

Ground States of the Ising Model on the Shastry-Sutherland Lattice and the Origin of the Fractional Magnetization Plateaus in Rare-Earth-Metal Tetraborides

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(Received 22 April 2012; published 16 October 2012)

A complete and exact solution of the ground-state problem for the Ising model on the Shastry-Sutherland lattice in an applied magnetic field is found. The magnetization plateau at one third of the saturation value is shown to be the only possible fractional plateau in this model. However, stripe magnetic structures with $1/2$ and $1/n$ ($n > 3$) magnetization, observed in the rare-earth-metal tetraborides RB_4 , occur at the boundaries of the three-dimensional regions of the ground-state phase diagram. These structures give rise to new magnetization plateaus if interactions of longer range are taken into account. For instance, an additional third-neighbor interaction is shown to produce a $1/2$ plateau. The results obtained significantly refine the understanding of the magnetization process in RB_4 compounds, especially in TmB_4 and ErB_4 , which are strong Ising magnets.

DOI: 10.1103/PhysRevLett.109.167202

PACS numbers: 75.60.Ej, 05.50.+q, 75.10.-b

Geometric frustrations in lattice systems result in a rich variety of phenomena in both classical and quantum models. The investigation of such models, and even of their ground states is, however, a difficult problem. The first two-dimensional frustrated quantum model whose ground states have been determined exactly was introduced by Shastry and Sutherland in 1981 [1]. The Shastry-Sutherland (SS) lattice [Fig. 1(a)] is topologically equivalent to the Archimedean lattice $3^2.4.3.4$ [Fig. 1(b)]. In 1999, the SS model was shown to describe the magnetic properties of the compound $SrCu_2(BO_3)_2$ [2] synthesized in 1991 [3]. Some time later, other quasi-two-dimensional compounds with magnetic atoms of each layer located on a lattice topologically equivalent to the SS one have been discovered. In particular, this concerns the rare-earth-metal tetraborides RB_4 ($R = La - Lu$) [4–7]. Some of these are regarded to be classical systems, since the magnetic moments of the magnetic ions are large. Moreover, if the crystal field effects are strong enough, then the compounds can be described in terms of an effective spin-1/2 SS model under strong Ising anisotropy. This is the case of TmB_4 [4,5], ErB_4 [5,6], and HoB_4 [5], where the easy-magnetization axis is normal to SS planes.

SS magnets exhibit fascinating and puzzling sequences of fractional magnetization plateaus. For instance, plateaus at $m/m_s = 1/2, 1/7, 1/8, 1/9, \dots$ of the saturated magnetization m_s have been observed in TmB_4 for temperatures below 4 K when the field was normal to the SS planes; the $1/2$ plateau is the major one. To explain the origin of this sequence of plateaus is a challenging task. Some efforts were made to do this through the Ising model on the SS lattice since TmB_4 is a strong Ising magnet, as well as ErB_4 [5], where a single $1/2$ plateau was observed [6]. Because of the strong frustration, the solution of the ground-state problem for this model is difficult to obtain; therefore, in Ref. [1] the Ising limit was analyzed only for the zero field.

There is still no analytical solution for nonzero fields. There are only some numerical results showing the existence of a single fractional plateau at $m/m_s = 1/3$. However, the results of numerical simulations cannot be considered as absolutely reliable since an inappropriate finite lattice size can lead to erroneous conclusions. For instance, in Ref. [4] a single magnetization plateau at $m/m_s = 1/2$ was found for a system of 16 spins. More precise calculations, however, did not confirm this conclusion [8–10].

