

Sequence of first-order quantum phase transitions in a frustrated spin half dimer-plaquette chain

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We study the frustrated dimer-plaquette quantum spin chain for ferromagnetic dimer bonds. This quantum system undergoes a series of first-order ground-state phase transitions driven by frustration or by a magnetic field. We find that the different nature of the ground-state phases has a strong influence on the magnetization curve as well as the low-temperature thermodynamics. In particular, the magnetization curve exhibits plateaus and jumps, the number of which depends on the strength of frustration. The temperature dependence of the susceptibility may either show an activated spin-gap behavior for small frustration or a Curie-like paramagnetic behavior for strong frustration.

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I. INTRODUCTION

Over the last years much attention has been concentrated on the physics of low-dimensional quantum spin systems. In particular, zero-temperature phase transitions and exotic magnetization curves in frustrated quantum magnets are in the focus of investigations. Besides continuous quantum phase transitions, remarkable first-order transitions can also be driven by frustration. Theoretical studies have benefited from recent experimental results on low-dimensional spin half antiferromagnets like quasi-one-dimensional ladder, zig-zag, and spin Peierls systems as well as gapped quasi-two-dimensional quantum magnets like CaV_4O_9 (Refs. 1 and 2) and $\text{SrCu}_2(\text{BO}_2)_3$.³⁻⁵

Among the various models for low-dimensional quantum magnets the spin half frustrated dimer-plaquette chain (FDPC) has been discussed in a series of papers in recent years. This FDPC was introduced in Ref. 6, and its Hamiltonian reads (see also Fig. 1)

$$H = H_{dp} + H_f = J_d \sum_{n=1}^{N_p} \mathbf{S}_\alpha^n \mathbf{S}_\beta^n + J_p \sum_{n=1}^{N_p} (\mathbf{S}_a^n + \mathbf{S}_b^n)(\mathbf{S}_\beta^n + \mathbf{S}_\alpha^{n+1}) + J_f \sum_{n=1}^{N_p} \mathbf{S}_a^n \mathbf{S}_b^n, \quad (1)$$

where $N_p = N/4$ is the number of plaquettes (unit cells) and N is the number of spins. We consider finite chains of N spins with periodic boundary conditions. The Hamiltonian fulfills the important relation $[H, (\mathbf{S}_a^n + \mathbf{S}_b^n)^2]_- = 0$. Hence the vertical dimer spins \mathbf{S}_a^n and \mathbf{S}_b^n form a composite spin $\mathbf{S}_{ab}^n = (\mathbf{S}_a^n + \mathbf{S}_b^n)$ with eigenvalues $(\mathbf{S}_{ab}^n)^2 = S_{ab}^n(S_{ab}^n + 1)$ and $S_{ab}^n = \{1, 0\}$. The physics of this model was discussed in the literature for antiferromagnetic couplings $J_d, J_p, J_f \geq 0$.⁶⁻¹⁰

In the unfrustrated version ($J_f = 0$, $H = H_{dp}$) the model may serve as the one-dimensional counterpart of the 1/5-depleted square lattice of CaV_4O_9 .^{6,7,11} In this case ($J_f = 0$) all composite spins have eigenvalues $S_{ab}^n = 1$ in the singlet ground state as well as in the first triplet excitation, and the ground-state physics of Eq. (1) is determined by the competition of dimer (J_d) and plaquette (J_p) bonds. In the limit $J_d \gg J_p$ the low-energy physics of the model is that of the

Haldane chain with an effective coupling $J_{eff} = J_p^2/2J_d$, whereas for $J_p \gg J_d$ a plaquette phase is realized.⁷ The singlet-triplet excitation gap is always finite for $J_p > 0$. A special feature of the model is the existence of a class of exact product eigenstates, for which the composite spins of certain plaquettes form a vertical dimer singlet (i.e. $S_{ab}^n = 0$ for certain n). The finite strips (fragments) between two vertical dimer singlets contain vertical dimer triplets $S_{ab}^n = 1$ and are decoupled from each other. Therefore, these product eigenstates correspond to a fragmentation of the chain.

