
STRUCTURE AND DYNAMICS OF GRANULAR MATERIALS PERTURBED BY EXTERNAL FIELDS

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The problem of the description of a local structure of granular materials is discussed within the concept of existence of structural invariants. The superimposition of this approach with the probabilistic arguments concerning the crossover to the kinetics of a transition between states with different measures gives a relevant picture of the evolution with a reminiscence of the first-order phase transition between states with different structural orderings.

1. Introduction

Granular materials are large assemblies of solid macroscopic particles. If they are non-cohesive, the forces between them are strictly repulsive. The particles are usually surrounded by a fluid, most often air, which may play a role in the dynamics of a system. Examples of such materials include sand, stones, soil, ores, pharmaceuticals, and variety of chemicals. The global behavior of a large number of grains that only interact through collisions and friction is typically nonlinear and different from that of the other standard and familiar forms of matter. Vibrated granular materials are perfect examples of this multiscale nonlinearity. Despite the long-term tradition among engineers, granular media have attracted a considerable amount of attention within the physics community over the last decade. This is partly due to the realization that some methods of statistical mechanics under appropriate circumstances could be useful in the understanding of certain phenomena which are observed in granular materials. The unique status of granular materials is based on two characteristics: the ordinary temperature plays no role, and the interaction between grains are dissipative because of the static friction and the inelasticity of collisions. There are no long-range interactions between individual grains or between individual grains and the walls of a confining container. Yet despite this seeming simplicity, a granular material behaves itself differently from any of the other familiar forms of matter – solids, liquids, or gases. For instance, one can cite internal stress fluctuations, strain localization, non-Newtonian rheology, spontaneous clusteriza-

tion, size segregation, or creation of spatial patterns. All these phenomena have no equivalent in classical solid- or liquid-state physics. Therefore, granular material could be even considered as an additional state of matter in its own right [1–5].

Attempts toward understanding and controlling both static and dynamic properties of granular materials are thus of the highest interest to many fields of physics (both theoretical and experimental), applied science, and engineering. We are focused here on the understanding of the ways of description of the local structures of granular materials and the crossover between the macroscopic behavior of granular materials (g.m.) and their microstructures.

The present state of the theory of physical processes which occur in g.m. when they are subjected to gentle internal perturbations of different types is mainly based on the intuitive concepts of the isomorphic character of the physical processes which occur in g.m. and in typical gases, liquids, and solids.

To study the local structure of g.m., contrary to typical condensed matter objects, like liquids or solids, it is not enough to know the positions of the centers of grains which would allow one to reconstruct their space arrangements.

Even being at rest, g.m. have a complex topology of the interparticle space (free volume) which contributes to the formation of a local order in the form of coexistence of the clusters of a different symmetries on the appropriate scales.

When being weakly shaken, the conglomeration of beads shows a complex dynamic behavior which is characterized by transitions between the different symmetries in local arrangements of grains within certain scaled-domains.

In addition of the grain shape, dimension of the system, and boundary conditions, the important role in the complex dynamic behavior of g.m. is played the effects of inelastic intergrain collisions, the presence of intergrain media (vacuum, water, another species, etc.), and magnetization.

The complex structure of g.m. makes their description impossible in terms of one structural parameter only. In what follows, we are going to show a possible way to introduce the statistical-like measure for a local order in g.m., which leads to a respective kinetics of the processes with a transformation of local structures of different types.

Note that the modern concepts of a local structure of condensed phases of matter include also a diversity of models which have mostly an intuitive (*ad hoc*) origin. The majority of such approaches has a quite solid background, i.e., they are based on the idea of that the order is convenient, and the information about a crystalline structure can be obtained in a very compact way, by retaining only the information about a single fundamental cell plus some translational symmetry operations.

2. Definition of the Order Parameter in Terms of Structural Invariants

We now restrict our analysis, by considering the discrete set $\{G_i\}$ ($i = 0, 1, 2, \dots$) of points with coordinates $r^{(i)}$ representing the centers of particles (grains) which surround the central one located at the origin of the coordinate frame.

We will follow the common paradigm that the geometrical structure of $\{G_i\}$ can be determined by comparing of $\{G_i\}$ with an alternative set of points $\{\Gamma_i\}$ which form a collection of familiar ideal (ordered) structure patterns (say, fcc, hcp, *etc.*).

