Magnetization Curves of Antiferromagnetic Heisenberg Spin-$\frac{1}{2}$ Ladders

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Magnetization processes of spin-$\frac{1}{2}$ Heisenberg ladders are studied using strong-coupling expansions, numerical diagonalization of finite systems, and a bosonization approach. We find that the magnetization exhibits plateaux as a function of the applied field at certain rational fractions of the saturation value. Our main focus is on ladders with three legs where plateaux with magnetization one third of the saturation value are shown to exist.   [S0031-9007(97)04782-0]

Recently there has been considerable interest in coupled Heisenberg antiferromagnetic (HAF) chains, so-called "spin ladders," where one of the fascinating discoveries is that the appearance of a gap depends on the number of chains being even or odd (for recent reviews see, e.g., [1]). In this Letter we study ladder systems at zero temperature in a strong uniform magnetic field. This issue has so far only been addressed for *two* coupled chains with a magnetization experiment on $Cu_2(C_5H_{12}N_2)_2Cl_4$ [2] and theoretically using numerical diagonalization [3], series expansions [4], and a bosonization approach [5]. These studies found a plateau at zero magnetization whose width is given by the spin gap in the otherwise smooth magnetization curve. In this Letter we extend the theoretical approaches to three and more coupled chains using strong-coupling expansions, numerical diagonalization, and a bosonization approach. We find that in general the magnetization curves exhibit plateaux also at certain nonzero quantized values of the magnetization.

To be precise, we concentrate on the zero-temperature behavior of the following HAF spin ladder with N legs (kept fixed) and length L (taken to infinity):

$$H^{(N)} = J' \sum_i \sum_{x=1}^{L} \vec{S}_{i,x} \vec{S}_{i+1,x} + J \sum_{i=1}^{N} \sum_{x=1}^{L} \vec{S}_{i,x} \vec{S}_{i,x+1}$$
$$- h \sum_{i,x} S_{i,x}^z, \qquad (1)$$

where the $\vec{S}_{i,x}$ are spin-$\frac{1}{2}$ operators and h is a dimensionless magnetic field. We assume periodic boundary conditions along the chains but investigate both open (OBC) and periodic boundary conditions (PBC) along the rungs. The magnetization $\langle M \rangle$ is given by the expectation value of the operator $M = \frac{2}{LN} \sum_{i,x} S_{i,x}^z$.

Our main result is that $\langle M \rangle$ as a function of h has plateaux at the quantized values

$$\frac{N}{2}(1 - \langle M \rangle) \in \mathbf{Z}. \qquad (2)$$

This condition also appears in the Lieb-Schultz-Mattis theorem [6] and its generalizations [7,8]. There it is related to gapless *nonmagnetic* excitations, but plateaux in

magnetization curves appear if there is a gap to *magnetic* excitations.

However, there are other arguments which lead to (2) as the quantization condition for $\langle M \rangle$ at a plateau. A particularly simple one is given by the limit of strong coupling along the rungs $J' \gg J$ (which has also proven useful in other respects [9]). At $J = 0$ one has to deal only with Heisenberg chains of length N and the only possible values for the magnetization are precisely the solutions of (2): $\langle M \rangle \in \{-1, -1 + 2/N, \ldots, 1 - 2/N, 1\}$.

For $N = 2$ and $J' \gg J$ this consideration predicts a plateau at $\langle M \rangle = 0$. The boundary of this plateau is related to the spin gap simply by $h_c = \Delta$. The strong-coupling series for this gap reads [10]

$$\Delta = J' - J + \frac{J^2}{2J'} + \frac{J^3}{4J'^2} - \frac{J^4}{8J'^3} + \mathcal{O}(J^5), \quad (3)$$

where we have extended the result of [10] to fourth order (this further order is crucial to obtain a zero of the gap for $J' \geq 0$).

