CHAOTIC LATTICE - GAS MODEL

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Abstract

A nonuniform system is considered consisting of two phases with different densities of particles. At each given time the distribution of the phases in space is chaotic: each phase filling a set of regions with random shapes and locations. A chaotic diffusion process intermixes these regions by varying their shapes and locations in a random way. To investigate the statistical properties of such a system, it is exemplified by a lattice–gas model. Conditions are analysed when this chaotic lattice–gas model can become thermodynamically more stable than the usual model describing a pure one–phase system.
1 Introduction

According to the space distribution of particles, there are three types of stable matter: uniform, periodic and quasiperiodic. The uniform matter is presented by gases and fluids. This can be also called plasma if it consists of charged particles. Periodic systems are crystals and quasiperiodic ones are incommensurate crystals and quasicrystals [1].

Amorphous solids, or glasses, are not stable, but they are metastable. Metastable systems, contrary to stable ones, have finite lifetimes, although the latter can be quite long, as it is, e.g., for the window glass. A more detailed description of metastable systems can be found in reviews [2,3].

The dependence of system characteristics on space variables can be analysed from the point of view of dynamical theory by analogy with the dependence of trajectories on time. Basing on this analogy, amorphous solids could be called turbulent crystals [4]. The breaking of a regular periodicity in crystals by defects can be named the defect turbulence [5]. And an irregular distribution of particles, which is neither uniform nor periodic or quasiperiodic, is called chaotic [6].

As far as amorphous solids, having chaotic distribution of particles in space, are metastable, their consideration in time is limited by their lifetime. The state, existing during a finite time $\tau_f$ that is much longer than an observation time $\tau_{obs}$ is called frozen ($\tau_{obs} \ll \tau_f$). A frozen chaotic system, after its lifetime, transforms, as time goes to infinity, into either uniform or periodic (quasiperiodic) system. This can be equivalently said that a frozen chaotic system is thermodynamically unstable.

The question arises whether only uniform, periodic and quasiperiodic space structures can be stable, or a spatially chaotic system also can exist an infinite time. An analogous question has been put forward by Ruelle [4] who has asked whether there can exist a frozen turbulent structure with infinite lifetime. What is known now about the real as well as model systems offers the negative answer to the Ruelle question, although this does not prove, of course, that such a situation is principally impossible. The question that we pose here is more general: we ask whether there can exist a thermodynamically stable chaotic system (not necessarily frozen).

Let us concretize this question. Suppose $\rho(\vec{r}, t)$ is the distribution of particle density as a function of the real-space vector, $\vec{r}$, and time, $t$. Assume that the limit

$$\lim_{t \to \infty} \rho(\vec{r}, t) = \rho(\vec{r})$$

exists. If the resulting state of the system with the density $\rho(\vec{r})$ is stable against
small perturbations and has infinite lifetime, it is called thermodynamically stable. One can define a thermodynamic potential enjoying the property of convexity and being minimal for a thermodynamically stable system. The latter usually has well defined symmetry properties governing the behavior of the density $\rho(\vec{r})$ as a function of space coordinates $[1,3,7]$. If the system is uniform, then the density is translationally invariant with respect to an arbitrary space vector $\vec{r} \in \mathbb{R}^3$, that is

$$\rho(\vec{r}) = \rho(0). \quad \text{(uniformity)}$$

A periodic structure, or a crystal, is described by a basis $\{\vec{b}_\alpha \mid \alpha = 1, 2, 3\}$ of generating lattice vectors $\vec{b}_\alpha$, with respect to which the density is periodic,

$$\rho(\vec{r}) = \rho(\vec{r} + \vec{b}_\alpha), \quad \text{(periodicity)}$$

implying that $\rho(\vec{r})$ is not constant.

A quasiperiodic structure is characterized by a basis $\{\vec{b}_\alpha \mid \alpha = 1, 2, \ldots, p; p > 3\}$ of integrally independent vectors $\vec{b}_\alpha$ such that the density can be expressed as a $p$-fold Fourier series

$$\rho(\vec{r}) = \sum_{n_1 \ldots n_p} \rho_{n_1 \ldots n_p} e^{i(n_1 \vec{g}_1 + n_2 \vec{g}_2 + \ldots + n_p \vec{g}_p) \cdot \vec{r}}, \quad \text{(quasiperiodicity)}$$

in which $n_\alpha$ are integers and the set $\{\vec{g}_\alpha\}$ is the reciprocal basis defined by the equations

$$\vec{g}_\alpha \cdot \vec{b}_\alpha = 2\pi \quad (\alpha = 1, 2, \ldots, p > 3).$$

