

Ground states of lattice-gas models on the triangular and honeycomb lattices: Devil's step and quasicrystals

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We propose a method for determining the ground states of lattice-gas (or Ising) models. The method makes possible to find all types of ground states, including chaotic and ordered-but-aperiodic ones, and to identify the first-order phase transitions between them. Using this method, we prove the existence of an infinite series of ground states (the so-called “devil’s step”) in the lattice-gas model on the triangular lattice with up to third nearest-neighbor interactions and we study the effect of the interactions up to 19th neighbors on this series. To our best knowledge, this is only the second example of the devil’s step at zero temperature in the lattice-gas models with one kind of particles.

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I. INTRODUCTION

Recently, in “Nature,” a paper was published where a new kind of system was considered experimentally and theoretically [1]. The paper deals with buckled colloidal monolayers. The authors claimed that such a system is analogous to Wannier’s antiferromagnetic Ising model on the triangular lattice. Among other things, they study the ground states of the system and observe the formation of zigzagging stripes.

Starting from the paper by Wannier [2], Ising models on the triangular lattice (or equivalent lattice-gas models) were studied very intensively and by various methods. Great interest in these models was awoken by their multiple applications: from magnetic phenomena [3] to the adsorption of particles on crystal surfaces [4] or on carbon nanotubes [5] and the intercalation of particles between atomic layers of some compounds [6]. Much research was devoted to the ground states, since the latter often can be determined exactly and they provide an idea of the low-temperature behavior of the models.

Ground states of the simplest Ising model on the triangular lattice (with only nearest-neighbor interaction and with no field) were studied by Wannier. However, the intensive study of the ground states of complex lattice-gas models started only after the publication of a paper by Kanamori, where the method of geometrical inequalities was proposed [7]. Thereafter, many other methods have been developed; some of them were elaborated in connection with the problem of the ground states of quantum Falicov-Kimball model. Although the pioneer work by Kanamori appeared 40 years ago, until now there is no easy general algorithm for determining ground states of lattice-gas models.

Here we revisit the ground-state problem of the lattice-gas model on the triangular lattice with nearest- and next-nearest-neighbor interactions. A number of authors have already addressed this issue. These states were calculated by Metcalf in 1974 [8]. Using the Monte Carlo method, he found six ordered structures. In the same year, these structures were rigorously determined by Kaburagi and Kanamori

with the help of their method of geometrical inequalities [9]. They also found an additional structure which was overlooked by Metcalf. In 1986, using the method closely related to the one of Kanamori, Brandt and Stolze confirmed the results of earlier studies [10]. Hence, one would think that the problem of the ground states of this model is completely solved long ago. This is not the case, however. Here, we develop an alternative approach, initiated in our previous work [11], and apply it to this old problem. This provides a comprehensive idea of the ground states of this model and allows one to identify easily the first-order phase transitions.

With the help of our approach, we show that the addition of sufficiently small third nearest-neighbor repulsive interaction generates an infinite series of periodic structures with unit cells as large as we want. It is an example of the so-called zero-temperature “devil’s step.” The existence of devil’s step at zero temperature was proven by Kanamori for the lattice-gas model on the honeycomb lattice with up to third nearest-neighbor interaction.

The zero-temperature devil’s step is related to the infinite adaptivity that was discovered by Anderson in a group of crystalline materials (see Ref. [12] and references therein). In 1978, Kittel suggested that long-range repulsive interactions can account for such an infinite adaptivity. The example by Kanamori proves that the range of these interactions does not need to be very long. But it seems that Kanamori’s example is the only known example of zero-temperature devil’s step in lattice-gas models with one kind of particles.

We investigate how the pairwise interactions up to 19th neighbors affect the devil’s step and show that some interactions favor its formation only in the case of repulsion, the others, in the case of attraction, and still others, in none of the cases. Our method provides a possibility to demonstrate clearly the role of topology of the lattice in the formation of ground-state structures. To do so, we consider also the ground state of the lattice-gas model on the honeycomb lattice with nearest- and next-nearest-neighbor interactions in the case of attractive nearest-neighbor interaction and demonstrate that ordered structures without translational symmetry, as well as partially disordered ones, can have minimal energy.

The paper is organized as follows. In Sec. II, the method for determining the ground states is illustrated by the example of the lattice-gas model on the triangular lattice with nearest- and next-nearest-neighbor interactions. In Sec. III, the analogous model on the honeycomb lattice is considered (in the case of attractive nearest-neighbor interaction) and quasicrystal-like ground states are found. In Sec. IV, it is shown how to identify first-order phase transitions between ground states and it is proven the existence of zero-temperature devil's step in the lattice-gas model on the triangular lattice with up to third nearest-neighbor interaction. In Sec. V, the effect of pairwise interactions up to 19th neighbors on the devil's step is investigated and some necessary conditions for the existence of the devil's step are found. The final Sec. VI is devoted to the most important concluding remarks.

II. METHOD FOR DETERMINING THE GROUND STATES

We will explain our method for determining the ground states by the example of the lattice-gas model on the triangular lattice with nearest- and next-nearest-neighbor interactions. Here is the Hamiltonian of this model:

$$H_{lg} = I_1 \sum_{NN} c_i c_j + I_2 \sum_{NNN} c_i c_j - \mu_{lg} \sum_i c_i, \quad (1)$$

where c_i are the lattice-gas occupation variables ($c_i=1$ if the i th site is occupied and $c_i=0$ if it is empty), I_1 and I_2 are the nearest- and next-nearest-neighbor couplings, respectively, and μ_{lg} denotes the chemical potential of particles.

It is well known that this model is mathematically equivalent to the following spin model:

$$H_I = J_1 \sum_{NN} \sigma_i \sigma_j + J_2 \sum_{NNN} \sigma_i \sigma_j - h \sum_i \sigma_i, \quad (2)$$

with the couplings $J_1 = \frac{I_1}{4}$, $J_2 = \frac{I_2}{4}$, and the external field $h = \frac{\mu_{lg}}{2} - 6(J_1 + J_2)$, where $\sigma_i = 2c_i - 1 = \pm 1$ are the spin variables. Since the phase diagram of the latter is symmetric with respect to the field inversion $h \leftrightarrow -h$, we will construct the ground-state phase diagram for the spin model, although for the sake of clarity we will use “the language of particles” and not spins.

Our method of building the ground-state phase diagram is based on the obvious fact that the ground states are the same not only for the two Hamiltonians (1) and (2) but for the entire family of Hamiltonians

$$H = V_1 \sum_{NN} (a_1 \sigma_i + b_1)(a_1 \sigma_j + b_1) + V_2 \sum_{NNN} (a_2 \sigma_i + b_2)(a_2 \sigma_j + b_2) - \mu \sum_i (a_0 \sigma_i + b_0), \quad (3)$$

given that the couplings and “chemical potential” are related to the parameters of model (2) in the following way:

$$V_1 = \frac{J_1}{a_1^2}, \quad V_2 = \frac{J_2}{a_2^2}, \quad \mu = \frac{1}{a_0} [h + 6(a_1 b_1 V_1 + a_2 b_2 V_2)], \quad (4)$$

where $a_i \neq 0$ and b_i are arbitrary quantities independent of spin variables.

Now, we will consider the four-site clusters in the form of trefoil. Then Hamiltonian (3) can be rewritten in the form that contains the *single* sum over *all possible* trefoils

$$H = \sum_{\text{trefoil}} H_i = \sum_{\text{trefoil}} \left\{ \frac{V_1}{2} \sigma_{i0}^1 (\sigma_{i1}^1 + \sigma_{i2}^1 + \sigma_{i3}^1) + \frac{V_2}{2} (\sigma_{i1}^2 \sigma_{i2}^2 + \sigma_{i2}^2 \sigma_{i3}^2 + \sigma_{i3}^2 \sigma_{i1}^2) - \frac{\mu}{2(\alpha_1 + 3\alpha_2)} [\alpha_1 \sigma_{i0}^0 + \alpha_2 (\sigma_{i1}^0 + \sigma_{i2}^0 + \sigma_{i3}^0)] \right\}, \quad (5)$$

where the following notation is introduced:

$$\sigma_{ij}^k = a_k \sigma_{ij} + b_k, \quad k = 0, 1, 2. \quad (6)$$

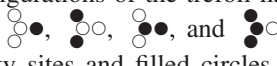

Here, the first index at spin variables σ_{ij} denotes host trefoil and the second one counts the sites within the trefoil (the central site is marked as zero). Every bond between a pair of nearest or next-nearest neighbors belongs to two different trefoils and is counted twice therefore the factors $\frac{1}{2}$ are introduced in the first and the second terms. Arbitrary factors α_1 and α_2 ($\alpha_1 \neq -3\alpha_2$ and at least one of them is nonzero) take account of the fact that every site belongs to two different trefoils as the central site and to six other trefoils as the lateral one. Let us note that the Hamiltonian H does not depend on α_1 and α_2 , although the local Hamiltonian H_i depends on them. It will be shown below that such representation of the Hamiltonian enables one to build complete ground-state phase diagram for the $J_1 < 0$ case.

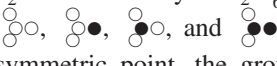
The following obvious statement holds true: if in a point of the space (V_1, V_2, μ) one can choose such a set of parameters a_k , b_k , and α_k that some trefoil configurations (one or more) have equal energy H_i and this energy is less than the energy of other configurations and if, in addition, *they can fill the lattice*, then these configurations generate all ground states in this point. (More exactly, all ground states without defects, that is such ground states where there are no configurations with greater energy). Without loss of generality, the coefficients a_k may be chosen equal, for instance, to $1/2$.


The methods for determining the ground states of lattice-gas models based on the representation of the Hamiltonian in the form of a single sum over some clusters are known as “ m -potential methods” [13]. These methods were used to determine the ground states of the Falicov-Kimball model [14,15]. Our method differs from the known ones by the way of constructing such m -potentials. In addition, we construct m -potentials not for entire regions but only for so-called “special points” including “infinite” ones (i.e., infinitely dis-

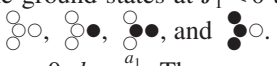
tant from the origin of the coordinates) and we consider not only periodic structures but all possible structures including disordered and ordered-but-a-periodic ones.


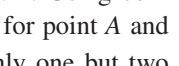
We will present the ground-state phase diagram in the $(\frac{h}{|J_1|}, \frac{J_2}{|J_1|})$ plane. Since every region of the diagram which corresponds to certain structure should be *convex*, it will be sufficient to find the points where the boundaries of these regions meet each other and to determine the ground states at these points. However, some regions meet at infinity, hence, we also have to consider the points infinitely distant from the origin of the coordinates. We will refer to the points of two-dimensional ground-state phase diagram where the regions meet as “special points.”

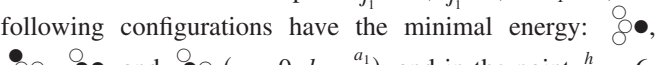
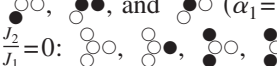
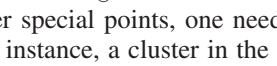
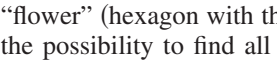
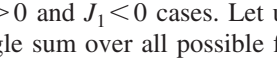

There are four infinite special points. If $J_2 \rightarrow +\infty$, $\alpha_1 = 0$, and the remaining parameters are fixed and arbitrary, then the following configurations of the trefoil have the minimal and equal energy:  (here, open circles represent empty sites and filled circles represent particles). These are just the configurations that determine the ground state at the point infinitely distant in the positive direction of the ordinate axis. Any configuration of the entire triangular lattice where there is no trefoil configurations, except these four, will be a ground state in this point. If $J_2 \rightarrow -\infty$, $\alpha_1 = 0$, then the following configurations have the minimal energy: . They determine the ground state at the point infinitely distant in the negative direction of the ordinate axis.

