state is spontaneously dimerized, we look for a dimerized solution by starting with alternating expectation values $\langle \mathbf{S}_r \cdot \mathbf{S}_{r+1} \rangle = C_S (-1)^r \delta_S$ and $\langle \tau_r^\pm \tau_{r+1}^\mp + \tau_r^\mp \tau_{r+1}^\pm \rangle = C_\tau (-1)^r \delta_\tau$, where $\delta_{S/\tau}$ is the alternation parameter in the corresponding channel. The mean-field Hamiltonian then describes Heisenberg and XY chains with alternating bond strengths. Now, for Heisenberg and XY chains with alternating exchange $J[1 + (-1)^r \epsilon]$, the scalings of the gap and of the alternation parameters as a function of ϵ are well known.^{21–24} Up to logarithmic corrections, they are given by $\Delta_S \propto J(\epsilon)^{2/3}$, $\delta_S \propto \epsilon^{1/3}$, $\Delta_\tau \propto J\epsilon$, and $\delta_\tau \propto \epsilon$. The mean-field decoupling then leads to a J of order one and $\epsilon \propto \alpha \delta_\tau$ for the spin part, and to $J \propto \alpha$ and $\epsilon \propto \delta_S$ for the chirality. Self-consistency then requires that $\delta_S \propto \alpha^{1/2}$ and $\Delta_\tau \propto \alpha^{3/2}$. This last scaling is in very good agreement with our DMRG result for the chirality gap, for which no logarithmic correction could be extracted within our numerical accuracy. As a further check, we have also extracted δ_S and δ_τ as a function of α . They are not strictly proportional, but we believe this is due to logarithmic corrections. Indeed, the ground-state energy for small α reads

$$e_{eff}^{MF} = A \alpha \delta_S^2 (1 + B \ln \delta_S) + C (\alpha \delta_\tau)^{4/3} (\ln D \alpha \delta_\tau)^{-1} - \alpha \delta_S \delta_\tau + \text{const.} \quad (2)$$

A , B , C , and D are constants²⁵ independent of α . The alternation parameters δ minimizing the ground-state energy obey the relation $\delta_\tau \propto \delta_S (\log \delta_S + \text{const})$, leading to a very good fit of our numerical results (not shown).

In order to illustrate the influence of the chirality degrees of freedom, we have calculated the specific heat density c as a function of temperature using exact diagonalization of small systems. We have used weighted means between systems with an even and odd number of sites²⁶ to extrapolate the curves to the bulk limit. Extrapolations for systems with

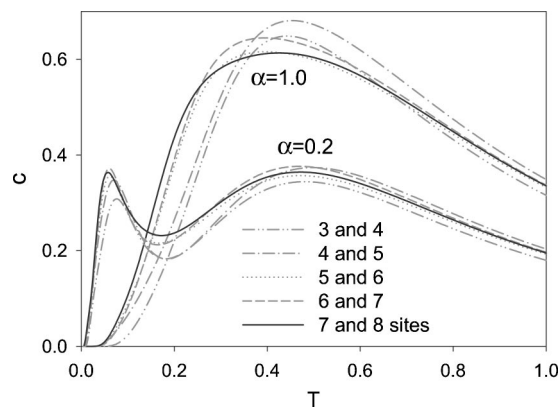


FIG. 4. Specific heat per site as a function of the temperature T for $\alpha=0.2$ and 1.0 calculated by exact diagonalization of systems with up to eight sites.

up to eight sites and two particular values of α are shown in Fig. 4. The double-peak structure for small α is due to the well-separated low-lying chirality excitations. The distinct low-temperature peak progressively disappears as the chirality excitations get closer to the spin excitations and is completely absent at $\alpha \approx 1$. We also note that these low-lying chirality excitations, which are singlets, should be detectable

in Raman spectroscopy. Both predictions are expected to apply to $\text{Na}_2\text{V}_3\text{O}_7$ if the absence of any detectable zero-field spin gap in that compound is indeed a consequence of frustration.

