Effects of biquadratic exchange on the spectrum of elementary excitations in spin ladders

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We investigate the influence of biquadratic exchange interactions on the low-lying excitations of a $S=\frac{1}{2}$ ladder using perturbation theory, numerical diagonalization of finite systems, and exact results for ladders with matrix product ground states. We consider in particular the combination of biquadratic exchange interactions corresponding to ring exchange on the basic ladder plaquette. We find that a moderate amount of ring exchange reduces the spin gap substantially and makes equal bilinear exchange on legs and rungs consistent with experimentally observed spectra. [S0163-1829(99)05425-9]

I. INTRODUCTION

Two-legged spin ladders with spins $\frac{1}{2}$ have attracted considerable interest over the past years, both as ideal models for quasi-one-dimensional materials and as a theoretical model for a spin liquid characterized by an excitation gap. From the geometric structure of the two-legged spin ladder as shown in Fig. 1(a) it is clear that the main exchange interactions are expected along the legs and across the rungs; if the most general interaction for a plaquette formed by four spins $S=\frac{1}{2}$ on two neighboring rungs is considered, exchange along the two diagonals (corresponding to next-nearest-neighbor interactions) and biquadratic exchange interactions appear in addition.

This general plaquette Hamiltonian for $S=\frac{1}{2}$ is formulated as

$$H=\sum_n \{J_{rung}\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n}+J_{leg}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{1,n+1}+\mathbf{S}_{2,n}\cdot\mathbf{S}_{2,n+1}) + J_{diag}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n+1}+\mathbf{S}_{2,n}\cdot\mathbf{S}_{1,n+1})+V_{RR}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n}) \times(\mathbf{S}_{1,n+1}\cdot\mathbf{S}_{2,n+1})+V_{LL}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{1,n+1})(\mathbf{S}_{2,n}\cdot\mathbf{S}_{2,n+1}) + V_{DD}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n+1})(\mathbf{S}_{2,n}\cdot\mathbf{S}_{1,n+1})\}. \quad (1)$$

For simplicity we have assumed equal exchange interactions J_{leg} on the two legs and J_{diag} on the two diagonals, a natural assumption for the symmetric structure of Fig. 1 which we will consider in the following. Recently, theoretical studies have demonstrated that this generalized Hamiltonian (i) allows us to formulate models interpolating smoothly between the dimer and Haldane limits for the ground state[1] and (ii) has a parameter space sufficiently large to allow us to study quantum phase transitions.[2] The typical Hamiltonian for both applications has some nonvanishing biquadratic terms. In the past, only little attention has been paid to biquadratic exchange terms, although finite strength of these terms was, e.g., found in the spectra of small clusters of magnetic ions.[3]

Recently, the possible importance of biquadratic exchange for some properties of low-dimensional spin systems has been pointed out: Honda, Kuramoto, and Watanabe[4] have argued for finite ring exchange (corresponding to a particular combination of biquadratic terms) on the basic CuO_2 plaquette. The relevance of ring exchange for magnetization plateaus has been discussed for the spin ladder by Sakai and Hasegawa[5] and for solid He^3 films by Momoi, Sakamoto, and Kubo[6] In this paper we study the effect of the biquadratic terms in the Hamiltonian (1) on the low-lying excitations of this ''generalized spin ladder'' by both analytical and numerical approaches. The motivation for this work is twofold: First, we want to extend the knowledge obtained analytically in Ref. 2 to a wider range of strengths of the biquadratic interactions.

Second, we want to investigate the relevance of ring exchange of finite strength J_{ring} as introduced by Honda, Kuramoto, and Watanabe[4] on the determination of coupling constants from ladder spectra. The standard analysis of experimental data on quasi-one-dimensional ladder systems starts from Eq. (1) with J_{diag} and all biquadratic couplings set equal to 0 and results in $J_{leg}\approx 2J_{rung}$ from inelastic neutron-scattering experiments on $Sr_{14}Cu_{24}O_{41}$ (Ref. 7) and $La_6Ca_8Cu_{24}O_{41}$,[8] as well as from NMR experiments on these substances.[9–11] This large value for the ratio J_{leg}/J_{rung} is not understood at present: It is not expected from the geometric structure of the ladder, from electronic structure calculations[12] a value somewhat larger than unity is obtained

