On the Violation of Marshall-Peierls Sign Rule in the Frustrated $J_1$-$J_2$ Heisenberg Antiferromagnet.

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Abstract. – We present a number of arguments in favour of the suggestion that the Marshall-Peierls sign rule survives the frustration in the square-lattice Heisenberg antiferromagnet with frustrating next-nearest-neighbour (diagonal) bonds ($J_1$-$J_2$ model) for relatively large values of the parameter $J_2/J_1$. Both the spin-wave analysis and the exact-diagonalization data concerning the weight of Marshall states support the above suggestion.

Many years ago Marshall proved, using a Lemma due to Peierls, the well-known theorem determining the amplitude phases of the set of Ising states building the ground-state wave function of the spin-$\langle 1/2 \rangle$ Heisenberg antiferromagnet on bipartite lattices[1]. Later on, Lieb and Mattis generalized the theorem to arbitrary site spins and bipartite lattices without translational symmetries[2,3].

Assigned to the $J_1$-$J_2$ model

$$H = H_1 + \alpha H_2 \equiv \sum_{nn} S_i S_j + \alpha \sum_{nnn} S_i S_j, \quad J_1 \equiv 1, \quad \alpha \equiv J_2/J_1 = J_2,$$

(nn and nnn mean that the summations run over the nearest-neighbour and next-nearest-neighbour, diagonal, bonds on the square lattice, respectively), the theorem, applicable to the case $\alpha \leq 0$, says:

i) The lowest eigenstate of the Hamiltonian (1) in each subspace determined by the eigenvalue $M$ of the spin operator $S_{\text{total}}^z$ reads

$$\psi^M = \sum_m c_m^{(M)} |m\rangle, \quad c_m^{(M)} > 0.$$
Here the Ising states $|m\rangle$ are defined by

$$|m\rangle \equiv (-1)^{s_{\lambda}-M_{\lambda}} |m_1\rangle \otimes |m_2\rangle \otimes \ldots \otimes |m_N\rangle,$$

(3)

where $|m_i\rangle$, $i = 1, \ldots, N$, are the eigenstates of the site spin operator $S_i (-s_i \leq m_i \leq s_i)$, $S_A = \sum_{i \in A} s_i$, $M_{A(B)} = \sum_{i \in A(B)} m_i$, $M = M_A + M_B$. The lattice consists of two equivalent sublattices $A$ and $B$. $s_i \equiv s$, $i = 1, \ldots, N$, are the site spins. The summations in eq. (2) are restricted by the condition $\sum_{i=1}^{N} m_i = M$, $N$ being the number of sites.

A direct result of the property $c^{(M)}_m > 0$, valid for each $m$ from the basis set (3), is the non-degeneracy of $\Psi_M$, since it is impossible to build up other orthonormal states without using negative amplitudes $c^{(M)}_m$.

ii) The total spin of $\Psi_M$ is $S = |M|$. Since each $M$ subspace contains only eigenstates with $S \geq |M|$, a result of ii) is the relation

$$E(S) < E(S + 1), \quad S \geq 0,$$

(4)

with $E(S)$ being the lowest energy eigenvalue belonging to $S$. It is worth noticing that the property ii) is characteristic for a larger set of Hamiltonians (1), because the relation i) $\Rightarrow$ ii) can always be proved.

In what follows we address the model (1) with frustrating diagonal bonds, i.e. $\alpha > 0$. According to Lieb-Mattis definition [3], if $\alpha > 0$, the square lattice is not bipartite because it is not possible to find a positive constant $c > 0$ such that the conditions $J_{AA}, J_{BB} \leq c, J_{AB} \geq c$ are satisfied for all bonds of the square lattice. The goal of the present letter is to give arguments in favour of the suggestion that the Marshall-Peierls sign rule i) can survive the frustration for relatively large positive parameters $\alpha$.

