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Universal sequence of ground states validating the classification of frustration in antiferromagnetic rings with a single bond defect

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The sequence of $2s + 1$ ground states in the frustrated antiferromagnetic rings with odd number n of local spins s resulting from a single bond defect strength is determined, and occurrence of the Lieb-Mattis energy level ordering, proven for the bipartite systems only, is confirmed for the rings in question. The sequence is universal, provides the theoretical basis for the recent classification of spin frustration in molecular magnets, and indicates that the Lieb-Mattis theorem (LMT) consequences exist for the nonbipartite rings. The states in the sequence are characterized by the total spin S fulfilling the constraint $S \leqslant s$ and are separated by $2s$ Kahn degenerate frustration points. The possible LMT consequences in other systems are discussed.

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Frustration in magnetic systems is a subject of intensive research which has been reinforced in the framework of molecular magnetism, providing excellent examples of quantum spin systems described by the Heisenberg model [1–6]. A class of chromium-based complexes with the nearest-neighbor antiferromagnetic interactions between magnetic ions with spin $s = 3/2$ is particularly interesting and one of the most studied [7–11]. The approaches, however, have been used in a fragmented way focused on solving a given problem, e.g., frustration effects in the Cr_9 rings [8,9] or analysis of the frustration signatures which leads to qualitative conclusions, e.g., reduction of the total or local magnetization [1,9]. A more fundamental approach which clarifies the piecemeal results and predicts new properties is still missing.

In classical systems frustration is directly related to the bond configuration. A given system is frustrated if it contains some loops with an odd number of antiferromagnetic bonds. This geometric property implies that in such a layout the ground state (GS) energy is higher than the sum of energies of all the saturated bonds [12–15].

In quantum spin systems, the GS energy [16–18] is also higher than the sum of the GS energies for the constituent geometrically satisfied bonds due to *quantum frustration* originating from the entanglement [19–21]. Here we consider the *geometric frustration* which exists in systems with competing interactions and is an analog of the classical frustration. We analyze its effects in relation to bipartiteness [1,3] defined by Lieb and Mattis (LM) [22].

Following LM, a finite system of quantum spins $\{\boldsymbol{s}_j\}$ coupled by bilinear interactions $J_{jk}\boldsymbol{s}_j \cdot \boldsymbol{s}_k$ is bipartite if it can be decomposed into two separable parts (or sublattices A and B) and there exists a number $g^2 \geqslant 0$ being simultaneously the lower bound of the exchange integrals J_{jk} for spins from different subsystems and the upper bound of couplings J_{jk} for spins in the same sublattice. According to this definition, e.g., a lattice system is bipartite with $g^2 = 0$ if all the exchange couplings connect two sites residing on different subgroups only and they are antiferromagnetic.

In the nonuniform chromium-based $s = 3/2$ rings Cr_9 which are frustrated, the $S = s$ GS's typical for the unfrustrated rings were predicted [9,23] and observed [8]. To account for this unexpected result, a new classification of spin frustration was proposed [8], using the classical concepts. The first type was attributed to the Kahn frustration [1,24] which occurs if the GS is a superposition of degenerated states. At least one of them should be described by a nonzero spin quantum number. For the Cr_9 molecule this condition is fulfilled if all the couplings are equal. Then there is a pair of degenerate $S = 1/2$ GS's. The second type of frustration is realized in the domain of bond defects yielding the GS with $S = 1/2 < s$ [8]. The third type is characterized by the $S = 3/2 = s$ GS which was viewed as a relic of the GS of the corresponding nonfrustrated ring.

We bring the theoretical grounds for this classification, considering a variety of the odd-numbered antiferromagnetic rings. We predict for them the universal sequence of GS's in terms of the total spin S, calculate the coordinates of their boundaries which coincide with the Kahn degenerate frustration points, and determine the domains assigned to the second and third types of frustration. The sequence explains qualitatively some experimental findings for a number of rings. We check that the Lieb-Mattis level ordering (LMLO) [25], proven for the bipartite systems [22], is fulfilled for any bond defect strength [i.e., $E(S') < E(S' + 1)$ for $S' \geqslant S$, where $E(S')$ and $E(S' + 1)$ are the lowest energies belonging to the levels with the total spins S' and $S' + 1$, respectively]. Our conclusions arise from the Lieb-Mattis theorem (LMT) [22,26] and algebraic calculations for the bipartite systems with the size $n = 3$. Their validity for the nonbipartite systems with the size $n > 3$ is confirmed by precise numerical calculations within the resources available.