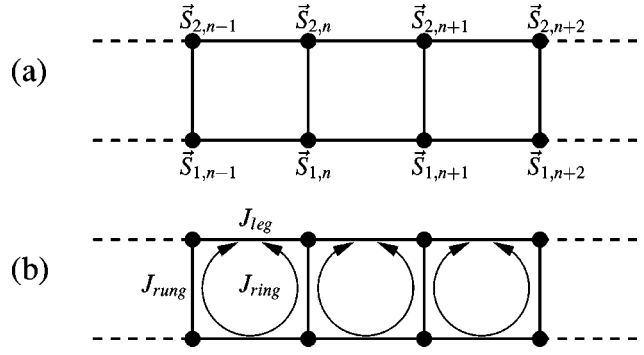

FIG. 1. (a) Structure of the two-leg ladder; (b) possible ring exchange around the basic plaquette.

and interladder interactions,[13] which have recently been considered, do not resolve the discrepancy.

In the following we show that even a moderate amount of ring exchange J_{ring} is relevant for the determination of the coupling constants in the two-leg ladder from the energies of elementary excitations. We define the strength J_{ring} on a plaquette of four spins i,j,k,l in terms of the permutation operator P_{ijkl} by the symmetric contribution

$$H_{ring}=\frac{1}{2}J_{ring}\sum_{plaquettes}(P_{ijkl}+P_{lkji})$$

to the Hamiltonian. Since we deal with spins $S=\frac{1}{2}$, H_{ring} is identical to a special choice of constants in Eq. (1), namely

$$V_{RR}^{ring}=V_{LL}^{ring}=-V_{DD}^{ring}=2J_{ring},\quad J_{rung}^{ring}=J_{ring},$$

$$J_{leg}^{ring}=J_{diag}^{ring}=\frac{1}{2}J_{ring}.$$

A natural Hamiltonian for the geometric structure of the spin ladder then is obtained by bilinear exchange terms and ring exchange terms, leading to the Hamiltonian of Eq. (1) with

$$J_{rung}=J_{rung}^{bl}+J_{ring},\quad J_{leg}=J_{leg}^{bl}+\frac{1}{2}J_{ring},$$

$$J_{diag}=\frac{1}{2}J_{ring},$$

$$V_{RR}=V_{LL}=-V_{DD}=2J_{ring}. \tag{2}$$

In the following we will present results on the influence of $J_{ring}\neq 0$ on the dispersion of low-lying excitations for antiferromagnetic ladders using perturbation theory (Sec. II), numerical calculations covering the case of experimental interest (Sec. III) and discussing the relevance for the dispersion of exactly known excitation branches (in the case of special choices for the strength of biquadratic exchange).

II. PERTURBATION THEORY

We have calculated the dispersion of the lowest triplet excitation in an expansion in the neighborhood of the dimer point for a chain of L rungs with periodic boundary conditions.[14] At the dimer point J_{rung} is the only nonvanishing exchange constant and the lowest excited states are obtained by exciting any of the L dimers from singlet to triplet state, giving an L-fold degenerate state with energy J_{rung}. In the following we discuss the evolution of these basic triplet excitations for the Hamiltonian of experimental interest,

$$H=\sum_n\{\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n}+J_{leg}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{1,n+1}+\mathbf{S}_{2,n}\cdot\mathbf{S}_{2,n+1})\}+H_{ring}, \tag{3}$$

with small values of J_{leg},J_{ring} in perturbation theory (we will use $J_{rung}=1$ as unit of energy). Switching on these interactions the basic triplet starts to propagate and the L excitations which are degenerate in the dimer limit are distinguished by wave vector k, which is a good quantum number due to translational invariance. Therefore straightforward nondegenerate Rayleigh-Schrödinger perturbation theory can be applied. In nth order nonvanishing values of J_{leg},J_{ring} lead to intermediate states with n excited dimers which are coupled to total spin $S=1$. Including terms of third order in J_{leg},J_{ring} the following spectrum is obtained for the lowest triplet excitation:

