Stable Quasicrystalline Ground States

Jacek Miękisz
Institut de Physique Théorique
Université Catholique de Louvain
1348 Louvain-la-Neuve, Belgium

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Abstract. We give a strong evidence that noncrystalline materials such as quasicrystals or incommensurate solids are not exceptions but rather are generic in some regions of a phase space. We show this by constructing classical lattice gas models with translation-invariant finite range interactions and with unique quasiperiodic ground states which are stable against small perturbations of finite range potentials.

1 Introduction

One of the important problems in physics is to understand why matter is crystalline at low temperatures [1, 2, 3, 4, 5, 6, 7]. It is traditionally assumed (but has never been proved) that at zero temperature minimization of the free energy of a system of many interacting particles can only be obtained by their periodic arrangements (a perfect crystal) which at nonzero temperature is disrupted by defects due to entropy. Recently, however, there has been a growing evidence, that this basic phenomenon, the crystalline symmetry of low temperature matter, has exceptions; in particular incommensurate solids [8] and, more recently, quasicrystals [9]. It is very important to find out how generic are these examples. In other words, is nonperiodic order present in these systems stable against small perturbations of interactions between particles?

The problem of stability of quasiperiodic structures was studied recently in continuum models of particles interacting through a well, Lennard-Jones, and other potentials [10, 11, 12, 13, 14, 15]. However, no final conclusion was reached. After all one has to compare chosen quasiperiodic structure with all possible arrangements of particles in the space, a really formidable task.

Here we will present two classical lattice gas (toy) models with unique stable nonperiodic ground states. More precisely, every site of the simple square lattice can be occupied by one of several different particles. The particles interact through two-body finite range translation-invariant potentials. Our models has only nonperiodic ground state configurations (infinite lattice configurations minimizing the energy density of the system). However, they all look the same: there is a unique translation-invariant ground state measure supported by them and which is a zero temperature limit of the grand canonical ensemble. We will prove that if one perturbs our models a little bit, by introducing sufficiently small chemical potentials or two-body translation-invariant interactions, their ground states do not change. It means that there is an open ball in the space of finite range interactions (with a fixed range) without periodic ground states. This constitutes a first generic counterexample to the crystal problem which is an attempt to deduce, within statistical mechanics, periodic order in systems of many interacting particles.

In Section 2 we introduce a strict boundary property for local ground states and explain why it implies stability of our nonperiodic ground states. In Section 3 we describe the main features of Robinson's nonperiodic tilings of the plane, construct a classical lattice gas model out of it, and show why its unique ground state is not stable against small perturbations of nearest neighbor potentials. In Section 4 we present a modification of Robinson's tilings which allows us to construct models with unique nonperiodic ground states satisfying the strict boundary property. Section 5 contains a short discussion.

2 Classical Lattice Gas Models and Nonperiodic Ground States

A classical lattice gas model is a system in which every site of a lattice Z^d can be occupied by one of n different particles. An infinite lattice configuration is an assignment of particles to lattice sites, that is an element of $\Omega = \{1, ..., n\}^{Z^d}$. Particles can interact through two-body finite range translation-invariant potentials $f(\underline{x} - \underline{y})$. Configurations of particles minimizing the potential energy density of a Hamiltonian $H = \sum_{(\underline{x},\underline{y})} f(\underline{x} - \underline{y})$ are called ground state configurations. For any potential the set of ground state configurations is nonempty but it may not contain any periodic configurations [16, 17, 18, 19, 20, 21, 22, 23]. We restrict ourselves to systems in which, although all ground state configurations are nonperiodic, there is a unique translation-invariant measure (called a ground state) supported by them. The unique ground state is then inevitably a zero temperature limit of an infinite volume grand canonical probability distribution. This is in analogy with the Ising antiferromagnet, where there are two alternating ground state configurations but only one ground state measure which is just their average. Such ground state configurations have then necessarily uniformly defined frequencies for all finite arrangements of particles. More precisely, to find a frequency of a finite arrangement in a given configuration we first count the number of times it appears in a box of size l and centered at the origin of the lattice, divide it by l^d, and then take the limit $l \to \infty$. If the convergence is uniform with respect to the position of the boxes then we say that the configuration has a uniformly defined frequency of this arrangement. It is a main point of this paper to claim that stability of nonperiodic ground states is intimitely connected with the rate of this convergence.