The isosceles triangles (Fig. 1) represent the most simple models of the rings considered here. Assigning the spin $\boldsymbol{s}_2$ to one subsystem and the remaining two spins to the other one [Fig. 1(a)], we can choose $g^2 = 0$ for $J_{31} = \alpha \leqslant 0$ to validate the bipartiteness. When $0 < \alpha \leqslant 1$ [Fig. 1(b)], the decomposition into subsystems is the same as in Fig. 1(a), but now the condition for bipartiteness is satisfied provided that $\alpha \leqslant g^2 \leqslant 1$. The system is bipartite also for $\alpha > 1$, assigning the spins $\boldsymbol{s}_1$ and $\boldsymbol{s}_3$ linked by the defected bond

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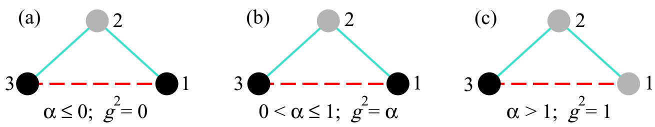

FIG. 1. (Color online) Bipartiteness of a quantum triangle ($J_{12} = J_{23} = 1$, $J_{31} = \alpha$) with possible choices of g^2. Spins denoted by gray and black circles form different subsystems.

to *different subsystems* and putting constant $g^2 = 1$ [see Fig. 1(c)]. Therefore, the system represented by the isosceles triangle can be *frustrated* and *bipartite* with $g^2 > 0$. We note that this compelling statement is also valid for squares with the competing diagonal interactions [27].

Graph theory implies [28] that the geometrical frustration *is excluded* in bipartite systems with $g^2 = 0$ but not in those with $g^2 > 0$. The statement does not support the suggestion that frustration is the opposite of bipartiteness propounded on the unreliable basis $g^2 = 0$ [1].

It is crucial to distinguish implications of the value $g^2 = 0$ and $g^2 > 0$ for a bipartite system. For $g^2 > 0$ the outcome of LMT [22,26] determines the upper bound $\mathcal{S} = |S_A - S_B|$ for the total spin S of GS (i.e., the allowed spin value $S \leqslant \mathcal{S}$), whereas for $g^2 = 0$ it implies the value $S = \mathcal{S}$, where $S_{A(B)} = \sum_{j\in A(B)} s_j$.

As a consequence, LMT provides the strong constraint on the possible GS spin values. For the triangles in Fig. 1 with arbitrary spin s, the value $\mathcal{S} = s$ and the LMT predicts the unique GS with $S = s$ if $\alpha < 0$, otherwise the GS spin $S \leqslant s$. The latter stands for an upper bound for S and proves that the value $S = s$ attributed to the third type of frustration is fully *allowed* in the GS of the frustrated triangle.

The rings in question are described by the isotropic Heisenberg Hamiltonian

$$\mathcal{H} = \sum_{j=1}^{n-1} \boldsymbol{s}_j \cdot \boldsymbol{s}_{j+1} + \alpha \boldsymbol{s}_n \cdot \boldsymbol{s}_1, \tag{1}$$

where the bond defect α is arbitrary and the size $n \geqslant 3$, so that the total spin S and the total spin component M are good quantum numbers. As the model (1) is symmetric with respect to the reflection ρ in the plane perpendicular to the defected bond, the eigenstates $|S\,r\rangle$ are classified by the additional quantum number $r = \pm 1$ ($\rho|S\,r\rangle = r|S\,r\rangle$) which distinguishes the *symmetric* and the *antisymmetric* states, respectively. If $\alpha \leqslant 0$, the model (1) is bipartite with $g^2 = 0$. In this case LMT predicts unambiguously GS $|S = s, r = +1\rangle$, irrespective of the size n of the ring and the local spin value s.