The integral independence of $\vec{b}_\alpha$ means that the linear combination

$$n_1 \vec{b}_1 + n_2 \vec{b}_2 + \ldots + n_p \vec{b}_p \neq 0$$

is nonzero for any nontrivial $p$-tuple of integers $(n_1, n_2, \ldots, n_p)$, i.e. when at least one of $n_\alpha$ is nonzero. Note that the structure is quasiperiodic if and only if the rank $p$ of the basis $\{\vec{b}_\alpha\}$ is greater than the space dimensionality: $p > 3$. Although, probably, the majority of quasiperiodic structures are metastable, they also can indeed form stable equilibrium states [8], with free energies lower than those of the conventional crystalline structures. Recall that if $\rho(\vec{r})$ is chaotic in real space, as it is for different glasses, e.g. metallic glasses, vitreous silica or amorphous polymers [9,10], then such a system is always thermodynamically metastable.
Suppose now that the particle density does not tend to a stationary limit as time tends to infinity,
\[ \rho(\vec{r}, t) \to \rho_c(\vec{r}, t); \quad t \to \infty, \]
but displays spatiotemporal chaos \[5\]. The average density of the whole system is assumed to be fixed,
\[ \frac{1}{V} \int \rho(\vec{r}, t) d\vec{r} = \frac{1}{V} \int \rho_c(\vec{r}, t) d\vec{r} = \rho; \]
here \( V \) is the system volume. A function \( \rho(\vec{r}, t) \) for a given \( \vec{r} \), exhibits temporal chaos if its time motion is locally unstable. We may say that the motion is locally unstable, or locally chaotic, if a local Lyapunov exponent is positive:
\[
\lambda(\vec{r}) \equiv \lim_{t \to \infty} \lim_{\delta \rho(\vec{r}, t_0) \to 0} \frac{1}{t} \ln \left| \frac{\delta \rho(\vec{r}, t_0 + t)}{\delta \rho(\vec{r}, t_0)} \right| > 0.
\]

The case when \( \rho(\vec{r}, t) \), under fixed \( \vec{r} \), tends to a time-independent limit \( \rho(\vec{r}) \), as \( t \to \infty \), describes the passage of a system to an equilibrium state. In the language of dynamical theory such a state corresponds to an attractor of fixed-point type \[3\]. Thus, the equilibrium uniform, spatially periodic and spatially quasiperiodic states are fixpoint attractors.

The situation when a system tends, as time increases, to a chaotic state, may be associated with the existence of a chaotic attractor. In a system with dissipation the chaotic attractor is fractal, or strange \[11\]. As far as the equations of motion for \( \rho(\vec{r}, t) \) are partial differential equations involving derivatives with respect to space variables as well as with respect to time, we have to speak about spatiotemporal chaos. Such evolution equations correspond to what is called infinite-dimensional dynamical systems \[12-14\].

A chaotic state can be averaged with respect to time. For example, for the particle density we may define
\[
\bar{\rho}(\vec{r}) \equiv \lim_{\tau \to \infty} \frac{1}{\tau} \int_0^\tau \rho(\vec{r}, t) dt.
\]
The average state, independent of time, can be put into correspondence with an effective equilibrium state, or the state equilibrium on the average \[3\], for which an effective free energy can also be defined. When the latter is lower than the free energy of any equilibrium state, whether uniform, spatially periodic or spatially
quasiperiodic, then such a state is thermodynamically, or globally, stable, although
dynamically, or locally, it is unstable.

The question we suggest in this paper is as follows: *Can a statistical system with
spatiotemporal chaos be globally stable?* We construct and analyse a model giving
the positive answer to this question.

2 Two-density system

Before considering a particular realization of a chaotic statistical system it is worth
paying some attention to terminology. Since we are dealing not with simple
dynamical systems but with statistical systems containing infinitely many degrees of
freedom, we can call the chaos appearing in the latter the *statistical chaos*. This
chaotic state evolves in a system automatically, without any special fine-tuning
of external fields; hence this can be named the *self-organized chaos*. The time
duration of chaotic fluctuations and their spatial sizes are distributed in a quite
large scale, somewhat similar to microscopic critical fluctuations of equilibrium sys-
tems, thus, one could use the term the *critical chaos*. This should be distinguished
from the self-organized criticality [15] which is a generalization of the equilibrium
critical state to nonequilibrium systems. The main feature distinguishing the chaos
we consider is that, although each chaotic fluctuation can have any duration, size,
and shape, but there can be defined, nevertheless, their characteristic, or average,
lifetime, $\tau_f$, and linear size, $l_f$, satisfying the inequalities

