Spinon signatures in the critical phase of the \((1, \frac{1}{2})\) ferrimagnet in a magnetic field

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We propose an effective theory for the critical phase of a quantum ferrimagnetic chain with alternating spins 1 and \(\frac{1}{2}\) in an external magnetic field. With the help of the matrix product variational approach, the system is mapped to a spin-\(\frac{1}{2}\) XXZ chain in an (effective) magnetic field; as a byproduct, we obtain an excellent description of the optical magnon branch in the gapped phase. Recent finite-temperature DMRG results for the low-temperature part of the specific heat are well described by the present approach, and the “pop-up” peaks, developing near the critical field values and in the middle of the critical phase, are identified with the contributions from two different spinon bands of the effective spin-\(\frac{1}{2}\) chain. The effect should be as well observable in other spin-gap systems in an external field, particularly in spin ladders.

75.10.Jm, 75.50.G, 75.40.Cx, 75.40.Mg

Introduction. Recently, there has been a considerable interest in the properties of one-dimensional (1D) quantum ferrimagnets. A generic example of a 1D ferrimagnet is the Heisenberg spin chain with antiferromagnetic nearest neighbor interaction and alternating spins 1 and \(\frac{1}{2}\), described by the Hamiltonian

\[ \hat{H} = J \sum_n (S_n \tau_n + \tau_n S_{n+1}) - h \sum_n (S_n^z + \tau_n^z), \]  

where \(S_n\) and \(\tau_n\) are respectively spin-1 and spin-\(\frac{1}{2}\) operators in the \(n\)-th elementary magnetic cell, and \(h = g\mu_B H\), where \(H\) is the external magnetic field. According to the Lieb-Mattis theorem, \(h = 0\) the ground state of the system has total spin \(S_{tot} = L/2\), where \(L\) is the number of unit cells, and thus is necessarily long-range ordered, making the problem amenable to the spin wave theory (SWT) approach. Since the elementary cell consists of two spins, SWT yields two types of magnons: a gapless “acoustical,” or “ferromagnetic” branch with \(S^2 = L/2 - 1\), and a gapped “optical,” or “antiferromagnetic” branch with \(S^2 = L/2 + 1\). The optical magnon gap was numerically found to be \(\Delta_{opt} = 1.759J\). The existence of two magnon branches manifests itself in various thermodynamic quantities.

In magnetic field the acoustic branch acquires a gap, while the optical gap decreases with the field. If the field exceeds the critical value \(h = h_{c1} = \Delta_{opt}\), the optical gap closes, and the system enters the critical phase, which is expected to be of the Luttinger-liquid type, in analogy with other models (e.g., spin-1 chain) gapped spin-\(\frac{1}{2}\) chains and ladders). This phase extends up to the second critical field \(h = h_{c2} = 3J\), where there is an other transition to the saturated ferromagnetic phase.

In the gapless regime \(h_{c1} < h < h_{c2}\), the temperature dependence of the specific heat has revealed a puzzling behavior with a single well-pronounced low-\(T\) peak which pops up around \(T \approx 0.2J\) when \(h\) is in the middle between \(h_{c1}\) and \(h_{c2}\); when \(h\) is shifted towards \(h_{c1}\) or \(h_{c2}\), the peak becomes flat and develops a shoulder with another ill-pronounced peak centered at about \(0.05J\).

In this paper, we show that in the critical phase the model \(i\) can be mapped to an effective spin-\(\frac{1}{2}\) XXZ chain in an external field, which yields quite an accurate description of all the features of the low-temperature part of the specific heat. The “pop-up” peak structure described above can be explained by the presence of two different spinon bands of the effective spin-\(\frac{1}{2}\) chain. This effect is rather general and should be observable in other gapped 1D spin models, e.g., spin-\(\frac{1}{2}\) ladders.