Another two infinite special points which are symmetric with respect to the field inversion are obtained if both h and J_2 tend to infinity. If $J_2 = \frac{h}{6} \rightarrow -\infty$, $\alpha_1 = 0$, then configurations  have the minimal energy. In the symmetric point, the ground state is constructed with the trefoil configurations where open circles are substituted by filled ones and vice versa.

In the $J_1 < 0$ case, there are only three finite special points. If $h = 0$, $J_2 = -\frac{J_1}{2}$, $b_1 = 0$, and $\alpha_1 = 0$, then at $J_1 < 0$, the following configurations of the trefoil have the minimal energy: . They determine the ground

states in the point $\frac{h}{|J_1|} = 0$, $\frac{J_2}{|J_1|} = \frac{1}{2}$ for $J_1 < 0$. In the point $\frac{h}{|J_1|} = -2$, $\frac{J_2}{|J_1|} = 1$, the ground states at $J_1 < 0$ are constructed with configurations . They have the minimal energy at $\alpha_1 = 0$, $b_1 = \frac{a_1}{3}$. The ground states in the symmetric point $\frac{h}{|J_1|} = 2$, $\frac{J_2}{|J_1|} = 1$ are constructed with the configurations where open circles are substituted by filled ones and vice versa.

The ground-state phase diagram for $J_1 < 0$ is shown in Fig. 1. To build it, only the ground states at three finite and four infinite points were needed. All the ground states in the line which connects two special points are generated by those configurations of the trefoil which are common for both points due to convexity of the regions. In the triangle between three points, the ground states are constructed with the configurations of the trefoil which are common for all three points. For instance, configurations  generate the stripe structure (a) depicted in Fig. 1. Using configurations  and two infinite points, one can construct not only one but two (and only two) structures (b) and (c) (Fig. 1). There is a twofold degeneracy that cannot be removed by any isotropic two-particle interaction [10]. Hence, in the $J_1 < 0$, $h < 0$ case, we have three structures in addition to the “empty” one.

Now, let us consider the $J_1 > 0$ case. As it will be clear later, in this case, there are four finite special points at $h \leq 0$, but only in two of them the ground states can be constructed of trefoils. In the point $\frac{h}{J_1} = -4$, $\frac{J_2}{J_1} = 1$, at $J_1 > 0$, the following configurations have the minimal energy: , , and  ($\alpha_1 = 0$, $b_1 = \frac{a_1}{3}$), and in the point $\frac{h}{J_1} = -6$, $\frac{J_2}{J_1} = 0$: , , and  ($b_1 = a_1$). To construct ground states in two other special points, one needs to consider a bigger cluster, for instance, a cluster in the form of a “flower” (hexagon with the central site). Such a cluster gives the possibility to find all special points and to construct the ground states in both $J_1 > 0$ and $J_1 < 0$ cases. Let us rewrite Hamiltonian (3) as a single sum over all possible flowers

$$\begin{aligned}
 H &= \sum_{\text{flowers}_i} H_i \\
 &= \sum_{\text{flowers}_i} \left\{ \frac{V_1}{2(\beta_1 + \beta_2)} [\beta_1 \sigma_{i0}^1 (\sigma_{i1}^1 + \sigma_{i2}^1 + \sigma_{i3}^1 + \sigma_{i4}^1 + \sigma_{i5}^1 + \sigma_{i6}^1) \right. \\
 &\quad + \beta_2 (\sigma_{i1}^1 \sigma_{i2}^1 + \sigma_{i2}^1 \sigma_{i3}^1 + \sigma_{i3}^1 \sigma_{i4}^1 + \sigma_{i4}^1 \sigma_{i5}^1 + \sigma_{i5}^1 \sigma_{i6}^1 + \sigma_{i6}^1 \sigma_{i1}^1)] \\
 &\quad + \frac{V_2}{2} (\sigma_{i1}^2 \sigma_{i3}^2 + \sigma_{i3}^2 \sigma_{i5}^2 + \sigma_{i5}^2 \sigma_{i1}^2 + \sigma_{i2}^2 \sigma_{i4}^2 + \sigma_{i4}^2 \sigma_{i6}^2 + \sigma_{i6}^2 \sigma_{i2}^2) \\
 &\quad \left. - \frac{\mu}{\alpha_1 + 6\alpha_2} [\alpha_1 \sigma_{i0}^0 + \alpha_2 (\sigma_{i1}^0 + \sigma_{i2}^0 + \sigma_{i3}^0 + \sigma_{i4}^0 + \sigma_{i5}^0 + \sigma_{i6}^0)] \right\}, \tag{7}
 \end{aligned}$$

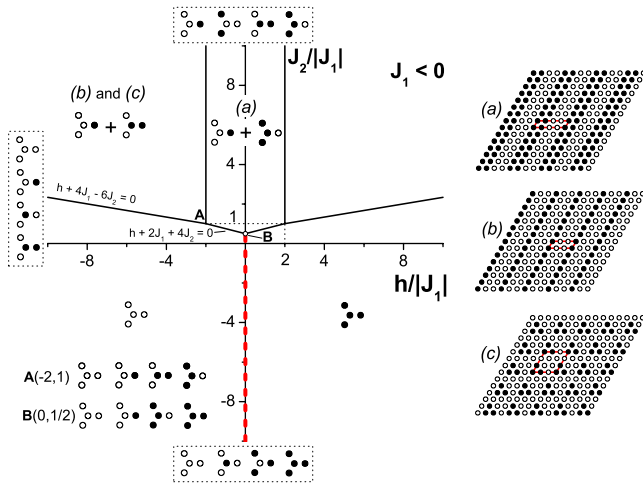


FIG. 1. (Color online) Ground-state phase diagram for Ising model on the triangular lattice with nearest- and next-nearest-neighbor interactions in the $J_1 < 0$ case. The configurations of the trefoil for three infinite and two finite points (A and B) as well as for different regions are indicated. Dashed red line is the line of the first-order phase transitions.

where the following notations are introduced:

$$\sigma_{ij}^1 = a_1 \sigma_{ij} + b_1, \quad \sigma_{ij}^2 = a_2 \sigma_{ij} + b_2, \quad \sigma_{ij}^0 = a_0 \sigma_{ij} + b_0. \quad (8)$$

Here, as in the case of the trefoil, the first index at spin variables σ_{ij} denotes host flower and the second one counts the sites within the flower (the central site is marked as zero). Arbitrary factors α_1 and α_2 are introduced to take into account that every site belongs to one flower as the central one and to six other flowers as the lateral one. In a similar man-

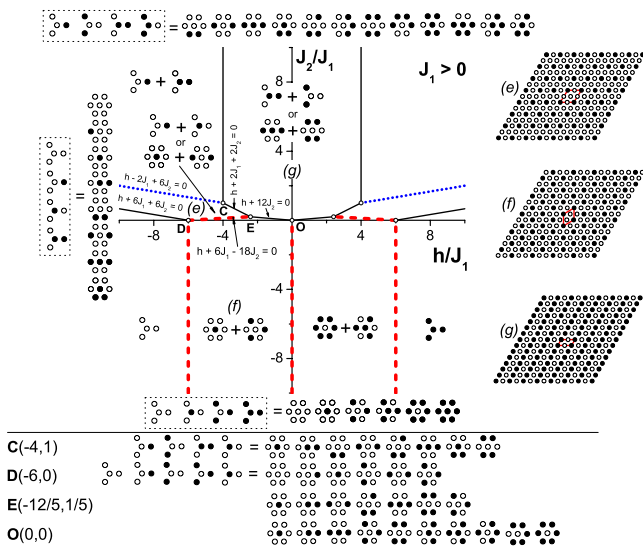


FIG. 2. (Color online) Ground-state phase diagram for Ising model on the triangular lattice with nearest- and next-nearest-neighbor interactions in the $J_1 > 0$ case. The configurations of the trefoil and/or of the flower are indicated for special points and different regions. Three new structures [(e), (f), and (g)] for this case are depicted. Dashed red lines are the lines of first-order phase transitions; dotted blue lines are the lines of multiphase points.

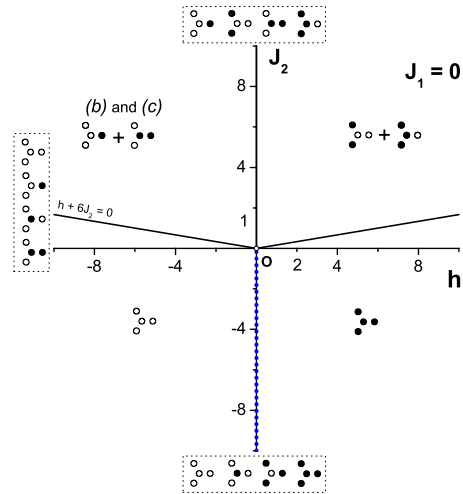


FIG. 3. (Color online) Ground-state phase diagram for Ising model on the triangular lattice with nearest- and next-nearest-neighbor interactions in the $J_1 = 0$ case. The configurations of the trefoil are indicated for infinite points and different regions. Dotted blue line is the lines of multiphase points.

ner, arbitrary factors β_1 and β_2 reflect the fact that every bond between nearest neighbors belongs to two flowers as the radial one and to two other flowers as the lateral one.

If $h = -\frac{12J_1}{5}$, $J_2 = \frac{J_1}{5}$, $b_1 = \frac{a_1}{3}$, $\alpha_1 = 0$, and $\beta_2 = \frac{2\beta_1}{3}$, then at $J_1 > 0$, the following flower configurations have the minimal energy: . If $h = 0$, $J_2 = 0$, $b_1 = 0$, and $\beta_2 = \frac{\beta_1}{2}$, then at $J_1 > 0$, flower configurations have the minimal energy.

Now we have the complete set of special points to construct ground-state phase diagram in the $J_1 > 0$ case. It is depicted in Fig. 2, where three new ground-state structures which appear in this case for $h < 0$ are indicated.

For the sake of completeness, we will construct the ground-state phase diagram for the $J_1 = 0$ case. It is easy to do since, in addition to the infinite points, being already found, there exists only one finite point. It is the origin of the coordinates. In this point, all configurations of the trefoil or the flower are the ground-state configurations. The ground-state phase diagram for $J_1 = 0$ is shown in Fig. 3.

III. ROLE OF TOPOLOGY OF THE LATTICE

As may be seen, the problem of determining the ground states can be divided into two interrelated parts: algebraic one (determining the configurations of a cluster which have minimal energy) and topological one (filling the lattice with these configurations). To demonstrate the role of topology in the formation of ground states, let us also find the ground states of Hamiltonian (2) on the honeycomb lattice. Consider only the $J_1 < 0$ case.