For $N = 3$ and small magnetic fields one has a degeneracy which makes already first-order perturbation theory nontrivial. For OBC and $|\langle M \rangle| \leq 1/3$, the low-lying spectrum is then given by a spin-$\frac{1}{2}$ chain in a magnetic field whose magnetization curve is well studied (see [11] and references therein). On the other hand, for PBC and $|\langle M \rangle| \leq 1/3$, the first-order low-energy effective Hamiltonian for (1) at $N = 3$ turns out to be (see also [12])

$$H_{\text{eff}}^{(III, p)} = \frac{J}{3} \sum_{x=1}^{L} (1 + \sigma_x^+ \sigma_{x+1}^- + \sigma_x^- \sigma_{x+1}^+) \vec{S}_x \vec{S}_{x+1}$$
$$- h \sum_{x=1}^{L} S_x^z, \qquad (4)$$

where the $\vec{S}_x$ are SU(2) operators acting in the spin space and $\sigma_x^{\pm}$ act on another two-dimensional space which comes from a degeneracy due to the permutational symmetry of the chains. In particular, the usual spin-rotation symmetry is enlarged by an U(1) coming from the

XY-type interaction in the space of the Fourier transforms along the rungs [first factor in (4)]. The Hamiltonian (4) encodes the magnetization curve in the strong-coupling limit of the three-leg ladder with PBC for $|\langle M \rangle| \leq 1/3$.

At least for OBC, the situation is a little more favorable for the $\langle M \rangle = 1/3$ plateau that one expects for $N = 3$. It turns out that one can use nondegenerate perturbation theory to compute the energy cost to flip one spin around this plateau and thus its lower and upper boundaries,

$$h_{c_1} = 2J - \frac{2J^2}{9J'} - \frac{94J^3}{243J'^2} + \mathcal{O}(J^4), \tag{5}$$

$$h_{c_2} = \frac{3}{2}J' - J + \frac{3J^2}{4J'} + \frac{2065J^3}{3888J'^2} + \mathcal{O}(J^4). \tag{6}$$

Finally, it is straightforward to exactly compute the upper critical field h_{uc} for the transition to a fully magnetized state for PBC, even L and arbitrary N;

$$h_{uc} = \begin{cases} 2(J + J'), & N \text{ even}, \\ [1 - \cos(\pi \frac{N-1}{N})]J' + 2J, & N \text{ odd}. \end{cases} \tag{7}$$

Actually, the result (7) at $N = 3$, $h_{uc} = 3/2J' + 2J$ also applies to OBC.

We now proceed with considerations that follow closely classical work on single Heisenberg spin chains [11] and the 2D triangular HAF [13]. We have numerically calculated the lowest eigenvalues as a function of the magnetization, wave vectors, and coupling constants on finite systems with up to a total of 24 sites.

Figure 1 illustrates our main results with a magnetization curve for $N = 3$ at $J'/J = 3$ with OBC. The thin lines denote curves at finite system size and clearly exhibit a plateau with $\langle M \rangle = 1/3$. The strong-coupling approximations (5) and (6) for the boundaries of this plateau are in good agreement with the finite-size data. Finite-size ef-
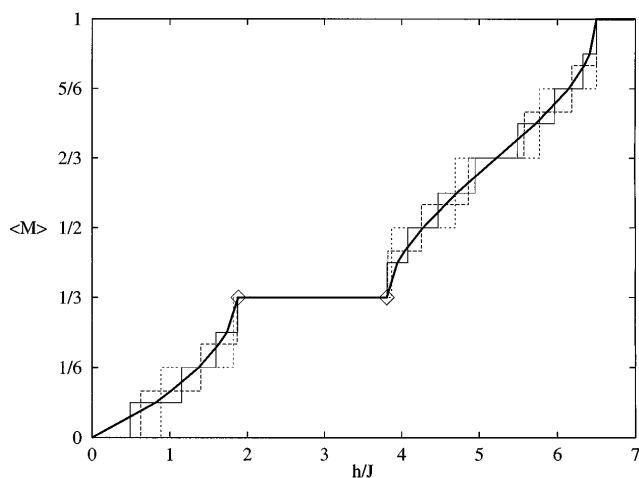
fects are also small for the midpoints of the steps in the magnetization curves [11]. From this one obtains an extrapolation to infinite L which is shown by the full line in Fig. 1.

A more compact representation is given by "magnetic phase diagrams" which show the projection of the conventional magnetization curves as in Fig. 1 onto the axis of the magnetic field. We illustrate this in Fig. 2 with the case of a two-leg ladder which has already been studied in [3]. On a finite lattice the possible values of $\langle M \rangle$ are quantized; the values of the magnetic field h/J where a transition from one such value occurs to the neighboring one are shown by lines as a function of J'/J. Regions without such lines denote plateaux in the magnetization curve while regions where they are close to each other correspond to smooth transitions in the thermodynamic limit.