Taking into account frustration $J_f > 0$ the formation of vertical dimer singlets becomes energetically more favorable and the system undergoes at a critical frustration $J_f^{crit} = f(J_d, J_p)$ a first-order quantum phase transition from the collective ground state with $S_{ab}^n = 1$ for all n to a fragmented dimer product ground state,

$$|\Psi_{0\dots 0}\rangle = \prod_{n=1}^{N_p} 2^{-1/2} (|\uparrow_a^n \downarrow_b^n\rangle - |\downarrow_a^n \uparrow_b^n\rangle) \prod_{n=1}^{N_p} |d_{\alpha\beta}^n\rangle, \quad (2)$$

with $S_{ab}^n = 0$ for all n . For $J_d > 0$ the dimer state $|d_{\alpha\beta}^n\rangle$ is a singlet and the energy of $|\Psi_{0\dots 0}\rangle$ is $E/N_p = -3J_f/4 - 3J_d/4$.

In the special limit $J_f = J_d$ the critical value is $J_f^{crit} = 1.2210J_p$.⁸ In this limit the FDPC has a close relation to the two-dimensional spin model for $\text{SrCu}_2(\text{BO}_3)_2$,^{13,8,9} and is called an orthogonal-dimer chain. Similar to the quasi-two-dimensional spin system $\text{SrCu}_2(\text{BO}_3)_2$,^{3,5} the FDPC exhibits nontrivial magnetization plateaus.^{8,9} An interesting

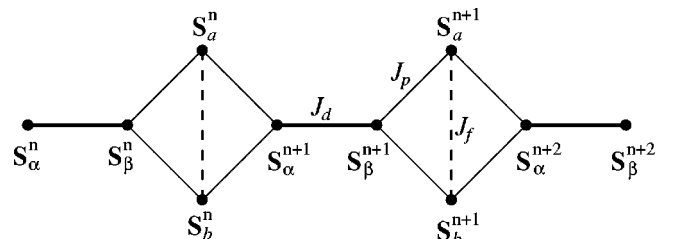


FIG. 1. The frustrated dimer-plaquette chain (FDPC).

property of the FDPC, not found so far in quantum spin systems to our knowledge, is the existence of an infinite sequence of plateaus.⁹

Very recently, the FDPC was considered for arbitrary spin quantum numbers $s \geq 1/2$ and a series of $2s$ first-order quantum phase transitions was found by Koga and Kawakami.¹⁰ Koga and co-workers argued that the first-order phase transitions described by the FDPC possess most of the essential features inherent in frustrated quantum spin systems.^{8,10} In that sense the FDPC may serve as a prototype model for spin systems showing first-order quantum phase transitions.

In the present paper we extend the model to ferromagnetic bonds J_d . We notice that the fragmentation of the chain due to vertical dimer singlets occurs for $J_d < 0$, too. In particular, the dimer product state [Eq. (2)] is also an eigenstate of Eq. (1) for $J_d < 0$; however, the dimer state $|d_{\alpha\beta}^n\rangle$ in $|\Psi_{0\dots 0}\rangle$ is a triplet. The energy of $|\Psi_{0\dots 0}\rangle$ is then $E/N_p = -3J_f/4 + J_d/4$.

II. GROUND-STATE PHASE DIAGRAM

We start from the energy eigenvalues E of model (1) and write the dependence of E on J_f in an explicit form,

$$E(J_p, J_d, J_f) = E_{dp}(J_p, J_d) + J_f \left(\frac{1}{4} N_t - \frac{3}{4} N_s \right), \quad (3)$$

where N_s is the number of vertical dimers with composite spin $S_{ab}^n = 0$ and N_t is the number of vertical dimers with composite spin $S_{ab}^n = 1$. We have $N_s + N_t = N_p$. Since every vertical dimer singlet leads to an energy gain of J_f in the second part of the energy, the frustration favors states with $S_{ab}^n = 0$, i.e., a fragmentation of the chain. The lowest state of these finite fragments has a total spin $S_{frag} = 1$ for $J_d < 0$. We define the length k of a fragment as the number of triplet composite spins $S_{ab}^n = 1$ between two singlet composite spins.