Such a collection $\{\Gamma_i\}$ can be chosen from alternative phenomenological sources of information about a local construction of the selected units. It is worth noting that there is a limited direct information, for example, about the local structure of typical liquids [6].

On the contrary, the local structure of g.m., especially in 2D case, and its transformations are observable almost by the naked eye. Furthermore, the problem of the analysis of a local structure is reduced to the direct observation and classification with respect to a set of ordered domains of given (crystallographic) symmetries. The mapping of the structure is realized by means of scaled superimpositions of resemblant figures, so it looks like a many-particle system structured on meso- and even macroscales.

Having made the mentioned construction, each state of the ensemble of grains which is stroboscopically observed can be quantitatively interpreted as fluctuational, i.e. as a deviation from the given set of $\{\Gamma_i\}$.

In other words, one can see the local structure as an excitation of one of the selected so-called “ideal” ordered states.

In the phase space, those picture looks like a division into domains, each of which represents the deformed state of one of the patterns $\{\Gamma_i\}$.

The formal quantitative description within the above-outlined approach can be made by means of introducing the relevant local order parameter [7, 8].

Consider the structure of a selected group consisting of a finite number of particles. The structure of such an object is uniquely determined by the set of the coordinates of its constituent particles (grains) or by the finite set of algebraically independent invariants.

Selecting the appropriate central particle, we direct the vectors $\{\mathbf{r}^{(\alpha)}\}$ centered in this grain to the particles in their neighborhood limited by the certain distance r_0 .

The role of r_0 could be played by the radii of coordinate shells (spheres) or experimentally observed domains which have some specific crystallographic ordering. Formally, the set $\{\mathbf{r}^{(\alpha)}\}$ is already the order parameter which describes the structural order.

Strictly speaking, $\{\mathbf{r}^{(\alpha)}\}$ is a strongly fluctuating value in gases. For the crystals, it has the same value for every vortex.

In the case of granular perturbed materials, one can adopt the conception typical of the physics of condensed matter: the fluctuations of $\{\mathbf{r}^{(\alpha)}\}$ are weak enough.

Moreover, the weak fluctuations are prescribed both for the length and for the relative angles between the vectors from the set $\{\mathbf{r}^{(\alpha)}\}$.

For the description of a structure on a larger scale, more convenient are tensor values which can be constructed as follows:

$$T_{\alpha_1 \dots \alpha_l}^{(t)} = \sum_{(\alpha)} \omega(\mathbf{r}^{(\alpha)}) t_{\alpha_1 \dots \alpha_l}^{(a)}, \quad (1)$$

and

$$T_{lm}^{(0)} = \sum \tilde{\omega}(\mathbf{r}^{(a)}) t_{lm}, \quad (2)$$

where

$$t_{\alpha_1 \dots \alpha_l}^{(a)} = \overline{\mathbf{r}_{\alpha_1}^{(a)} \dots \mathbf{r}_{\alpha_l}^{(a)}}, \quad (3)$$

and $\mathbf{r}_{\alpha_1}^{(a)} \dots \mathbf{r}_{\alpha_l}^{(a)}$ is a Cartesian tensor,

$$t_{lm}^{(a)} = Y_{lm}(\Omega^{(a)}), \quad (4)$$

$\Omega^{(a)} = \{\varphi^{(a)}, \theta^{(a)}\}$ denotes the polar and azimuthal angles that correspond to the direction $\mathbf{r}^{(\alpha)} / |\mathbf{r}^{(\alpha)}|$.

In (3) and (4), $\omega(\mathbf{r}^{(a)})$ and $\tilde{\omega}(\mathbf{r}^{(a)})$ are the weight functions which define the probabilities of the contributions to $T_{\alpha_1 \dots \alpha_l}^{(t)}$ from different coordination shells and to $T_{lm}^{(0)}$ from the different orientational configurations, respectively.