In the region $\alpha > 0$, only the spin system (1) with $n = 3$ is bipartite. The value of the total spin S in the GS is then subject to the LMT constraint $S \leqslant \mathcal{S} = s$, whereas the value r remains not determined. However, in this case the quantum numbers S and r can be found from direct algebraic calculations based on the sublattice model [29–31]. Their details are presented in the Supplemental Material [27] together with plots of the low-lying energy levels as a function of α for $s \leqslant 3/2$. When $\alpha > 0$ increases monotonically the *regular sequence* of GS's $|S\,r\rangle$ occurs. It starts from the total spin $S = s$ and $r = 1$, i.e., the state with ferromagnetically ordered dimer $\boldsymbol{\sigma} = \boldsymbol{s}_1 + \boldsymbol{s}_3$.

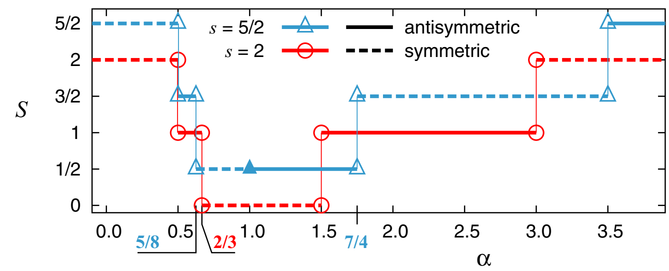

FIG. 2. (Color online) The total spin S in GS as a function of α for $n = 3$ in the cases $s = 2$ and $s = 5/2$. The symmetry of the state $S = 1/2$ is changed for $\alpha = 1$ which is marked by a full triangle.

For larger values of $\alpha > 0$ the dimer total spin σ decreases, whereas the total spin is given as $S = |s - \sigma|$, so the following sequence is produced:

$$S = s, s-1, \ldots, \left\{ \begin{array}{c} 2,1,0,1,2 \\ 3/2,1/2,1/2,3/2 \end{array} \right\}, \ldots, s-1, s, \tag{2}$$

where the upper (lower) row corresponds to the integer (half-integer) spin number s, respectively. Since the quantum number r alternates, the final GS in this sequence is $|S = s, r = (-1)^{2s}\rangle$.

Hence, there are $2s + 1$ different GS's separated by $2s$ Kahn frustration points. Interestingly, the spectrum of the GS total spins covers the full range of the allowed values arising from LMT, and the LMLO is strictly fulfilled for all α. The sequences occurring for $s = 2$ and $s = 5/2$ are illustrated in Fig. 2, whereas those for $s < 2$ are included in the Supplemental Material [27].

We have performed vast numerically exact diagonalizations of the model (1) in the domain $\alpha > 0$ and considered a number of system sizes $3 < n \leqslant 13$ as well as the spin values $1/2 \leqslant s \leqslant 7$. From these results we have confirmed both the universality of the sequence obtained and the tight preservation of the LMLO. Although for $n > 3$ and $\alpha > 0$ the rings are nonbipartite, nevertheless their sequences of GS's coincide with that given by Eq. (2), and the LMLO survives too. We can state that the rings inherit the features of their prototype bipartite triangle. The sequences and the LMLO property are illustrated in Fig. 3 for $n = 5$, $s = 3/2$, and $\alpha \leqslant 2$.

Referring to the classification of frustration [8], we can generalize its applicability to arbitrary rings in question. In the frustrated region $\alpha > 0$, there exist domains with the GS $S = s$ which is characteristic for the nonfrustrated rings. In these domains of interactions, the rings are subject to the third

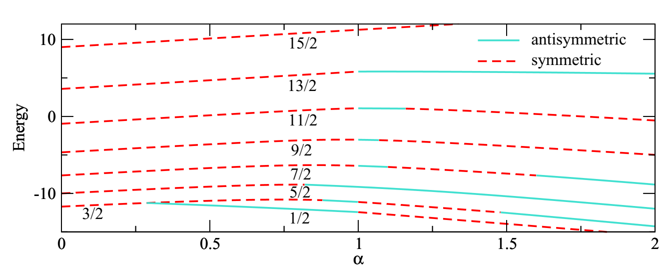

FIG. 3. (Color online) The sequence of the ground states and the Lieb-Mattis level ordering in the spin $s = 3/2$ pentagon for $\alpha \leqslant 2$. The irrelevant energy levels are omitted for clarity.