According to the so-called linear programming scheme the extrema of the energy belong to states with fragments of identical length k .¹² In what follows we use the notation $\langle k \rangle$ for a fragmented state consisting of identical fragments of length k . The dimer product state [Eq. (2)] corresponds to fragments of length $k=0$, i.e., $|\Psi_{0\dots 0}\rangle = \langle 0 \rangle$. It is evident that the system undergoes at least one transition from the nonfragmented ground state $\langle \infty \rangle$ with $S_{ab}^n = 1$ ($n = 1 \dots N_p$) at small J_f to the dimer product ground state $\langle 0 \rangle$ with $S_{ab}^n = 0$ ($n = 1 \dots N_p$) for large J_f (see Fig. 2). As mentioned above, for antiferromagnetic J_d we indeed have a direct transition $\langle \infty \rangle \rightarrow \langle 0 \rangle$. However, for ferromagnetic J_d the question for the existence of intermediate ground states $\langle k \rangle$ with fragments of finite length $0 < k < \infty$ needs more attention.

We start from the ground state $\langle \infty \rangle$ for low frustration. The energy per unit cell of this state is

$$e_\infty^f = E_\infty^f / N_p = e_\infty + J_f/4, \quad (4)$$

where e_∞ is the energy of the ground state for the unfrustrated chain ($J_f = 0$). We determine e_∞ by a finite-size ex-

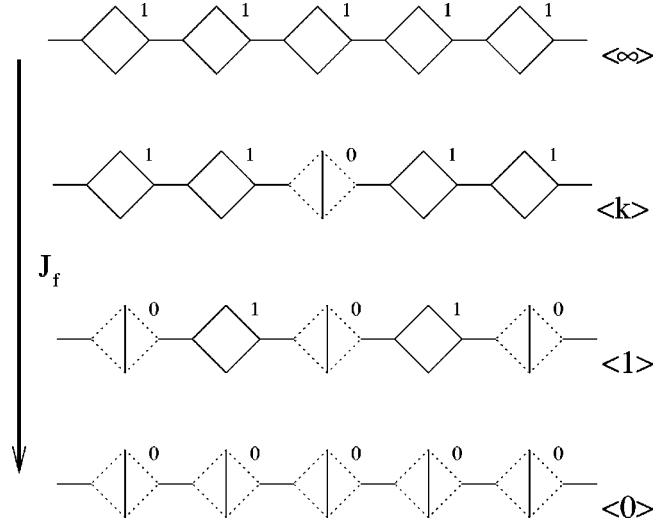


FIG. 2. Change of the ground state with increasing frustration J_f . For small frustration J_f all composite spins are in the triplet state and there is no fragmentation. Increasing J_f , some composite spins may prefer a vertical singlet state (indicated by vertical lines) and the chain may split into identical fragments of length k (the spin-spin correlation is zero along the J_p bonds neighboring a vertical singlet as indicated by dotted lines). For large J_f all composite spins are in the singlet state and the dimer product state [Eq. (2)] is the ground state.

trapolation of chains of length $N = 8, 16, 24$, and 32 as well as perturbation theory. The energy per unit cell e_k^f of a fragmented state $\langle k \rangle$ is given by the sum of the energies $E_k + kJ_f/4$ of the fragments and of the energies $-3J_f/4$ of the singlets separating the fragments:

$$e_k^f = \frac{E_k + (k-3)J_f/4}{k+1}. \quad (5)$$

E_k is the energy of the unfrustrated ($J_f = 0$) fragment of length k consisting of k plaquettes and $k+1$ dimer bonds J_d , and corresponds to the energy of a finite dimer-plaquette chain with $4k+2$ spins and open boundary conditions.