The tensors $T_{\alpha_1 \dots \alpha_l}^{(t)}$ and $T_{lm}^{(0)}$ can be treated as translational and orientational components of the global order parameter which characterize the considered system. Moreover, $t_{\alpha_1 \dots \alpha_l}^{(a)}$ and $T_{\alpha_1 \dots \alpha_l}$ are the linear combinations of $t_{lm}^{(a)}$ and T_{lm} , respectively. The tensor $T_{\alpha_1 \dots \alpha_l}$ belongs to the basis of the 3D rotational symmetry group O_3 . The translational and orientational order parameters given by Eqs. (1) and (2) adopt the construction of $2(l-1)$ independent (structural) invariants $\{\Psi_l^{(k)}\}$, $l = 0, 1, \dots, 2(l-1)$.

Furthermore, the structural invariants characterize the relative positions of all the particles of the structure.

Note that the value of $T^{(0)}$ gives us the number density of particles which is averaged over V_0 . The quantity

$$T_{ij}^{(2)} = \tilde{T}_{ij}^{(2)} - \frac{1}{3} \delta_{ij} \tilde{T}_{ij}^{(2)} \quad (5)$$

is the density of quadrupole moments.

The quantities $T_{\lambda_1 \lambda_2 \lambda_3 \lambda_4}^{(4)}$ and $T_{\lambda_1 \lambda_2 \lambda_3 \lambda_4}^{(5)}$ describe the ordered states in the systems of given crystallographic symmetries.

The definitions given by Eqs. (1)–(4) have been constructed under the assumption that all the particles-grains are equivalent to each other. If this condition does not hold, one has to determine a relevant set of tensor values ${}^\kappa T^{(n)}$ for every component κ ($\kappa = 1, 2, \dots$).

The intensive experimental investigations of the structure of granular materials show that both ordered and disordered domains coexist in most cases. Undergoing the external perturbations, these domains possess a plenty of transformations which can be described as transitions between quasistationary states.

3. Probabilistic Analysis and Recognition Problems in Studying a Local Structure in the Space of Structural Invariants

$2(l-1)$ independent invariants $\{\Psi_l^k\}$ constructed from the order parameters given by (1) and (2) create a structural invariants in the phase space. These structural invariants characterize the relative positions of the grains (without taking their spatial orientation into account).

Considering the sensitivity of structural invariants to the scale-hierarchy of fluctuations and concluding that

the high-rank invariants are not significant for the structural analysis as compared with the low-rank ones, we expect that, for each value of l , there exists a characteristic scale ξ_l of the grain displacement which corresponds to the selected deviation of $\{\Psi_l^k\}$. In the semiquantitative scenario of fluctuations of the structural invariants, ξ_l decrease with increase in l to the border, where ξ becomes comparable with a relevant ξ_l . Under this condition, the respective invariant $\{\Psi_l^k\}$ fluctuates so strongly that the system is able to move to other states.

For instance, let us consider a 3D cluster with the fcc, hcp, and icosahedral symmetries as possible ones for structured domains. Each cluster include 1 central and 12 “inner outer” granules equidistantly positioned from the central one. The expected scenario of the evolution looks as follows. Initially, each of 12 particles which belong to the considered domain are randomly displaced relative to the surface of a sphere with radius ξ around the central grain.

Along the line discussed in Section 2, we can introduce two order parameters, namely, the space order-parameter R_{lm} defined as

$$R_{lm} = \frac{1}{N} \sum_a Y_{lm}(\Omega^{(a)}) \left| \mathbf{r}^{(a)} \right|^l, \quad (6)$$

and the orientational order-parameter Q_{lm} :

$$Q_{lm} = \frac{1}{N} \sum_a Y_{lm}(\Omega^{(a)}). \quad (7)$$

The relevant structural invariants are

$$R_l^2 = \frac{4\pi}{2l+1} \sum_{m=-l}^l |R_{lm}|^2, \quad (8)$$

$$Q_l^2 = \frac{2\pi}{2l+1} \sum_{m=-l}^l |Q_{lm}|^2, \quad (9)$$

where Y_{lm} are spherical harmonics, and the angle $\Omega^a \equiv \{\varphi^{(a)}, \theta^{(a)}\}$ fixes the direction $\mathbf{r}^{(a)} / |\mathbf{r}^{(a)}|$.

The nonfluctuating group of grains ($R_l = Q_l$) are characterized by the values collected in Table.