Definition: Let ρ be a unique ground state measure of a finite range potential. Let $X(A)$ be a configuration on a finite region A of the lattice enclosed by a perimeter P and such that all interactions in A attain their minimal values. $X(A)$ is then a local ground state configuration but might not be extendable to any infinite lattice ground state configuration in the support of ρ. We say that a model satisfies a strict boundary property for local ground states if for any particle or a nearest neighbor pair of particles (any finite arrangement in general) the number of its appearance n in A satisfies the following inequality: $|n - wA| < CP$, where w is a frequency of the arrangement in ρ (frequency in any infinite lattice ground state configuration in the support of ρ), and C is a constant which depends only on the arrangement. $\square$

Now, we claim that the unique ground state of the model satisfying the strict boundary property for local ground states is stable in the space of nearest neighbor (finite range in general) interactions. For the proof assume otherwise, that a perturbed model has a different ground state. This implies the existence of a ground state configuration with a nonzero density of nearest neighbor pairs of particles with nonminimal energies of interactions - the so called broken bonds. The absence of broken bonds is a complete characterization of the original ground state as explained in Sec.3. Now, every broken bond introduces a positive bounded below energy, say 1. It is a unit of a boundary of a local ground state configuration which can decrease energy by an amount at most proportional to the strength of a perturbation which is a consequence of the strict boundary property. It follows that for sufficiently small perturbations the energy of a configuration is proportional to the total length of broken bonds (a Peierls condition is satisfied). Hence for sufficiently small perturbations the hypothetical ground state has an energy density bigger than the original one which is a contradiction.

3 Robinson's Tilings and Unstable Ground States

To construct our classical lattice gas model we will use Robinson's nonperiodic tilings of the plane [24, 25]. He designed 56 square-like tiles such that using an infinite number of their copies one can tile the plane only in a nonperiodic fashion. We describe now the Robinson tiles; we follow [24] closely. There are five basic tiles represented symbolically in Fig.1. The rest of them can be obtained by rotations and reflections. The first tile on the left is called a cross; the rest are called arms. All tiles are furnished with one of the four parity markings shown in Fig.2. Crosses can be combined with the parity marking at the lower left in Fig.2. Vertical arms (the direction of long arrows) can be combined with the marking at the lower right and horizontal arms with the marking at the upper left. All tiles may be combined with the remaining marking. Two tiles "match" if arrow heads meet arrow tails separately for the parity markings and the markings of crosses and arms.

We now describe main features of Robinson's nonperiodic tilings. Let us observe that if the plane is tiled with tiles with the above parity markings then these must alternate both horizontally and vertically in the manner shown in Fig.2. Now, every odd-odd position on the Z^2 lattice (if columns and rows are suitably numbered) is occupied by crosses in relative orientations as in Fig.3. (crosses are denoted there by $\lfloor$ with line segments corresponding to double arrows). They form a periodic configuration with period 4. Then in the center of each "square" matching rules force a cross such that the previous pattern reproduces but this time with period 8. Continuing this procedure (the so called principle of expanding squares) infinitely many times we obtain a nonperiodic configuration. It has built in periodic configurations of period 2^n, $n \geq 2$ with tiles on sublattices of Z^2 as shown in Fig.4.

Now using Robinson's tiles we construct a lattice gas model [16, 17, 18, 19, 20, 21, 22, 23]. Every site of the square lattice can be occupied by one of the 56 different particles-tiles. Two nearest neighbor particles which do not "match" contribute a positive energy, say 1; otherwise the energy is zero. Such a model obviously does not have periodic ground state configurations. In fact, there is a one-to-one correspondence between infinite tilings and configurations in the support of the unique ground state measure. It has been proven [19] that the unique ground state of this model is not stable against small perturbations of chemical potentials. Really, let us introduce a small negative chemical potential h for arms with a single long arrow (called later single arms). Notice that every ground state configuration contains an arbitrarily long sequence of arms with two long arrows (called later double arms). If we change such sequence of length n into a sequence of single arms we lower the energy by nh paying only at two endpoints of the sequence (the model does not satisfy the strict boundary property). So no matter how small is h one can always change a ground state configuration locally to lower its energy. This shows, in fact, that densities of single and double arms change continuously with respect to their chemical potentials. This continuous change in stoichiometries at zero temperature is a behavior not present in any previously known models and real alloys.