Using the Lanczos algorithm we can exactly calculate the energy of the chain fragments E_k up to $k=8$ (i.e., $N=34$). For larger fragment lengths and for the state $\langle \infty \rangle$ we need approximations. In the limits $|J_d/J_p| \ll 1$ and $|J_p/J_d| \ll 1$ perturbation theory is appropriate since for $J_d = 0$ and $J_p = 0$ the ground states are known as simple product states. Here we use Rayleigh-Schrödinger perturbation theory up to third order. The comparison of perturbation theory with the Lanczos results shows that the perturbation theory yields reliable results even for $|J_d/J_p| \approx 1$. Figure 3 shows the lowest energies of the state $\langle k \rangle$ with $k=0, 1, 2, 3, \dots$ and $k=\infty$ versus J_f for several J_d . The intersection points of the lowest lines determine the positions of first-order quantum phase transitions. It becomes obvious that the number of phase transitions changes from one to four, increasing the strength of ferromagnetic J_d . The existence of further transitions can be excluded by analyzing the k -dependence of E_k according to Niggemann *et al.*¹² The transition point $J_f^{k_1, k_2}$ between

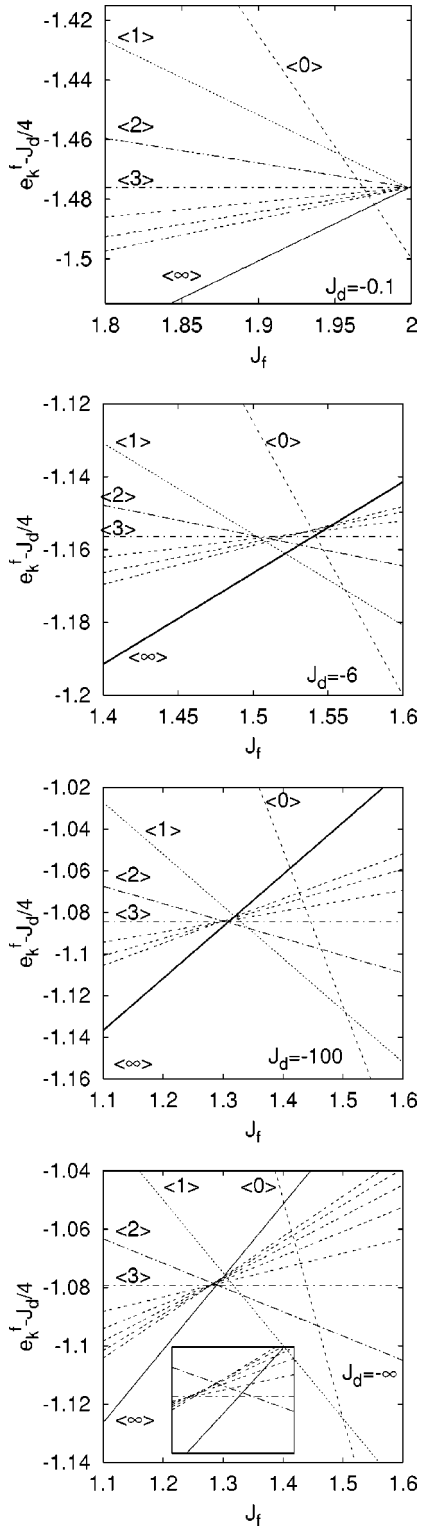


FIG. 3. Energies e_k^f [Eq. (5)] vs J_f for various strengths of ferromagnetic dimer bonds ($J_d = -0.1$, $J_d = -6$, $J_d = -100$, and $J_d = -\infty$) and fixed $J_p = 1$. For better comparison we have subtracted $e(J_p=0, J_f=0) = J_d/4$ from e_k^f . Curves are presented for $k=0, \dots, 7$ (exact results) and for $k=\infty$ (finite-size extrapolation). The accuracy of the $k=\infty$ results corresponds to the thickness of the solid line. We have labeled by $\langle k \rangle$ only the most relevant curves with $k=0, 1, 2, 3, \infty$. The inset in the figure for $J_d = -\infty$ shows the transition region between $\langle \infty \rangle$, $\langle 3 \rangle$, and $\langle 2 \rangle$ with an enlarged scale.