Effective model. The general idea of any mapping to an effective model is to reduce somehow the Hilbert space of the problem, keeping only a few “most important” states per the elementary cell. For example, in the strong-coupling limit of the spin-\(\frac{1}{2}\) ladder one keeps for each rung only the singlet and the lowest-energy triplet component. For the \((1, \frac{1}{2})\) ferrimagnet the set of the cell wave functions \(\psi_{jm}\) consists of a doublet \(j = \frac{3}{2}\) and a quartet \(j = \frac{5}{2}\). One would naively expect that in a strong field it is now necessary to keep three states with \((j, m) = (\frac{1}{2}, \frac{1}{2}), (\frac{3}{2}, \frac{1}{2}), (\frac{3}{2}, \frac{3}{2})\); it is clear, however, that there is only one ground state and one excitation becoming gapless at \(h = h_{c1}\), so that the interplay should be effectively between two cell wavefunctions being “proper” linear combinations of \(\psi_{jm}\). Thus, the problem is how to identify those proper combinations. The first step is to describe accurately the optical magnons, since the critical phase is formed by their condensation into the ground
The linear SWT captures the essential physics of the model at the qualitative level, however, for a quantitative description one has to go far beyond the linear approximation, keeping the higher-order terms in $1/S$ e.g., in linear SWT $\Delta_{opt} = J$ and the corrections from the leading $1/S$ term yield $1.676J$ instead of the numerical value $1.754J$. Thus we use a different scheme which has proved to be very successful for the $\frac{3}{2}$- $\frac{1}{2}$ ferrimagnet, namely the variational matrix product (MP) approach. In Ref. 3 the following (non-normalized) variational ground state wave function was proposed:

$$\Psi_0 = \text{tr} (g_1g_2 \cdots g_L),$$

$$g_n = uM^{0 \frac{1}{2}} + vM^{1 \frac{1}{2}} + M^{1 \frac{1}{2}}, \quad M^{0 \frac{1}{2}} = \mathbb{1} \psi^0 \frac{1}{\sqrt{2}},$$

$$M^{1 \frac{1}{2}} = -\frac{1}{\sqrt{3}} \sigma^0 \psi^0 \frac{1}{\sqrt{2}} + \frac{1}{\sqrt{2}} \sigma^+ \psi^+ \frac{1}{2},$$

$$M^{1 \frac{1}{2}} = \frac{1}{\sqrt{2}} \sigma^- \psi^- \frac{1}{2} + \frac{1}{\sqrt{6}} \sigma^0 \psi^0 \frac{1}{2} - \frac{1}{\sqrt{3}} \sigma^0 \psi^0 \frac{1}{2}.$$ (2)

Here $\sigma^0 = \sigma_z$ and $\sigma^\pm = \frac{1}{\sqrt{2}}(\sigma_x + i\sigma_y)$ are the Pauli matrices, $\mathbb{1}$ is the $2 \times 2$ unit matrix, $\psi_{jm}$ denote spin states of the $n$-th cell, and $u, v$ are the variational parameters whose optimal values minimizing the energy of $\Psi_0$ at $h = 0$ were found to be $u = 1.3026, v = 1.0788,$ with the variational ground state energy per unit cell $E_{0\text{var}} = -1.449J$ (to compare with the numerical value $E_0 = -1.455$). It was shown that $\Psi_0$ possesses the correct quantum numbers of $S_{tot} = S_{tot} = L/2$ and provides a very good description of the ground state correlations. Any quantum averages for MP wave functions are readily calculated using the transfer matrix technique; this nice formalism was first proposed in Refs. 1, 2. The MP approach is especially well suited to this problem since the fluctuations are extremely short-ranged, with the correlation radius smaller than one unit cell length.

Now we construct the simplest trial wave function for the optical magnon with the momentum $k$:

$$|k\rangle = \sum_n e^{i\delta n} |n\rangle, \quad |n\rangle = \text{tr} (g_1 \cdots g_{n-1} \bar{g}_n g_{n+1} \cdots g_L),$$

$$\bar{g}_n = \mathbb{1} \psi^0 \frac{1}{\sqrt{2}} + w \sigma^0 \psi^0 \frac{1}{\sqrt{2}} + \sqrt{5/3} f Q^{1 \frac{1}{2}},$$

$$Q^{1 \frac{1}{2}} = \sqrt{3/5} \sigma^0 \psi^0 \frac{1}{\sqrt{5}} - \sigma^+ \psi^+ \frac{1}{\sqrt{2}}.$$ (3)