Structure invariants $\{Q_i\}$

	Q_3	Q_4	Q_5	Q_6	Q_7	Q_8	Q_9	Q_{10}
fcc	0	0.1909	0	0.5745	0	0.4039	0	0.0129
hcp	0.0761	0.0972	0.2516	0.4848	0.3108	0.3170	0.1379	0.0102
ics	0	0	0	0.6633	0	0	0	0.3629

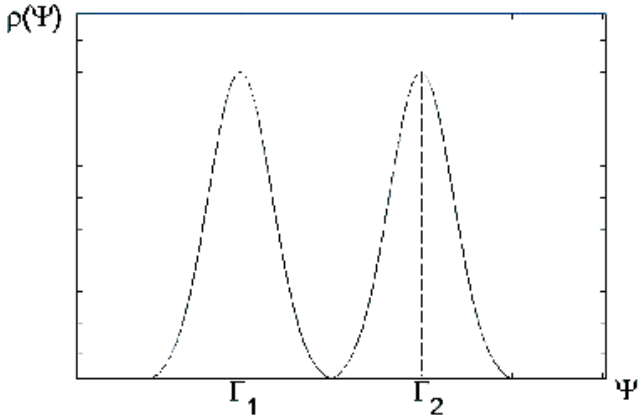


Fig. 1. Nonoverlapping distributions for Γ_1 and Γ_2

The analysis of data presented in Table shows that, indeed, Q_6 is very sensitive to any considered kind of crystallization (ordering).

If we look for the statistics of independent fluctuations of grain positions, then it is clear that the probability density of fluctuations of the invariants is different from the statistics of thermal fluctuations in the molecular systems, where correlations are present due to the interparticle interaction.

4. General Concept of a Quasistatistical Approach

In order to study a finite collection $\{\Gamma_i\}$ of selected patterns, we describe the probability distribution of fluctuations of the invariants Ψ_l $\rho(\Psi)$ in the phase space. Let the distribution $\rho(\Psi)$ have the neighboring distribution $\rho_n(\Psi)$ at some distance that can represent the current observable states (which can be interpreted as deformed patterns with selected symmetries of their distributions). Along this line, by definition, we have

$$dW = \rho_n(\Psi^{(0)}; \xi) d\Psi^{(0)}. \tag{10}$$

Let us fix that Eq. (10) gives the probability to find the values of $\Psi_l (l = 0, \dots)$ which represent, in turn, fluctuations of the selected pattern $\{\Gamma_n\}$ in a vicinity of the space volume $d\Psi^{(0)} = \prod_{(l)} d\Psi_l^{(0)}$ which surround the point $\Psi^{(0)}$.

Then, the collection of inequalities

$$\rho_n(\Psi) < \text{const}, \quad n = 1, 2, \dots, \tag{11}$$

defines the domains in the phase space which represent the observable current states (deformed of $\{\Gamma_n\}$).

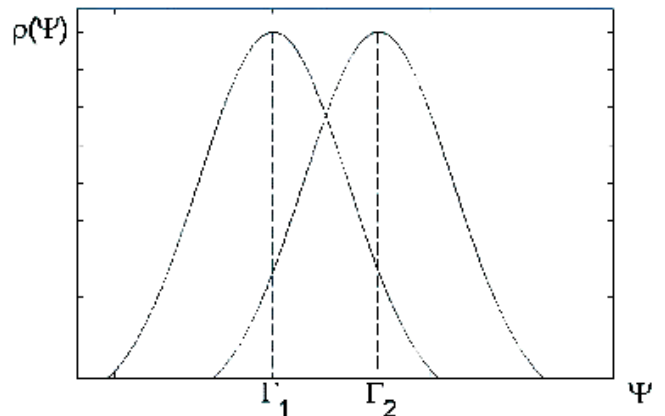


Fig. 2. Overlapping distributions for Γ_1 and Γ_2

A further possible scenario of the above-described probabilistic analysis could be qualitatively the following one.

When the radius ξ is small, the distributions do not overlap with one another (Fig. 1).

In such a case, each point Ψ represents a deformed state of some selected pattern (namely, denoted as Γ_1 and Γ_2). Moreover, each pattern Γ_i cannot simultaneously correspond to another one $\Gamma_j (i \neq j)$.

As ξ increases, two neighboring distributions at the appropriate values of $\xi > \xi$ start to overlap with each other (Fig. 2).