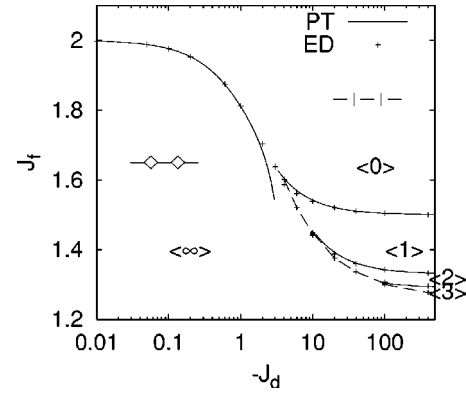


FIG. 4. Ground-state phase diagram for ferromagnetic $J_d < 0$ and fixed $J_p = 1$ (see the text). Solid line, perturbation theory; crosses, exact diagonalization.

two ground-state phases $\langle k_1 \rangle$ and $\langle k_2 \rangle$ is obtained by the relation $e_{k_1}^f = e_{k_2}^f$. Using Eqs. (4) and (5) we find

$$J_f^{k,\infty} = E_k - (k+1)e_\infty, \quad J_f^{k-1,k} = (k+1)E_{k-1} - (k)E_k. \quad (6)$$

The corresponding phase diagram is shown in Fig. 4. For completeness and comparison we have reconsidered the case of antiferromagnetic J_d (Refs. 6 and 7) including perturbation theory and an enlarged exact-diagonalization data set; see Fig. 5. For a small strength of the dimer coupling $|J_d| \lesssim 3$ the situation is similar for both ferromagnetic and antiferromagnetic J_d , i.e., we have a direct transition $\langle \infty \rangle \rightarrow \langle 0 \rangle$. Increasing the strength of ferromagnetic dimer coupling to $J_d \approx -4$ the situation is changed and we find an additional intermediate phase $\langle 1 \rangle$. Further increasing $|J_d|$ we find another intermediate phase $\langle 2 \rangle$, and finally we have a sequence of transitions $\langle \infty \rangle \rightarrow \langle 3 \rangle \rightarrow \langle 2 \rangle \rightarrow \langle 1 \rangle \rightarrow \langle 0 \rangle$ at a very strong ferromagnetic J_d . In this extreme limit $J_d \rightarrow -\infty$ the FDPC can be mapped onto the frustrated diamond chain and our results are in agreement with the results reported in Ref. 12.

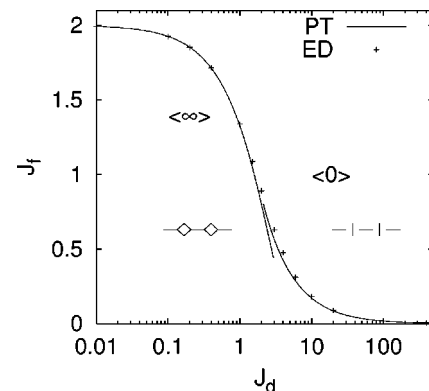


FIG. 5. Ground-state phase diagram for antiferromagnetic $J_d > 0$ and fixed $J_p = 1$ (see the text). Solid line, perturbation theory; crosses, exact diagonalization.

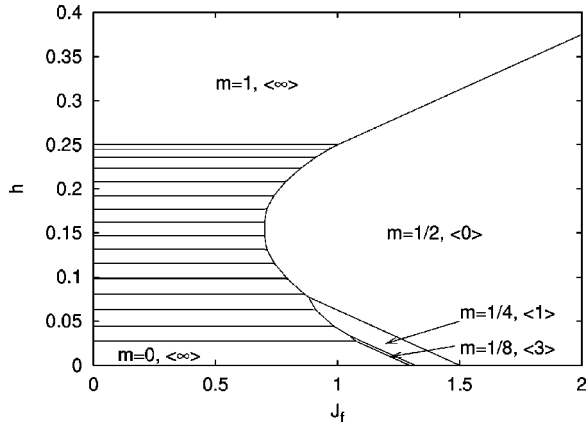


FIG. 6. Ground-state phase diagram for the FDPC of $N=32$ spins in a finite magnetic field h with $J_p=1$ and large ferromagnetic $J_d \rightarrow -\infty$.

