Zero-temperature magnetic-field-induced phase transition between two ordered gapped phases in spin-ladders with ferromagnetic legs

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Abstract

We suggest that under an increase of magnetic field a spin-ladder with ferromagnetic legs does not pass without fail through an incommensurate phase but possibly in a straight way turns into a fully polarized ferromagnetic phase. The spin gap remains finite at the transition point. This scenario of zero-temperature first order phase transition is demonstrated for two special solvable spin-ladder models.

1 Introduction

Low-temperature magnetic phase transitions in spin-ladders were intensively studied in the last decade both theoretically [1]-[8] and experimentally [8]-[10]. In all these papers (even in the Ref. [6] devoted especially to magnetic behavior of spin-ladders with ferromagnetic legs) the theoretical scenario was quite identical. At zero magnetic field the ladder has a non-magnetic gapped ground state. An increase of magnetic field entails a decrease of the gap. At the critical value of the magnetic field $h_c$ the gap closes and the system turns into a gapless incommensurate magnetic phase. A further increase of the magnetic field from $h_c$ up to the saturation field $h_s$ entails a continuous change of
the gapless ground state and an increase of magnetization up to the maximum saturated value corresponding to the full polarization. Any further increase of the magnetic field does not change the vacuum entailing only an appearance and increase of a gap.

This picture is based on the following theoretical argumentation [1]-[8]. In zero magnetic field the ladder has the factorized singlet-rung ground state:

\[ |\text{vac}\rangle_0 = \prod_n |0\rangle_n, \]

where \( |0\rangle_n \) is the singlet state associated with \( n \)-th rung. All low-lying excitations originate from sparse rung-triplets in the rung-singlet sea. Induced by magnetic field Zeeman splitting entails the decrease of the gap with the rate proportional to the total spin of the state. The gaps corresponding to high-spin sectors decrease more rapidly, however the low-spin sectors have more narrow gaps. By the latter reason it was always assumed in [1]-[10] that the one-magnon gap closes faster than the multi-magnon ones. Under this assumption the critical value of magnetic field may be expressed from the one-magnon spin gap according to the formula,

\[ g\mu_B h_c = E_{\text{gap}}^{\text{magnon}}. \]

However as it will be shown below these arguments fail for the spin-ladders with ferromagnetic legs. Really let us first consider a spin-ladder with zero coupling along the legs or equivalently a set of isolated dimers. For this system \( h_c = h_s \). This means that at \( h = h_c \) the system turns into the fully polarized ferromagnetic ground state,

\[ |\text{vac}\rangle_s = \prod_n |1\rangle_n. \]

Here \( |1\rangle_n \) is the \( S = 1 \) triplet associated with \( n \)-th rung. An addition of a ferromagnetic coupling along legs decreases the energy of the fully polarized state and prompts it to reach the zero-energy level first. The system will turn into the state (3) escaping a commensurate-incommensurate transition and still remaining gapped at the transition point.

In other words ferromagnetic legs favour the magnon attraction and in this case an incommensurate phase is exotic. However this phase is favorable for antiferromagnetic legs with repulsion of magnons.

This general argumentation may be confirmed by exact results obtained for two exactly solvable models. The first one was studied in [5]. The corresponding Hamiltonian is the
following:

\[
\mathcal{H} = \sum_n J_\perp S_{1,n} S_{2,n} + J_\parallel (S_{1,n} S_{1,n+1} + S_{2,n} S_{2,n+1}) \\
+ 4(S_{1,n} S_{1,n+1})(S_{2,n} S_{2,n+1}) - g\mu_B h(S_{1,n} + S_{2,n}),
\]

(4)

where \( S_{i,n} (i = 1, 2; n = -\infty, \ldots, \infty) \) are spin-\( \frac{1}{2} \) operators associated with cites of the ladder.

As it was shown in [5] the corresponding values of critical and saturation magnetic fields are

\[
g\mu_B h_c = J_\perp - 4J_\parallel, \quad g\mu_B h_s = J_\perp + 4J_\parallel,
\]

(5)

It follows from (5) that \( J_\parallel < 0 \) entails \( h_s < h_c \), so the incommensurate phase does not appear.

Another solvable model of spin-ladder with cyclic (ring) exchange and exact singlet-rung ground state is considered in the next section.

2 Magnetic behavior of a spin-ladder with exact singlet-rung ground state

The model of spin-ladder with cyclic exchange and exact singlet-rung ground state in magnetic field \( h \) was first suggested in [11] and studied in more detail in [12]. The corresponding Hamiltonian \( \mathcal{H} \) has the following form:

\[
\mathcal{H} = \sum_{n=-\infty}^{\infty} H_{n,n+1},
\]