The magnetization curves of [2,3] with $J'/J = 5$ are located somewhat beyond the right border of Fig. 2. Our figure contains additional information about the dependence on J'/J. Note that the numerically obtained values for the transition to full magnetization do indeed match with the version of (7) for the ladder: $h_{uc} = 2J + J'$. For the boundary of the plateau at $\langle M \rangle = 0$ one observes that finite-size effects are not substantial for $J'/J \gtrsim 3/2$, and in this region one observes also good agreement with the approximation (3) for the transition value of the magnetic field $h_c = \Delta$. At weak coupling, field theoretic arguments [14] predict the gap to open linearly, $\Delta \sim J'$. This is compatible with Fig. 2, though due to the large finite-size effects in this region much longer chains are needed to confirm this linear behavior conclusively [15].

Figures 3 and 4 show the magnetic phase diagrams for $N = 3$ with OBC and PBC, respectively (Fig. 1 is a section of Fig. 3 at $J'/J = 3$). Both figures clearly exhibit a plateau with $\langle M \rangle = 1/3$ at least in the region $J' \geq 2J$. The strong-coupling series (5) and (6) for the
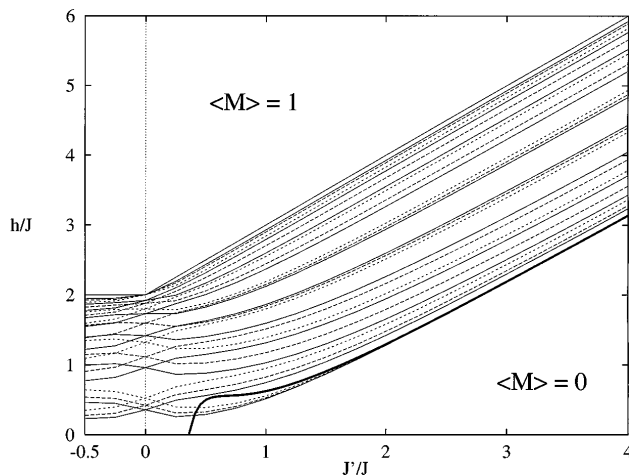

FIG. 1. Magnetization curve for $N = 3$ at $J'/J = 3$ with OBC. The thin full lines are for $L = 8$, the long-dashed lines for $L = 6$, and the short-dashed lines for $L = 4$. The thick full line indicates the expected form in the thermodynamic limit. The two diamonds denote the series (5) and (6) for the boundaries of the plateau.

FIG. 2. Magnetic phase diagram of the ladder with $N = 2$ legs. The thin full lines are for $L = 12$, the long-dashed lines for $L = 10$, and the short-dashed lines for $L = 8$. The thick full line shows the gap (3).
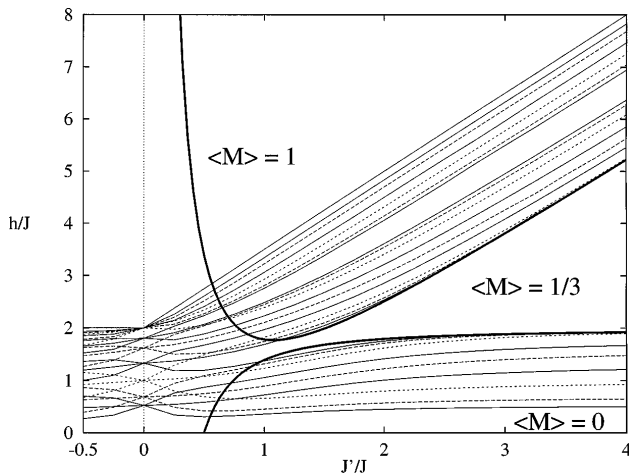
FIG. 3. Magnetic phase diagram of $N = 3$ chains with OBC. The thin full lines are for $L = 8$, the long-dashed lines for $L = 6$, and the short-dashed lines for $L = 4$. The two thick full lines denote (5) and (6), respectively.

boundaries of this plateau are also shown in Fig. 3, and in the aforementioned region $J' \gtrsim 2J$, where finite-size effects are small, one observes good agreement between the expansions and the numerical results.