III. INFLUENCE OF A MAGNETIC FIELD ON THE GROUND-STATE PHASES

As discussed in Refs. 8 and 9 the FDPC with antiferromagnetic J_d exhibits nontrivial magnetization plateaus. Therefore we now consider Hamiltonian (1) including an external magnetic field

$$H = H_{dp} + H_f - h S_{tot}^z, \quad (7)$$

where the z component of the total spin is given by $S_{tot}^z = \sum_i S_i^z$. For ferromagnetic J_d we have a more complex zero-field ground-state phase diagram, and we can expect interesting effects caused by a magnetic field. One important difference from the antiferromagnetic case $J_d > 0$ consists of the circumstance that, for ferromagnetic J_d , any fragment of length k carries a finite total spin $S_{frag} = 1$ in its lowest state for $h=0$, whereas the total spin S_{frag} for $J_d > 0$ is zero for even k . Hence the fragmentation is favored by the magnetic field, whereby short fragments are more favorable than long fragments. As already discussed above, the extrema of the energy belong to states with fragments of identical length k . For $J_d < 0$ in zero field the fragmented state $\langle k \rangle$ (k finite) can carry total spin $S_{tot}^z = N/(4k+4)$. From the numerical inspection of the excitation spectrum of finite fragments we argue that their excited states with $S_{frag} > 1$ are well separated from the lowest state with $S_{frag} = 1$. Hence we can expect for parameter points in the zero-field phase diagram (Fig. 4) not too far from a transition line that the first-order transitions can also be driven by an external magnetic field.

To be more precise we have studied the ground-state phases of a FDPC in a magnetic field with $N=32$ sites in the limit $J_d \rightarrow -\infty$. The corresponding phase diagram is shown in Fig. 6. We mention, that for this chain length the fragmented state $\langle 2 \rangle$ does not fit the periodic boundary conditions and is therefore missing. (A finite-size calculation including all zero-field ground-state phases would require $N=48$ sites which is currently beyond the available computer facilities.) We argue that except for missing the $\langle 2 \rangle$ phase the other ground-state phases should be well described by the $N=32$ system, since the correlation length in all phases is

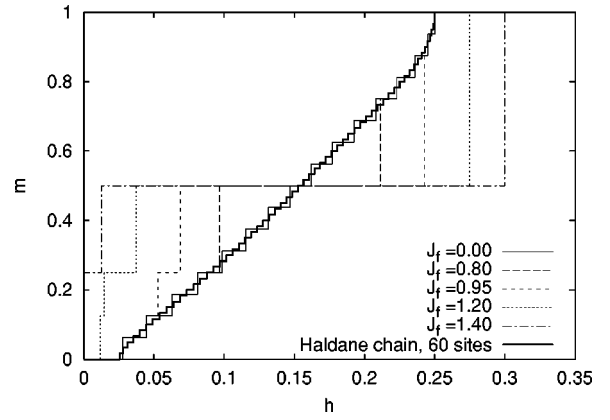


FIG. 7. Magnetization curves $m(h)$ for the FDPC of $N=32$ spins with $J_p=1$, large ferromagnetic $J_d \rightarrow -\infty$, and various strengths of frustration J_f .

rather small. In particular, the transition lines between $m=1/4$ and $1/2$ and between $m=1/2$ and 1 presented in Fig. 6 are exact, since these ground-state phases belong to product states correctly described for $N=32$.

Similar to the zero-field phase diagram there is no fragmentation for small frustration. However, supported by the field the fragmentation sets in already at $J_f \approx 0.71J_p$ for $h \approx 0.13$ instead of $J_f \approx 1.30J_p$ for $h=0$. In general, the phase transitions shown in Fig. 4 are shifted to lower values of frustration J_f .