In this case, Ψ can correspond with a certain probability to two different patterns, and this real physical situation happens for gently shaken granular materials.

For the numerical analysis, the classification of the types of local structures can be made, for instance, by adopting some statistical (recognition) hypotheses. This can be done by means of introducing the error function of recognition among two selected patterns, say, Γ_1 and Γ_2 :

$$E = \int \min\{\rho_1(\Psi); \rho_2(\Psi)\} d\Psi. \tag{12}$$

The integral in (12) is defined over the whole phase space of invariants, and the densities $\rho_i(\Psi) (i = 1, 2)$ characterize fluctuations of the patterns Γ_1 and Γ_2 . The total error function E given by (12) minimizes a mistake in the recognition.

The statistics of fluctuations of the invariants is governed by the effective Hamiltonian. Adopting the general concepts of the theory of fluctuations in vicinities of the selected collection of states $\{\Psi_i^{(0)}\}$ which are supposed to be stationary ones, we write the probability density

for fluctuations in the form

$$\rho(\Psi) \sim \exp\{-F(\Psi)\}. \tag{13}$$

One can expand the function $F(\Psi)$ in a Taylor series in vicinities of the set of states $\{\Psi^{(0)}\}$ up to the terms associated with the most probabilistic ones:

$$F(\Psi) = F(\Psi^{(0)}) + \frac{1}{2!} \sum_{k,l} \beta_{kl} \Psi'_k \Psi'_l + \frac{1}{4!} \sum_{k,l,m,n} \gamma_{klmn} \Psi'_k \Psi'_l \Psi'_m \Psi'_n. \tag{14}$$

Terminating expansions (14) by quadratic terms and substituting (14) in (13) yields the multidimensional Gaussian distribution

$$\rho_i(\Psi) = \prod_{(l)} \rho_i^{(l)}(\Psi_l);$$

$$\rho_i^{(l)}(\Psi_l) = \frac{1}{\sqrt{2\pi\sigma_i^{(l)}}} \exp\left\{-\frac{(\Psi_l - \langle\Psi_l\rangle_i)^2}{2(\sigma_i^{(l)})^2}\right\}, \tag{15}$$

where $\sigma_i^{(l)}$ are the mean-root-square deviations, $\Psi' = \Psi - \Psi^{(0)}$, β_{kl} and γ_{klmn} are the matrices, whose eigenvalues can be extracted from the experimental data (observation) related to measurements of the local structure.

The trivial fluctuation kinetics which describes the relaxation of the structural invariants introduced above can be viewed as follows.

The simplest kinetic equation for fluctuations of the structural invariants Ψ in a vicinity of the stationary state $\Psi^{(0)}$ based on concept (13)–(15) looks as follows:

$$\frac{\partial\Psi'}{\partial t'} = \rho\{-F(\Psi')\} \sim \exp\{-F(\Psi')\}. \tag{16}$$

Equations (16) and (14) bring us to the relevant type of kinetics which is governed by the differential equations familiar from the fluctuation theory of phase transitions [9,10].

5. Free Volume Kinetic Model

The possibilities of the approach given above can be estimated by its comparing with the mean-field free-volume kinetic scenario [17]. Namely, in order to model a compact gently agitated granular material, a free-volume kinetics is often in use. In this framework, one can describe

the process, where a solid particle (associated with a grain) with volume ω can jump into a hole of the appropriate size Ω distributed with a certain distribution function $f(\omega/\Omega)$. The simplest rate equation of type (16) which describes such a free volume kinetics in this case takes a form

$$\frac{d\eta}{dn} = kf, \tag{17}$$

where the variable η is the packing fraction, and k is a kinetic coefficient. Introducing the limiting maximum values for the compactivity η_m and estimating the simplest Poisson distribution for the free volume

$$\Omega = \omega \left(\frac{1}{\eta} - \frac{1}{\eta_m} \right), \tag{18}$$

one has

$$\frac{d\eta}{dn} = k \exp\left(-\frac{\eta_m \eta}{\eta_m - \eta}\right). \tag{19}$$