$$\omega_{S=1}(k)=(t_0+t_1\cos k+t_2\cos 2k+t_3\cos 3k), \tag{4}$$

$$t_0=1-2J_{ring}+\frac{3}{4}(J_{leg}-J_{ring})^2+\frac{3}{8}(J_{leg}-J_{ring})^2(J_{leg}+5J_{ring}),$$

$$t_1=J_{leg}+J_{ring}-\frac{1}{4}(J_{leg}-J_{ring})^2(J_{leg}+J_{ring}),$$

$$t_2=-\frac{1}{4}(J_{leg}-J_{ring})^2-\frac{1}{4}(J_{leg}-J_{ring})^2(J_{leg}+J_{ring}),$$

$$t_3=\frac{1}{8}(J_{leg}-J_{ring})^2(J_{leg}+J_{ring}).$$

Since this is an expansion in powers of J_{leg},J_{ring}, it will be applicable for sufficiently small values of these exchange constants as long as no phase boundary is crossed, i.e., as long as the ground state is obtained by continuous deformation from the rung dimer ground state. For a quantitative comparison we refer to Fig. 2(a) (see below).

The leading coefficient for the dispersion is $t_1\propto J_{leg}+J_{ring}$ in first order. This perturbative result shows that when $J_{ring}>0$ is present, an analysis of the spectra in terms of bilinear exchange only will lead to an effective value of J_{leg} which is increased in comparison to the value found for $J_{ring}=0$. We will see in the next section from numerical diagonalization that this result of perturbation theory continues to be qualitatively true for larger values of J_{leg}.

III. THE SYMMETRIC LADDER WITH BIQUADRATIC EXCHANGE

Motivated by the results of recent inelastic neutron-scattering experiments on the quasi-one-dimensional ladder material $La_6Ca_8Cu_{24}Sr_{41}$,[8] we have calculated numerically excitation spectra for spin ladders with the Hamiltonian of Eq. (2) using periodic boundary conditions for a finite number of spins. We have used the Lanczos method for ladders with a total of 24 spins, i.e., 12 rungs. Because of space inversion the number of wave vectors k with different energies is 7, $k=p\pi/6$, $p=0,1\cdots 6$. In Figs. 2(a)–(c) we present the results for the lowest negative parity excitation (parity with respect to interchange of the legs). This is the lowest excitation with the ladder gap at $k=\pi$; it is very likely a triplet excitation, since it is found with identical energies for both $S_{tot}^z=1$ and $S_{tot}^z=0$ ($S_{tot}>1$, however, is not formally excluded). We show the variation of the dispersion with J_{ring} between the limits $-0.3\cdots+0.3$ for three values of J_{leg}. For the smallest value, $J_{leg}=0.5$, we also show the perturbative result, see Eq. (4) for comparison (full lines). The agreement is satisfying for $J_{ring}<0.2$, whereas for larger values of J_{ring} a phase transition to a different