The ground-state phase diagram in finite magnetic field leads to interesting magnetization curves (Fig. 7). We define the magnetization as $m = 2S_{tot}^z/N$; m is zero in a singlet state but unity in the fully polarized ferromagnetic state. Then the fragmented ground-state phases $\langle \infty \rangle$, $\langle 3 \rangle$, $\langle 2 \rangle$, $\langle 1 \rangle$, and $\langle 0 \rangle$ in Fig. 4 correspond to $m=0, 1/8, 1/6, 1/4$, and $1/2$, respectively.

For small $J_f \lesssim 0.70J_p$ we have a magnetization curve $m(h)$ typical for unfrustrated gapped spin systems, like the Haldane chain.^{14,16,15} Due to the gap the curve starts with a plateau at zero field but then it goes continuously to saturation. For the finite system considered the continuous part is staircase like with small steps (see the thin solid line in Fig. 7) which is related to the corresponding line of a $N=16$ Haldane chain. For comparison we added the density matrix renormalization group (DMRG) data for the Haldane chain of 60 sites (see the thick solid line in Fig. 7) taken from Ref. 15. For intermediate frustration $0.71J_p \lesssim J_f \lesssim 1.08J_p$ we have jumps, plateaus, and continuous parts in the $m(h)$ curve. However, for large enough $J_f \gtrsim 1.08J_p$ we have no continuous parts in the curve, but only plateaus connected by jumps. For instance, for $J_f = 1.2J_p$ (dotted line in Fig. 7) we have the same sequence of phase transitions as in Fig. 4, but now driven by increasing the magnetic field from $h=0$ to $h \approx 0.05$. As a result the magnetization m jumps from $m=0$ to $m=1/8$ to $1/6$ to $m=1/4$ to $1/2$ and further increasing h to saturation $m=1$ (notice that $m=1/6$ is missing for $N=32$ in Fig. 7). For large frustration $J_f \leq 1.5J_p$ we have an extreme $m(h)$ curve consisting only of one plateau at $m=1/2$ followed by a jump to saturation $m=1$.

We remark that the jump to the saturation $m=1$ present

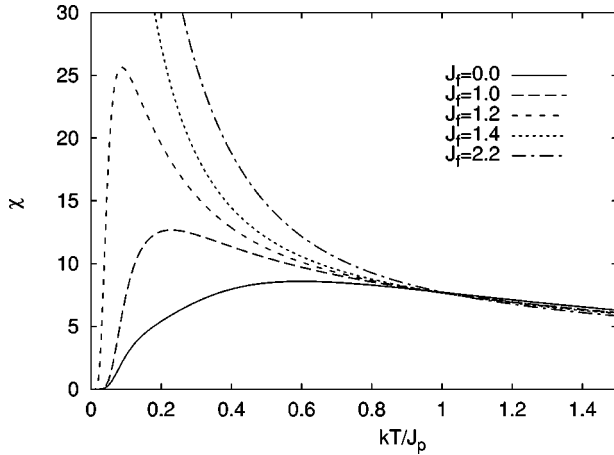


FIG. 8. Magnetic susceptibility χ of the FDPC of $N=16$ spins with $J_p=1$, ferromagnetic $J_d=-400$, and various strengths of frustration J_f .

for $J_f \gtrsim 0.97J_p$ is related to independent magnon excitations versus the fully polarized ferromagnetic state.¹⁷ We notice that our result is in accordance with the general rule of Oshikawa *et al.*¹⁸ that $n(s-\bar{m})$ has to be an integer. In our case the period n of a fragmented ground state $\langle k \rangle$ is $4k+4$, the spin s is one half and the magnetization per site \bar{m} is $1/(4k+4)$ (the magnetization per site \bar{m} corresponds to one half of the magnetization m used so far).