Here, we determine a complete solution of the ground-state problem for the Ising model on the SS lattice using our own method [11,12]. We rigorously prove the existence of a single $1/3$ plateau. As to magnetic structures with other fractional values of m/m_s , they exist at the boundaries of full-dimensional regions of the ground-state phase diagram. (A region in the parameter space of a model is full-dimensional if its dimension is equal to the dimension of the space). However, as the range of interactions increases, these structures become full-dimensional one by one and, hence, give rise to new plateaus. For example, we show that an additional third-neighbor interaction produces a $1/2$ plateau. A similar result was obtained numerically in Ref. [13], but we have found three different phases with

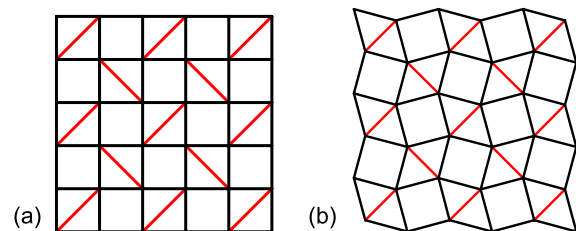


FIG. 1 (color online). (a) Shastry-Sutherland and (b) Archimedean $3^2.4.3.4$ lattices. They are topologically equivalent.

$m/m_s = 1/2$; one of these is expected to be realized in ErB_4 .

Consider the spin-1/2 Ising model on the SS lattice in the magnetic field

$$H = J_1 \sum_{\langle ij \rangle_1} \sigma_i \sigma_j + J_2 \sum_{\langle ij \rangle_2} \sigma_i \sigma_j - h \sum_i \sigma_i. \quad (1)$$

Here, $\sigma_i, \sigma_j = \pm 1$ are the spin variables; J_1 and J_2 are the interaction constants (for TmB_4 and ErB_4 , $J_1 \approx J_2 > 0$); $\langle ij \rangle_1$ and $\langle ij \rangle_2$ denote the summation over edges and diagonals [not all, see Fig. 1(a)] of the squares, respectively; h is the magnetic field.

The ground-state phase diagram for any Ising-type model is a set of convex polyhedral cones in the parameter space [11]. (A polyhedral cone is the linear hull, that is, all linear combinations with nonnegative coefficients—the so-called conic hull—of a set of vectors. It is fully determined by its edges or vectors along them). The full-dimensional

polyhedral cones fill the parameter space without gaps and overlaps. Herein, we refer to a structure, which is a ground-state structure in a full-dimensional polyhedral cone, as *full-dimensional* and to the corresponding edges (vectors) as *basic rays (vectors)* [11,12]. The convexity property (i.e., if a structure is a ground-state one in two points of the parameter space, then this structure is a ground-state one in the entire line segment connecting these points) makes it possible to find the ground-state structures in any point of the parameter space if all basic rays are known, as well as the ground-state structures in them.

Consider the rays listed in Table I. We shall shortly see that these form a complete set of basic rays for the model under consideration. To determine the ground-state structures in these rays, let us rewrite the Hamiltonian equation (1) as a single sum over all clusters in the form of a right-angled triangle with an SS diagonal being the hypotenuse, that is,

$$H = \sum_{\text{triangle}} H_i = \sum_{\text{triangle}} \left\{ J_1 (\sigma_{i0} \sigma_{i1} + \sigma_{i0} \sigma_{i2}) + \frac{J_2}{2} \sigma_{i1} \sigma_{i2} - h \left[\alpha \sigma_{i0} + \frac{1-\alpha}{2} (\sigma_{i1} + \sigma_{i2}) \right] \right\}. \quad (2)$$

The numbers σ_{i0} , σ_{i1} , and σ_{i2} define a configuration (among the six unique possible ones) of the i th triangle; σ_{i0} is the spin value in the vertex of the right angle. With each site being a vertex for three triangles, an arbitrary number α accounts for the fact that the energy contribution of a site can be distributed among these triangles in various ways.

A structure is generated by a set of triangle configurations if each triangle in the structure has a configuration belonging to the set. If, at a point (h, J_1, J_2) , all configurations of the set have the same energy, which is lower than the energies of all the remaining configurations, then the structures generated by the configurations of the set are ground-state ones at this point. Now, for each basic ray, we can find a set of triangle configurations which generate all ground-state structures in this ray. We call such sets basic sets of configurations [11]. They are given in the second

column in Table I, and in the fourth column such a value of the *free* coefficient α is indicated, for which all configurations from the basic set have the same energy H_i , which is lower than the energies of all the remaining configurations. Herein, solid and open circles denote spins up and down, respectively.