In the same way as for the triangular lattice, let us consider the family of Hamiltonians that have the same ground states and let us write them in the form of a single sum over trefoils

$$\begin{aligned}
 H &= \sum_{\text{o.o.}_i} H_i \\
 &= \sum_{\text{o.o.}_i} \left\{ \frac{V_1}{2} \sigma_{i0}^1 (\sigma_{i1}^1 + \sigma_{i2}^1 + \sigma_{i3}^1) \right. \\
 &\quad + V_2 (\sigma_{i1}^2 \sigma_{i2}^2 + \sigma_{i2}^2 \sigma_{i3}^2 + \sigma_{i3}^2 \sigma_{i1}^2) \\
 &\quad \left. - \frac{\mu}{\alpha_1 + 3\alpha_2} [\alpha_1 \sigma_{i0}^0 + \alpha_2 (\sigma_{i1}^0 + \sigma_{i2}^0 + \sigma_{i3}^0)] \right\}, \tag{9}
 \end{aligned}$$

where

$$\sigma_{ij}^k = a_k \sigma_{ij} + b_k, \quad k = 0, 1, 2. \tag{10}$$

These Hamiltonians are connected to the Ising Hamiltonian (2) using the following relations:

$$V_1 = \frac{J_1}{a_1^2}, \quad V_2 = \frac{J_2}{a_2^2}, \quad \mu = \frac{1}{a_0} (h + 3a_1 b_1 V_1 + 6a_2 b_2 V_2). \tag{11}$$

The ground-state phase diagram for the Ising model on the honeycomb lattice with nearest- and next-nearest-neighbor interactions in the $J_1 < 0$ case can be constructed in the same way as for the triangular lattice. It is depicted in Fig. 4. As one can see, it is almost the same as the corresponding diagram for the triangular lattice. The only difference is that the coordinates of points *A* and *B* for the honeycomb lattice are twice as large as for the triangular one. However, constructing ground-state structures with the same configurations for two lattices, one will reveal a striking difference. Thus, trefoil configurations $\circ\bullet$ and $\bullet\circ$ form no

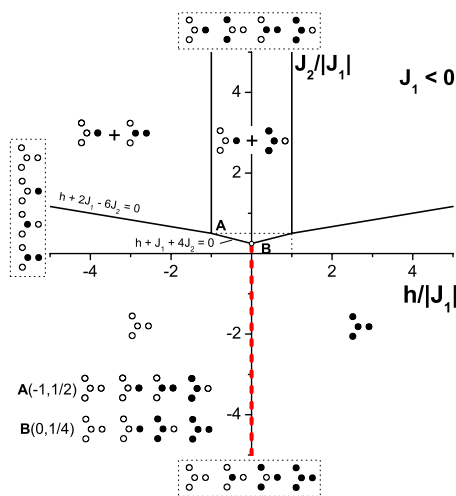


FIG. 4. (Color online) Ground-state phase diagram for Ising model on the honeycomb lattice with nearest- and next-nearest-neighbor interactions in the $J_1 < 0$ case. The configurations of the trefoil for special points as well as for different regions are indicated. Dashed red line is the line of the first-order phase transitions.

longer two but only one structure on the honeycomb lattice while configurations $\circ\bullet$ and $\bullet\circ$, which form a unique structure on the triangular lattice, generate infinite number of structures on the honeycomb lattice. These are the structures depicted in Figs. 5(b) and 5(c) and their mixture in arbitrary ratio (Fig. 6). Moreover, on the honeycomb lattice, these trefoil configurations generate two six-domain structures

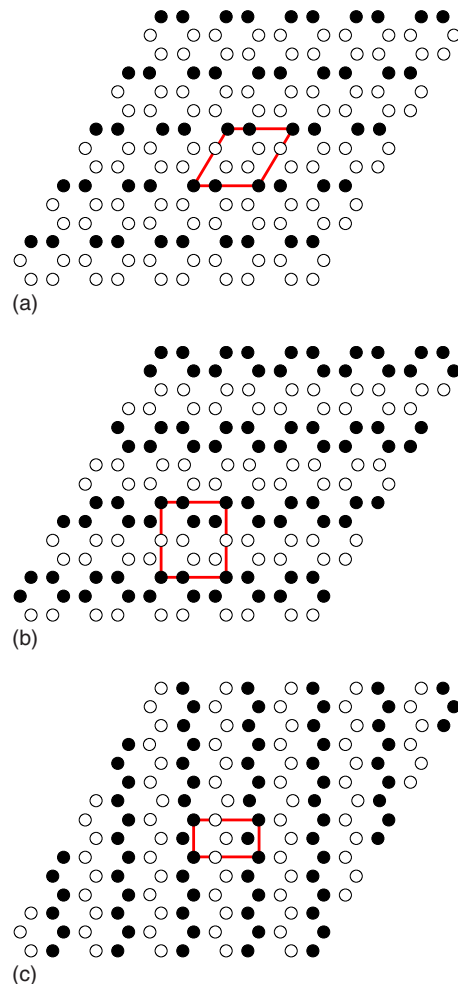


FIG. 5. (Color online) Ground-state structures for Ising model on the honeycomb lattice with nearest- and next-nearest-neighbor interactions in the $J_1 < 0$ case.

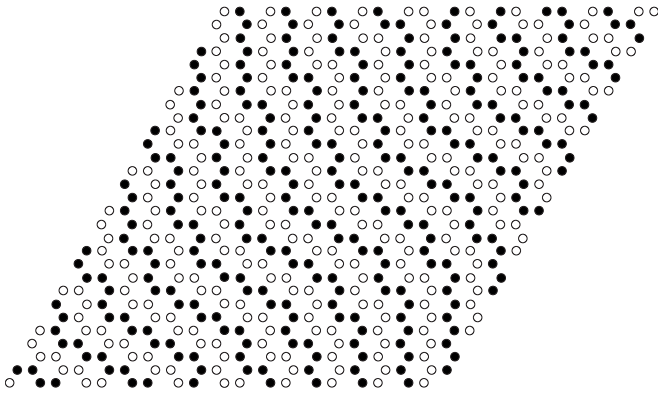


FIG. 6. A mixture of two last structures, depicted in the previous figure.

whose domain boundaries have zero formation energy (Fig. 7). They appear in the case where a closed loop of particles is formed. These ordered structures are similar to quasicrystals since they do not possess a global translational symme-

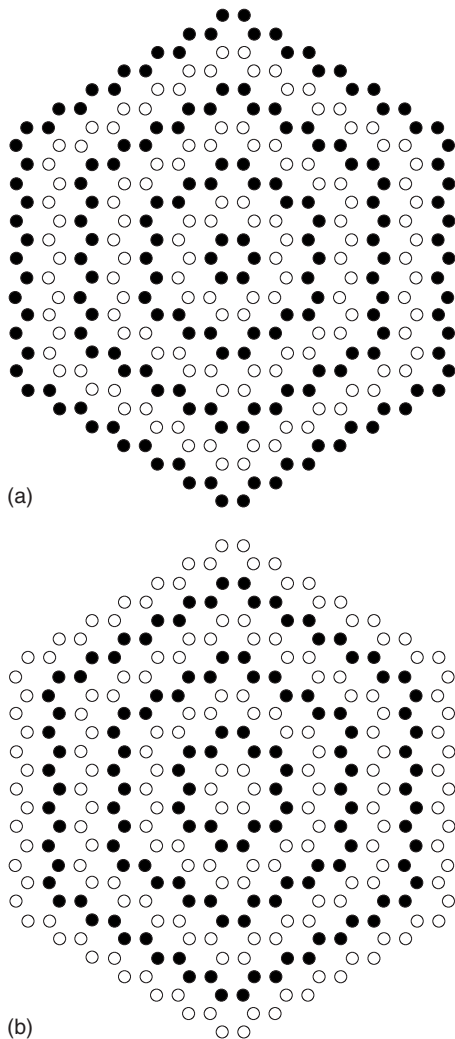


FIG. 7. Six-domain ground-state structures for the lattice-gas model on the honeycomb lattice with nearest- and next-nearest-neighbor interactions.

try. They are immiscible between themselves and with previous structures.

Although quasicrystals have been intensively studied since 1984, there is no clear understanding how they formed (see Ref. [16] and references therein and also Ref. [17]). Despite the fact that the quasicrystal-like structures that we have obtained are unstable (even infinitesimal third nearest-neighbor interaction will destroy them), our investigation can contribute to understanding the mechanism of formation of quasicrystalline and disordered structures, especially on two-dimensional lattices [18,19]. Like the authors of Ref. [16] do, we demonstrate that local interactions lead to the formation of an ordered structure without global translation symmetry.

We consider lattice models but we can reject the constraints imposed by the lattice and ask the following question: what structures can be formed, for instance, with blocks $\circ\bullet$ and $\bullet\circ$? It is clear that the answer is the following: “triangular” structure (a) and “honeycomb” structures, depicted in Figs. 6 and 7. Hybrid “triangular-honeycomb” structures cannot be formed.

IV. FIRST-ORDER PHASE TRANSITIONS AND DEVIL’S STEP

To answer the question what structures will have minimal energy if not the chemical potential but concentration of particles is fixed, one has to consider the ground-state structures also at the boundaries between regions. Moreover, it will provide the possibility to identify first-order phase transitions driven by the chemical potential in the lattice-gas model or by the field in the corresponding Ising model.

At the boundary between the “empty” phase (d) and phase (a), there is a partially disordered phase formed of configurations $\circ\circ$, $\circ\bullet$, and $\bullet\circ$. It is the mixture of both phases in arbitrary ratio [Fig. 8(a)]. At the boundary between the “empty” and “completely filled” phases, the ground states are generated only by two configurations: $\circ\circ$ and $\bullet\bullet$. These configurations are *incompatible* (in other words, *immiscible*) and are able to generate only two structures: “empty” one and “completely filled” one. There are no intermediate ground states without defects between these two and there will be a chemical potential (or field in the Ising model) driven first-order phase transition between these phases.

There is a twofold degeneracy at the boundary between the “empty” phase and phase (f) because the configuration $\circ\circ\circ$ and the set of configurations $\{\circ\circ\circ, \bullet\circ\circ\}$ which generates phase (f) are incompatible. The same occurs at the boundary between phase (f) and the phase which is symmetric to it (particle-hole symmetry) because the sets of configurations $\{\circ\circ\circ, \bullet\circ\circ\}$ and $\{\bullet\bullet\bullet, \circ\bullet\bullet\}$ are also incompatible. Hence, if the next-nearest-neighbor interaction is attractive ($J_2 < 0$), then all phase transitions are of the first order. This agrees with the results of Monte Carlo simulations at non-zero temperature [20]. However, there is the first-order phase transition between phases (e) and (f), if both interactions are repulsive ($0 < J_2 < \frac{J_1}{5}$), as far as the sets of configurations $\{\circ\circ\circ, \bullet\circ\circ\}$ and $\{\circ\circ\circ, \bullet\circ\circ\}$ are incompatible. Indeed, the

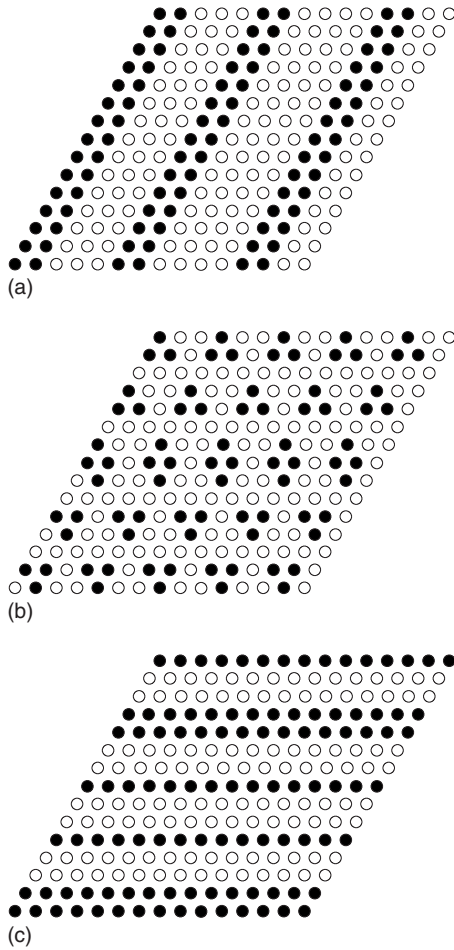


FIG. 8. (a) Chaotic mixture of phases (a) and (d), (b) two-domain structure at the boundary of phases (a) and (b) [(c)], and (c) chaotic mixture of phases (a) and (b).