The form of $\bar{g}_n$ in (3) is dictated by the requirement that $|\psi\rangle = |n\rangle$ is the state with $S_{tot} = S_{tot} = L/2 + 1$, then, according to the general formalism presented in Ref. 3 $\bar{g}_n$ should carry the “hypermagnon” quantum numbers $(\frac{1}{2}, \frac{1}{2})$, while each $g_n$ carries $(\frac{1}{2}, \frac{1}{2})$. Here $f, w$ are still free (variational) parameters. Note that generally the states $|n\rangle$ are not orthogonal to each other, but are orthogonal to $\Psi_0$. For the following, it is important to note that $\bar{g}_n$ can be represented in the form

$$\bar{g}_n = f - \frac{1}{\sqrt{2}} g_n \sigma^+ + \frac{1}{\sqrt{2}} g_n \sigma^+ - w \sigma^+ \psi^0 \frac{1}{\sqrt{2}},$$

so that $|k\rangle$ in fact depends only on $w = \{ w + \sqrt{2}(u + \frac{1}{\sqrt{3}}) \}$. Thus, one parameter in (3) is redundant, and we fix that by choosing

$$f = \{ z + w \sqrt{3} (v - u \sqrt{3}) \} / (2 + z),$$

$$z = 2 \sqrt{3} w + (3 + 12u^2 (u^2 + 1))^{1/2},$$ (4)

which yields a remarkable property: the set of one-magnon states $\{|n\rangle\}$ becomes mutually orthogonal, considerably simplifying further calculations. Moreover, one can show that arbitrary $N$-magnon states $|n_1, \ldots, n_N\rangle$ become orthogonal; however, they are not normalized, and, generally, $\langle n, n + m|n, n + m\rangle$ does not coincide with $\langle n|n\rangle^2$, though it tends very fast to the latter value as $m$ increases. The one-magnon norm and the matrix elements of the Hamiltonian (1) are given by

$$\begin{align*}
N_0 &\equiv \langle n|n\rangle = 0.1173 w^2 - 0.0337 w + 0.1432, \\
\langle n|H|n\rangle' &\equiv J x^{n-n'|A + B \delta_{n-n'|L} + C \delta_{n\text{var}}}, \\
x &\equiv 0.2147, \quad A = -0.0483 w^2 + 0.0808 w - 0.0284, \\
B &\equiv -0.0839 w^2 + 0.0379 w + 0.0010, \\
C &\equiv 0.3359 w^2 - 0.0495 w + 0.5860,
\end{align*}$$

where $\bar{H} = \bar{H} - L E_{0\text{var}}$. Now one can calculate the dispersion by minimizing the excitation energy $\varepsilon(k) = \langle k|\bar{H}|k\rangle / \langle k|k\rangle$. (5)

$$\varepsilon(k) = \frac{J}{N_0}\left\{ C + 2 B x \cos k + A \frac{1 - x^2}{1 + x^2 - 2 x \cos k} \right\}. \quad (6)$$

In principle, the optimal value of $w$ should be calculated separately for each $k$. One might first try to minimize the gap $\Delta_{opt} = \varepsilon(k = 0)$, which yields $w = w_0 = -3.8605$, with $\Delta_{opt} \approx 1.754 J$, the resulting dispersion for $w = w_0$ being in excellent agreement with the exact diagonalization data (see Fig. 1).

Now, we can obtain the desired mapping by introducing effective spin-$\frac{1}{2}$ states $|\alpha_n\rangle$ at each cell, $\alpha_n = \pm \frac{1}{2}$, and making the identification $|\alpha_1 \alpha_2 \cdots \alpha_L\rangle = \text{tr} (R_1 R_2 \cdots R_L)$, where $R_n = \bar{g}_n$, $g_n$ for $\alpha_n = \frac{1}{2}, -\frac{1}{2}$, respectively. Introducing the spin-$\frac{1}{2}$ operators $s_n^\pm$, and restricting all effective interactions to nearest neighbors only, one can write down the effective Hamiltonian for the critical phase of the (1, $\frac{3}{2}$) ferrimagnet (1):

$$\tilde{H}_0 = \sum_n \frac{J_{xy}}{2} (s_{n}^+ s_{n+1}^- + s_{n+1}^- s_{n}^+) + J_{zz} s_{n}^z s_{n+1}^z - 2 J_{zz} s_{n}^z ,$$

$$J_{xy} = -2 t_1, \quad J_{zz} = U_1, \quad h_n = (h - t_0 - U_1)/2, \quad (7)$$

$$t_m = \frac{\langle n|\bar{H}|n+m\rangle}{N_0}, \quad U_m = \frac{\langle n+m|\bar{H}|n+m\rangle}{N_0} - 2 t_0.$$ (8)

Here a remark is in order. From (3) one can see that the hopping amplitudes $t_m \propto x^m$ are very small for $m \geq 2$, $t_2 = -0.0242, t_3 = -0.0052, \ldots$ which justifies keeping only the magnon type only up to $m = 1$ in (3).
The same reasoning holds for neglecting other interaction terms like $(s_n^+ s_{n+1}^- s_{n-1}^- s_{n+2}^+), (s_n^+ s_{n+2}^- s_n^- s_{n+3}^+)$, etc.