(6)

where \( H_{n,n+1} = H_{\text{stand}}^{n,n+1} + H_{\text{frust}}^{n,n+1} + H_{\text{cyc}}^{n,n+1} + H_{\text{Zeeman}}^{n,n+1} + J_{\text{norm}}, \) and

\[
H_{\text{stand}}^{n,n+1} = \frac{J_\perp}{2}(S_{1,n} \cdot S_{2,n} + S_{1,n+1} \cdot S_{2,n+1}) + J_\parallel (S_{1,n} \cdot S_{1,n+1} + S_{2,n} \cdot S_{2,n+1}),
\]

\[
H_{\text{frust}}^{n,n+1} = J_{\text{frust}}(S_{1,n} \cdot S_{2,n+1} + S_{2,n} \cdot S_{1,n+1}),
\]

\[
H_{\text{cyc}}^{n,n+1} = J_c((S_{1,n} \cdot S_{1,n+1})(S_{2,n} \cdot S_{2,n+1}) + (S_{1,n} \cdot S_{2,n})(S_{1,n+1} \cdot S_{2,n+1}) \\
- (S_{1,n} \cdot S_{2,n+1})(S_{2,n} \cdot S_{1,n+1})),
\]

\[
H_{\text{Zeeman}}^{n,n+1} = -g\mu_B (S_{1,n}^3 + S_{2,n}^3).
\]

(7)

The constant term \( J_{\text{norm}} \) is added only for normalization to zero the ground state energy at \( h = 0 \).
As it was shown in [11] the following restriction:

\[ J_{\text{frust}} = J_\parallel - \frac{1}{2} J_c, \]  

guarantees that the Hamiltonian (6),(7) commutes with the triplon-number operator \( Q = \frac{1}{2} \sum_n (S_{1,n} + S_{1,n+1})^2 \). In this case the Hilbert space separates on sectors corresponding to different eigenvalues of \( Q = 0, 1, 2, ... \). The sector \( Q = 0 \) is generated by the single vector (1). The additional restrictions:

\[ J_{\text{norm}} = \frac{3}{4} J_\perp - \frac{9}{16} J_c, \quad J_\perp > 2J_\parallel, \quad J_\perp + J_\parallel > \frac{3}{4} J_c, \]  

guarantee that the state (1) is the zero-energy ground state of the Hamiltonian (6),(7) separated by a finite gap from the other states.

The following formulas corresponding to zero magnetic field,

\[
\begin{align*}
H_{n,n+1}|0\rangle_n|1\rangle_{n+1}^{j} &= \left( \frac{1}{2} J_\perp - \frac{3}{4} J_c \right)|0\rangle_n|1\rangle_{n+1}^{j} + \frac{J_c}{2} |1\rangle_n|0\rangle_{n+1}, \\
H_{n,n+1}|1\rangle_n^j|0\rangle_{n+1} &= \left( \frac{1}{2} J_\perp - \frac{3}{4} J_c \right)|1\rangle_n^j|0\rangle_{n+1} + \frac{J_c}{2} |0\rangle_n|1\rangle_{n+1}^j, \\
H_{n,n+1}|1\rangle_n^1|1\rangle_{n+1}^j &= (J_\perp + J_\parallel - \frac{3}{4} J_c)|1\rangle_n^1|1\rangle_{n+1}^j, \\
H_{n,n+1}|1\rangle_n^0|1\rangle_{n+1}^1 &= (J_\perp - \frac{J_c}{2})|1\rangle_n^0|1\rangle_{n+1}^1 + (J_\parallel - \frac{J_c}{4})|1\rangle_n^0|1\rangle_{n+1}^1, \\
H_{n,n+1}|1\rangle_n^1|0\rangle_{n+1}^0 &= (J_\perp - \frac{J_c}{2})|1\rangle_n^1|0\rangle_{n+1}^0 + (J_\parallel - \frac{J_c}{4})|1\rangle_n^1|0\rangle_{n+1}^1,
\end{align*}
\]

are useful for calculation of magnon dispersions.

If we suppose that the system does not pass through the incommensurate phase then the energy gap between the states (1) and (3) will be closed at the saturation field \( h_s \). Then as it follows from (11)

\[ g\mu_B h_s = J_\perp + J_\parallel - \frac{3}{4} J_c. \]  

The simplest one-magnon excitation of the ground state (1) has the following "triplon" form:

\[ |k\rangle_{\text{trip}} = \sum_n e^{ikn} ... |0\rangle_{n-1}|1\rangle_n^j|0\rangle_{n+1}... \]. It has the following dispersion \( E_{\text{trip}}(k) = J_\perp - 3/2J_c + J_c \cos k \) [11]. Positivity of the one-triplon gap at \( h = h_s \) entails the following condition:

\[ E_{\text{trip}}(h_s) = -(J_\parallel + \frac{3}{4} J_c + |J_c|) > 0. \]  

From (14) follows that \( J_\parallel < 0 \) irrespective to \( J_c \).
Excitations near the state $|vac\rangle_s$ are also gapped at $h = h_s$. We shall calculate only the gaps corresponding to $\Delta S^z = -1$ sectors with minimal increase of magnetic energy. There are two types of these states. The first one ”singlon”

$|k\rangle_{\text{sing}} = \sum_n e^{ikn} ... |1\rangle_n |1\rangle_{n-1} |0\rangle_n |1\rangle_{n+1} ...$, originates from ”annihilations” of rung-triplets into the corresponding rung-singlets. Standard calculation based on (10),(11) gives the following dispersion law for these states:

$E_{\text{singl}}(k) = -J_\parallel - 3/4 J_c + J_c \cos k$. The corresponding energy gap coincides with (14).