Monte Carlo simulations indicate a first-order phase transition in this region [21].

Let us also consider the boundary between regions (a) and (b) [(c)]. The ground states at this boundary are generated by the set of trefoil configurations $\{\circ\bullet\circ, \bullet\circ\bullet, \circ\bullet\bullet\}$ or by the equivalent set of flower configurations $\{\circ\circ\bullet, \bullet\circ\circ, \circ\bullet\circ, \bullet\bullet\circ, \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet\}$. The two last configurations cannot be realized in such a set. Configuration $\circ\circ\bullet$ (with other configurations) can only generate the two-domain structure shown in Fig. 8(b). This structure is ordered but it does not possess complete translational symmetry. Hence, it can be considered as a quasicrystal-like structure. Other configurations, in addition to structures (a), (b), and (c), generate infinite number of structures which represent a mixture of phases (a) and (b) in arbitrary ratio [Fig. 8(c)]. Thus, one of degenerated and immiscible phases (b) and (c), notably phase (b), is miscible with phase (a) and another is immiscible.

Finally, let us proceed to the ground states at the boundary of phases (b) [(c)] and (e). They are generated by the set of trefoil configurations $\{\circ\bullet\circ, \bullet\circ\bullet, \circ\bullet\bullet\}$ or by the equivalent set of flower configurations $\{\circ\circ\bullet, \bullet\circ\circ, \circ\bullet\circ, \bullet\bullet\circ, \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet\}$.

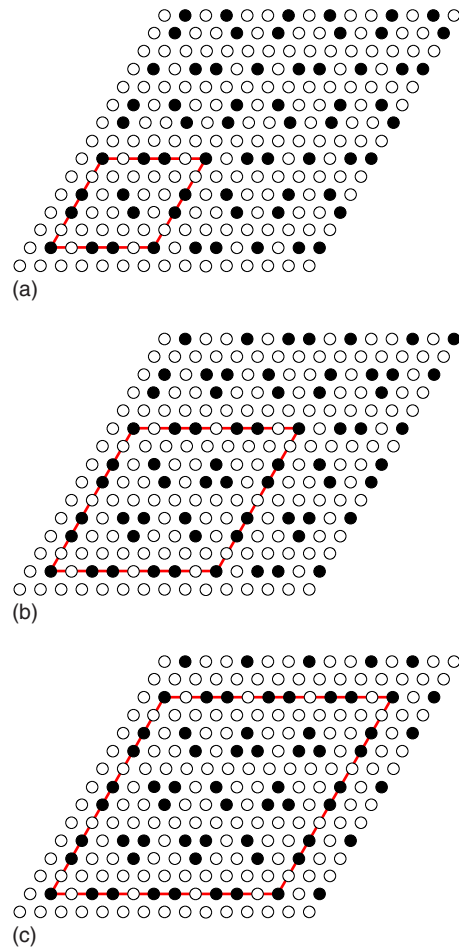


FIG. 9. (Color online) Structures S_1 (a), S_2 (b), and S_3 (c) from the infinite series S_m .

If one starts with configuration $\circ\circ\bullet$, then only structure (b) can be obtained. In contrast, other configurations of the set generate an infinite series of structures S_m ($m=0-\infty$). Structure number zero in this series is structure (e). The next three structures of the series are shown in Fig. 9.

In notations of Kaburagi and Kanamori (see Ref. [22]), every structure is characterized by a set of numbers

$$p_0, p_1, p_2, p_3, \dots, \tag{12}$$

where p_0 is the number of particles *per site* [which changes from $\frac{1}{4}$ ($m=0$) to $\frac{1}{3}$ ($m=\infty$)] and p_1, p_2, p_3, \dots provide the numbers of pairs of particles (*per particle*) that are first, second, third, ... neighbors, respectively. We will denote all structures with the same numbers p_i ($i=0, 1, 2, \dots$) by

$$S(p_0; p_1, p_2, p_3, \dots). \tag{13}$$

Numbers p_0, p_1, p_2, \dots allow one to calculate the energy density of corresponding structure. Energy per site reads

$$E = \sum_i p_0 p_i I_i - p_0 \mu_{lg}, \quad i = 1, 2, \dots \tag{14}$$

Here, I_i and μ_{lg} are couplings and the chemical potential of the lattice-gas Hamiltonian. One can switch to the Ising Hamiltonian using the following relations:

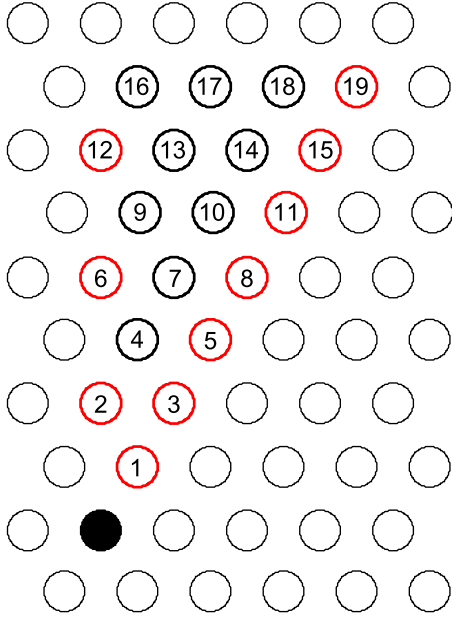


FIG. 10. (Color online) Neighbors on the triangular lattice. Neighbors with coordination number 6 are marked in red (light gray).

$$I_i = 4J_i, \quad \mu_{lg} = 2h + 2 \sum_k z_i J_i, \quad (15)$$

where z_i is the coordination number of the i th neighbors (see Fig. 10)

$$z_i = \begin{cases} 6 & \text{for } i = k^2 + k - 1, i = k^2 + k, i = k^2 + 2k \quad (k = 1, 2, 3, \dots) \\ 12 & \text{for other values of } i. \end{cases} \quad (16)$$

For structures S_m , we have calculated the numbers p_i up to $i=19$ (a method to calculate p_i is described in Appendix A)

$$\begin{aligned} p_0 &= \frac{3m^2 + 3m + 1}{(3m + 2)^2}, & p_1 &= \frac{3m^2}{3m^2 + 3m + 1}, \\ p_2 &= 0, & p_3 &= \frac{3(m+1)^2}{3m^2 + 3m + 1}, \\ p_4 &= \frac{6m(m+1)}{3m^2 + 3m + 1}, & p_5 &= \frac{3m(3m+1)}{3m^2 + 3m + 1}, \\ p_6 &= \frac{3(m+1)}{3m^2 + 3m + 1}, & p_7 &= \frac{6m(m-1)}{3m^2 + 3m + 1}, \\ p_8 &= \begin{cases} 3 & \text{for } m = 0 \\ \frac{3(m^2 - 2m + 2)}{3m^2 + 3m + 1} \end{cases}, & p_9 &= \frac{6m(m+3)}{3m^2 + 3m + 1}, \end{aligned}$$

$$p_{10} = \frac{6m}{3m^2 + 3m + 1}, \quad p_{11} = \begin{cases} 0 & \text{for } m = 0 \\ \frac{3(m^2 + 4m + 2)}{3m^2 + 3m + 1} \end{cases},$$

$$p_{12} = \frac{3m(3m-1)}{3m^2 + 3m + 1}, \quad p_{13} = \begin{cases} 6 & \text{for } m = 0 \\ \frac{6(m^2 - 1)}{3m^2 + 3m + 1} \end{cases},$$

$$p_{14} = \begin{cases} 0 & \text{for } m = 0 \\ \frac{6(m^2 + 2m - 2)}{3m^2 + 3m + 1} \end{cases}, \quad p_{15} = \begin{cases} 3 & \text{for } m = 0 \\ \frac{3(3m^2 - m - 1)}{3m^2 + 3m + 1} \end{cases},$$

$$p_{16} = \begin{cases} 0 & \text{for } m = 0 \\ \frac{6(m^2 - 3m + 4)}{3m^2 + 3m + 1} \end{cases}, \quad p_{17} = \begin{cases} 0 & \text{for } m = 0 \\ \frac{12(m+1)}{3m^2 + 3m + 1} \end{cases},$$

$$p_{18} = \begin{cases} 0 & \text{for } m = 0, 1 \\ \frac{6m(m-2)}{3m^2 + 3m + 1} \end{cases}, \quad p_{19} = \begin{cases} 0 & \text{for } m = 0 \\ \frac{3(m^2 - 4m + 8)}{3m^2 + 3m + 1} \end{cases}. \quad (17)$$

The structures depicted in Fig. 9 were found from completely different considerations by Nakanishi and Shiba and were called “triangular domain structures” [23]. In the model with only nearest- and next-nearest-neighbor interactions, they occur at the boundary between the structure S_0 [or (e)] and the structure (c) and there is no other pure ground-state structures at this boundary. They are *incompatible (immiscible)*. The nonzero temperature will remove the degeneracy at the boundary where these structures occur and they will form the (top) devil’s step [23,24], i.e., an infinite series of phases with the unite cell as large as we want. However, not only the temperature but also *the repulsive third neighbor interaction* removes the degeneracy and at $J_3 > 0$, we have the devil’s step even at zero temperature (see Fig. 11).

To prove it, we shall explain how, on a two-dimensional ground-state phase diagram, new regions (phases) appear when a new term is introduced into the Hamiltonian. If a new interaction is small enough, then new regions can grow only from the boundaries between existing regions, where corresponding structures are already present. Otherwise the convexity principle (the region corresponding to a structure should be convex) would be broken down. From this requirement follows the conclusion than only structures S_m can grow from the boundary between phases (e) and (c) [(b)] when interaction J_3 (or any other interaction) becomes non-zero.

To answer the question what interaction strength J_3 should be for the devil’s step to exist, let us add to Hamiltonian (7) the term that represents the third nearest-neighbor interaction

$$V_3(\sigma_{i1}^3 \sigma_{i4}^3 + \sigma_{i2}^3 \sigma_{i5}^3 + \sigma_{i3}^3 \sigma_{i6}^3), \quad (18)$$

where the following notations are introduced:

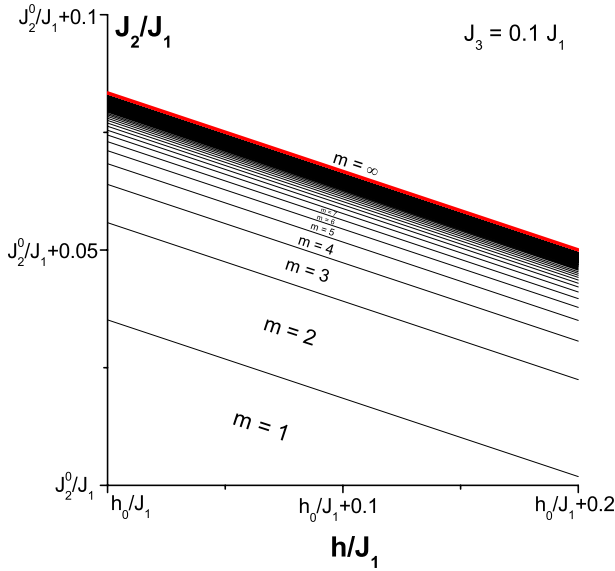


FIG. 11. (Color online) Regions in the $(\frac{h}{J_1}, \frac{J_2}{J_1})$ plane, which correspond to the infinite series of phases S_m at $J_3 = 0.1J_1$ and large enough and negative $\frac{h_0}{J_1}$. The red line is the boundary of the series. The figure was obtained through the comparison of the energies of structures S_m . h_0 is introduced because we are not able to find the values of h where the boundaries between phases S_m end (except the boundary between S_0 and S_1).

$$\sigma_{ij}^3 = a_3 \sigma_{ij} + b_3. \tag{19}$$

The expressions for switching to the simple spin model will be as follows:

$$V_1 = \frac{J_1}{a_1}, \quad V_2 = \frac{J_2}{a_2}, \quad V_3 = \frac{J_3}{a_3},$$

$$\mu = \frac{1}{a_0} [h + 6(a_1 b_1 V_1 + a_2 b_2 V_2 + a_3 b_3 V_3)]. \tag{20}$$

Whatever additional interactions are introduced into the Hamiltonian, infinite special points will remain the same in the $(\frac{h}{|J_1|}, \frac{J_2}{|J_1|})$ plane (it follows from their definition); only finite special points can change. Let us find some finite special points to be able to study the conditions for the existence of the devil's step in the case of nonzero third neighbor interaction. In the sets of flower configurations for finite points listed below, the configurations which are placed before semicolons are realized in the line connecting the corresponding point and infinite point $J_2 = \frac{h}{6} \rightarrow -\infty$.