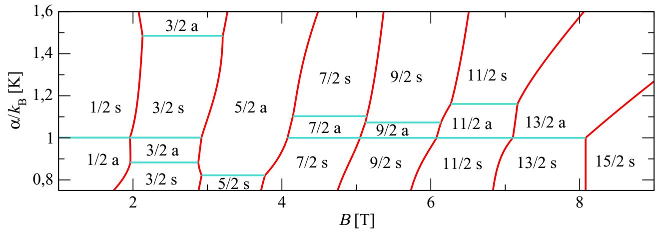

FIG. 4. (Color online) The ground state energy diagram in the presence of field for the spin $s = 3/2$ pentagon illustrating the LMLO and the size of the magnetization steps for $J/k_B = 1$ K and $g = 1$ in the corresponding Zeeman term. The domains are characterized by the total z component M of spin and the symmetry index r denoted by s or a. For a fixed α, the quantum number M increases systematically by 1.

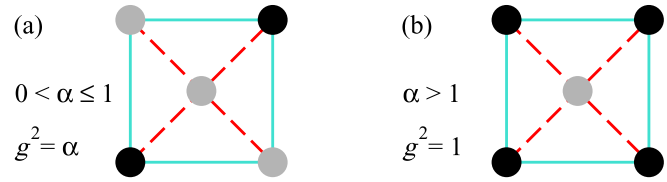

FIG. 5. (Color online) A bipartite centered square with fixed ($J = 1$) antiferromagnetic interactions between the neighboring peripheral spins and their couplings with the central spin parametrized by α. Partitioning of the system for $0 < \alpha \leqslant 1$ and $\alpha > 1$ is shown in panels (a) and (b), respectively.

type of frustration. In the remaining domains, the GS spin $S < s$ which is a signature of the second type of frustration.

The coordinates of the corresponding level crossings $\alpha_c^{(j)}$ $(1 \leqslant j \leqslant 2s)$ define the boundaries of a given type of GS and are exemplified in the Supplemental Material [27]. The value $\alpha_c^{(1)}$ terminates the domain of the third type of frustration characterized by GS $|s,+\rangle$ which starts from $\alpha = 0$. This type of frustration occurs also for $\alpha > \alpha_c^{(2s)}$ with GS $|s,(-1)^{2s}\rangle$. For $s = 1/2$ the third type of frustration is spread over the entire domain $\alpha > 0$ irrespective of the ring size n, excluding $\alpha_c^{(1)} = 1$ which is the single Kahn frustration point. The domain of the second type of frustration with the GS spin $S < s$ exists for $s > 1/2$ and $\alpha_c^{(1)} < \alpha < \alpha_c^{(2s)}$. It is split into $2s - 1$ intervals with different GS's determined by the sequences (2) and separated by the Kahn degenerate frustration points.

The LMLO property leads to a diagram showing areas of the GS's with a fixed value of M in the presence of a magnetic field. For a pentagon with $n = 5$ and $s = 3/2$ in Eq. (1), the diagram is shown in Fig. 4. For any α, the value M in the GS is enhanced only by 1 with increasing field, implying the in-field magnetization stairs with the magnetization steps or plateaus changing according to the data of the corresponding GS diagram.

For half-integer spins the point $\alpha_c^{(s+1/2)} = 1$ locates the level crossing of the states $|1/2\pm\rangle$, so that the anticipated fourfold degeneracy is recovered [32]. In particular, for α slightly higher than 1 in the spin $s = 1/2$ rings the energy levels of GS's $|1/2\pm\rangle$ should be close, which explains qualitatively a very small gap between the two Kramers' doublets observed in the heptanuclear vanadium ring [5]. For integer spins, the coordinates $\alpha_c^{(s)} < 1$ and $\alpha_c^{(s+1)} > 1$ encompass the value $\alpha = 1$, constitute the limits for the $S = 0$ GS domain, and imply the nondegenerate GS for $\alpha = 1$, as expected [24,32].

For the bipartite systems with $g^2 = 0$, the Marshall sign rule [25] should be satisfied, and this is true for the rings with the defect bond $\alpha < 0$. However, the rule is not systematically obeyed beyond this region of α. It is violated even for the triangle in the region $\alpha_c^{(1)} < \alpha < \alpha_c^{(2s)}$, where the system is bipartite with $g^2 > 0$.