$$\tau_{loc} \ll \tau_f \ll \tau_{obs}, \quad a_0 \ll l_f \ll L,$$

in which $\tau_{loc}$ is called the local equilibrium time; $\tau_{obs}$, the observation time, $a_0$ is
the average distance between the objects composing the system, and $L$ is a linear
size of the latter [3]. Because of this intermediate, or mesoscopic, character, it is
logical to title the considered phenomenon the *mesoscopic chaos*. One of the most
important observable quantities characterizing the properties of a system is the order
parameter, generally, being a function of space and time variables. The examples
are: the particle density $\rho(\vec{r},t)$ discussed above, the local magnetization, polar-
ization or superconducting gap. In chaotic systems, the mesoscopic fluctuations of
order parameters are clearly interpreted as fluctuating nuclei, or gems, of competing
phases. Therefore, such a mesoscopic chaos can be called as well the *heterophase
chaos*. 


Now, let us return to the concrete case when the chaos in a system is related to mesoscopic fluctuations of the particle density $\rho(\vec{r},t)$. Suppose that at each fixed time the function $\rho(\vec{r},t)$ describes a random distribution in space of two regions with different densities, $\rho_1$ and $\rho_2$, such that

$$\rho_1 > \rho_2 \quad (\rho_\nu \equiv \frac{N_\nu}{V_\nu}),$$

(1)

where $N_\nu$ and $V_\nu$ are the total number of particles and the volume, respectively, corresponding to a $\nu$-phase ($\nu = 1, 2$). One can distinguish the regions occupied by different phases owing to the mesoscopic nature of the density fluctuations considered. The latter are accompanied, of course, by the fluctuations of other physical quantities, for example, by rare large energy fluctuations [16,17]. However, we shall speak in what follows mainly about the mesoscopic density fluctuations since it is just density that plays the role of a natural parameter distinguishing different phases.

A physical meaning which we could ascribe to these phases with two different densities is by calling the denser phase liquid and the less dense one gas. Then, we could imagine that our system is a kind of fog consisting of liquid droplets inside gas. Imagining this, we should not identify our chaotic fog with the real fog corresponding rather to a frozen droplet configuration, as the average lifetime of real droplets is of the order or longer than the observation time. As is discussed in the Introduction, such a system with a frozen space disorder is metastable, as the real fog is. However, some thermodynamic peculiarities of our chaotic fog can be related to the so-called droplet singularities [18,19] of liquid-gas systems.

When a persistent chaotic state develops in a system, this means that such a state is a chaotic attractor. In dissipative systems, chaotic attractors are fractal, or strange [11,12,20]. A chaotic attractor, being metrically indecomposable (metrically transitive) allows to define an invariant probability measure. The property of metric transitivity for the considered case of a system with heterophase chaos is what has been called [3] the heterophase quasiergodicity. For a metrically transitive system, time averages can be replaced by ensemble averages. To this end, we have to introduce a probability measure describing the distribution of phases in space. The differential probability measure can be written as

$$D\mu(\xi) = \hat{\rho}_{loc}(\xi) D\xi,$$

where $\hat{\rho}_{loc}(\xi)$ is a locally-equilibrium density matrix, and $D\xi$ is a functional
differential over a set
\[ \xi = \{ \xi_\nu(\vec{r}) | \nu = 1, 2, \ldots; \vec{r} \in V \} \]
of the manifold indicator functions
\[ \xi_\nu(\vec{r}) = \begin{cases} 1, & \vec{r} \in V_\nu \\ 0, & \vec{r} \notin V_\nu \end{cases} \]
\( V \) and \( V_\nu \) meaning the whole volume occupied by the system, and a region of the volume filled by a \( \nu \)-phase, respectively [3]. The averaging over the ensemble of phase configurations makes it possible to define an effective phase-replica system that is equilibrium on the average [3]. The procedure of overaging over phase configurations with all foundations and details has been expounded in a series of papers [21-24] and reviewed in [3]. As a result, we can define an effective phase-replica Hamiltonian which for the two-phase case takes the form
\[ \tilde{H} = H_1 \oplus H_2, \]
\[ H_\nu = w_\nu \int \psi_\nu^\dagger(\vec{r}) \left[ K(\vec{r}) - \mu \right] \psi_\nu(\vec{r}) d\vec{r} + \]
\[ \frac{1}{2} w_\nu^2 \int \psi_\nu^\dagger(\vec{r}) \psi_\nu^\dagger(\vec{r'}) \Phi(\vec{r} - \vec{r'}) \psi_\nu(\vec{r'}) \psi_\nu(\vec{r}) d\vec{r} d\vec{r'}, \]
where \( K(\vec{r}) \) is the kinetic-energy operator including external fields, if any; \( \mu \), chemical potential; \( \Phi(\vec{r} - \vec{r'}) \), interaction potential; and \( \psi_\nu(\vec{r}) \) is a field-operator representation for the \( \nu \)-phase. The renormalizing factors \( w_\nu \equiv V_\nu/V \) are the geometric probabilities of the corresponding phases, satisfying the condition
\[ w_1 + w_2 = 1, \quad 0 \leq w_\nu \leq 1, \]
and given by the minimization of the thermodynamic potential
\[ y = -\frac{1}{N} \ln Tr e^{-\beta \tilde{H}} \quad (\beta \Theta \equiv 1), \]
in which \( N \) is the total number of particles in the system, and \( \Theta \) is temperature. The total number of particles is
\[ N = N_1 + N_2; \quad N_\nu \equiv -< \frac{\partial H_\nu}{\partial \mu} >, \]
where \(<\ldots>\) implies the statistical average with the phase-replica Hamiltonian (2). The number of particles in each of the phases is