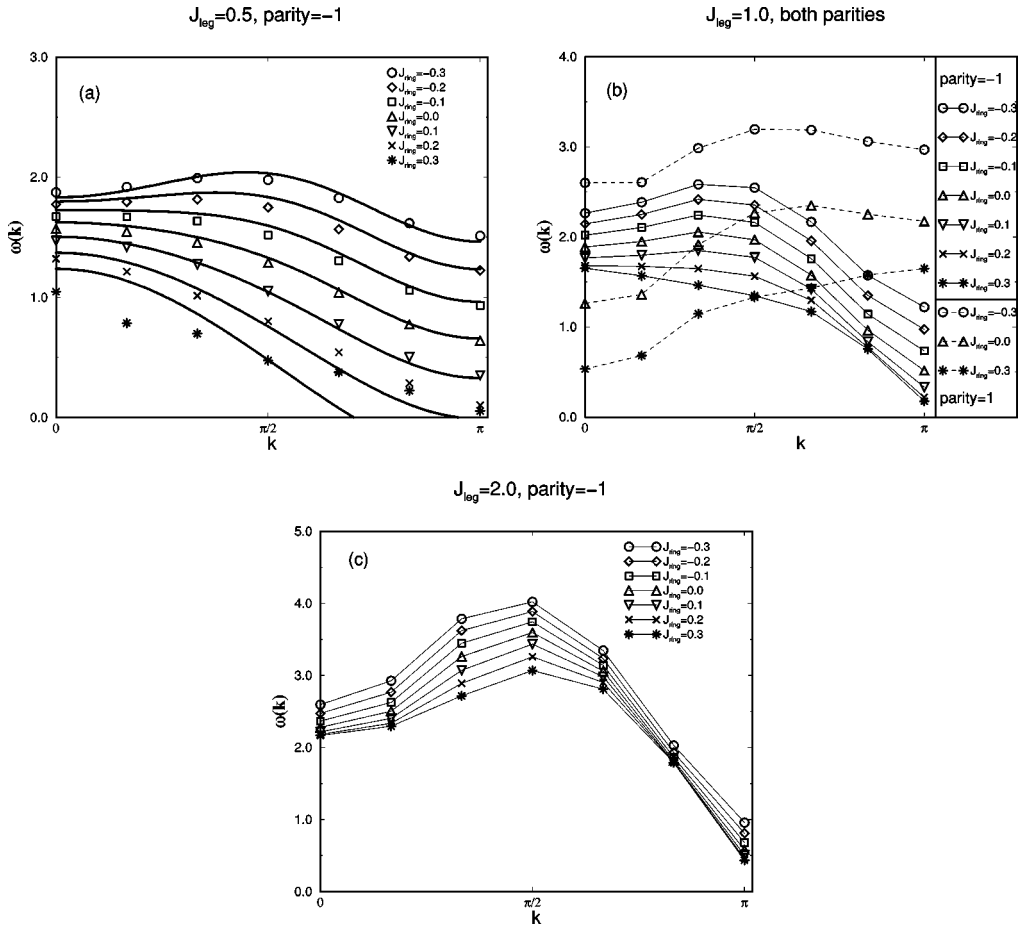

FIG. 2. Calculated ladder spectra (lowest negative parity triplet) for $J_{ring}=-0.3\cdots0.3$ and $J_{leg}=0.5$ (a); 1.0 (b); 2.0 (c). For $J_{leg}=0.5$ the result from perturbation theory [Eq. (4)] is given for comparison (full lines). For $J_{leg}=1.0$ the lowest positive parity excitation (singlet) is also shown (dashed lines). Full lines for $J_{leg}=1.0,2.0$ (negative parity excitations) are guides to the eye.

ground state has probably taken place and the perturbation theory of Sec. II is no more applicable. It should be noted that the unit of energy is the bilinear rung exchange J_{rung}^{bl} (without the effective contribution J_{rung}^{ring} resulting from the ring exchange). J_{rung}^{bl} should be fixed by the magnitude of the gap.

For a simple quantitative presentation of the effect of ring exchange on the spectra we have considered the ratio

$$R=\frac{\omega\left(\frac{2\pi}{3}\right)}{\omega(\pi)}. \tag{5}$$

This quantity may conveniently serve for comparison with experimental data since the most accurate spectra are obtained for $k>\pi/2$ and it is here where $\omega(k)$ varies most. The curves in Fig. 3 show the dependence of the ratio R on J_{ring} for $J_{leg}/J_{rung}=0.5,1.0,1.5,2.0$. The analysis of the experiments gives $R\approx5$, which may be realized for different combinations of J_{leg} and J_{ring}: Two examples are $J_{leg}\approx1.8$, $J_{ring}\approx0$ and $J_{leg}\approx1.0$, $J_{ring}\approx0.14$. Thus we conclude from the data shown in Fig. 2 the tendency that a finite $J_{ring}>0$ tends to simulate the effect of an increased bilinear exchange J_{leg} on the legs. This is in agreement with perturbation theory as discussed in Sec. II. We have checked that the perturbative result of Eq. (3) is a good approximation to the numerical data for $J_{leg}=0.5$ for $J_{ring}<0.1$; for larger ring exchange a transition to a new phase occurs (see below) such that perturbation theory no longer applies.