The rigorous analytical solution of this equation which connects the packing fraction η with the number of taps n can be obtained in the following functional form:

$$(\eta_m - \eta_1) \exp\left(-\frac{\eta_m^2}{\eta_m - \eta_1}\right) + \eta_m^2 E_1\left(-\frac{\eta_m^2}{\eta_m - \eta_1}\right) -$$

$$-(\eta_m - \eta) \exp\left(-\frac{\eta_m^2}{\eta_m - \eta}\right) - \eta_m^2 E_1\left(-\frac{\eta_m^2}{\eta_m - \eta}\right) =$$

$$= k \exp(\eta_m)n. \tag{20}$$

Here, η_1 is the initial compactivity, and $E_1(z)$ is an integral exponent. If the initial state is already densified, i.e. $\eta_1 \rightarrow \eta_m \left(\frac{\eta_m^2}{\eta_m - \eta_1} \gg 1, \frac{\eta_m^2}{\eta_m - \eta} \gg 1 \right)$, Eq. (20) yields

$$\eta = \eta_m - \frac{\eta_m^2}{\ln\gamma + \ln\left(\frac{a}{\gamma} + n\right)}, \tag{21}$$

where

$$a = \left| (\eta_m - \eta_1) \exp\left(-\frac{\eta_m^2}{\eta_m - \eta_1}\right) + \eta_m^2 E_1\left(-\frac{\eta_m^2}{\eta_m - \eta_1}\right) \right|,$$

and $\gamma = k \exp(\eta_m)$. This logarithmic law for the granular compaction has been reported many times either by experimentalists or by theoreticians [1–3, 5–17]. When the system is initially diluted, so that $\eta_m^2/(\eta_m - \eta_1) < 1$ and $\eta_m^2/(\eta_m - \eta) < 1$, we obtain

$$\eta = \eta_1 + \gamma n, \tag{22}$$

i.e. the behavior deviates strongly from a logarithmic one.

We note that if $\eta_m^2/(\eta_m - \eta_1) < 1$, but $\eta_m^2/(\eta_m - \eta) > 1$, Eq. (20) again yields the logarithmic law given by (21). Independently from how the system was initially prepared (diluted or densified), the model gives the slow logarithmic growth rate of η in the limit when η tends to η_m .

This simple argumentation demonstrates the basic possibility to get different kinetic scenarios, i.e. the logarithmic dynamics and more faster stages of compaction, already known in the framework of mean-field arguments, including the free-volume kinetic model. Different scenarios are found in experiments, and a mixture of those dynamics is more appropriate for describing the experimental data. This will be discussed in the next sections.

6. A Quasistatistical Model of Inherent States

In order to connect the dynamics of granular systems with the geometrical and topological properties of their local configurations, we write firstly the configurational partition function Q for N undistinguishable particles in a volume V as a sum over contributions associated with a set of ideal states " α " which was introduced in Section 2, as the measure of a local structure. This collection of states $\{\Gamma_\alpha\}$ can be associated with local minima of the energy of a granular system $U(\mathbf{r}_1, \dots, \mathbf{r}_N)$. One can observe these states experimentally by means of simple stroboscopic observations of gently shaken granular systems. Every stop-picture which escorts the granular dynamics makes our system be frozen in some state. The nature of these states is normally considered quasistationary [11–14]. This phenomenological evidence forces us to adopt the concept that the probability to find the particles-granules in the states which correspond to given configurations is locally maximum:

$$Q = \sum_{\alpha} e^{-\beta F^{\alpha}} = \sum_{\alpha} \frac{1}{N! \Lambda^{3N}} \int_{\{\Gamma_{\alpha}\}} d\mathbf{r}_1 \dots \int d\mathbf{r}_N e^{-\beta U(\mathbf{r}_1, \dots, \mathbf{r}_N)}, \quad (23)$$

where $\beta = (k_B T)^{-1}$, $\Lambda = h/(2\pi m k_B T)^{1/2}$, m is the granule mass, and the integral in Eq. (23) is taken over the regions $\{\Gamma_{\alpha}\}$ which are characterized by the volumes in the configuration space defined around the states $\{\mathbf{r}_1, \dots, \mathbf{r}_N\}$. In the limit when $\{\Gamma_{\alpha}\}$ do not overlap

one another, relation (23) can be considered as a configuration integral.