If $J_2 = 0, h = -6J_1 - 6J_3$, then at $J_1 > 0, J_3 > 0$ ($\alpha_1 = 0, \beta_1 = 0$), configurations $\circ\circ\circ, \bullet\circ\circ, \circ\bullet\circ; \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet, \bullet\bullet\bullet$ and $\bullet\bullet\bullet$ have the minimal energy but the last three ones cannot be realized in this set.

If $J_2 = 2J_3, h = -6J_1 - 4J_3$, then at $0 < J_3 < \frac{J_1}{4}$ ($\alpha_1 = 0, \beta_1 = \frac{J_1 - 2J_3}{2J_3} \beta_2, b_1 = \frac{3J_1 - 8J_3}{3(J_1 - 2J_3)} a_1$), configurations $\bullet\circ\circ, \circ\circ\bullet, \circ\bullet\bullet; \bullet\circ\bullet, \bullet\bullet\circ, \bullet\bullet\bullet$ have the minimal energy (the last one cannot be realized).

If $J_2 = J_1 - 4J_3, h = -4J_1$, then at $0 < J_3 < \frac{J_1}{6}$ ($\alpha_1 = 0, \beta_1 = \frac{J_1 - 2J_3}{2J_3} \beta_2, b_1 = \frac{J_1}{3(J_1 - 2J_3)} a_1$), the following configurations have the minimal energy: $\circ\circ\circ, \bullet\circ\circ, \circ\bullet\circ, \bullet\circ\bullet; \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet$ and $\bullet\bullet\bullet$ (the last one cannot be realized in this set). If, in addition, $J_3 = \frac{J_1}{6}$, then configurations $\circ\circ\circ$ and $\bullet\bullet\bullet$ are added to the previous ones.

If $J_2 = 2J_3, h = 2J_1 - 36J_3$, then at $\frac{J_1}{6} < J_3 < \frac{J_1}{4}$ ($\alpha_1 = 0, \beta_1 = \frac{J_1 - 2J_3}{2J_3} \beta_2, b_1 = \frac{J_1}{3(J_1 - 2J_3)} a_1$), the following configurations have the minimal energy: $\circ\circ\circ, \bullet\circ\circ, \circ\bullet\circ, \bullet\circ\bullet; \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet$ and $\bullet\bullet\bullet$. If, in addition, $J_3 = \frac{J_1}{4}$, then configuration $\circ\circ\circ$ is added to the previous ones.

If $J_2 = J_1 - 2J_3, h = -4J_1 + 2J_3$, then at $0 < J_3 < \frac{J_1}{4}$ ($\alpha_1 = 0, \beta_1 = \frac{J_1 - 2J_3}{2J_3} \beta_2, b_1 = \frac{a_1}{3}$), the following configurations have the minimal energy: $\circ\circ\circ, \bullet\circ\circ, \circ\bullet\circ, \bullet\circ\bullet, \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet$ and $\bullet\bullet\bullet$, but the last one cannot be realized in this set. If, in addition, $J_3 = \frac{J_1}{4}$, then configurations $\circ\circ\circ, \bullet\circ\circ$, and $\circ\bullet\circ$ are added to the previous ones (the last one cannot be realized).

If $J_2 = 2J_3, h = 2J_1 - 22J_3$, then at $\frac{J_1}{4} < J_3 < \frac{J_1}{2}$ ($\alpha_1 = 0, \beta_1 = \frac{J_1 - 2J_3}{2J_3} \beta_2, b_1 = \frac{a_1}{3}$), configurations $\circ\circ\circ, \bullet\circ\circ, \circ\bullet\circ, \bullet\circ\bullet, \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet$ and $\bullet\bullet\bullet$ have the minimal energy (the last one cannot be realized). If, in addition, $J_3 = \frac{J_1}{2}$, then configuration $\circ\circ\circ$ is added to the previous ones.

If $J_2 = \frac{1}{2}J_1, h = -\frac{11}{2}J_1 - 6J_3$, then at $0 < \frac{J_1}{4} < J_3$ ($\alpha_1 = 0, \beta_1 = \beta_2, b_1 = \frac{2}{3}a_1$), the following configurations have the minimal energy: $\circ\circ\circ, \bullet\circ\circ, \circ\bullet\circ, \bullet\circ\bullet; \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet$.

If $J_2 = 2J_3, h = -5J_1 - 8J_3$, then at $\frac{J_1}{4} < J_3 < \frac{J_1}{2}$ ($\alpha_1 = 0, \beta_1 = \frac{J_1 - 2J_3}{2J_3} \beta_2, b_1 = \frac{2}{3}a_1$), the following configurations have the minimal energy: $\circ\circ\circ, \bullet\circ\circ, \circ\bullet\circ, \bullet\circ\bullet; \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet$.

If $J_2 = J_1, h = -6J_1 - 6J_3$, then at $0 < \frac{J_1}{2} < J_3$ ($\alpha_1 = 0, \beta_1 = 0$), the following configurations have the minimal energy: $\circ\circ\circ, \bullet\circ\circ, \circ\bullet\circ, \bullet\circ\bullet; \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet$ (the last one cannot be realized in this set).

If $J_2 = 2J_3, h = -4J_1 - 10J_3$, then at $0 < \frac{J_1}{2} < J_3$ ($\alpha_1 = 0, \beta_1 = \frac{J_1 - 2J_3}{2J_3} \beta_2, b_1 = \frac{a_1}{3}$), the following configurations have the minimal energy: $\circ\circ\circ, \bullet\circ\circ, \circ\bullet\circ, \bullet\circ\bullet, \circ\circ\bullet, \bullet\circ\bullet, \circ\bullet\bullet$ and $\bullet\bullet\bullet$ (the last one cannot be realized).

These special points make it possible to find the sequence of regions (phases) on the ground-state phase diagram in the $(\frac{h}{|J_1|}, \frac{J_2}{|J_1|})$ plane at $0 < J_3 < \frac{J_1}{4}$ which converge in the infinite point $J_2 = \frac{h}{6} \rightarrow -\infty$ and have the form of parallel stripes (except the first one and the last one). It is the following sequence of phases: "empty" phase, phase $S(\frac{1}{7}; 0, 0, 0)$, generated by configurations $\circ\circ\circ$ and $\bullet\bullet\bullet$ (see Fig. 12), the infinite series of phases $S_m (m=0-\infty)$, and degenerated phases (b) and (c).

In the case $\frac{J_1}{4} < J_3 < \frac{J_1}{2}$, phase S_0 does not exist: after phase $S(\frac{1}{7}; 0, 0, 0)$ goes the phase $S(\frac{1}{5}; \frac{1}{2}, 0, 0)$, generated by configurations $\bullet\circ\circ, \circ\bullet\circ$, and $\circ\circ\bullet$ (Fig. 12). As can be seen from Fig. 12, this phase is disordered. One should men-

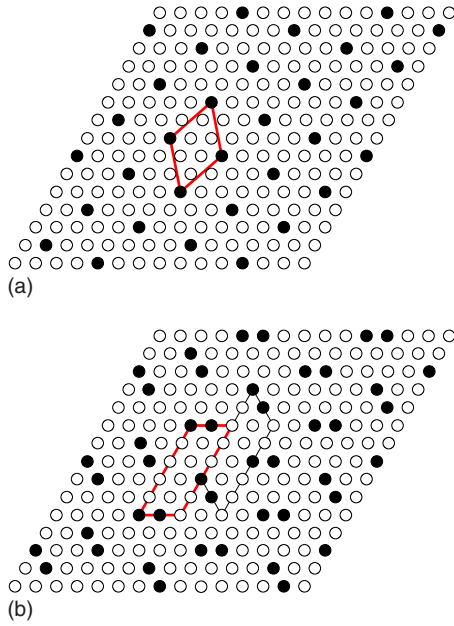


FIG. 12. (Color online) (a) Phase $S(\frac{1}{7}; 0, 0, 0)$ and (b) disordered phase $S(\frac{1}{5}; \frac{1}{2}, 0, 0)$. A unit cell is indicated for the first one and a motif for the second one.

tion that in Ref. [22] (where ground states of lattice-gas model on the triangular lattice with up to third nearest-neighbor interaction were investigated), nothing has been said about chaotization of phases despite the fact that a considerable part of phases found in this paper are disordered.

The phase that succeeds the phase $S(\frac{1}{5}; \frac{1}{2}, 0, 0)$ is generated by configurations $\begin{matrix} \bullet & \circ & \circ \\ \circ & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$, $\begin{matrix} \bullet & \bullet & \circ \\ \circ & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$, $\begin{matrix} \circ & \circ & \circ \\ \bullet & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$, and $\begin{matrix} \bullet & \bullet & \bullet \\ \circ & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$ (all or some of them) which have the minimal energy at the boundary of this phase. These configurations generate an infinite

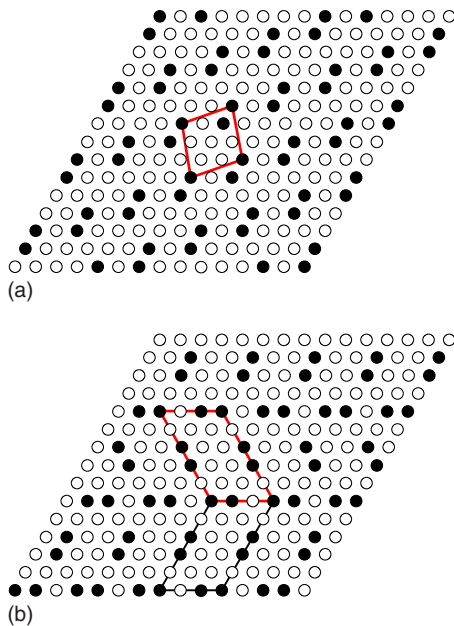


FIG. 13. (Color online) (a) Structure $S(\frac{1}{4}; \frac{1}{2}, 0, 1)$ (a unit cell is indicated) and (b) disordered phase $S(\frac{4}{15}; \frac{1}{2}, 0, \frac{5}{4})$ (a motif is indicated).