4 Stable Quasiperiodic Ground States

We will construct models for which introducing a chemical potential for one kind of arms (any two-body interaction in general) does not change their ground states. Simply getting rid of, say, double arms will not work because these are exactly double arms which create squares at whose centers crosses are forced. The absence of double arms will destroy the principle of expanding squares and we will no longer have a unique nonperiodic ground state. However, let us notice that the forcing is done by four arms located exactly in the middle between crosses forming a square. Now, the main idea of our construction is to force similar arms in the middle of every side of every square on every scale as in the Robinson original tiling, and to do it not by sequences of single and double arms but by sequences of new markings. These will be sequences of repeated A, B, and C markings (explained later in detail) propagating from crosses. Whenever two B or C markings meet at the middle of a side of a square then our special arms (described later) are forced there. Observe, that we do not have long sequences of different arms anymore, just three translates of $...ABCABC...$ lines. Obviously, it is impossible to distinguish between the translates by a chemical potential and also by two-body interactions because of the "incommensurability" of 2^n "periods" of Robinson's tilings and period 3 of the lines, and the symmetry of tiles.

MODEL I

Let us now describe tiles of our first model. We have modified crosses and arms, and we have the parity markings as before. In addition, we introduce two levels of letter markings. Each side of each tile is marked by A, B, or C and by T, O, M, E, or K (not all choices are present as explained below). All possible markings of crosses are shown in Fig.5. Let us stress at this point that we allow all rotations of crosses but we do not allow any reflections which makes our model somewhat different from the original Robinson one. Now, we have 15 types of arrows present in arms: all combinations of $A \rightarrow B$, $B \rightarrow C$, $C \rightarrow A$ and $T \rightarrow O$, $O \rightarrow M$, $M \rightarrow E$, $E \rightarrow K$, $K \rightarrow T$. Line segments present in arms are marked $A - A$, $B - B$, or $C - C$; $B - B$ and $C - C$ segments are also marked $T - O$ or $O - T$ as shown in Fig.6. Again, we do allow rotations but not reflections. Now, two tiles match if in addition to previous requirements letter markings match separately on both levels. We also assume that line segments $A - A$ can be put next to any letter marking of the second level (this is not a tiling condition). Our matching rules can be translated as before into a lattice gas model with nearest neighbor interactions. In addition we have to take care of the fact that letter markings of the second level which are opposite nearest neighbors of $A - A$ line segments are not forced to be correlated. We introduce a two-body interaction of range 2 between two arrows facing one another such that the energy is zero if one of them has T marking and the other one is marked by O; otherwise the energy is 1.

Now we will show that the only ground state configurations of our model are those of Robinsons's type shown in Fig.4. More precisely, looking at crosses and ignoring letters and A arrows on every sublattice $2^n Z^2$ with an odd n and C arrows for an even n we see the self-similar structure of expanding squares shown in Fig.4.

Proposition 1 *The unique ground state measure of Model I is of Robinson's type as described above.*

Proof: First, let us recall that the parity markings force crosses to occupy every site of a $2Z^2$ sublattice. Letter markings of the second level allow there only crosses which are rotations of the cross at the upper left in Fig.5. Now, arms with the upper left and lower right parity marking force relative orientations of these crosses to be such that squares of length 2 are created as shown in Fig.3 (letters, arrows with A markings and arms are not drawn). The above arms with $C-C$ line segments force crosses to appear in the center of every square just created. New crosses (four rotations of the cross at the upper right in Fig.5) form a $4Z^2$ sublattice. Now, the nature of our interactions is such that there are sequences of arrows propagating from every cross and meeting half-way through line segments of arms. For the crosses on a $2^n Z^2$ sublattice with an odd n these are alternating $C-C$ and $A-A$ line segments and alternating $B-B$ and $A-A$ ones for an even n. $C-C(B-B)$ segments, located in the middle of four sides of the squares they have created, force crosses in their centers and together with $A-A$ segments force them to have relative orientations on every sublattice as in Fig.3. Crosses on succesive sublattices have to be chosen in the clockwise order from Fig.5. The upper left corner of every square has to be occupied by an appropriate cross from Fig.5 and other corners by its clockwise rotations. Inductively an infinite nonperiodic configuration is created. □