\[
N_\nu = w_\nu \int <\psi_\nu^\dagger(\vec{r})\psi_\nu(\vec{r})> d\vec{r}.
\] (6)

The mean density of particles in the system is a linear combination

\[
\rho \equiv \frac{N}{V} = w_1 \rho_1 + w_2 \rho_2
\] (7)

of densities

\[
\rho_\nu = \frac{1}{V} \int <\psi_\nu^\dagger(\vec{r})\psi_\nu(\vec{r})> d\vec{r},
\] (8)

which, by definition (1), are different in the two phases.

For a system equilibrium on average we can define all observable quantities as standard statistical averages with the Hamiltonian (2). Thus, for the internal energy per particle we have

\[
E \equiv \frac{1}{N} \left( <\tilde{H}> + \mu N \right) = \frac{1}{N} \left( <H_1> + <H_2> \right) + \mu,
\]

\[
<H_\nu> = w_\nu \left( K_\nu - \mu \frac{\rho_\nu}{\rho} \right) N + w_\nu^2 B_\nu N,
\] (9)

where the notation is used for the mean kinetic energy

\[
K_\nu = \frac{1}{N} \int <\psi_\nu^\dagger(\vec{r})K(\vec{r})\psi_\nu(\vec{r})> d\vec{r},
\] (10)

and mean potential energy

\[
B_\nu = \frac{1}{2N} \int <\psi_\nu^\dagger(\vec{r})\psi_\nu^\dagger(\vec{r}')\Phi(\vec{r} - \vec{r}')\psi_\nu(\vec{r})\psi_\nu(\vec{r}')> d\vec{r}
\] (11)

per particle in each of the phases.

Recall that speaking here about the system equilibrium on average we imply an effective system obtained after averaging over phase configurations which is equivalent to the averaging with respect to time. The actual system describing chaotic motion of two phases is, of course, nonequilibrium [3].
To define the phase probabilities, \( w_\nu \), providing the minimum for the thermodynamic potential (4), under the normalization condition (3), it is convenient to use the notation

\[
w \equiv w_1, \quad w_2 \equiv 1 - w. \tag{12}\]

Then, the extremum of (4) with respect to \( w \) implies

\[
\frac{\partial y}{\partial w} = \frac{1}{N} < \frac{\partial \tilde{H}}{\partial w} > = 0. \tag{13}\]

This, with the Hamiltonian (2), yields

\[
w = \frac{2B_2 + K_2 - K_1 - (\mu/\rho)(\rho_2 - \rho_1)}{2(B_1 + B_2)}. \tag{14}\]

Equation (14), in compliance with notation (12) and assumption (1), defines the probability of the dense phase. The probability of the diluted phase is \( w_2 = 1 - w \).

The phase probabilities given by (12) and (14) provide the minimum of (4) if

\[
\frac{\partial^2 y}{\partial w^2} > 0. \tag{15}\]

The latter inequality is equivalent to

\[
\left\{ < \frac{\partial^2 \tilde{H}}{\partial w^2} > \leq \frac{1}{T} < \left( \frac{\partial \tilde{H}}{\partial w} \right)^2 \right\} > 0. \tag{16}\]

Conditions (15), or (16), are conditions of heterophase stability. The second term in (16) is always non-negative, thence we can write a simplified version of (16),

\[
\left\{ < \frac{\partial^2 \tilde{H}}{\partial w^2} > \right\} > 0, \tag{17}\]

which is a necessary conditions of heterophase stability. The latter, taking account of (2), yields

\[
B_1 + B_2 > 0. \tag{18}\]