For a given state α , we divide the volume V into N cells $C_1^{\alpha} \dots C_N^{\alpha}$ constructed around the positions $\mathbf{r}_1 \dots \mathbf{r}_N$. They are characterized by a set of parameters n_i^{α} which carry the complete information about the geometrical and topological properties.

The volume of the cell C_i^{α} is $v(n_i^{\alpha}) = \int_{C_i^{\alpha}} d\mathbf{r}$, and the total volume is $V = \sum_i v(n_i^{\alpha})$.

The configuration integral (23) is taken over the accessible packing configurations associated with each state α . In each configuration, the number of particle in a given cell can vary within the interval $(m_i = 0, N)$, with the constraint $\sum_i m_i = N$. Note that these cells are constructed around particular positions that maximize the probability to find one particle in that given region. Therefore, we assume that the configurations with the cells singly occupied would maximally contribute to F^{α} . Superimposing the concepts of the free volume with the probability of finding the distinguished cell singly occupied, we have reduced the expression for the free energy to a sum over the contributions from each cell $i|j$ which is fully characterized by its geometrical and topological parameters n_i^{α} . As a consequence, we obtain

$$F^{\alpha} = F(\{N^{\alpha}(n)\}),$$

where $N^{\alpha}(n)$ is the number of cells, each of which characterized by the set of n . Different α are associated with distinct cellular partitions.

Therefore, Q can be written as

$$Q = \sum_{\{N(n)\}} \Omega(\{N(n)\}) e^{-\beta F(\{N(n)\})}, \quad (24)$$

where $\Omega(\{N(n)\})$ define the number of distinct space partitions made with the set $\{N(n)\}$.

The cells from the states associated with disordered (noncrystalline) structures have, in general, different shapes and volumes. For instance, for cells identical to the fcc crystalline packing, $\Omega = 1$.

Note that, rigorously speaking, this quantity can be only modeled or calculated numerically to estimate its extrema, but it is not evaluated in general.

Indeed, the maximum number of distinct configurations (different arrangements of N cells distributed in groups of $N(n)$) is

$$\Omega(\{N(n)\}) = \frac{N!}{\prod_{(n)} N(n)}. \quad (25)$$

This maximal value cannot be achieved generally, and the total number of distinct configurations can only be approximated by its quality.

Since the total number of distinct configurations must correspond to the total number of minima of the potential energy, the configuration energy per particle can be given by $S_c = \frac{\ln \Omega}{N}$.

Therefore, in order to calculate the dynamics within the outlined approach, one must firstly to extract (from the experiment or alternative sources) a set of attainable domain-cell shapes associated with the above-described so-called inherent states and then to estimate their combinations in numbers and sizes with the help of (24) which maximally contribute to the partition function.

As for the patterns of a given selected symmetry which follows from the experimental observations of a granular packing [15-17], it can be shown that the relevant dynamics of the relaxation in vicinities of quasistationary states follows the Vogel-Fulcher behavior.

The analysis of possibilities of the above-described approach and its attitude to the experimental observations of granular kinetics will be the matter of a separate paper.

7. Random Close Packing of Hard Spheres and Phase Transitions in Granular Materials

Experimental researches [15-17] discovered a first-order phase transition in a 2D granular fluid excited by vibration from below and confined by a floating plate above. The steady-state transition occurs between a crystal and a gas and is characterized by discontinuous changes in the density and the temperature. It shows a rate-dependent hysteresis and obeys the Lindemann criterion for a melting. Although the granular systems do not conserve the energy, and the concept of free energy is not properly applicable even at steady-state situations, since there is a constant flow energy through the system, the idea of applying the thermodynamic ideas to a nonequilibrium steady-state is at the forefront of current statistical physics.

The phenomenon of random close packing is normally associated with samples produced by shaking. Moreover, it is known that the volume fraction beyond the value 0.64 can be attained by cyclic shearing, and this phenomena also accompanied by the appearance of small crystal-like domains. For instance, during the horizontal shaking, the volume fractions up to 0.70 accompanied by a crystalline ordered domains have been observed [17]. It is commonly observed in experiments that, above a volume fraction of 0.64, the boundary between two regimes (states) is observed: at volume fractions below 0.64, the

structures of g.m. are random. Above 0.64, there appears some ordering in the form of domains with a crystalline order. To the best of our knowledge, no experiments with g.m. just above a volume fraction of 0.64 were reported till now. The numerous experimental observations and the numerical analysis yield the idea of the random close packing as a phase transition. This phase transition seems to occur by the following scenario. Let the distribution $p(m)_\eta$ describe the mixed phase which corresponds, as follows from experimental data, to the interval of volume fractions $0.49 \leq \eta \leq 0.54$.