The main physics behind the importance of J_{ring} for the

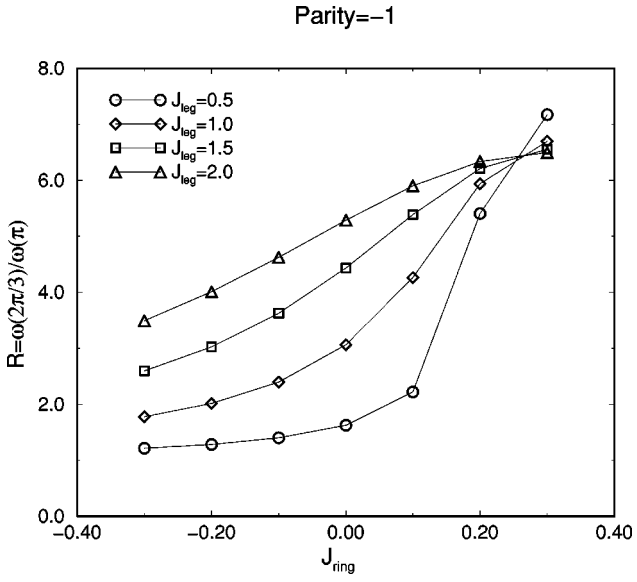

FIG. 3. Ratio R [see Eq. (5)] characterizing the steepness of the dispersion near wave vector $k=\pi$.

determination of bilinear exchange constants is the following: the influence of J_{ring} on the gap (at $k=\pi/2$) is relatively much stronger than on the dispersion at intermediate wave vectors. Actually, for $J_{leg}<1$, the gap appears to vanish for a critical magnitude of J_{ring}, indicating a phase transition. This phase transition may also be responsible for the irregular behavior of the dispersion curves for $J_{leg}=0.5$, $J_{ring}=0.3$; this system may actually be in a different phase. It is the neighborhood of this phase transition also for values $J_{leg}\propto O(1)$ which explains the strong influence of the biquadratic exchange terms. We expect this phase transition to be of the same type as discussed before for generalized spin ladders,[2,15,16] leading to a spontaneously dimerized ground state.

The energy of the spin gap is the basic input for the determination of the energy scale in ladder systems. Owing to the large effect of finite J_{ring} on the spin gap the values of the exchange constants in CuO_2 planes will have to be renormalized considerably. For $J_{ring}\approx 0.14J_{rung}$, $J_{leg}\approx J_{rung}$ the spin gap is $\Delta\approx 0.28J_{rung}$ (compared to $\Delta\approx 0.51J_{rung}$ in the case $J_{ring}=0$). This implies an energy scale which is larger by a factor close to 2 owing to the ring exchange terms. Thus the basic exchange energy J_{rung} between two Cu ions changes from $\approx$800 to $\approx$1400 K, comparable to the magnitude of the main exchange constant in the two-dimensional (2D) material La_2CuO_4.

If the k-dependent dispersion is considered in more detail, the situation is of course more complex: In particular the presence or absence of a maximum between $k=0$ and $k=\pi$ is affected by the value of J_{ring}. At present, however, this is a point of little relevance for experimental results. In Fig. 2(b) we have also included results for the lowest excitation energy with positive parity for $J_{ring}=0,\pm 0.3$: these data illustrate our general observation that the effect of biquadratic exchange on the positive parity excitation energies is less spectacular.

IV. EXACT DISPERSION RELATIONS FOR SPIN LADDERS WITH SPECIAL VALUES OF THE EXCHANGE CONSTANTS

It was observed in Ref. 2 that the general plaquette Hamiltonian for the ladder structure allows some combinations of parameters which result in either a rung dimer (singlet) ground state with exactly known triplet excitations or a Haldane-liquid-like ground state with exactly known singlet excitations, both with negative parity. In both cases the dispersion is a pure cosine dispersion. The physical picture of these exact excited states is simple: An excited rung triplet (singlet) propagates to the neighboring rungs both on its right and left side without exciting states containing two or three quanta. The interesting and natural question, however, whether these exact excitations are of interest, in particular whether they are the lowest excitations in the corresponding ladder, can only be answered by numerical methods.