In addition, the second property in (3) gives

\[
\frac{\mu}{\rho} (\rho_1 - \rho_2) - 2B_1 \leq K_1 - K_2 \leq 2B_2 + \frac{\mu}{\rho} (\rho_1 - \rho_2). \tag{19}\]
Besides, it is worth checking that the system satisfies the usual conditions of thermodynamic stability, according to which the specific heat and isothermal compressibility are to be positive:

\[ C_\nu = -\Theta \frac{\partial^2 f}{\partial \Theta^2} > 0, \]
\[ \kappa_T = \left( \frac{\partial P}{\rho \partial \rho} \right)^{-1} > 0, \]

(20)

here the free energy per particle, \( f \), and the pressure, \( P \), are related to the thermodynamical potential (4) by the equations

\[ f = y\Theta + \mu, \quad P = \rho^2 \frac{\partial f}{\partial \rho}. \]

The system is called absolutely stable in the thermodynamic sense if its thermodynamic potential is the lowest possible and all stability conditions described above are valid. For brevity, we may call this type of absolute thermodynamic stability the global stability, as compared with the local dynamic instability of chaotic motion. The heterophase chaotic system is dynamically (locally) unstable, while it can be thermodynamically (globally) stable.

3 Lattice - gas model

To get more details of statistical properties for a system with heterophase chaos, let us pass to an approximation leading to what is termed lattice - gas model. To this end, we divide the whole volume of the system, \( V \), into \( N_L \) lattice cells, the density of lattice sites being

\[ \rho_0 \equiv \frac{N_L}{V}. \]

(21)

This quantity can be used for defining the dimensionless densities

\[ n_\nu \equiv \frac{\rho_\nu}{\rho_0}, \quad n \equiv \frac{\rho}{\rho_0} = \frac{N}{N_L}. \]

(22)

With notation (22), we can recast (7) to the equation

\[ n = w_1 n_1 + w_2 n_2, \]

(23)
which may be used for defining the chemical potential $\mu$ as a function of density and temperature, $\mu(n, \Theta)$.

Introduce a set $\{\vec{a}_i \mid i = 1, 2, \ldots, N_L\}$ of vectors corresponding to lattice sites and define the Wannier functions $\varphi_n(\vec{r} - \vec{a}_i)$ with $n$ indexing quantum states. The field operator $\psi_\nu(\vec{r})$ can be written as an expansion

$$\psi_\nu(\vec{r}) = \sum_{i=1}^{N_L} \sum_{n} c_{in\nu} e^{i\nu} \varphi_n(\vec{r} - \vec{a}_i),$$

in which $e_{\nu} = 1, 0$ is a variable telling whether the $i$-lattice cell inside the $\nu$-phase is occupied by a particle or empty, respectively. Assume that each cell can accept not more than one particle, which results in the homeopolarity condition

$$\sum_n c_{in\nu} c_{jn\nu} = 1.$$  \hspace{1cm} (25)

A standard supposition for a lattice-gas approximation [25] is that intercell and interlevel transitions are prohibited. This can be expressed in two ways. One possibility [26] is to require the diagonality of the matrix elements

$$<m|K|n> = \delta_{mn} \delta_{ij} K_i,$$

$$<m, m'|\Phi|n, n'> = \delta_{mn} \delta_{m'n'} \delta_{ij} \Phi_{ij}.$$  

Equivalently, we can define the operators $c_{\nu}$ on a restricted space of states with the restriction

$$\sum_n c_{in\nu} c_{jn\nu} = 1.$$  \hspace{1cm} (25)

Then, (24), (25) and (26) permit to cast each term of (2) into the form

$$H_\nu = w_\nu \sum_{i=1}^{N_L} (K_i - \mu) e_{\nu} + \frac{1}{2} w^2_\nu \sum_{i \neq j} \Phi_{ij} e_{\nu} e_{\nu}.$$  \hspace{1cm} (27)

Thus, we obtain the effective phase-replica Hamiltonian $\overline{H} = H_1 \oplus H_2$, with terms (27), describing the statistical properties of a chaotic lattice-gas model.

In the considered case of heterophase chaos, the system consists of two randomly distributed phases with different densities. The latter are given by (8) which for the lattice gas become

$$\rho_\nu = \frac{1}{V} \sum_{i=1}^{N_L} <e_{\nu}>.$$  \hspace{1cm} (28)
As the dimensionless densities (22) we get
\[ n_\nu = \frac{1}{N_L} \sum_{i=1}^{N_L} <e_{i\nu}>. \] (29)

From the occupation - of - cell variable \( e_{i\nu} = 1, 0 \) one can pass to the spin variable \( \sigma_{i\nu} = \pm 1 \) by using the transformation
\[ e_{i\nu} = \frac{1}{2} (1 + \sigma_{i\nu}), \quad \sigma_{i\nu} = 2e_{i\nu} - 1. \] (30)