By definition, we adopt that the distribution $p(m)_\eta$ is represented by the average of the distributions of pure phases 1 and 2:

$$p(m)_\eta = cp_1 + (1 - c)p_2, \quad (26)$$

where p_1 corresponds to a distribution of the highest density phase ($\eta = 0.54$) and p_1 is a distribution of the lowest density phase ($\eta = 0.49$). Respectively, $0 \leq c \leq 1$ is such that $p(m)_\eta$ corresponds to volume fraction η . In a certain sense, relation (26) is merely a statement of the fact that distinct phases are separated, when coexisting in equilibrium, and each phase occupies a definite volume. The role of distributions p_1 and p_2 can be played by the probability functions described in Section 5.

Furthermore, the distribution $p(m)_\eta$ is for the infinite system, and this means that the fraction c of samples would represent an infinite system at the volume fraction, say, $\eta_1 = 0.54$. Respectively, the fraction $1 - c$ would represent an infinite system at the volume fraction $\eta_2 = 0.49$. Along this line, we assume, as generally believed, that the value $\eta_1 = 0.49$ is the volume fraction of "freezing" (i.e. "highest random density"), and the solid phase could have a crystalline order (e.g., fcc). Intuitively, at any volume fraction above the freezing point, there is a nonzero probability to see an infinite ordered crystal. It is the use of the infinite-volume limit together with the probabilistic formalism that produces a sharp phase transition between disorder and order in equilibrium statistical mechanics [9-10].

So, we emphasize that there is a sphere packing with the volume fraction $\eta = 0.49$ which might be random. However, the total of all such packings has probability zero as compared with the uniform distribution at the packing with volume fraction η .

We note, while this hard-sphere model argumentation is not directly applicable to the granular matter which is our proper subject, it shows, nevertheless, that the intuitive notion of random close packing is not inherently inconsistent, as it was claimed some times.

At the same times, it is clear that traditional hard-sphere model does not include the effect of gravity and thus cannot represent properly the properties of granular matter. However, a slightly modified ensemble framework was proposed in [18] as a model for granular systems. This approach use a uniform distribution of static monodisperse sphere packings with a fixed volume fraction, which are mechanically stable under gravity.

8. Conclusions

The introduction of a local measure for the granular materials in a vicinity of the quasistationary state, being superimposed with a probabilistic measure for interstate transitions, has given the relevant kinetics which has a reminiscence of phase transitions. It is a wide spectra of structural invariants that can be used as elements for the classification of the symmetry of the stationary states. A numerous experimental evidences indicate the existence of certain features in the relaxation picture for the relevant order parameters which belong to the scenario of phase transitions [5,16,19].

It is shown that the kinetics of the formation of compacted states in vibrated granular systems can be satisfactorily described within a mean-field free-volume kinetic model [16]. The structural transformations in gently vibrated granular materials in vicinities of ordered domains show a strong visual analogy with a morphology of phase ordering systems [19], which is described in the present paper. As a next step, the quantitative parametrizations of the experimentally observed crystallization and the pattern formation in terms of stereospecific parameters within the above-proposed approach are strongly required.

The role played by inelasticity in the determination of a local structure and the selective dispersive kinetics should be examined separately.

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СТРУКТУРА ТА ДИНАМІКА ГРАНУЛЬОВАНИХ МАТЕРІАЛІВ ЗА НАЯВНОСТІ ЗОВНІШНІХ ЗБУРЮЮЧИХ ПОЛІВ

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Р е з ю м е

Проблема опису локальної структури гранульованих матеріалів запропонована до розгляду з концепцією існування структурних інваріантів. Доповнений імовірнісним сценарієм переходів між квазістаціонарними станами підхід дає можливість опису зв'язків між параметрами локальної структури у вигляді інваріантів з кінетикою змін станів впорядкування, що має ознаки фазових перетворень першого роду.