The condition for an exact dimer ground state with exact triplet excitations for the Hamiltonian of Eq. (1) according to Ref. 2 reads

$$J_{leg}-J_{diag}=\frac{1}{4}(V_{LL}-V_{DD}). \tag{6}$$

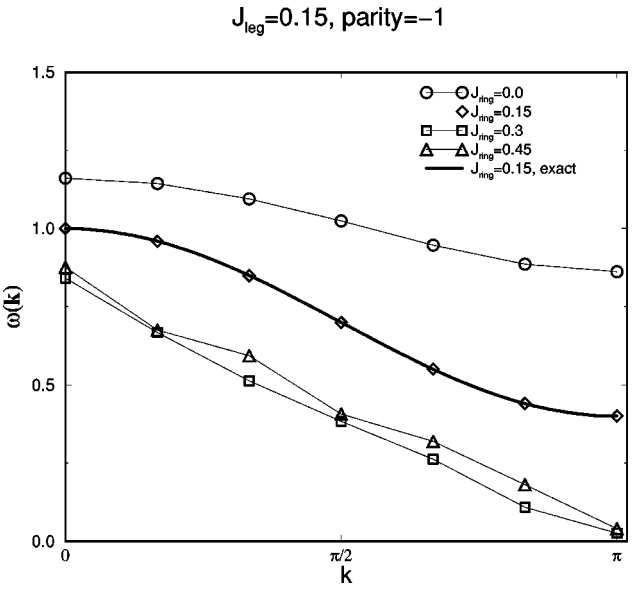

FIG. 4. Basic negative parity triplet for $J_{leg}^{bl}=0.15$ and $J_{ring}=0\cdots 0.45$. For $J_{ring}=0.15$ the numerical results reproduce the exactly known dispersion curve.

It has to be supplemented[2] by the inequalities which guarantee the stability of the dimer ground state. For the Hamiltonian of Eqs. (1) and (2) the conditions reads

$$J_{leg}^{bl}=J_{ring}\leqslant\frac{1}{5}.$$

We illustrate this situation in Fig. 4 where we plot the lowest negative-parity excitation in the subspace $S_{tot}^{z}=1$ for $J_{leg}^{bl}=0.15$ and increasing values of J_{ring}. We find that for $J_{ring}=J_{leg}^{bl}=0.15$ the exact excitation is indeed the lowest excited state for $S_{tot}^{z}=0$ as well as for ± 1. The numerical spectra reproduce perfectly the exact dispersion law. Beyond the stability limit the exact excitation energy is between two highly excited states (since the rung dimer state then has an energy above the new ground state proportional to the number of rungs) and is therefore of no more interest.

It is interesting to note that an exact dimer ground state with an exact triplet excitation is also realized in 2D and 3D structures. The condition is that Eq. (6) is fulfilled separately for the exchange interactions in each spatial direction.

A second example where the exact ground state and exact excitation spectra can be explicitly given is the generalized Bose-Gayen model as introduced in Sec. V of Ref. 2. This model is defined by the following choice of constants in the Hamiltonian of Eq. (1):

$$J_{rung}=y_1,\quad J_{leg}=1,\quad J_{diag}=y_2,$$

$$V_{RR}=0,\quad V_{LL}=\frac{4}{5}(3-2y_2),\quad V_{DD}=\frac{4}{5}(3y_2-2). \tag{7}$$

Varying the two parameters y_1, y_2 a number of phases, distinguished by different ground states, are realized, among them the rung dimer and the valence bond (AKLT, see Ref. 2) phases. In Ref. 2 exact triplet (singlet) excitations in the rung dimer (AKLT) phase with negative parity have been

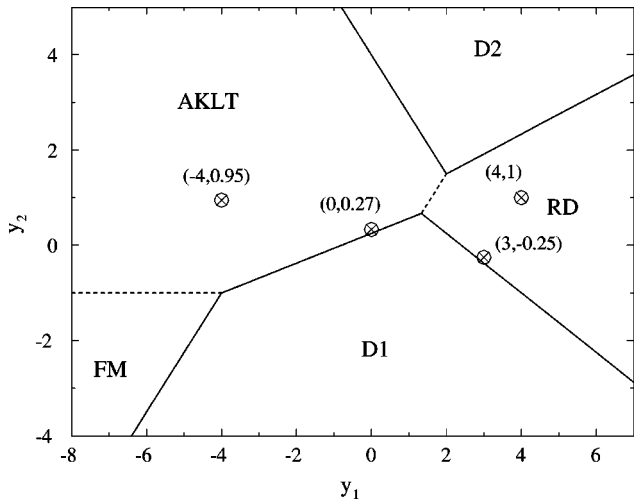

FIG. 5. Phase diagram of the generalized Bose-Gayen model (see Ref. 2). The points specify the parameters used for the calculations of the spectra shown in Fig. 6.