For references, let us firstly recall the important steps leading to the sign rule i) [3]. We suppose that the ground state of (1) in the subspace $M$ (with an eigenvalue $E_M$) is written in the form (2). The Schrödinger equation for the amplitudes $c^{(M)}_m$ reads

$$\sum_n A_{nm} c^{(M)}_n = (\varepsilon_m - E_M) c^{(M)}_m,$$

(5)

where

$$A_{nm} = B_{nm} - \alpha C_{nm} \equiv -\langle n | H^{xy} | m \rangle - \alpha \langle n | H^{xy} | m \rangle, \quad B_{nm}, C_{nm} \geq 0.$$  

(6)

$\varepsilon_m$ ($\varepsilon_m > E_M$) is the eigenvalue of $H^s$ in the Ising state $|m\rangle$.

Further, notice that the variational function $\Phi_M = \sum_m |c^{(M)}_m | |m\rangle$, producing the Schrödinger equation

$$\sum_n A_{nm} |c^{(M)}_n | = (\varepsilon_m - E_M) |c^{(M)}_m |,$$

(7)

should also have an eigenvalue $E_M$. Let us suppose that the matrix elements $A_{nm}$ satisfy

$$A_{nm} = B_{nm} - \alpha C_{nm} \geq 0.$$  

(8)

Then eqs. (5),(7) lead to

$$| \sum_n A_{nm} c^{(M)}_n | = \sum_n | A_{nm} | |c^{(M)}_n |,$$

(9)

which is possible if, and only if, $c^{(M)}_m \geq 0$. Finally, the chance of some $c^{(M)}_m$ to vanish is
excluded, for if it were so, eqs. (5), (9) would imply that \( c_m^{(M)} = 0 \) for all \( m \). These are the main steps leading to the sign rule i).

Now, notice that for \( \alpha > 0 \) the above reasoning is not applicable, since inequality (8) will be violated for some matrix elements \( A_{nm} \). Nevertheless, for small enough frustration parameters \( \alpha > 0 \), one can argue that the Marshall-Peierls sign rule survives the frustration for every finite \( N \). Indeed, on rather general variational grounds the amplitudes \( c_m^{(M)}(\alpha = 0^+) \leq 0 \). Then in the limit \( \alpha \downarrow 0^+ \), (8) will imply

\[
\lim_{\alpha \downarrow 0^+} A_{nm} = B_{nm} \geq 0 ,
\]

showing that inequality (8) is satisfied at \( \alpha = 0^+ \). Thus, the same reasoning as in the case \( \alpha \leq 0 \) implies that the above assumption is wrong, i.e. all the amplitudes should be positive, \( c_m^{(M)}(\alpha = 0^+) > 0 \). Finally, since all the possible Ising configurations take part in the ground state at \( \alpha = 0^+ \) with finite positive amplitudes, it is clear that the same property conserves in some region of positive \( \alpha \), as well. Notice that in the limit \( N = \infty \) such a consideration does not ensure the survival of the discussed region. In principle, one could have different qualitative behaviour in \( D = 1 \) and \( D = 2 \).

**Spin-wave analysis.** – The self-consistent spin-wave approach used below[4,5] is predominantly addressed to low-dimensional spin systems. The conventional spin-wave technique is supplemented with a condition for zero sublattice magnetization, thus fulfilling, by hand, Mermin-Wagner’s theorem[6] at finite temperatures, or the same requirement on finite lattices. The \( J_1 - J_2 \) model has also been analysed by this method in a number of recent papers (for a review, see, e.g., ref.[7] and references therein). Here we omit the details and directly present the spin-wave ansatz due to the theory mentioned above

\[
\psi_{sw} \sim \exp \left[ \sum_k w_k \hat{a}_k^+ \hat{\delta}_k^+ \right] |\text{Néel}\rangle .
\] (11)

Here \( |\text{Néel}\rangle \) is the classical Néel state. The weight factors \( w_k \) are defined by \( w_k = v_k / u_k \), \( v_k \) and \( u_k \) being the well-known Bogoliubov coefficients

\[
2v_k^2 = (1 - \gamma_k^2)^{-1/2} - 1 , \quad 2u_k^2 = (1 - \gamma_k^2)^{-1/2} + 1 ,
\] (12)

\[
\gamma_k = \frac{\Gamma_k}{1 + \mu - \alpha U(1 - \Gamma_k)} , \quad \gamma_k = \frac{1}{2} (\cos k_x + \cos k_y) , \quad \Gamma_k = \cos k_x \cos k_y .
\] (13)