Proposition 2 *Model I satisfies the strict boundary property for local ground states if in counting particles and pairs of particles we do not distinguish between their different rotations and reflections.*

Proof: Let a broken bond be a unit segment on the dual lattice separating two nearest neighbor particles with a positive interaction energy (a common side of two nearest neighbor tiles which do not match). Let us divide A into connected components without broken bonds (two lattice sites are connected if they are nearest neighbors) such that on every component crosses having relative orientations shown in Fig.3 form a $2Z^2$ sublattice. This can be achieved by paths on the lattice dual to Z^2 with lengths smaller than 16 and joining broken bonds. When one considers only $2Z^2$ sublattices and lines of arms joining their sites the deviation of the number of the appearance of a given particle from the number of its appearance in a global ground state configuration is not bigger than 16 times the number of the above paths. Now, we decompose further every connected component into connected components such that crosses form there $4Z^2$ sublattices. This time it is achieved by paths on the dual lattice with lengths smaller than 32 and again joining broken bonds. Now, when one considers $4Z^2$ sublattices and lines of arms joining their sites, the additional deviation of the occurence of our particle is not bigger than 16 times the number of new paths plus 16/2 times the number of old paths. At the $n-th$ step we divide every connected component constructed so far into connected components with crosses forming 2^nZ^2 sublattices and using paths with lengths smaller than 2^{n+3}. Again, the additional deviation of the occurence of our particle is bounded by 16 times the number of new paths plus 1/2 of the deviation at the $n-1th$ step. We repeat this process m times, where m is the smallest number such that $diamA < m$. Now, the total number of all paths is bounded by 3 times the number of broken bonds (broken bonds and paths can be regarded as vertices and edges of a planar graph and our bound follows from Euler's formula). This shows that $|n - wA| \leq 96P$, where w is a frequency of the particle in a global ground state and n the number of its appearance in $X(A)$. Similarly one can obtain the same bound for any two-particle arrangements. □

Let us notice that our model does not satisfy the strict boundary property for three-particle arrangements which leads to instability in the space of three-body interactions. To see this let us look at a line of arms with the length k. T and O markings of arrows of tiles immediately above form a nonperiodic sequence with the following property: if a certain fraction of particles is ignored the rest of a ground state configuration is periodic; the smaller the fraction, the larger the period. When one just looks at two-particle arrangements than the above proof applies because for every T above the line there O below it and vice versa, and in counting arrangements we do not distinguish between reflected ones. Let us look now at two consecutive markings and create a nonperiodic sequence of two symbols a and b, where a corresponds to TT and OO, and b to OT and TO in the original sequence. Shifting the line of arms we may then increase up to $\log k$ (one for every sublattice $2^n Z^2$, $2^n \leq k$ the number of, say, $A \rightarrow B$ arrows with the a symbol immediately above them. The shift creates only two contours hence it follows that the strict boundary property is not satisfied.

Our first zero temperature stability result will be stated in the space of covariant interactions. That it is to say that a chemical potential of a particle-tile or a two-body interaction between a pair of particles should be the same for any of their rotations and reflections. The following theorem follows directly from the strict boundary property as shown in the introduction.

Theorem 1 *The unique nonperiodic ground state of Model I is the unique ground state for any sufficiently small two-body covariant perturbation.*

MODEL II

Let us now describe tiles of our second model. We have modified crosses with A and C arrows shown in Fig.7, arms with $A \rightarrow B$, $B \rightarrow C$, and $C \rightarrow A$ arrows and $A - A$, $B - B$, and $C - C$ line segments shown in Fig.8 and 9. Arms in Fig.7 are combined with the upper right parity marking and in addition an arm at the bottom left can be combined with the lower right or upper left parity marking. Arms in Fig.8 can be combined with the lower right or upper left parity marking. Crosses are combined with the parity markings as before. We allow all rotations and reflections of our new tiles as in the original Robinson model.

We do not have anymore the previous second level of letter markings. $B - B$ and $C - C$ line segments, not located in the middle of arms anymore, still force crosses to create squares but symmetric $A - A$ arms cannot prevent these squares going out of phase; a fault shown in Fig.10 can be created. The presence of such a fault does not produce any broken bonds if and only if all line segment on it are of the $A - A$ form. It follows that in any tiling there may be at most one fault so they have zero probability of occurence with respect to the unique translation-invariant measure supported by tilings. For exactly the same reasons such faults are present in some of the original Robinson tilings. Therefore as in the previous model the unique ground state of Model II is of the Robinson type. Looking at crosses and ignoring letter markings and A arrows on every sublattice $2^n Z^2$ with an odd n and C arrows with an even n we again see the self-similar structure of expanding squares shown in Fig.4.