Then, for the density (29) we have
\[ n_\nu = \frac{1}{2} (1 + m_\nu), \] (31)

where
\[ m_\nu = \frac{1}{N_L} \sum_{i=1}^{N_L} <\sigma_{i\nu}>. \] (32)

According to (1), the first phase is dense and the second one is diluted, which for (31) and (32) means that
\[ n_1 > n_2, \quad m_1 > m_2. \] (33)

In the quasispin representation the Hamiltonian (27) becomes
\[ H_\nu = U_\nu + \frac{1}{8} w_\nu^2 \sum_{i \neq j}^{N_L} \Phi_{ij} \sigma_{i\nu} \sigma_{j\nu} - \frac{1}{2} w_\nu \sum_{i=1}^{N_L} h_{i\nu} \sigma_{i\nu}, \] (34)

with the notation
\[ U_\nu = \frac{1}{2} N_L w_\nu \left( K - \mu + \frac{1}{4} w_\nu \Phi \right), \]
\[ K \equiv \frac{1}{N_L} \sum_{i=1}^{N_L} K_i, \quad \Phi \equiv \frac{1}{N_L} \sum_{i \neq j}^{N_L} \Phi_{ij}, \]
\[ h_{i\nu} = \mu - K_i - \frac{1}{2} w_\nu \Phi. \]

The operator from of (34) corresponds to the Ising model with an external field. Such a model, as is known, is difficult to treat. For low dimensionality, there exists a numerical transfer - matrix method based on a real - space Trotter decomposition.
of the statistical operator [27]. A numerical transfer - matrix study based on finite - size scaling [28] is applicable to the Ising model in two and three dimensions. However, our aim here is not to present some extra accurate calculations but to clarify the main physical features of a new model. Therefore, at this stage it would be unreasonable to sink into clumsy technical details. Instead, we can think that $\Phi_{ij}$ is a long - range interaction for which the mean - field approximation works well. The latter gives simple expressions for correlation functions, such as

$$<\sigma_i \sigma_j> = m^2_\nu (i \neq j).$$

For arbitrary lattice sites we can write

$$<\sigma_i \sigma_j> = \delta_{ij} + (1 - \delta_{ij})m^2_\nu.$$  \hspace{1cm} (35)

Using (35), we are able to continue the explicit analytical investigation of our model.

First of all, let us check the conditions of heterophase stability. For quantities (10) and (11) we now have

$$K_\nu = \frac{K}{2n} (1 + m_\nu) = K \frac{n_\nu}{n},$$

$$B_\nu = \frac{\Phi}{8n} (1 + m_\nu)^2 = \Phi \frac{n^2_\nu}{2n},$$ \hspace{1cm} (36)

As far as in the mean - field approximation

$$<\left( \frac{\partial H}{\partial w} \right)^2 = 0,$$

the conditions of stability (16) and (17) coincide with each other and with (18). The latter, taking account of (36), leads to the condition of heterophase stability

$$\Phi > 0.$$ \hspace{1cm} (37)

This shows that to make the heterophase system stable, the particle interactions are to be repulsive on average.

For the free energy we obtain

$$f = \frac{1}{2n} (K - \mu) + \frac{1}{8n} \Phi \sum_\nu w^2_\nu (1 - m^2_\nu) + \mu -$$

14
\[-\frac{\Theta}{n} \sum_{\nu} \ln \left[ 2 \cosh \frac{\Phi (1 + m_\nu) w_\nu^2 + 2(K - \mu)w_\nu}{4\Theta} \right]. \quad (38)\]

The mean quasispin (32) becomes

\[m_\nu = -\tanh \frac{\Phi w_\nu^2 (1 + m_\nu) + 2w_\nu(K - \mu)}{4\Theta}. \quad (39)\]

The equation for the phase probabilities can be obtained either from (13), that is from the minimization of (38) with respect to \( w \), or by substituting (36) into (14). In the both cases we get, of course, the same equation

\[w_1 \Phi (1 + m_1)^2 + 2m_1(K - \mu) = w_2 \Phi (1 + m_2)^2 + 2m_2(K - \mu), \quad (40)\]

to which we have to add (23) to define the chemical potential.