IV. FINITE TEMPERATURES

In this section we discuss some consequences of the ground-state phase diagram on the low-temperature thermodynamics. The different nature of the nonfragmented phase $\langle \infty \rangle$ and the fragmented phases $\langle k \rangle$ (k finite) has an important impact on thermodynamics. While the the ground state $\langle \infty \rangle$ is a singlet with gapped triplet excitations, the fragmented ground states $\langle k \rangle$ consist of independent paramagnetic units. As a consequence, in the former case both the susceptibility χ and the specific heat c are thermally activated and decay exponentially to zero if the temperature T goes to zero. Varying J_f the low-temperature behavior of χ changes basically, crossing the transition line between $\langle \infty \rangle$ and $\langle 3 \rangle$. For a fragmented ground state the susceptibility shows a paramagnetic Curie-like (i.e., $\chi=A/T$) divergency for $T \rightarrow 0$. Increasing the temperature we find deviations from the simple Curie-like behavior due to the existence of nonfragmented (collective) eigenstates. The Curie constant A is proportional to $S_{frag}^2 N/(4k+4)$, where k is the fragment length in the ground state, $N/(4k+4)$ is the number of independent paramagnetic units (fragments) and $S_{frag}=1$ is the total spin of these units. Therefore, $A(J_f)$ shows a jump crossing the phase transition line between two fragmented phases. The behavior for the specific heat near the phase boundaries can be more complex, since close to these boundaries we have quasidegenerated low-lying levels which may lead to additional low-temperature peaks in c .

To illustrate this behavior, in Figs. 8 and 9 we present the susceptibility and the specific heat for a FDPC of $N=16$ sites

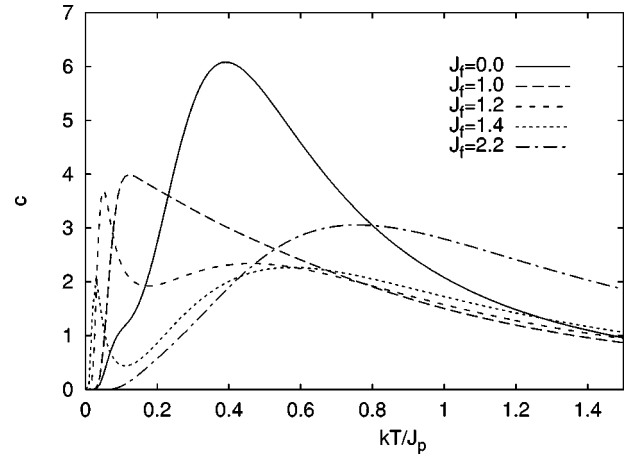


FIG. 9. Specific heat c of the FDPC of $N=16$ spins with $J_p=1$, ferromagnetic $J_d=-400$, and various strengths of frustration J_f .

obtained by full diagonalization of the Hamiltonian matrix. The transition from spin-gap to paramagnetic behavior is clearly seen in Fig. 8 between $J_f=1.2$ and 1.4 . In the same parameter region the specific heat shows a characteristic double-peak structure due to the quasidegeneracy of states near the transition line. This effect should survive in the thermodynamic limit.

V. SUMMARY

In conclusion, we have found a series of first-order quantum phase transitions in a frustrated one-dimensional quantum spin system which is closely related to the possibility of fragmentation of the considered chain. This series of phase transitions can be driven either by frustration or by a magnetic field. We emphasize that this fragmentation and the related consequences for physical properties are pure quantum effects not present in classical spin systems. The existence of different ground states has a strong impact on the magnetization curve and the low-temperature thermodynamics of the spin system. In dependence on the frustration the magnetization curve shows plateaus, jumps as well as continuous parts.

Though the possibility of fragmentation has been observed also for frustrated two-leg spin ladder^{19,15}, in the spin ladder system only very simple ground states seem to be relevant for its magnetization curve. The low-temperature thermodynamics can be either spin-gap-like for small frustration or paramagnetic for large frustration. Furthermore an additional low-temperature peak in the specific heat can appear.

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