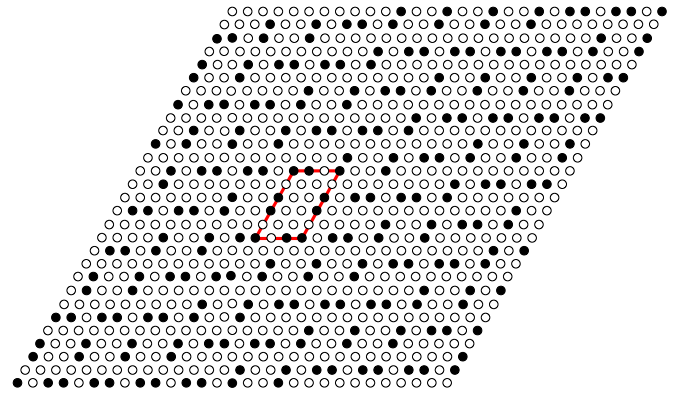


FIG. 14. (Color online) Another way of chaotization of phase $S(\frac{4}{15}; \frac{1}{2}, 0, \frac{5}{4})$ (a motif is indicated).

number of structures which are realized at the boundary of phase $S(\frac{1}{5}; \frac{1}{2}, 0, 0)$. This phase is not succeeded by the phase $S(\frac{1}{4}; \frac{1}{2}, 0, 1)$ as follows from Ref. [22] (it exists only at the boundary) but by disordered phase $S(\frac{4}{15}; \frac{1}{2}, 0, \frac{5}{4})$ (Figs. 13 and 14). At $J_3 = \frac{9}{26}J_1$, the latter completely supplants the phase S_1 . Although we do not know the configurations of a cluster which are realized at the boundary between these two phases, we can prove that there is no first-order phase transition between them (see Appendix B).

We do not know whether new phases appear when J_3 continues to increase. To answer this question, one needs to consider successively bigger clusters. Not counting the symmetry of a cluster, the number of its states is equal to 2^n , where n is the number of cluster's sites. Hence, the amount of calculations increases exponentially with n . However, one can state that the devil's step completely disappears only at $J_3 = \frac{J_1}{2}$ because only then a new phase begins to grow from the boundary line of the infinite series of phases. It is the line $J_2 = -\frac{1}{6}h + \frac{1}{3}J_1 - \frac{5}{3}J_3$ that goes from $J_2 = \max(2J_3, J_1 - 2J_3)$ to the infinity. If $0 < J_3 < \frac{J_1}{2}$, then the ground states in this line [structures (b) and (c)] are generated by configurations $\begin{matrix} \bullet & \circ & \circ \\ \circ & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$, $\begin{matrix} \bullet & \bullet & \circ \\ \circ & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$, $\begin{matrix} \circ & \circ & \circ \\ \bullet & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$, and $\begin{matrix} \bullet & \bullet & \bullet \\ \circ & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$. If $J_3 = \frac{J_1}{2}$, then also configuration $\begin{matrix} \bullet & \bullet & \circ \\ \circ & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$ has the same energy and further increasing of J_3 leads to the emergence of new phase $S(\frac{1}{4}; 1, 0, 0)$ that grows from the boundary line of the infinite series of phases. It is a disordered phase generated by configurations $\begin{matrix} \bullet & \circ & \circ \\ \circ & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$, $\begin{matrix} \bullet & \bullet & \circ \\ \circ & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$, and $\begin{matrix} \circ & \circ & \circ \\ \bullet & \circ & \circ \\ \circ & \circ & \circ \end{matrix}$ (Fig. 15). Hence, we have proven the existence of zero-temperature devil's step in the model of the lattice gas on the

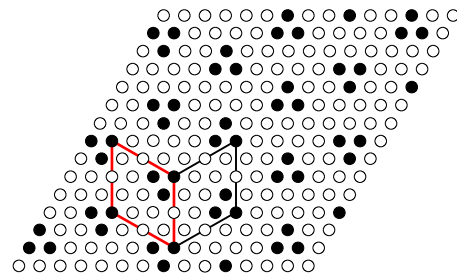


FIG. 15. (Color online) Disordered phase $S(\frac{1}{4}; 1, 0, 0)$ (a motif is indicated).

triangular lattice with up to third nearest-neighbor interaction or in the corresponding Ising model at the conditions $J_1 > 0$, $0 < J_3 < \frac{J_2}{2}$.

In 1983, zero-temperature devil's steps were first discovered by Kanamori in the lattice-gas model on the honeycomb lattice with first, second, and third neighbor interactions [25]. It seems that until now, it has remained a single example of the zero-temperature devil's step in the lattice-gas models with one kind of particles. Kanamori and Kaburagi have not found it on the triangular lattice [22]. Though, in 1990, together with Tonegawa, they found an infinite series of ground states in a lattice-gas model with *two* kinds of particles on the triangular lattice with first and second-neighbor interactions [26]. But the presence of just two kinds of particles is substantial there.

Complete devil's staircase and devil's step as its part are not only among the most interesting objects in theoretical physics; they exist in real physical systems [27]. For instance, recently devil's staircase was revealed in the system of lead atoms adsorbed on the crystal silicon surface Pb/Si(111) [28]. As the authors write, devil's staircase "shows a high degree of self-organization driven by a repulsive long-range adatom interaction."

The existence of devil's step in the model that we consider as well as in the model considered by Kanamori [25] means that even the short-range interactions (in two- and three-dimensional models) can generate an infinite number of phases in the ground-state phase diagram. Devil's step is also related to the problem of infinite adaptivity discovered experimentally by Anderson (see Ref. [12] and references therein). The infinite adaptivity implies that, within certain composition limits, every possible composition can attain a unique, fully ordered structure without defects. The zero-temperature devil's step is just an example of such an infinite adaptivity. In 1978, Kittel suggested that long-range repulsive interactions can account for it. Our example, as well as the example of Kanamori, shows that the range of interactions need not be large, much less infinite. In the next section, we will show that the interactions need even not be purely repulsive.

V. EFFECT OF PAIRWISE INTERACTIONS UP TO 19TH RANGE ON THE DEVIL'S STEP

The question arises of whether pairwise interactions of a range greater than 3 remove the degeneracy on the boundary between the phases (*e*) and (*c*) [(*b*)]. The expressions (17) allow us to answer this question easily. Let J_1 be greater than zero and let all the interactions of the range equal to or greater than 3 be zero except the interaction of the range r . Then one can prove that interactions of ranges $r = 6, 7, 8, 12, 16, 18, 19$ generate the devil's step in the case of repulsion, interactions of ranges $r = 4, 5, 9, 10, 14$ generate it in the case of attraction, and interactions of ranges $r = 11, 13, 15, 17$ do not generate it in any case (they generate only the phase S_1).

To explain why pairwise interactions of some ranges generate the devil's step in the case of attraction, the others, in the case of repulsion, and still others, in none of the cases, let

us considered the expression for the energy of the structure S_m . It reads as follows:

$$E_m = 2p_0(m) \left\{ \sum_i [2p_i(m) - z_i] J_i - h \right\}. \quad (21)$$

Now let us write the equation of the boundary between phases S_m and S_{m+1} ,

$$E_{m+1} - E_m = 2p_0(m+1) \left\{ \sum_i [2p_i(m+1) - z_i] J_i - h \right\} - 2p_0(m) \left\{ \sum_i [2p_i(m) - z_i] J_i - h \right\} = 0 \quad (22)$$

or

$$h = \frac{2 \sum_i [p_0(m+1)p_i(m+1) - p_0(m)p_i(m)] J_i}{p_0(m+1) - p_0(m)} - \sum_i z_i J_i. \quad (23)$$

The number of i th neighbors per one particle for the structure S_m is equal to

$$p_i(m) = \frac{a_i(m)m^2 + b_i(m)m + c_i(m)}{3m^2 + 3m + 1}. \quad (24)$$

If the interaction range is finite, then there is such a number m_0 that for all $m \geq m_0$, the coefficients in the numerator do not depend on m ,

$$\begin{aligned} a_i(m) &= a_i(m_0) = a_i(\infty) = a_i, \\ b_i(m) &= b_i(m_0) = b_i(\infty) = b_i, \\ c_i(m) &= c_i(m_0) = c_i(\infty) = c_i. \end{aligned} \quad (25)$$

The limit of Eq. (23) at $m \rightarrow \infty$ gives the equation of the boundary of infinite series S_m ,

$$h = \sum_i \frac{8a_i - 6b_i - 3z_i}{3} J_i. \quad (26)$$

If maximal range of interaction is equal to 19, then this equation takes the following form:

$$\begin{aligned} h &= 2J_1 - 6J_2 - 10J_3 - 8J_4 + 12J_5 - 12J_6 + 16J_7 + 14J_8 - 32J_9 \\ &\quad - 24J_{10} - 22J_{11} + 24J_{12} + 4J_{13} - 20J_{14} + 24J_{15} + 40J_{16} \\ &\quad - 36J_{17} + 28J_{18} + 26J_{19}. \end{aligned} \quad (27)$$

If the interaction range is large, we are not able to find the conditions when the devil's step exists in the $(\frac{h}{|J_1|}, \frac{J_2}{|J_1|})$ plane, but we can write "internal" (only in the frame of the infinite series of phases S_m) conditions of its existence (from some m to the infinity). They have the following form:

$$E_\infty < E_m \text{ for } h = h(\infty), \quad m = 0, 1, 2, \dots, \quad (28)$$

where

$$h(\infty) = \lim_{m \rightarrow \infty} h(m) = \sum_i \frac{8a_i - 6b_i - 3z_i}{3} J_i. \quad (29)$$

These conditions are equivalent to the following ones:

$$\sum_i \{3m^2[a_i(\infty) - a_i(m)] + 3m[b_i(\infty) - b_i(m)] + b_i(\infty) - 3c_i(m)\}J_i < 0. \quad (30)$$

They become identical starting from $m=m_0$,

$$\sum_i \frac{b_i - 3c_i}{3} J_i < 0. \quad (31)$$

If the maximal range of interactions is equal to 19, then we have three ‘‘internal’’ conditions:

$$\begin{aligned} & -J_3 + 2J_4 + J_5 - 2J_6 - 2J_7 - 8J_8 + 6J_9 + 2J_{10} - 2J_{11} - J_{12} \\ & + 6J_{13} + 16J_{14} + 2J_{15} - 30J_{16} - 8J_{17} - 4J_{18} - 28J_{19} \\ & < 0 \quad (\text{for } m > 1), \end{aligned} \quad (32)$$

$$\begin{aligned} & -J_3 + 2J_4 + J_5 - 2J_6 - 2J_7 - 8J_8 + 6J_9 + 2J_{10} - 2J_{11} - J_{12} \\ & + 6J_{13} + 16J_{14} + 2J_{15} - 30J_{16} - 8J_{17} - 10J_{18} - 28J_{19} \\ & < 0 \quad (\text{for } m = 1), \end{aligned} \quad (33)$$

$$\begin{aligned} & -J_3 + 2J_4 + J_5 - 2J_6 - 2J_7 - 5J_8 + 6J_9 + 2J_{10} + 4J_{11} - J_{12} \\ & - 6J_{13} + 4J_{14} - 4J_{15} - 6J_{16} + 4J_{17} - 4J_{18} - 4J_{19} \\ & < 0 \quad (\text{for } m = 0). \end{aligned} \quad (34)$$

We have found only necessary, internal, conditions for devil’s step existence in the case of pairwise interactions up to $r=19$. (As follows from the convexity principle, the internal conditions are not only necessary but also sufficient for devil’s step existence if $|J_i|$ ($i=3,4,\dots$) are small enough in comparison to $J_1 > 0$.) If the coefficients at J_i are negative (positive) in all three conditions, then the corresponding interaction favors the formation of the devil’s step in the case of repulsion (attraction). If the coefficient at J_i is negative in some conditions and positive in another, then the corresponding interaction does not favor the formation of the devil’s step in any case.