Table I represents a complete solution of the ground-state problem for the Ising model on the SS lattice. Using this table, we should firstly determine the full-dimensional regions and structures. Here is an example to explain how this is done: The Néel structure (Fig. 4, structure 3) is generated by the set of configurations $\text{⦿} \text{⦿} \text{⦿} \text{⦿}$ and $\text{⦿} \text{⦿} \text{⦿} \text{⦿}$. This set is a subset of the basic sets of configurations for \mathbf{r}_1 , \mathbf{r}_3 , \mathbf{r}_5 , and \mathbf{r}_5^- . Hence, the Néel structure is the ground-state one in these rays and, by virtue of the convexity property, in the whole polyhedral cone generated by these four vectors. A structure is full-, i.e., three-dimensional, if it is

TABLE I. Basic rays and basic sets of configurations for the Ising model on the SS lattice.

| Basic ray \mathbf{r}_i (h, J_1, J_2) | Basic set of configurations | Full-dimensional regions | α |
|--|---------------------------------------|--------------------------|-----------|
| \mathbf{r}_1 (0, 0, -1) | $\text{⦿} \text{⦿} \text{⦿} \text{⦿}$ | 1, $\bar{1}$, 3 | Arbitrary |
| \mathbf{r}_2 (0, -1, 2) | $\text{⦿} \text{⦿} \text{⦿} \text{⦿}$ | 1, $\bar{1}$, 2 | Arbitrary |
| \mathbf{r}_3 (0, 1, 2) | $\text{⦿} \text{⦿} \text{⦿} \text{⦿}$ | 2, 3, 4, $\bar{4}$ | Arbitrary |
| \mathbf{r}_4 (1, 0, 1) | $\text{⦿} \text{⦿} \text{⦿} \text{⦿}$ | 1, 2, 4 | 0 |
| \mathbf{r}_4^- (-1, 0, 1) | $\text{⦿} \text{⦿} \text{⦿} \text{⦿}$ | $\bar{1}$, 2, $\bar{4}$ | 0 |
| \mathbf{r}_5 (4, 1, 0) | $\text{⦿} \text{⦿} \text{⦿} \text{⦿}$ | 1, 3, 4 | 1/2 |
| \mathbf{r}_5^- (-4, 1, 0) | $\text{⦿} \text{⦿} \text{⦿} \text{⦿}$ | $\bar{1}$, 3, $\bar{4}$ | 1/2 |

TABLE II. Basic rays and generating configurations for the full-dimensional ground-state regions (top) and for their faces (bottom).

| Region | Configurations | Basic rays |
|--------------|----------------|--|
| 1 | | $\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_4, \mathbf{r}_5$ |
| $\bar{1}$ | | $\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_4^-, \mathbf{r}_5^-$ |
| 2 | | $\mathbf{r}_2, \mathbf{r}_4, \mathbf{r}_3, \mathbf{r}_4^-$ |
| 3 | | $\mathbf{r}_1, \mathbf{r}_5, \mathbf{r}_3, \mathbf{r}_5^-$ |
| 4 | | $\mathbf{r}_3, \mathbf{r}_4, \mathbf{r}_5$ |
| $\bar{4}$ | | $\mathbf{r}_3, \mathbf{r}_4^-, \mathbf{r}_5^-$ |
| Boundary | Configurations | Basic rays |
| 1, $\bar{1}$ | | $\mathbf{r}_1, \mathbf{r}_2$ |
| 1, 2 | | $\mathbf{r}_2, \mathbf{r}_4$ |
| 1, 3 | | $\mathbf{r}_1, \mathbf{r}_5$ |
| 1, 4 | | $\mathbf{r}_4, \mathbf{r}_5$ |
| 2, 4 | | $\mathbf{r}_3, \mathbf{r}_4$ |
| 3, 4 | | $\mathbf{r}_3, \mathbf{r}_5$ |