Excitation of the second type ”ferromagnon”: $|k\rangle_{\text{fmagn}} = \sum_n e^{ikn} ... |1\rangle_n |1\rangle_{n-1} |0\rangle_n |1\rangle_{n+1} ...$, originates from rotations of single rung-triplets. Its dispersion $E_{\text{fmagn}}(k) = J_\perp - J_\parallel - J_c/4 + 2(J_\parallel - J_c/4) \cos k$ may be easily calculated from (11),(12) and corresponds to the following gap:

$E_{\text{gap}}(h_s) = J_\perp - J_\parallel - J_c/4 - 2|J_\parallel - J_c/4|$. (15)

It may be easily proved using (15) and (9) that $E_{\text{gap}}^{\text{sing}} < E_{\text{gap}}^{\text{fmagn}}$. This fact has a clear interpretation. In both the cases an increase of magnetic energy is equal. However singlons have an additional energy decrease originated from the triplet-singlet annihilation. On the other hand ferromagnons have an additional energy increase caused by the destruction of the ferromagnetic order. By this reason the energy gap corresponding to ferromagnons must be bigger than the one corresponding to singlons.

### 3 Magnetic behavior at various temperatures

It was argued that for spin-ladders with ferromagnetic legs an increase of external magnetic field inspires direct first order transition between two gapped phases: non-magnetic singlet-rung and ferromagnetic fully polarized. This transition occurs without passing throw a gapless incommensurate phase. In other words the transition interval between $h_c$ and $h_s$ turns to zero for these materials. Here we discuss some characteristic features of the corresponding experimental behavior.

Of course the step-like jump of magnetization will be effectively noticeable only for $T < E_{\text{gap}}(h_s)$. For $T > E_{\text{gap}}(h_s)$ contributions from excited states will smooth the magnetic curve getting it similar to the one corresponding to passing through an incommensurate phase. For these temperatures it will be difficult to distinguish a spin ladder with low $E_{\text{gap}}(h_s)$ from the one with low $h_s - h_c$.

Since the effective difference between magnetic curves for spin-ladders with weak ferromagnetic and antiferromagnetic legs disappears for $T > E_{\text{gap}}(h_s)$ it is necesssary to use
some other experimental approaches to study the nature of these compounds. Probably
a good criterion may be obtained from an independent measuring of the $E_{\text{gap}}$ (from mag-
netic susceptibility, neutron or Raman scattering data e.t.c.). If at low temperatures the
magnetic curve reduces to the step-like form as well as it was previously exactly deter-
mined that $E_{\text{gap}} > h_c$ then we have a right to content that a nonmagnetic gapped phase
turns directly into a gapped ferromagnetic. We suggest that this fact also indicates a
ferromagnetism along legs.

It was reported in [9] that the spin-ladder compound (5IAP)$_2$CuBr$_4$·2H$_2$O has a weak
antiferromagnetic interaction along legs ($J_{||}/J_{\perp} = 0.077$). Its magnetic behavior is very
similar to the one discussed above. Really the magnetization curve at $T = 0.4$ K has a
pronounced step-like form and almost vertical slope compared to the $T = 4.35$ K case.
This kind of behavior drastically differs from the one corresponding to another spin-ladder
compound MgV$_2$O$_5$ [8] where the slope of the magnetic curve does not change significantly
under a decrease of temperature.

Analysis of the temperature dependence of magnetic susceptibility for (5IAP)$_2$CuBr$_4$·
2H$_2$O reported in [9] gives $E_{\text{gap}} = h_c = 12.23$ K, however an analysis of the magnetization
curve gives $h_c = 11.90$ K. If we suppose that these results indicate a weak ferromagnet-
ism along legs then we have to conclude that at the transition point the system has an
extremely little gap $E_{\text{gap}}(h_s) = 0.33$ K. Of course this conjecture may be confirmed only
in a more precise experiment at $T < 0.33$K.

4 Conclusions

In this paper we have suggested that spin-ladders with ferromagnetic legs have different
magnetic behavior than the ones with antiferromagnetic legs. We have confirmed our
qualitative arguments by considerations of two exactly solvable models. This effect will
be really noticeable only at low temperatures ($T \ll E_{\text{gap}}(h_s)$). For rather big temperatures
the real difference of magnetization curves for spin-ladders with weak ferromagnetic or
antiferromagnetic legs must be negligible. We also have noticed that the spin-ladder
compound (5IAP)$_2$CuBr$_4$·2H$_2$O may be discussed in this context.

Spin-ladders with ferromagnetic legs have a special theoretical interest (see [6] and
references therein). However the characteristic type of their magnetic behavior suggested
in the present paper was not previously discussed.

Field-induced first order phase transitions in spin-ladders were also studied in [7].
However the corresponding phase diagram was different from the one suggested in the present paper. In non-magnetic and fully polarized phases are separated by the phase with half polarization.

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