## 4 Thermodynamic properties

Now we have in hands all information needed for investigating thermodynamic properties of the chaotic lattice-gas model, whose phase probabilities and chemical potential are given by equations (40) and (23). For convenience, we write down these equations in the form

\[w = \frac{\Phi n_2^2 + (K - \mu)(n_2 - n_1)}{\Phi (n_1^2 + n_2^2)}, \quad (41)\]

\[n = wn_1 + (1 - w)n_2. \quad (42)\]

First, let us show that the considered chaotic state is thermodynamically more profitable than a homophase state. For zero temperature, this can be readily shown by comparing the internal energies of the chaotic two-phase system and of a homogeneous one-phase system. Thus, substituting (36) into (9), we find

\[E = \frac{K}{n}(w_1n_1 + w_2n_2) + \frac{\Phi}{2n} \left( w_1^2 n_1^2 + w_2^2 n_2^2 \right). \]

Invoking (42), we obtain

\[E = K + \frac{\Phi}{2} \frac{n}{n} - \frac{\Phi}{n} \\left( w_1 w_2 n_1 n_2 \right). \quad (43)\]
In the case of a homophase system, when one of the phase probabilities is zero, we have

\[ E_{\text{hom}} = K + \Phi \frac{n}{2}. \]  

(44)

The comparison of (43) and (44) proves that

\[ E < E_{\text{hom}} \quad (\Phi > 0). \]  

(45)

At finite temperatures, we have to compare (38) with

\[ f_{\text{hom}} = \frac{K - \mu_0}{2n} + \frac{1 - n}{2} \Phi + \mu_0 - \frac{\Theta}{n} \ln \left( 2 \cosh \frac{\Phi n + K - \mu_0}{2\Theta} \right), \]

where

\[ \mu_0 = K + \Phi n + 2\Theta \arctanh(2n - 1). \]

Numerical analysis shows that there always exists a range of temperatures when \( f < f_{\text{hom}} \).

To investigate in more details the temperature behavior of thermodynamic characteristics of the chaotic lattice - gas model, and to check its stability conditions, we proceed in what follows to a numerical investigation. For this purpose it is convenient introduce a dimensionless chemical potential and temperature,

\[ \mu_* \equiv \frac{\mu - K}{\Phi}, \quad T \equiv \frac{\Theta}{\Phi}. \]

(46)

Let us also use the notation

\[ a \equiv n_1, \quad b \equiv n_2 \quad (a > b). \]

(47)

Eq.(38), with the help of (46) and (47), can be rewritten as

\[ \frac{f - \mu}{\Phi} n = \frac{a}{2} (1 - a) w^2 + \frac{b}{2} (1 - b) (1 - w)^2 - \frac{1}{2} \mu_* - T \ln \left[ 4 \cosh \frac{aw^2 - \mu_* w}{2T} \cdot \cosh \frac{b(1 - w)^2 - \mu_* (1 - w)}{2T} \right]. \]  

(48)

The quantities \( a \) and \( b \) play the role of order parameters distinguishing phases with different densities. The equations for these order parameters follow from (39) giving

\[ 2a - 1 = - \tanh \frac{aw^2 - \mu_* w}{2T} \]
\[ 2b - 1 = - \tanh \frac{b(1-w)^2 - \mu^*(1-w)}{2T}. \] (49)

And the equations for \( w \) and \( \mu^* \) are (41) and (42). From (41) we can express the chemical potential
\[ \mu^* = \frac{a^2 w - b^2 (1-w)}{a-b}, \] (50)

and from (42), the phase probability
\[ w = \frac{n-b}{a-b} \quad (a > b). \] (51)

Substituting (51) into (50), we get
\[ \mu^* = \frac{a^2 (n-b) + b^2 (n-a)}{(a-b)^2}. \] (52)

Equations (49), with the use of (52), become
\[ 2a = 1 - \tanh \left[ b(n-b) \frac{2ab - n(a+b)}{2T(a-b)^3} \right]. \]
\[ 2b = 1 + \tanh \left[ a(n-a) \frac{2ab - n(a+b)}{2T(a-b)^3} \right]. \] (53)

In this way, if we solve (53) with respect to \( a > b \), then we can calculate (51) and (52).

Note that (51), together with the property of the phase probability, \( 0 < w < 1 \), impose the following condition
\[ b < n < a \] (54)
on the solution of (53). Numerical investigation of (53) shows that the solutions \( a > b \) exist provided that
\[ 0 < n < 0.323. \] (55)

These solutions are plotted in fig. 1.

Figs. 2 and 3 display the temperature behaviour of the dense - phase probability (51) and of the dimensionless chemical potential (52), respectively. As we see, the probability of the dense phase diminishes with temperature, becoming zero at some \( T_c(n) \).
The pressure of the system is drawn in fig. 4. And figs. 5 and 6 demonstrate that the stability conditions (20) for the specific heat and isothermal compressibility are satisfied.