generated by a set of triangle configurations that is a subset of at least three basic sets. All full-dimensional regions are listed in Table II. In its first, second, and third columns we see, respectively, the notation of the region, the set of triangle configurations generating the ground-state structures in this region, and the basic rays, which are the edges of the region. The bar over the number of a region (structure) means that this region (structure) is symmetric to the region (structure) with the same number but without a bar in regard to field inversion (the flip of all spins). Now we can see that the vectors listed in Table I are basic vectors indeed and constitute a complete set. This is so, since the full-dimensional regions, determined from this set, fill the whole parameter space without gaps and overlaps (see Figs. 2 and 3).

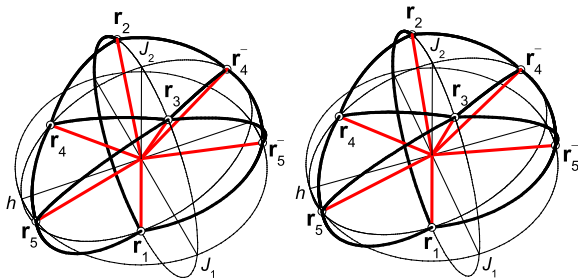


FIG. 2 (color online). Stereoscopic ground-state phase diagram for the Ising model on the SS lattice (see also Tables I and II). Edges of the polyhedral cones (basic rays) are depicted in red (gray); faces of cones are bordered by arcs of the unit sphere (heavy black lines). Unit circles (thin lines) in the three coordinate planes are figured for better visualization. This figure is a 3D image.

With the generating configurations for the full-dimensional ground-state structures being known, one can easily construct the latter. These are shown in Fig. 4, except for the fully polarized structure 1 (see also Fig. 3). Phase 2 is a disordered phase of Ising dimers (opposite spins on the SS diagonals). The disorder is two-dimensional; i.e., an extensive entropy occurs ($k_B \ln 2/2$ per site [1]). Phase 3 is the Néel phase. The magnetization of phases 1, 2, 3, and 4 is equal to 1, 0, 0, and $1/3$, respectively. Hence, there is a single fractional plateau at $m/m_s = 1/3$, which confirms the numerical results of Refs. [8–10].

An important advantage of our method is that it makes it possible to determine the ground-state structures not only in full-dimensional regions but also at their boundaries. This enables us to analyze the effects of longer-range interactions. The sets of generating configurations for the boundary structures are given in Table II. The sets are obtained as intersections of corresponding basic sets. The structures observed in TmB_4 emerge at the boundary between Néel and $1/3$ -plateau phases. They are generated by the configurations and . In addition to structures 3 and 4, these configurations generate a sequence of stripe structures and their mixtures (Fig. 5). The stripes contain an even number $2n$ of antiferromagnetic chains and are bordered by the ferromagnetic ones. We denote the structures composed of one type of stripe only by $(3, 4)_n$ ($n = 1$ for structure 4 and $n = \infty$ for structure 3). The magnetization of structure $(3, 4)_n$ is equal to $\frac{1}{2n+1}$. The periodic mixture with the smallest unit cell of consecutive structures $(3, 4)_n$ and $(3, 4)_{n+1}$ [we denote it by $(3, 4)_{n,n+1}$; see Fig. 5] has a magnetization equal to $\frac{1}{2n+2}$. In a future paper [14], we will rigorously show that the structure $(3, 4)_n$ ($n \geq 2$) can become full-dimensional (i.e., produce a plateau) if the interaction range is not less than the distance between the successive ferromagnetic chains of the structure $(3, 4)_{n-1}$. At the boundary considered, there can also occur structures where there is the first ferromagnetic chain on the left and/or the last one on the right, which can then form a right angle.