given. We have investigated by exact diagonalization of ladders with 24 spins to what extent these exact excitations are the lowest ones for representative points in the y_1-y_2 phase diagram as indicated in Fig. 5. Typical results of these calculations are shown in Fig. 6. We find that close to the phase boundary of both the rung dimer phase [Fig. 6(a)] and of the AKLT phase [Fig. 6(b)] the exact excitation is the lowest one for wavevectors in some neighborhood of $k=\pi$ (wave vector of the gap). This implies that they are exactly known critical modes as strongly suggested by physical intuition. Sufficiently far away in phase space from the transition line, however, a crossover occurs and there may exist lower modes of both negative and positive parity: this is the situation close to the line $y_2=1$, where the exact excitations are dispersionless. Dispersionless behavior is reproduced for all wave vectors for the point $(y_1=4, y_2=1)$ in the rung dimer phase, whereas a deviation from the exact solution for small wave vectors only is shown to occur in Fig. 6(c) for a point in the AKLT phase.

V. CONCLUSIONS

We have investigated biquadratic exchange interactions in their influence on the low-lying excitations of $S=\frac{1}{2}$ ladders with various coupling constants. From both analytical and numerical calculations we conclude that a quantum phase transition (which is likely to lead to a spontaneously dimerized ground state) occurs for small values of the ring exchange $J_{ring}>0$. At this phase transition the spin gap vanishes, therefore even small values of J_{ring} imply large variations of the spin gap and have a strong influence on the