The numerical values for the parameters $t_0, t_1, U_1$, which determine our effective model, are $t_0 = 2.3370, t_1 = -0.2608, U_1 = 0.1187$, so that the spin-$\frac{1}{2}$ chain is in a gapless XX phase with $\Delta = J_{xy}/J_{x} \simeq 0.227$. At the critical value of the field $h_c = h_{c,c} = \frac{1}{2}J_{xy}(1 + \Delta)$ the spin-$\frac{1}{2}$ chain undergoes a transition into the saturated ferromagnetic phase. The obvious symmetry $h_c \rightarrow -h_c$ corresponds for the ferrimagnet to the symmetry around $h = h_0/2$. Such a symmetry was indeed observed in Ref. 9 for the behavior of the specific heat, though for the $h < h_0$ side it was somewhat plagued by the low-$T$ contribution of the acoustical magnon branch. In what follows, we compare our results only with the data for $h > h_0$. Because of cutting out non-nearest neighbor interactions, the critical fields for the model are slightly different from the real ones: $h_c = \pm h_{c,c}$ corresponds to $h = h'_{c,c} \simeq 1.81$ and $h = h'_{c,c} \simeq 3.09$ instead of the correct values listed above. Thus, for a comparison to the numerical data below, we have linearly rescaled the field interval $(h'_{c,c}, h''_{c,c})$ onto $(h_{c,c}, h_{c,c})$.

The specific heat. The temperature dependence of the specific heat for the spin-$\frac{1}{2}$ chain can be calculated using Klümper’s version of the thermodynamic Bethe ansatz; this amounts to solving numerically the following system of two coupled nonlinear integral equations:

$$y_1 = \frac{\pi \beta}{\gamma \cosh \frac{\pi \beta}{\gamma}} - \frac{\pi \beta h}{2(\pi - \gamma)} - \int_{-\infty}^{+\infty} dx \{ K(x - x') L(y_1) - K(x - x' - i\gamma + i\delta) L(y_2) \},$$

the other equation being of the same form with $i \rightarrow -i$, $y_1 \leftrightarrow y_2$, and $h \rightarrow -h$. Here $L(y) = \ln(1 + e^{-y})$, $\gamma = \cos \gamma$, $\delta$ is an infinitesimal positive number, $h = 4h_c/(J_{xy}\sin \gamma)$, $\beta = 2T/(J_{xy}\sin \gamma)$, and the kernel $K(x)$ is defined as follows:

$$K(x) = \frac{1}{4\pi} \int_{-\infty}^{+\infty} dq \sinh[(\pi/2 - \gamma)q] \cos qx \cosh(q\gamma/2) \sinh((\pi - \gamma)/q/2).$$

(Note that the last term in the integral contains a singularity!)

Then the specific heat $C(T)$ can be found as

$$C = \beta^2 \frac{\partial^2 f}{\partial \beta^2}, f = \frac{1}{2\gamma} \int_{-\infty}^{+\infty} dx \left[ L(y_1) + L(y_2) \cosh(\pi x/\gamma) \right].$$

In Fig. 2 we compare the results given by this integral equation with the results obtained within the present approach. The “pop-up” peaks observed near the critical field values are in the middle of the critical phase, is clarified: they can be identified with the contributions from two different spinon bands of the effective spin-$\frac{1}{2}$ chain. We believe these results should be accessible to experimental verification, and we would like to emphasize that this effect is general and should manifest itself in the critical regime of essentially any gapped 1D spin system in a magnetic field, as far as it is possible to construct the mapping to a spin-$\frac{1}{2}$ chain. Spin-$\frac{1}{2}$ ladder compounds like Cu$_2$(C$_5$H$_{12}$N$_2$)$_2$Cl$_4$ would be the natural candidates.