The condition of bipartiteness $J_{jk} \geqslant g^2 > 0$ is satisfied when *all* the spins from different subsystems are coupled antiferromagnetically. This may occur for small systems, for systems with a special topology, like the centered rings [4,33] (see Fig. 5), or for those with the properly tuned long-range RKKY interactions. The question arises if the sequence of GS's and existence of the LMLO are particular properties of the rings modeled by the Hamiltonian (1) or they can also be observed in other systems.

The prototype of the odd-numbered cluster of the centered rings is illustrated in Fig. 5. The interactions between the neighboring spins on the outer ring are fixed and equal 1, whereas those between the central spin and peripheral ones equal α. Choosing properly the sublattices (Fig. 5), the frustrated cluster is bipartite for $\alpha \geqslant 0$, and we deduce from LMT that the ground state spin S amounts to $S = s$, $S \leqslant s$, and $S \leqslant 3s$ for $\alpha = 0$, $0 < \alpha \leqslant 1$, and $\alpha > 1$, respectively.

The corresponding centered hexagon is bipartite for $\alpha = 0$ and $\alpha \geqslant 1$ only, implying the LMT constraints $S = s$ and $S \leqslant 5s$, respectively. Nevertheless for the $n = 7$ and $s = 5/2$ iron ring (Fig. 8 in Ref. [4]) the subsequence of the GS spins values $S = 5/2, 3/2, 1/2$ was found for the domain $0 \leqslant \alpha \leqslant 1$, which is in perfect agreement with the constraint $S \leqslant s$ predicted here for a *bipartite* centered square. Moreover, the $n = 7$ and $S = 5/2$ analog of the staircase shown in Fig. 2 was obtained [4]. Having that in mind, we guess that a universal sequence of the GS's as well as the LMLO might also be realized for the odd-numbered centered rings.

A bipartite square representing the frustrated even-numbered cluster on the square lattice is shown in Fig. S1 [27]. For this square the number $\mathcal{S} = 0$ which implies the unique GS $|S = 0\rangle$. This trivial $S = 0$ sequence was determined also for a number of nonbipartite clusters with interacting spins $s = 1/2$ [34]. We anticipate that for any even-numbered cluster the same GS $S = 0$ and the LMLO can be established.

The simplest prototypes of the antiferromagnetic systems with an impurity spin and nonuniform couplings such as Cr_8Ni [35–37] are bipartite. For the triangles illustrated in Figs. 1(b) and 1(c), where $s_1 = s_3 = 3/2$, $s_2 = 1$ (corresponding to the Ni ion) and the bonds are rescaled by a factor $\alpha' = 1/\alpha$, the sequence of the GS's in terms of increasing α' is given by expression (2), where the left and the right bound are given by $S = 1$ and $S = 2$, respectively. The noteworthy subsequence $S = 1,0$ of GS's was determined in a nonbipartite model of the Cr_8Ni molecule [10,36] for $0 \leqslant \alpha' \leqslant 1$. In addition, an insight into the energy spectrum confirms the LMLO. This partial result may suggest that the constraint for the GS spin $0 \leqslant S \leqslant 2$ exists for this type of molecular rings. A similar reasoning can be provided for the frustrated uniform even-numbered rings with the antiferromagnetic next-nearest-neighbor couplings (see the prototype ring in Fig. S1 of

Ref. [27]) or for nanomagnets built from magnetic atoms adsorbed on a nonmagnetic surface and coupled by RKKY exchange [38,39].

In conclusion, we have established the universal sequence of the GS's for antiferromagnetic rings with the odd number of the local spins s and a single bond defect α described by the model (1). The sequence is characterized by the total spin $S \leqslant s$ and contains all the numbers belonging to the interval allowed. For $S' \geqslant S$, the LMLO $E(S'+1) > E(S')$ is valid, where $E(S')$ is the lowest energy of the states described by the quantum number S'. The sequence validates the classification of frustration in this type of nanomagnets. Our calculations have revealed the unexpected features of the model in question: The rings with enlarged nonbipartite structure inherit the LMT consequences of their bipartite archetypes.

We have pointed out that the class of frustrated nanomagnets which inherits the GS energy structure from their bipartite counterparts can be broader. In these nanomagnets the values of the total spin S in their GS obey the corresponding constraint $S \leqslant \mathcal{S}$ arising from LMT. A knowledge of possible GS's in this type of systems may facilitate classification of frustration and the interpretation of experimental results in terms of the Heisenberg model.

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