Consider some other boundaries. The structures at the boundary between regions 1 and 3 (see Table II and Fig. 6)

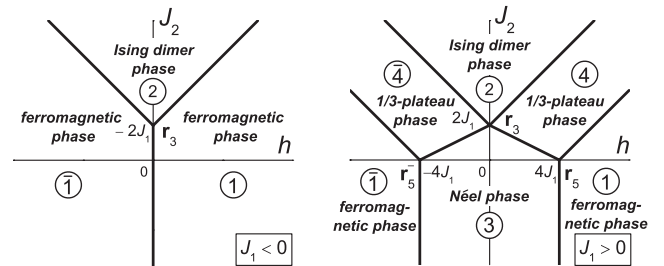


FIG. 3. Two-dimensional ground-state phase diagram for the Ising model on the SS lattice (see also Tables I and II and Figs. 2 and 4).

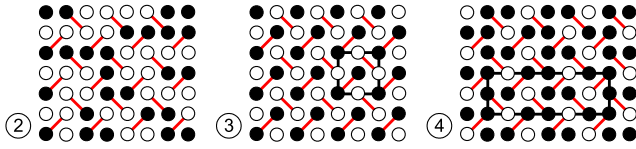


FIG. 4 (color online). Full-dimensional ground-state structures for the Ising model on the SS lattice. Phases 2, 3, and 4 are, respectively, the Ising dimer phase (opposite spins on the SS diagonals), the Néel phase, and the 1/3-plateau phase. Unit cells are shown. Solid and open circles represent spins up and down, respectively.

are constructed according to the following rule: all the spins at SS diagonals chosen arbitrarily, but without any nearest couples (i.e., belonging to diagonally joined squares), are orientated downwards; all the rest of spins are orientated upwards. The structures at the boundary between regions 1 and 4 (Fig. 6) are determined by a single condition: there cannot be two neighboring spins downwards either along SS diagonals or along the edges. If an additional antiferromagnetic (as in TmB_4 [5]) third-neighbor interaction J_3 along all the diagonals of *empty* (i.e., without SS diagonals) squares is included, then, from all possible structures at the boundary between regions 1 and 3 as well as 1 and 4, only those structures become full-dimensional in which all the *empty* squares have the configuration $\begin{smallmatrix} \bullet & \bullet \\ \bullet & \bullet \end{smallmatrix}$ (Fig. 6), since the energy should be as small as possible. These structures constitute disordered phases with 1/2 magnetization. One can show that the width of corresponding plateaus is equal to $4J_3$. Thus, contrary to the conclusion of Ref. [13], we can say that an additional third-neighbor interaction is sufficient for the stabilization of a 1/2 plateau. If $J_3 < 0$, then among all structures at the boundary between phases 1 and 4, only the ordered structure $(1, 4)_-$ (Fig. 6) becomes full-dimensional. It produces a 1/2 plateau as well. We suppose that this structure emerges in ErB_4 since J_3 is expected to be negative in this compound [5]. Interaction J_3 lifts also the degeneracy of the Ising dimer phase. If $J_3 > 0$ ($J_3 < 0$),

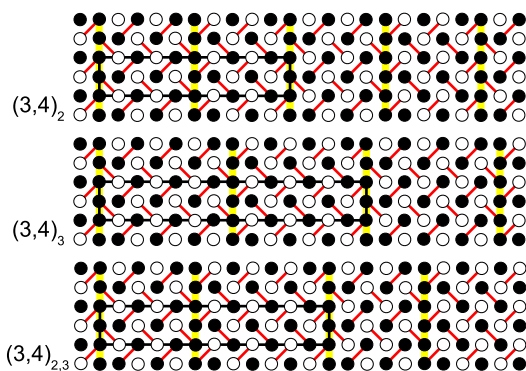


FIG. 5 (color online). Ground-state structures $(3, 4)_2$ and $(3, 4)_3$ ($m/m_s = 1/5$ and $1/7$) and their periodic mixture $(3, 4)_{2,3}$ with the smallest unit cell at the boundary between phases 3 and 4. The structures consist of ferro- [on the yellow (light gray) background] and antiferromagnetic chains. Unit cells are indicated.