Figures 3 and 4 differ at least in their details. For example, one observes that at the upper boundary of the $\langle M \rangle = 1/3$ plateau in Fig. 4 it becomes favorable to flip two spins at once rather than one for $J' \gtrsim 3J$ which may be interpreted as one signal of the frustration introduced into the system.

At strong coupling the excitations above the $\frac{1}{3}$ plateau in Fig. 4 are described by (4) with all spins aligned along the field ($S_x^z = \frac{1}{2}$). This is an XY chain and therefore massless, providing us with an example of a plateau in the magnetization curve with gapless nonmagnetic excitations above it.

The strong-coupling expansions as well as the numerical results obtained so far clearly show the existence
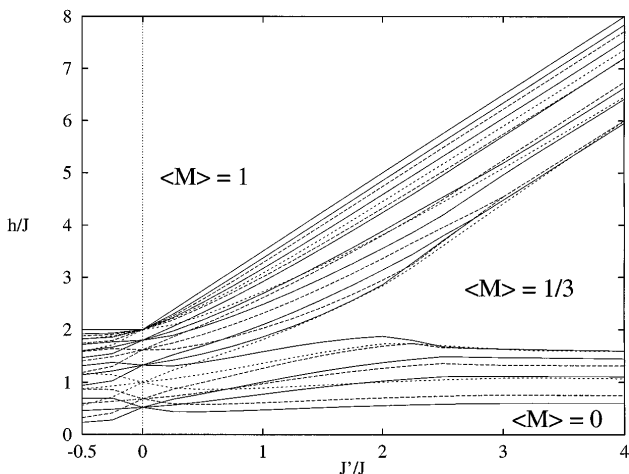

FIG. 4. Same as Fig. 3, but with periodic boundary conditions.

of plateaux for sufficiently strong coupling. To see if the plateau persists in the weak-coupling region $J' \ll J$ we use Abelian bonsonization (see, e.g., [16,17]). The computation to be presented below is similar to the ones performed recently in [8,18], so we refer the interested reader to these two references for more details.

The starting point for this weak-coupling expansion is the observation that at $J' = 0$ one has N decoupled spin-$\frac{1}{2}$ HAFs in a magnetic field whose low-energy properties are described by a $c = 1$ Gaussian conformal field theory [19]. This theory is characterized by the radius of compactification $R(\langle M \rangle, \Delta)$ which depends on the magnetization as well as the XXZ anisotropy Δ which we have here introduced for convenience. At zero magnetic field one has [17]: $R^2(0, \Delta) = \frac{1}{2\pi}(1 - \frac{1}{\pi}\cos^{-1}\Delta)$. In general, this radius can be computed using the Bethe-ansatz solution [19] for the Heisenberg chain. For a qualitative understanding it may be helpful to notice that the radius of compactification R decreases if either Δ becomes smaller or if the magnetization $\langle M \rangle$ increases.

Now the low-energy properties of the Hamiltonian (1) at small coupling are described by the following Tomonaga Hamiltonian with interaction terms:

$$\bar{H}^{(N)} = \int dx \left[\frac{\pi}{2} \sum_{i=1}^{N} \{\Pi_i^2(x) + R^2(\langle M \rangle, \Delta)[\partial_x \phi_i(x)]^2\} \right.$$
$$+ \frac{\lambda_1}{2\pi} \sum_i [\partial_x \phi_i(x)][\partial_x \phi_{i+1}(x)]$$
$$+ \sum_i \{\lambda_2 : \cos[4k_F x + \sqrt{4\pi}(\phi_i + \phi_{i+1})] :$$
$$+ \lambda_3 : \cos[\sqrt{4\pi}(\phi_i - \phi_{i+1})] :$$
$$\left. + \lambda_4 : \cos[\sqrt{\pi}(\tilde{\phi}_i - \tilde{\phi}_{i+1})] :\} \right], \qquad (8)$$

with $\Pi_i = \frac{1}{\pi}\partial_x \tilde{\phi}_i$, and $\lambda_i \sim J'/J$. It is convenient to rescale the fields by the radius of compactification R in (8), i.e., $\phi_i \rightarrow \sqrt{4\pi} R \phi_i$.