The Bose operators \( \hat{a}_k^+ , \hat{\delta}_k^+ \) come from the Dyson-Maleev transformation and live on \( A \) and \( B \) sublattices, respectively. The prime over sums means that \( k \) vectors run in the small Brillouin zone. \( \mu \) is the Lagrange multiplier used to imply the condition for zero sublattice magnetization. The renormalization factor \( U \), renormalizing the frustration parameter \( \alpha \) in the last equation, is a result of the Hartree-Fock decoupling corresponding to the theory (for details, see ref.[8-10]). Within the theory, \( U \) is a ratio of two short-range correlators determined by the self-consistent equations. It has been shown in a recent study[10] that the spin-wave ansatz (11) gives an excellent fit to the exact-diagonalization data for the relevant quantities of the model (1) up to \( \alpha \approx 0.45 \), provided the quasi-classical limit (large site spin
\( s_i \equiv s \) for the factor \( U \) (see, e.g., ref.[11,12])

\[
U = \frac{1 - 0.102/2s + O((2s)^{-2})}{1 + 0.158/2s + O((2s)^{-2})},
\]

(14)
is used. \( U = 1 \) corresponds to the linear spin-wave approximation[13].

Now, let us rewrite (11) in the form

\[
\varphi_{sw} \sim \exp \left[ - \sum_{R, r} w(r) \delta_{R, R+r} \right] |\text{Néel}>, \quad w(r) = \frac{2}{N} \sum_k w_k \cos kr,
\]

(15)

where \( w(r) \) is a pairing function. The vector \( r \) connects sites from different sublattices. From the structure of the latter state (15), it is clear that the sign rule i) breaks if, and only if, the pairing function \( w(r) \) changes its sign for some vector \( r \) connecting two spins living on different sublattices. For the \( 4 \times 4 \) lattice this is just the vector \( r = x + 2y \) (and the vectors related by the lattice symmetry). A numerical calculation demonstrates that \( w(x + 2y) \) changes sign at a point practically coinciding with the related \( N = \infty \) case \( (a_M = 0.323 \text{ for } U = 1) \). In the thermodynamic limit \( N = \infty \), we have checked numerically that it is just the function \( w(x + 2y) \) which firstly becomes negative when \( x \) increases. Unfortunately, we have not succeeded in finding any analytical proof for this observation. Finally, it is interesting to notice that the curve representing the exact weight of Marshall states vs \( x \) for the \( 4 \times 4 \) lattice is surprisingly well reproduced from the finite-size spin-wave analysis based on the ansatz (15)[10,14].

**Exact-diagonalization data.** – Here we present exact-diagonalization results for the weight of non-Marshall states (which do not follow the sign rule i)) in the exact ground-state wave function of the \( J_1-J_2 \) model, eq. (1), on \( 4 \times 4 \) and \( 6 \times 4 \) lattices in the interval \( 0 \leq x \leq 0.52 \), fig. 1 (a detailed and varied analysis of that matter for the \( S = 0 \) state is presented in ref.[14]). Here we study the ground-state wave functions corresponding to total-spin quantum numbers \( S = 0, 1, 2 \) and 3. These states are non-degenerate in the whole region presented in fig. 1. We found a relatively wide region where the non-Marshall states are lacking at all (for each of the studied states). For example, the exact upper bound \( a_M \) in the case \( N = 4 \times 4, S = 0 \) is \( a_M = 0.28 \) (this number seems to be slightly size-dependent as seen in fig. 1, \( 6 \times 4 \) lattice). It is worth noticing that even in the strongly frustrated region, \( x \approx 0.52 \), the weight of the Ising states violating the rule i) is very small as compared to the weight of states which fulfil the rule. Further, a well-pronounced tendency is that in the states with larger spin \( S \) the weight of non-Marshall states is smaller. In addition, a more detailed study also shows[14] that the exclusion of the non-Marshall states from the exact ground states does not have any drastic effect on the spin-spin correlators and the other relevant quantities characterizing the model (1). It is interesting to compare the above picture to the one produced by the \( J_1-J_2 \) antiferromagnetic chain, fig. 2. As is clearly seen, the behaviour is quite different: first, the range where the sign rule is exactly fulfilled is considerably reduced. Second, for states with larger spins the weight of non-Marshall states is larger, a tendency which is just the opposite to the one seen in the two-dimensional case. Precisely at \( x = 0.5 \) the exact ground state is known to be the twofold degenerate spin-Peierls dimer state.