It is much more difficult to find the sufficient conditions of devil’s step existence. Therefore, we will restrict our consideration to the fourth nearest neighbors. Let us consider the ten-site cluster depicted in Fig. 16 and let us write the Hamiltonian in the form of a single sum over such clusters

$$\begin{aligned} H &= \sum_{\text{cluster}} H_i \\ &= \sum_{\text{cluster}} \left\{ \frac{V_1}{2(2\beta_1 + \beta_2 + \beta_3 + 2\beta_4)} [\beta_1 \sigma_{i0}^1 (\sigma_{i1}^1 + \sigma_{i2}^1 + \sigma_{i3}^1 + \sigma_{i4}^1 + \sigma_{i5}^1 + \sigma_{i6}^1) \right. \\ &+ \beta_2 (\sigma_{i1}^1 \sigma_{i2}^1 + \sigma_{i3}^1 \sigma_{i4}^1 + \sigma_{i5}^1 \sigma_{i6}^1) + \beta_3 (\sigma_{i2}^1 \sigma_{i3}^1 + \sigma_{i4}^1 \sigma_{i5}^1 + \sigma_{i6}^1 \sigma_{i1}^1) \\ &+ \beta_4 (\sigma_{i7}^1 (\sigma_{i1}^1 + \sigma_{i6}^1) + \sigma_{i8}^1 (\sigma_{i2}^1 + \sigma_{i3}^1) + \sigma_{i9}^1 (\sigma_{i4}^1 + \sigma_{i5}^1))] \\ &+ \frac{V_2}{2(2\gamma_1 + \gamma_2)} [\gamma_1 (\sigma_{i1}^2 \sigma_{i3}^2 + \sigma_{i3}^2 \sigma_{i5}^2 + \sigma_{i5}^2 \sigma_{i1}^2 + \sigma_{i2}^2 \sigma_{i4}^2 + \sigma_{i4}^2 \sigma_{i6}^2 \\ &+ \sigma_{i6}^2 \sigma_{i2}^2) + \gamma_2 \sigma_{i0}^2 (\sigma_{i7}^2 + \sigma_{i8}^2 + \sigma_{i9}^2)] \\ &+ \frac{V_3}{2(\delta_1 + 2\delta_2)} [\delta_1 (\sigma_{i1}^3 \sigma_{i4}^3 + \sigma_{i2}^3 \sigma_{i5}^3 + \sigma_{i3}^3 \sigma_{i6}^3) + \delta_2 (\sigma_{i7}^3 (\sigma_{i2}^3 + \sigma_{i3}^3) \\ &+ \sigma_{i8}^3 (\sigma_{i1}^3 + \sigma_{i4}^3) + \sigma_{i9}^3 (\sigma_{i3}^3 + \sigma_{i6}^3))] \\ &+ \frac{V_4}{2} [\sigma_{i7}^4 (\sigma_{i3}^4 + \sigma_{i4}^4) + \sigma_{i8}^4 (\sigma_{i5}^4 + \sigma_{i6}^4) + \sigma_{i9}^4 (\sigma_{i1}^4 + \sigma_{i2}^4)] \\ &+ \frac{V_5}{2} [\sigma_{i7}^5 \sigma_{i8}^5 + \sigma_{i8}^5 \sigma_{i9}^5 + \sigma_{i9}^5 \sigma_{i7}^5] \\ &- \frac{\mu}{2(\alpha_1 + 6\alpha_2 + 3\alpha_3)} [\alpha_1 \sigma_{i0}^0 + \alpha_2 (\sigma_{i1}^0 + \sigma_{i2}^0 + \sigma_{i3}^0 + \sigma_{i4}^0 + \sigma_{i5}^0 + \sigma_{i6}^0) \\ &+ \alpha_3 (\sigma_{i7}^0 + \sigma_{i8}^0 + \sigma_{i9}^0)] \left. \right\}, \end{aligned} \quad (35)$$

where the following notations are introduced:

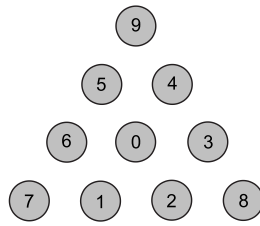


FIG. 16. Numeration of sites in a big triangular cluster.

$$\sigma_{ij}^4 = a_4 \sigma_{ij} + b_4, \quad \sigma_{ij}^5 = a_5 \sigma_{ij} + b_5, \quad (36)$$

where $\alpha_k, \beta_k, \gamma_k,$ and δ_k are arbitrary. Since every site has 12 fourth neighbors and six fifth neighbors, the expressions connecting this model with the Ising model are as follows:

$$V_i = \frac{J_i}{a_i^2}, \quad i = 1 - 5,$$

$$\mu = \frac{1}{a_0} [h + 6(a_1 b_1 V_1 + a_2 b_2 V_2 + a_3 b_3 V_3 + 2a_4 b_4 V_4 + a_5 b_5 V_5)]. \quad (37)$$

As one can see from the above formulae, the cluster in the form of a big triangle makes it possible to consider interactions up to fifth nearest neighbors, but we will restrict our consideration to the range $r=4$ ($J_5=0$). The number of configurations of the cluster is equal to 208. Hence, in this case (and even in the case of the “flower” for which the number of configurations is equal to 26), it is impossible to avoid the use of computer programs for analytical calculations.

The conditions for existence of the infinite series S_m in the $(\frac{h}{|J_1|}, \frac{J_2}{|J_1|})$ plane are equivalent to the conditions for existence of a two-domain structure at the boundary of this series. This structure is depicted in Fig. 17. It is generated by configurations $\circ\circ\bullet\bullet, \circ\circ\circ\bullet, \bullet\circ\circ\bullet,$ and $\bullet\circ\bullet\bullet$ or by triangle configurations depicted in Fig. 18(a).

If $-J_3+2J_4 < 0, -J_1+2J_3-2J_4 < 0, -J_1+2J_3+4J_4 < 0,$ then one can choose such a set of “free coefficients” in Eq. (35) then at the boundary $h=2J_1-6J_2-10J_3-8J_4$ of the infinite series $S_m,$ starting from certain value of $J_2,$ the triangle configurations depicted in Fig. 18(a) will have the minimal energy. These “free coefficients” can be for instance as follows: $\alpha_1=0, \alpha_2=\alpha_3, \beta_1=\frac{J_1-2J_3+2J_4}{2J_4}\beta_4, \beta_2=\beta_3=\frac{J_3-2J_4}{J_4}\beta_4,$ $\gamma_1=\gamma_2, \delta_1=\frac{J_3-2J_4}{J_4}\delta_2, a_i=\frac{1}{2}, b_2=\frac{-2J_1+J_2+4J_3-4J_4}{2J_2}, b_1=b_4=\frac{1}{2},$

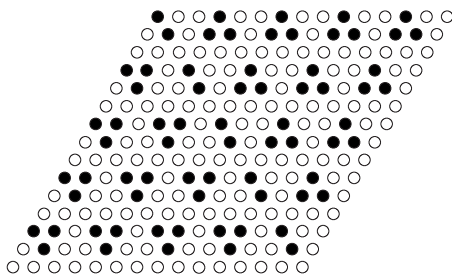


FIG. 17. Two-domain structure at the boundary of the infinite series $S_m.$

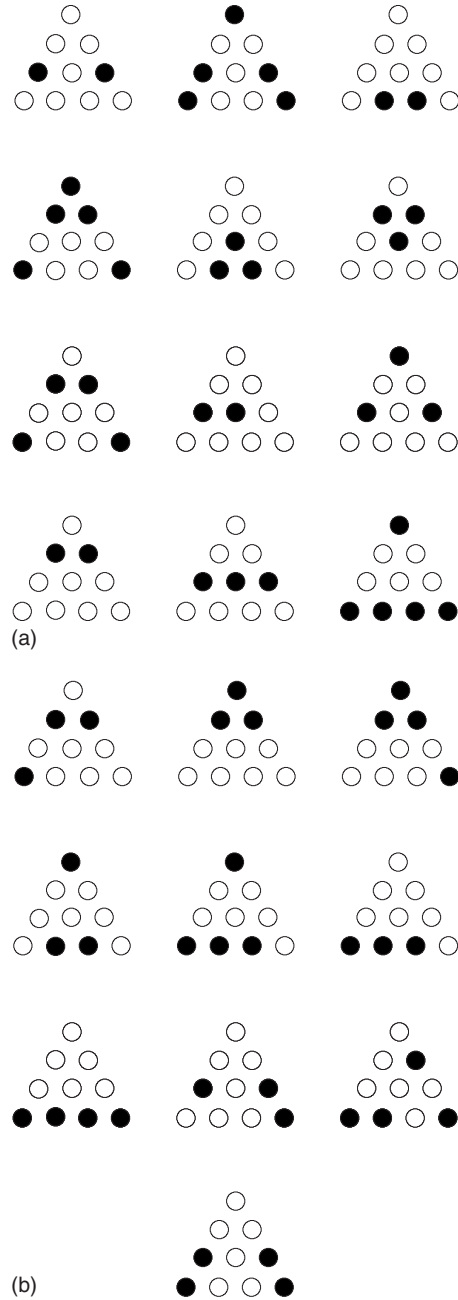


FIG. 18. (a) Configurations of the big triangle, which generate the two-domain structure (first three rows) and structure (b) (last row). (b) Configurations of the big triangle which, at the boundary of infinite series $S_m,$ have the same energy as the configurations depicted in (a).

$b_3 = \frac{-J_1-J_3+13J_4}{6(J_3-3J_4)}$. Although another ten configurations have the same energy [Fig. 18(b)], they cannot be realized in this set. It is easy to verify it directly, but we can avoid this as far as these configurations contain no other flower configurations than those which generate the two-domain structure and structure (b).

Then, we have sufficient conditions for existence of the infinite series of phases S_m in the model with up to fourth nearest-neighbor interaction. Here they are:

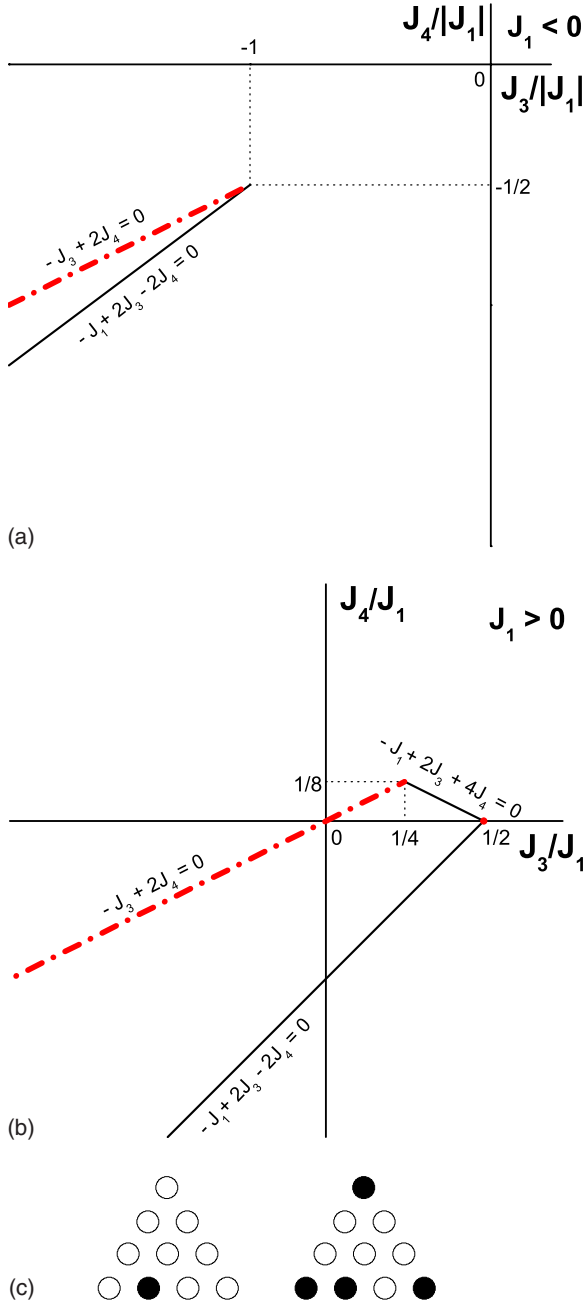


FIG. 19. (Color online) A part of the region where the devil's step exists for (a) $J_1 < 0$ and (b) $J_1 > 0$ in the case of pairwise interactions up to fourth nearest neighbors and (c) additional configurations of big triangle at boundaries of the indicated regions. Dash-and-dot red line shows the real boundary of the regions.

$$\begin{aligned}
 -J_3 + 2J_4 < 0, \quad -J_1 + 2J_3 - 2J_4 < 0, \\
 -J_1 + 2J_3 + 4J_4 < 0.
 \end{aligned} \tag{38}$$

These conditions define the regions shown in Fig. 19. As one can see, if the range of interactions is greater than 3, the devil's step can exist even in the case of attraction between nearest neighbors ($J_1 < 0$). The first of these conditions is also necessary because it is condition (32). The second (the third) condition is sufficient but not necessary because if it is

satisfied, then, at the boundary of the infinite series of phases, in addition to the configurations depicted in Fig. 18, only the first (second) configuration shown in Fig. 19(c) has the minimal energy. However, it does not yield new structures. To find the whole region where the devil's step exists (in the case of up to fourth nearest-neighbor interaction), one should consider bigger clusters.