In conclusion, we have shown that the low-energy properties of odd-legged spin tubes change dramatically if the coupling between spin and chirality degrees of freedom is reduced, as would be the case for frustrated spin tubes. While our results confirm that excitations in nonfrustrated or weakly frustrated spin tubes are unbound solitons, they show that solitons are bound into singlet bound states if the coupling between spin and chirality is sufficiently reduced by frustration. As a consequence, spin and chirality gaps are no longer equal, and the low-lying chirality excitations, which lie below the spin gap, are expected to lead to a number of interesting consequences, such as an additional low-temperature peak in the specific heat. We hope that these conclusions will encourage further experimental investigations of $\text{Na}_2\text{V}_3\text{O}_7$ and of other odd-legged spin tubes.¹²

It is a pleasure to thank J. Gavilano, H. R. Ott, J. Sirker, and O. Sushkov for valuable discussions. G. M. acknowledges the hospitality of IRRMA (Lausanne), and V. K. and F. M. acknowledge the support of the Swiss National Fund.

¹For a review, see E. Dagotto and T. M. Rice, *Science* **271**, 618 (1996).

²V. Subrahmanyam, *Phys. Rev. B* **50**, 16 109 (1994).

³H. J. Schulz, in *Correlated Fermions and Transport in Mesoscopic Systems*, edited by T. Martin, G. Montambaux, and J. Tran Than Van (Editions Frontiers, Gif-sur-Yvette, 1996), p. 81.

⁴K. Kawano and M. Takahashi, *J. Phys. Soc. Jpn.* **66**, 4001 (1997).

⁵D. C. Cabra, A. Honecker, and P. Pujol, *Phys. Rev. B* **58**, 6241 (1998).

⁶H.-T. Wang, *Phys. Rev. B* **64**, 174410 (2001).

⁷P. Millet, J. Y. Henry, F. Mila, and J. Galy, *J. Solid State Chem.* **147**, 676 (1999).

⁸J. L. Gavilano *et al.*, *Phys. Rev. Lett.* **90**, 167202 (2003).

⁹M.-H. Whangbo and H. J. Koo, *Solid State Commun.* **115**, 675 (2000).

¹⁰V. Mazurenko and V. Anisimov (private communication).

¹¹S. R. White, *Phys. Rev. Lett.* **69**, 2863 (1992), *Phys. Rev. B* **48**, 10 345 (1993).

¹²This seems to be the topology of a newly discovered, three-leg spin tube. See J. Schnack, H. Nojiri, P. Kögeler, G. Cooper, and L. Cronin, *cond-mat/0403548* (unpublished).

¹³C. K. Majumdar and D. P. Ghosh, *J. Math. Phys.* **10**, 1388 (1969).

¹⁴B. S. Shastry and B. Sutherland, *Phys. Rev. Lett.* **47**, 964 (1981).

¹⁵I. Affleck, D. Augier, and P. Poilblanc, *Phys. Rev. B* **58**, R14 701 (1998).

¹⁶A. Lüscher *et al.* (unpublished).

¹⁷F. Mila, B. Frischmuth, A. Deppeler, and M. Troyer, *Phys. Rev. Lett.* **82**, 3697 (1999).

¹⁸A. K. Kolezhuk, H.-J. Mikeska, and U. Schollwöck, *Phys. Rev. B* **63**, 064418 (2001).

¹⁹Y. Yamashita, N. Shibata, and K. Ueda, *J. Phys. Soc. Jpn.* **69**, 242 (2000).

²⁰S. K. Pati and R. R. P. Singh, *Phys. Rev. B* **61**, 5868 (2000).

²¹M. C. Cross and D. S. Fisher, *Phys. Rev. B* **19**, 402 (1979).

²²J. L. Black and V. J. Emery, *Phys. Rev. B* **23**, 429 (1981).

²³I. Affleck, D. Gepner, H. J. Schulz, and T. Ziman, *J. Phys. A* **22**, 511 (1989).

²⁴P. Pincus, *Solid State Commun.* **9**, 1971 (1971).

²⁵E. Orignac, *cond-mat/0403175* (unpublished).

²⁶J. C. Bonner and M. E. Fisher, *Phys. Rev.* **135**, A640 (1964).