**Concluding remarks.** – In conclusion, a number of arguments are given in favour of the suggestion that in the \( J_1-J_2 \) model the Marshall-Peierls sign rule survives the frustration in a
Fig. 1. – The weight of the non-Marshall Ising states \( \left( \sum_{i \in \mathbb{Z}^2} [c_i(M)]^2, M = S \right) \) in the lowest-energy normalized eigenstates with total spins: \( \square S = 0 \ (N = 16); \ \bigcirc S = 0 \ (N = 24); \ \triangle S = 1 \ (N = 16); \ \bigcirc S = 2 \ (N = 16); \ \star S = 2 \ (N = 16) \) vs. the frustration parameter \( J_2/J_1 \) for the \( s = 1/2 \) square-lattice Heisenberg antiferromagnet with frustrating diagonal bonds for \( 4 \times 4 \) and \( 6 \times 4 \) lattices.

Fig. 2. – The weight of the non-Marshall Ising states (see the caption of fig. 1) in the lowest-energy normalized eigenstates with total spins \( S = 0, 1, 2 \) and \( 3 \) vs. the frustration parameter \( J_2/J_1 \) for the \( s = 1/2 \) Heisenberg antiferromagnetic chain with frustrating next-nearest-neighbour bonds for \( N = 16 \) and \( N = 24 \). (Notations are the same as in fig. 1.)

wide region up to the limit \( \alpha_M \). Evidently, at this point some of the Ising amplitudes in the ground state should vanish and further become negative. This new quality of the ground state, however, does not sufficiently influence the quantities characterizing the long-range order in the system, which is rather natural in view of the picture drawn above. It is worth noticing that the number of non-Marshall states just after the Marshall point \( \alpha_M \), seen in the exact results, is very small, which is in agreement with the spin-wave picture showing that the violation of the sign rule starts from Ising states with a small number of reversed (with respect to the classical Néel state) sublattice spins. Further, the weight of Ising states violating the rule i) remains extremely small up to the strong frustration limit \( \alpha = 0.5 \). This fact is relevant for construction of variational wave functions in the frustrated region.

It is also clearly seen that the ground states with larger spins \( S \) are, in a sense, less destroyed exactly in that case (two-dimensional \( J_1-J_2 \) model) when the ground state is, most probably, macroscopically Néel ordered. In addition, it is interesting to notice that the sharp increase in the weight of non-Marshall Ising states with larger spins, fig. 1, is observed close to the same point \( \alpha = 0.5 \) where, according to the renormalized spin-wave ansatz (14), (15) (see ref. [10]), the two-sublattice long-range Néel order breaks down. Therefore, there are some indications in favour of the suggestion that the phenomenon discussed in the present letter may, in principle, be effectively used in the finite-size analysis concerning the mechanism of breaking of the global rotational symmetry of the Heisenberg Hamiltonians [15, 16]. Clearly, further work in this direction is needed. For such studies, the frustrated \( J_1-J_2 \) Heisenberg antiferromagnet seems to be quite an adequate model, for the whole process of destroying of the Néel order can be suitably studied by changing the continuous frustration parameter \( J_2/J_1 \).

\[ \sum_{i \in \mathbb{Z}^2} [c_i(M)]^2, M = S \]
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