VI. CONCLUSIONS

The method that we have proposed allows us to construct complete ground-state phase diagram for many lattice-gas models or equivalent Ising models. The method is especially effective for investigation of degeneracy at the boundaries between different regions in the space of parameters of the Hamiltonian. It gives the possibility to identify first-order phase transitions and to find infinite series of phases, i.e., zero-temperature devil's steps. In contrast to the method proposed by Brandt and Stolze, our method makes possible to find disordered phases and ordered-but-a-periodic ones (quasicrystalline).

Using our method, we have found ground states in all points of the parameter space of the lattice-gas (or Ising) Hamiltonian on the triangular lattice with nearest- and next-nearest-neighbor interactions. We have also identified the first-order phase transitions. We have proven that additional repulsive third nearest-neighbor interaction leads to appearance of zero-temperature devil's step. It is the most important result of the work. We have also found some necessary conditions for existence of the devil's step in the model with up to 19th nearest-neighbor interaction and sufficient conditions for existence of the devil's step in the model with up to fourth nearest-neighbor interaction. In this model, the devil's step can exist even in the case of attractive nearest-neighbor interaction.

At the end, it should be noted that our method is also applicable to the lattice-gas models with many kinds of particles (or to Ising-type models with spin larger than 1/2). However, despite interesting results owing to this method, it needs further developments and improvements to solve the problem of ground states of complex lattice-gas models.

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APPENDIX A: A WAY TO CALCULATE EXPRESSIONS FOR p_i

Here, we will give without proof (the proof is rather simple) a way to easily calculate p_i for the structures of the infinite series S_m [see Eq. (17)]. Starting from some values of m , expression for p_i ($i=1, 2, \dots$), which corresponds to the structure S_m , has the following form:

$$p_i = \frac{a_i m^2 + b_i m + c_i}{3m^2 + 3m + 1}, \tag{A1}$$

where coefficients a_i , b_i , and c_i do not depend on m . Coefficient a_i is equal to $2/3$ of the number of i th neighbors for any particle of structure (c) (Fig. 1). Coefficient b_i is determined as follows:

$$b_i = 3(\Delta_0 + \Delta_1 + \Delta_2 + \dots + \Delta_k) - (6k + 1)a_i, \quad (\text{A2})$$

where Δ_0 is the number of i th neighbors for a particle at the boundary between triangular domains (see Fig. 17), which is sufficiently distant from the vertex; Δ_l ($l=1, 2, \dots, k$) is the number of i th neighbors for a three-particle group of l th layer of these group; k is the number of three-particle group layers (on one side of the domain boundary) interacting with particles at the domain boundary.

An example: for eighth neighbors $a_8=3$, $k=1$, $\Delta_0=1$, $\Delta_1=4$, $b_8=3(1+4)-(6+1)3=-6$. To calculate c_i , it suffices to calculate p_i for a structure S_m , where m is big enough.

APPENDIX B: PROOF OF ABSENCE OF FIRST ORDER PHASE TRANSITION BETWEEN TWO PHASES

Let us consider phase $S(p_0; p_1, p_2, p_3, \dots)$ which is a mixture of two phases: $S(p_0^1; p_1^1, p_2^1, p_3^1, \dots)$ with the weight ε and $S(p_0^2; p_1^2, p_2^2, p_3^2, \dots)$ with the weight $1-\varepsilon$. If the boundary and the interaction between two identical motifs are the same as the boundary and the interaction between different motifs, i.e., if domain wall between domains of different phases has zero formation energy, then the expressions for p_0 and p_i are as follows:

$$p_0 = \frac{\varepsilon p_0^1 + \delta(1-\varepsilon)p_0^2}{\varepsilon + \delta(1-\varepsilon)},$$

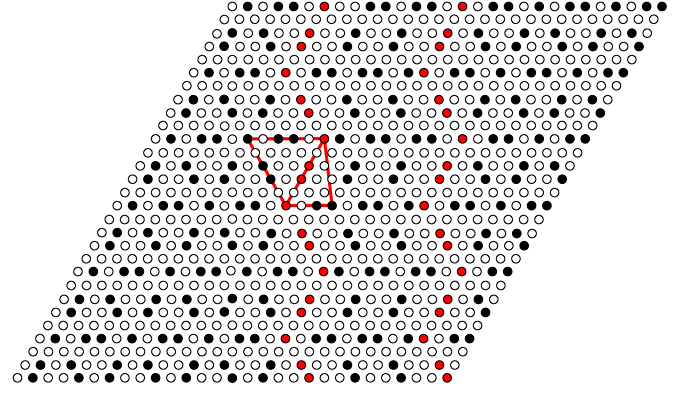


FIG. 20. (Color online) One of possible configurations at the boundary between phases $S(\frac{4}{15}; \frac{1}{2}, 0, \frac{5}{4})$ and $S(\frac{7}{25}; \frac{3}{7}, 0, \frac{12}{7})$. Red sites are filled sites of domain walls.

$$p_i = \frac{\varepsilon p_i^1 p_0^1 + \delta(1-\varepsilon)p_i^2 p_0^2}{\varepsilon p_0^1 + \delta(1-\varepsilon)p_0^2} \quad (i = 1, 2, 3, \dots). \quad (\text{B1})$$

Here, δ is the ratio of the number of sites in the motif of second phase to the number of sites in the motif of first phase.

The mixture of phase $S(\frac{4}{15}; \frac{1}{2}, 0, \frac{5}{4})$ with the weight ε and phase $S(\frac{7}{25}; \frac{3}{7}, 0, \frac{12}{7})$ with the weight $1-\varepsilon$ is phase $S(\frac{7-3\varepsilon}{5(5-2\varepsilon)}; \frac{3-\varepsilon}{7-3\varepsilon}, 0, \frac{12-7\varepsilon}{7-3\varepsilon})$ (see Fig. 20). At the line $J_2 = -\frac{4}{3}J_1 + \frac{8}{3}J_3 - \frac{1}{6}h$, i.e., at the boundary between phases $S(\frac{4}{15}; \frac{1}{2}, 0, \frac{5}{4})$ and $S(\frac{7}{25}; \frac{3}{7}, 0, \frac{12}{7})$, all mixtures of these phases have the same energy. Hence there is no first-order phase transition between these phases.

- [1] Y. Han, Y. Shokef, A. M. Alsayed, P. Yunker, T. C. Lubensky, and A. G. Yodh, *Nature (London)* **456**, 898 (2008); Y. Shokef and T. C. Lubensky, *Phys. Rev. Lett.* **102**, 048303 (2009).
- [2] G. H. Wannier, *Phys. Rev.* **79**, 357 (1950); *Phys. Rev. B* **7**, 5017 (1973).
- [3] R. Soto, G. Martínez, M. N. Baibich, J. M. Florez, and P. Vargas *Phys. Rev. B* **79**, 184422 (2009); Yuri B. Kudasov, *Phys. Rev. Lett.* **96**, 027212 (2006).
- [4] B. Mihura and D. P. Landau, *Phys. Rev. Lett.* **38**, 977 (1977).
- [5] Xiaobao Yang and Jun Ni, *Phys. Rev. B* **67**, 195403 (2003).
- [6] A. H. Thompson, *Phys. Rev. Lett.* **40**, 1511 (1978).
- [7] J. Kanamori, *Prog. Theor. Phys.* **35**, 16 (1966).
- [8] B. D. Metcalf, *Phys. Lett.* **46A**, 325 (1974).
- [9] M. Kaburagi and J. Kanamori, *Jpn. J. Appl. Phys., Suppl.* **2**, 145 (1974).
- [10] U. Brandt and J. Stolze, *Z. Phys. B: Condens. Matter* **64**, 481 (1986).
- [11] Yu. I. Dublenych, *Phys. Rev. B* **71**, 012411 (2005).
- [12] M. Sanati, L. G. Wang, and Alex Zunger, *Phys. Rev. Lett.* **90**, 045502 (2003).
- [13] J. Slawny, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. Lebowitz (Academic Press, New York, 1985), Vol. 11.
- [14] C. Gruber, J. Jędrzerewski, and P. Lemberger, *J. Stat. Phys.* **66**, 913 (1992).
- [15] T. Kennedy, *Rev. Math. Phys.* **6**, 901 (1994).
- [16] A. S. Keys and S. C. Glotzer, *Phys. Rev. Lett.* **99**, 235503 (2007).
- [17] P. J. Steinhardt, *Nature (London)* **452**, 43 (2008).
- [18] M. O. Blunt, J. C. Russell, M. C. Giménez-López, J. P. Garrahan, X. Lin, M. Schröder, N. R. Champness, and P. H. Beton, *Science* **322**, 1077 (2008).
- [19] R. Otero, M. Lukas, R. E. A. Kelly, W. Xu, E. Lægsgaard, I. Stensgaard, L. N. Kantorovich, and F. Besenbacher, *Science* **319**, 312 (2008).
- [20] D. P. Landau, *Phys. Rev. B* **27**, 5604 (1983).
- [21] M. Sandhoff, H. Pfnur, and H.-U. Everts, *Europhys. Lett.* **25**, 105 (1994); J. Glosli and M. Plischke, *Can. J. Phys.* **61**, 1515 (1983).
- [22] M. Kaburagi and J. Kanamori, *J. Phys. Soc. Jpn.* **44**, 718 (1978).
- [23] K. Nakanishi and H. Shiba, *J. Phys. Soc. Jpn.* **51**, 2089 (1982).
- [24] M. E. Fisher and W. Selke, *Philos. Trans. R. Soc. London, Ser.*

- A **302**, 1 (1981); Phys. Rev. Lett. **44**, 1502 (1980).
[25] J. Kanamori, J. Phys. Soc. Jpn. **53**, 250 (1984).
[26] T. Tonegawa, M. Kaburagi, and J. Kanamori, J. Phys. Soc. Jpn. **59**, 1660 (1990).
[27] P. Bak, Phys. Today **39**(12), 38 (1986).
[28] M. Hupalo, J. Schmalian, and M. C. Tringides, Phys. Rev. Lett. **90**, 216106 (2003); M. Yakes, V. Yeh, M. Hupalo, and M. C. Tringides, Phys. Rev. B **69**, 224103 (2004).