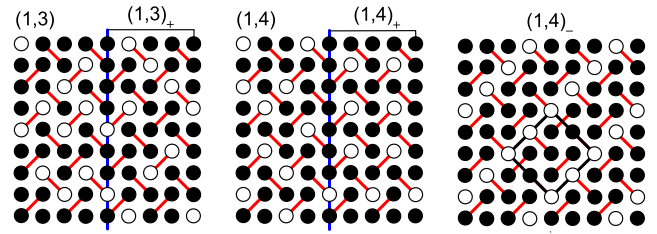


FIG. 6 (color online). Ground-state structures at the boundaries between regions 1 and 3 as well as 1 and 4 (see also Table II). First and second figures present disordered structures. The right-hand parts of these figures show the arrangements of spins (still disordered) if an additional third-neighbor interaction $J_3 > 0$ is included. All the *empty* squares have then the configuration $\begin{smallmatrix} \bullet & \bullet \\ \bullet & \bullet \end{smallmatrix}$. The third figure shows an ordered structure that becomes full-dimensional if $J_3 < 0$. All the structures $(1, 3)_+$, $(1, 4)_+$, and $(1, 4)_-$ produce a 1/2 plateau. The magnetization of the structures $(1, 3)$ and $(1, 4)$ can vary between 0 and 1 as well as 1/3 and 1, respectively.

then among all the structures of this phase, the structure with the minimum energy is the one for which spins at J_3 bonds are different (identical) (Fig. 7).

Some ground-state structures, obtained here analytically, coincide with those found numerically. Structure 4 was determined in Refs. [9,10,13,15], structure 3 (Néel phase) in Refs. [4,9,10], and structure 2 (Ising dimers) in Refs. [1,10]. It seems that structures with 1/4, 1/5, and 1/6 magnetization coincide with those determined in Ref. [15] for a spin-electron model (we cannot be sure, since SS bonds are not indicated there). This is quite clear because the spin-electron interaction lifts the degeneracy and some structures become full-dimensional. Our structures with $\frac{1}{2n+1}$ magnetization are different from those shown in Ref. [4]: all antiferromagnetic chains are shifted. The same concerns the structure with 1/3 magnetization shown in Ref. [8].

The main conclusion of our study is that an Ising model with additional long-range interactions is sufficient to explain the origin of fractional magnetization plateaus in rare-earth-metal tetraborides which are strong Ising magnets (TmB_4 and ErB_4). The long-range interactions are RKKY ones [4,13], since rare-earth-metal tetraborides are good metals. Here, we have studied the role of the first- and second-neighbor interactions and rigorously

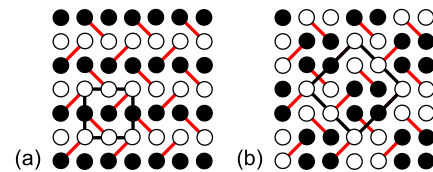


FIG. 7 (color online). Ground-state structures of the Ising model on the SS lattice with additional third-neighbor interaction: (a) $J_3 > 0$ (collinear phase), (b) $J_3 < 0$ (chessboard phase). The Ising dimer phase exists at the boundary between these phases.

proved that they produce a single fractional plateau at $1/3$ saturated magnetization. We have also partially analyzed the effect of the third-neighbor interaction and thus have shown that it produces at least five new phases; for three of these the magnetization is equal to $1/2$. One of these $1/2$ -plateau phases is expected to emerge in ErB_4 . Having analyzed the structures at the boundary between the Néel phase and the $1/3$ -plateau phase, we have also found the stripe structures with $m/m_s = 1/n$ ($n \geq 4$); some of these have been observed in TmB_4 . To finally explain the emergence of the sequence of magnetization plateaus in TmB_4 , the effect of further-neighbor interactions should be investigated.

We are grateful to T. Verkholyak for drawing our attention to the problem of fractional magnetization plateaus in SS compounds and for useful discussions.

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