To illustrate that the condition of heterophase stability (37) is directly related to the conditions of thermodynamic stability (20), we also considered the case $\Phi < 0$. The corresponding equations for the order parameters can be obtained from (53) after the change $\Phi \rightarrow -\Phi$. The resulting equations do possess nontrivial solutions depicted in figs. 7, 8 and 9. Even more, there appear three branches of these solutions. However, for no one of these branches the conditions of thermodynamic stability (20) hold. Thus, the conditions of heterophase and thermodynamic stability are either both valid or both broken.

5 Discussion

We have constructed a heterophase lattice-gas model consisting of two phases with different densities, one phase may be called dense; another, diluted. The dynamics of the phases is chaotic. However, the system, as a whole, can be globally stable. The density of particles in such a system, after averaging over phase configurations, or over time, has the form of a sum

$$\bar{\rho} (\vec{r}) = \sum_{\nu} w_{\nu} \rho_{\nu}(\vec{r}), \quad (56)$$

in which

$$\rho_{\nu}(\vec{r}) = \langle \psi_{\nu}(\vec{r})^\dagger \psi_{\nu}(\vec{r}) \rangle > . \quad (57)$$

The space averages of (56) and (57), i.e.,

$$\rho = \frac{1}{V} \int \bar{\rho} (\vec{r}) d \vec{r}, \quad \rho_{\nu} = \frac{1}{V} \int \rho_{\nu}(\vec{r}) d \vec{r}, \quad (58)$$

lead to the mean densities (7) and (8), respectively.

For the lattice-gas model considered, the density (56), in accordance with (57) and (24), is periodic. In this way, although at each phase configuration, or at any fixed time, there is no periodicity in the system because of random distribution of two phases is space, but after averaging over phase configurations, or over time, the averaged density of particles (56) happens to be periodic.

A straightforward generalization of the model could be if we would ascribe to different phases different lattices. Putting into correspondence to each phase $N_{L\nu}$...
cells with the attached lattice vectors \( \{ \vec{a}_{i\nu} \} \), we may write, instead of (24), the expansion
\[
\psi_\nu(\vec{r}) = \sum_{i=1}^{N_{L\nu}} \sum_n c_{i\nu n} e^{i\nu} \varphi_n(\vec{r} - \vec{a}_i).
\] (59)

Then, in place of (57) we would have
\[
\rho_\nu(\vec{r}) = \sum_{i=1}^{N_{L\nu}} \sum_n \langle e_{i\nu} \rvert c_{i\nu n} \rangle \langle c_{i\nu n} \rvert \varphi_n(\vec{r} - \vec{a}_{i\nu}) \rangle^2,
\] (60)

which is a periodic function,
\[
\rho_\nu(\vec{r} + \vec{a}_{i\nu}) = \rho_\nu(\vec{r}).
\]

If for a two-phase system the vectors \( \vec{a}_{1\nu} \) and \( \vec{a}_{2\nu} \) are integrally independent, as is defined in the Introduction, then the sum (56) of two periodic functions (59) is a quasiperiodic function. Similarly, we could construct a quasiperiodic linear combination (56) composed of an arbitrary number of periodic terms.

So, a system can seem, on average, to be quasiperiodic while actually it is chaotic. It is possible to speculate that some incommensurate structures could be of this kind. Whether an order in a system is frozen or is sustained only on average, can be checked by appropriate experiments. For example, the Mössbauer spectroscopy is very sensitive to structural features, and the Mössbauer factor displays specific anomalies in the presence of lattice instabilities, especially when the latter are connected with mesoscopic structural fluctuations as in the considered case [29,30]. Anyway, even if such systems would not exist in nature, the model investigated here presents an illustration of unexpected possibility when the onset of chaos, that is of local instability, makes the system globally more stable. However, we think that this possibility is not just a strangeness of an artificial model but rather a general phenomenon common for many physical systems, the phenomenon called [3] the spontaneous breaking of equilibrium.
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Figure captions

Fig.1. The dimensionless density of the dense phase (solid curves) and the diluted phase (dashed curves) vs. temperature for several mean densities. The curves end at the points where the system looses its stability.

Fig.2. The probability of the dense phase as a function of temperature.

Fig.3. The dimensionless chemical potential vs. temperature.

Fig.4. The temperature dependence of pressure.

Fig.5. The specific heat as a function of temperature.

Fig.6. The isothermal compressibility vs. temperature.

Fig.7. The dimensionless density of the dense phase (solid curves) and of the diluted phase (dashed curves) vs. temperature in the case of the unstable system with the mean density $n = 0.20$. Numbers 1, 2 and 3 enumerate the branches of solutions.

Fig.8
The same as in fig.7 but for the mean density $n = 0.26$.

**Fig.9**
The dependence of the densities of the dense phase (solid curve) and of the diluted phase (dashed curve) on the mean density in the case of the unstable system at fixed temperature $T = 0.03$. 

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