Proposition 3 *Model II satisfies the strict boundary property for local ground states if in counting particles and pairs of nearest neighbor particles we do not distinguish between their different rotations and reflections.*

Proof: The first step is the same as in the proof of Proposition 1. In the second step we divide further connected components such that on every component crosses form $4Z^2$ sublattices but not necessarily having relative orientations as in Fig.3; faults may be present. We may have an arbitrarily long fault along perpendicular $A-A$ line segments without any broken bonds except at the endpoints of the fault. The presence of two arms on the fault with line segments parallel to the fault forces a cross between them and then a broken bond is forced at a distace smaller than 16 from the cross as can be seen from Fig.10. Arms with $B-B$ or $C-C$ line segments perpendicular to the fault also force a cross on the fault. It follows that when one considers only $4Z^2$ sublattices and lines of arms joining their sites the deviation of the number of the appearance of a given particle from the number of its appearance in a global ground state configuration is not bigger than 16 times the number of paths in the present decomposition plus 16/2 times the number of paths in the first step. (there is no change of numbers of particles and pairs of nearest neighbors along a fault with $A-A$ line segments perpendicular to it). Now, we decompose further every connected component into connected components such that crosses form there $8Z^2$ sublattices which of course go out of phase along previous faults and they may also create they own faults. The induction based on self-similarity of our tilings can now be applied to obtain the same bound as before. $\Box$

The following theorem follows.

Theorem 2 *The unique nonperiodic ground state of Model II is a unique ground state for any sufficiently small nearest-neighbor covariant perturbation.*

Modifying slightly the previous argument involving three-body interactions it can be shown that the ground state of the second model is not stable against arbitrarily small perturbations of range 2.

Let us notice that particles of our models have shapes and interactions between them are covariant but not symmetric: an interaction between nearest neighbors RP is the same as between   but not necessarily the same as between PR. There is however a construction due to Radin [17] who by enlarging a family of possible particles makes them shapeless and interactions between them fully symmetric (dependent only on the distance between particles). He constructs in this way a classical lattice gas model with a finite range fully symmetric interactions and with unique nonperiodic ground state. His method requires the original family of particles-tiles to be both rotation and reflection invariant. Therefore it can be applied only to our second model. We use it to construct a model with shapeless particles and with a unique nonperiodic ground state which is stable in a space of fully symmetric interactions.

Theorem 3 *The unique ground state of a symmetric version of Model II is stable in the space of symmetric interactions with the range smaller than three lattice spacings.*

5 Conclusions

We have constructed finite range classical lattice gas models with unique quasiperiodic ground states which are stable against small perturbations of interactions.

In the construction of our models we used the presence of order in nonperiodic ground state configurations. Although nonperiodic they are not however completely chaotic. They exhibit long-range positional order in the sense that states at distant regions are correlated. In fact, our ground state configurations possess highly ordered structures: if a certain fraction of particles is ignored the rest of a ground state configuration is periodic; the smaller the fraction, the larger the period. It would be interesting to see if other, more disordered ground states could be stable. Modifying the principle of expanding squares responsible for nonperiodicity of Robinson's tilings a large family of models without periodic ground states was recently constructed [26, 23, 27]. In another class of models we have ones based on Penrose tilings but with particles on the simple square lattice. Low temperature behavior of such models was recently studied in [28]. Their stability would be especially interesting in connection with quasicrystals. It was proven recently [29] that Penrose matching rules satisfy the strict boundary property for finite regions of perfect infinite tilings. It is not known if this is also true for local tilings - ground states.

Finally, let us say few words about positive temperatures, where to construct stable equilibrium phases of a system of many interacting particles one has to minimize not the energy but the free energy of the system taking into account disruptions due to entropy. Here one of the important problems is to construct a quasiperiodic equilibrium phase for a finite range interaction - a microscopic model of a quasicrystal. We conjecture the existence of quasiperiodic equilibrium phases in a modified three-dimensional version of our second model.

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