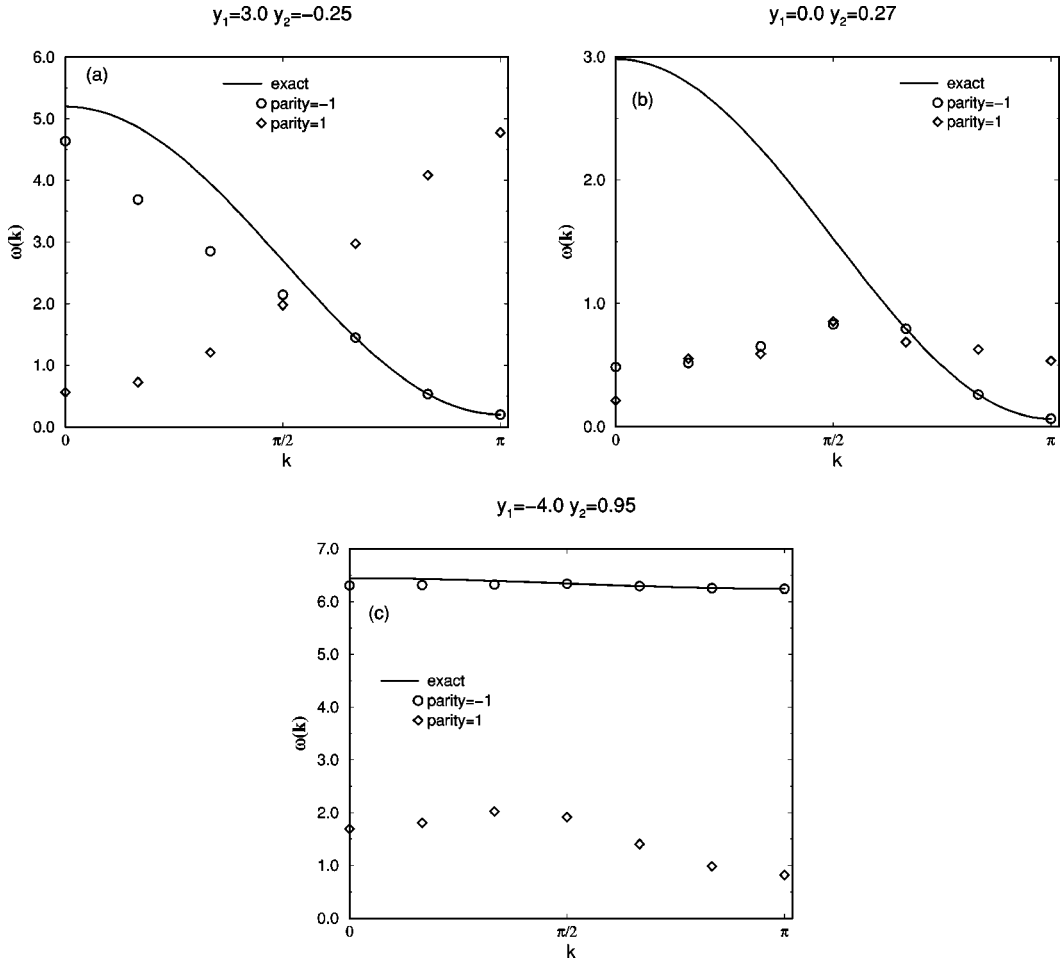

FIG. 6. Numerical and exact excitations in the generalized Bose-Gayen model: The exactly known triplet dispersions (full lines) are numerically reproduced as lowest negative parity excitations for $k>k_{min}$. (a) $k_{min}\geqslant 2\pi/3$; (b) $k_{min}\geqslant 2\pi/3$; (c) $k_{min}\geqslant \pi/2$.

determination of exchange parameters in the ladder in general. Quantitatively, the presence of a small amount of ring exchange is shown to be consistent with $J_{rung} \approx J_{leg}$ as suggested by the geometrical structure of the ladder and with a value of this basic exchange constant between two neighboring Cu ions which is close to the one found in the two-dimensional material La_2CuO_4.

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[1] A. K. Kolezhuk and H.-J. Mikeska, Phys. Rev. B **56**, R11 380 (1997).
[2] A. K. Kolezhuk and H.-J. Mikeska, Int. J. Mod. Phys. B **12**, 2325 (1998).
[3] U. Falk, A. Furrer, J. K. Kjems, and H. U. Güdel, Phys. Rev. Lett. **52**, 1336 (1984), **56**, 1956 (1986); U. Falk, A. Furrer, N. Furer, U. Güdel, and J. K. Kjems, Phys. Rev. B **35**, 4893 (1987).
[4] Y. Honda, Y. Kuramoto, and T. Watanabe, Phys. Rev. B **47**, 11 329 (1993).
[5] T. Sakai and Y. Hasegawa, cond-mat/9809291 (unpublished).
[6] T. Momoi, H. Sakamoto, and K. Kubo, cond-mat/9810118 (unpublished).
[7] R. S. Eccleston, M. Uehara, J. Akimitsu, H. Eisaki, N. Motoyama, and S. Uchida, Phys. Rev. Lett. **81**, 1702 (1998).
[8] M. Matsuda, K. Katsumata, and R.S. Eccleston (private communication).
[9] M. Takigawa, N. Motoyama, H. Eisaki, and S. Uchida, Phys. Rev. B **57**, 1124 (1998).
[10] K. Magishi, S. Matsumoto, Y. Kitaoka, K. Ishida, K. Asayama, M. Uehara, T. Nagata, and J. Akimitsu, Phys. Rev. B **57**, 11 533 (1998).
[11] T. Imai, K. R. Thurber, K. M. Shen, A. W. Hunt, and F. C. Chou, Phys. Rev. Lett. **81**, 220 (1998).
[12] T. F. A. Müller, V. Anisimov, T. M. Rice, I. Dasgupta, and T. Saha-Dasgupta, Phys. Rev. B **57**, R12 655 (1998); Y. Mizuno, T. Tohyama, and S. Maekawa, *ibid.* **58**, R14 713 (1998); cond-mat/9801195 (unpublished).
[13] S. Miyahara, M. Troyer, D. C. Johnston, and K. Ueda, cond-mat/9807127 (unpublished).
[14] M. Müller, Diploma thesis, University of Hannover, 1998.
[15] A. K. Kolezhuk and H.-J. Mikeska, Phys. Rev. Lett. **80**, 2709 (1998).
[16] A. A. Nersesyan and A. M. Tsvelik, Phys. Rev. Lett. **78**, 3939 (1997).