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O. Kahn, *Magnetism of the heteropolymetallic systems*, Structure and Bonding 68, 91 (Springer-Verlag, 1987); O. Kahn, Y. Pei, and Y. Journaux, in *Inorganic Materials*, edited by D. W. Bruce and D. O’Hare (Wiley, New York, 1995), p. 95.

A. K. Kolezhuk, H.-J. Mikeska, and Shoji Yamamoto, Phys. Rev. B 55, 8336 (1997).

S. Brehmer, H.-J. Mikeska, and Shoji Yamamoto, J. Phys.: Condens. Matter 9, 3201 (1997); S. K. Pati, S. Ramaseh, and D. Sen, Phys. Rev. B 55, 8894 (1997); J. Phys.: Condens. Matter 9, 8707 (1997).

M. Alcaraz and A. L. Malvezzi, J. Phys. A 30, 767 (1997).

S. Yamamoto and T. Fukui, Phys. Rev. B 57, R14008 (1998).

N. B. Ivanov, ibid. 14024 (1998).

S. Yamamoto, T. Fukui, K. Maisinger, and U. Schollwöck, Phys. Rev. B 55, 8894 (1997); S. K. Pati, S. Ramashasa, and D. Sen, Phys. Rev. B 55, 8894 (1997); J. Phys.: Condens. Matter 9, 8707 (1997).

M. Alcaraz and A. L. Malvezzi, J. Phys. A 30, 767 (1997).

S. Yamamoto and T. Fukui, Phys. Rev. B 57, R14008 (1998).

N. B. Ivanov, ibid. 14024 (1998).

S. Yamamoto, T. Fukui, K. Maisinger, and U. Schollwöck, J. Phys.: Condens. Matter 10, 11033 (1998).

M. Hagiwara, K. Minami, Y. Narumi, K. Tatani, and K. Kindo, J. Phys. Soc. Jpn. 67, 2209 (1998). A. Escuer, R. Vicente, M. S. Elfallah, M. A. S. Goher, and F. A. Maunter, Inorg. Chemistry 37, 4466 (1998); E. Belorizky, P. Rey, and D. Luneau, Molecular Phys. 94, 643 (1998).

K. Maisinger, U. Schollwöck, S. Brehmer, H.-J. Mikeska, and Shoji Yamamoto, Phys. Rev. B 58, 5908 (1998).

E. Lieb and D. Mattis, J. Math. Phys. 3, 749 (1962).

I. Affleck, Phys. Rev. B 41, 6697 (1990); A. Tsvelik, ibid. 42, 10499 (1990); M. Takahashi and T. Sakai, J. Phys. Soc. Jpn. 60, 760 (1991); Phys. Rev. B 43, 13383 (1991).

R. Chitra and T. Giamarchi, Phys. Rev. B 55, 5816 (1997).

F. Mila, cond-mat/9805029; T. Giamarchi and A. Tsvelik, cond-mat/9810219 (1998).

M. Fannes, B. Nachtergaele and R. F. Werner, Europhys. Lett. 10, 633 (1989); Commun. Math. Phys. 144, 443 (1992).

A. Klümper, A. Schadschneider and J. Zittartz, J. Phys. A 24, L955 (1991); Z. Phys. B 87, 281 (1992); Europhys. Lett. 24, 293 (1993).

No finite-size extrapolation was necessary due to a very short correlation length $\xi \approx 0.365$.

V. E. Korepin, N. M. Bogoliubov, and A. G. Izergin, *Quantum inverse scattering method and correlation functions*. (Cambridge University Press, 1993).

A. Klümper, Z. Phys. B 91, 507 (1993); Euro. Phys. J. B 5, 677, (1998); T. Nishino, J. Phys. Soc. Jpn. 64, L3598 (1995); R. J. Bursill, T. Xiang, and G. A. Gehring, J. Phys.: Condens. Matter 8, L583 (1996); N. Shibata, J. Phys. Soc. Jpn. 66, 2221 (1997); X. Wang and T. Xiang, Phys. Rev. B 56, 5061 (1997); K. Maisinger and U. Schollwöck, Phys. Rev. Lett. 81, 445 (1998).

G. Chaboussant, M.-H. Julien, Y. Fagot-Revurat, M. Hanson, L. P. Levy, C. Berthier, M. Horvatić, and O. Pi- ovesana, cond-mat/9811068, to appear in Euro. Phys. J. B.