For $N = 3$ chains with PBC we now change variables from the fields ϕ_1, ϕ_2, ϕ_3 to $(1/\sqrt{3})(\phi_1 + \phi_2 + \phi_3)$, $(1/\sqrt{2})(\phi_1 - \phi_2)$, $(1/\sqrt{6})(\phi_1 + \phi_2 - 2\phi_3)$. Following [16] one can show that the perturbation terms with coefficients λ_3 or λ_4 in (8) give rise to a mass for the latter two fields.

Let us now consider the remaining field $\phi_{\mathrm{diag}} := (1/\sqrt{N}) \sum_{i=1}^{N} \phi_i$. Radiative corrections to (8) generate the interaction term $: \cos(2\phi_{\mathrm{diag}}/R) :$ [20]. For $N = 3$ and at $\langle M \rangle = 0$ this operator is irrelevant (in the region of Δ close to 1), which confirms that three chains weakly coupled in a periodic manner are massless.

Now we address the question of the appearance of plateaux for $\langle M \rangle \neq 0$. Since this requires a gap for the (magnetic) excitations, we expect such a plateau to occur if the remaining field ϕ_{diag} acquires a mass. In fact, the additional interaction term

$$: \cos(\phi_{\mathrm{diag}}/R) : \qquad (9)$$

survives in the continuum limit on a plateau [21]. The operator (9) can appear only if (2) holds, as can easily be inferred from translational invariance of the original Hamiltonian (1) and the fact that $k_F = \frac{1}{2}(1 - \langle M \rangle)\pi$ and a one-site translation of the lattice Hamiltonian (1) translates into the internal symmetry transformation $\phi_{\text{diag}} \rightarrow \phi_{\text{diag}} + 2NRk_F$.

For $N = 3$, Eq. (2) requires that $\langle M \rangle = 1/3$. If one now estimates the radius of compactification $R(\frac{1}{3}, 1)$ following, e.g., [18], one finds that at $J' = 0$ the operator (9) is slightly irrelevant for $\Delta = 1$. The dimension of this operator decreases with J' implying that the $\frac{1}{3}$ plateau in Fig. 4 extends down into the region of small J'. It should be noted that the appearance of the plateau for a given small J' crucially depends on the value of Δ, explaining why the numerical evidence in Fig. 4 is not conclusive in this region. For simplicity we have concentrated on PBC. The case of OBC is qualitatively similar but more subtle in the details and will be discussed in [21].

In this Letter we have shown that spin ladders exhibit plateaux in their magnetization curves when subjected to strong magnetic fields. We have mainly concentrated on the plateau with $\langle M \rangle = 1/3$ in three coupled chains, but also other rational values can be obtained by varying the number of chains N. Quantum fluctuations (i.e., the choice of the spin $S = \frac{1}{2}$) do play an important role—the plateaux disappear in general when one inserts classical spins into (1). Nevertheless, by analogy to the two-dimensional triangular antiferromagnet [22] it seems likely that such plateaux would appear in first-order spin-wave theory. Also with Ising spins the behavior of (1) is different: At zero temperature the magnetization changes only discontinuously between plateaux values (i.e., no smooth transitions occur), and a gap opens for both odd and even numbers of chains.

It would be highly interesting to check experimentally whether the plateaux can indeed be observed, in particular since nowadays materials with a given number of legs can be engineered. The first nontrivial check would be to look for the $\langle M \rangle = 1/3$ plateau in a three-leg ladder. Here, the material $Sr_2Cu_3O_5$ [23] comes to mind, which is, however, not very well suited for these purposes since the necessary order of magnetic fields is not accessible today due to its large coupling constants. However, there are at least two-leg ladder materials such as the conventional $(VO)_2P_2O_7$ [24] or $Cu_2(C_5H_{12}N_2)_2Cl_4$ [2] with much weaker coupling constants. A three-leg analog of such materials could provide a testing ground for our predictions, in particular if such a material can be found with $J' \gtrsim 2J$ where we would expect a clearly visible plateau in the magnetization curve at sufficiently low temperatures (cf. Fig. 1).

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