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# THE KINETICS OF GRANULAR SEGREGATION

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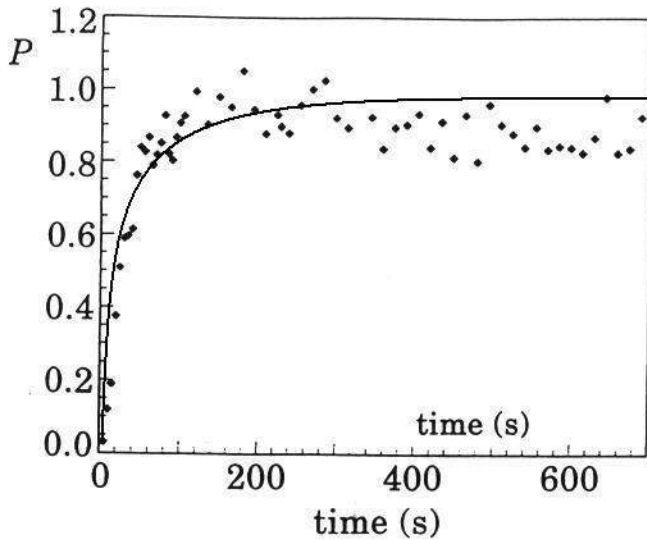
An analytical model of the radial segregation in driven containers filled by a dry granular mixture, which exhibits phenomena as a non-equilibrium phase transition to an asymptotically metastable stationary state (steady state) is developed. Nonlinear equations of motion for the relevant order parameter have been solved exactly in quasi-linear approximation in the vicinity of a steady state. The theoretical results obtained are in a good enough qualitative agreement with the experimental data.

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## Introduction

It is known since numerous experiments [1–6] that when granular materials are placed in a rotating bed, different flow dynamics are observed. In particular, when a mixture of particles which differ in size or density is placed, for example, in a rotating cylinder, the denser or smaller particles will concentrate in the central region close to the free surface after only a few rotations (a phenomenon which is normally termed as radial segregation). This phenomenon was studied experimentally and numerically for varying size ratios [1–6] and density ratios [7–9]. For instance, the experimental set-up for tracing the position of colored or illuminated particles by a video-camera, and evaluating the record by a digital processing program (like, for example, Visilog 4.14) on a workstation is easily accessible and highly automationable. The radial segregation is always observed in the continuous flow regime for any arbitrary small particle size difference, regardless of the filling fraction of the cylinder, and also no mixing of the components is necessary in order to obtain a radially segregated core [10]. The experimental evidence clearly shows a segregation mechanism of a single tracer particle based on size ratio. A large particle is pushed to the edges and a small one will rather stay at the center. Looking at the distribution probability for the trajectories of a single particle of the same

size, one can clearly see the presence of two competing regions: the center of the cylinder (drum) and the edge which have a tendency to attract the trajectory distributions. Recently, the three-dimensional Magnetic Resonance Imaging has been used to study the mixing and segregation processes in a special Turbula blender using binary mixtures of sugar beds [11]. This study by a noninvasive method shows that the kinetics of the segregation process includes two characteristic time scales and that the segregation process is one order faster than mixing. Moreover, in three-dimensional devices, simultaneous axial and longitudinal segregation routes are presented [13]. Particle percolation has been invoked to be responsible for axial segregation. And longitudinal segregation (called also radial segregation), occurring in a long cylinder, is currently related to differential surface flow properties. In the  $3D$  case, both parallel processes which are the segregation by percolation and the filtration of smaller particles in the voids of the porous network made by large particles, are present. On the contrary, in the  $2D$  drums, the segregation by percolation within the granular medium is absent (the pores are not connected in a dense  $2D$  packing of grains). Another traditional advantage of  $2D$  geometry, namely, visualization facilities, is not so important anymore since the three-dimensional Magnetic Resonance Imaging was successfully used to study the mixing and segregation processes. It is interesting to note that the parameters of segregation (like the segregation rate from [11]) in  $2D$  and  $3D$  rotating drums are very similar. We stress that the segregation in  $3D$  (especially in turbula blender geometry) is quite efficient, which is relatively surprising since a better mixing was expected than that with a typical drum because of the  $3D$  nature of a flow. Generally speaking, many parameters are involved in the process of segregation, such as the above-mentioned size ratio, also shape, mass, friction forces, rotating velocity, filling of the drum, etc. And it is not quite clear in advance, which of them should be more important than



Variation of the order parameter  $P(t)$  defined by Eq.(1) for segregation in driven granular mixtures (data from [3]) vs time, solid line denote our theoretical results

other. Thus, the question naturally arises: what is the simplest model representing the essential physics. On that way, we have chosen to discuss and to study here the specifically radial segregation (as an example of fragmentational processes in driven granular systems of different dimensions). After all discussed *vide supra*, one can say the size segregation mechanism can be seen as a more pronounced influence of one attractor compared to the other. This is a reminiscence of non-equilibrium phase transitions, the control parameter being the size ratio (nevertheless, here the non-trivial point is the importance of fluctuations, which are of the size of the system [12, 13]). If we accept this, we may describe the evolution of such a kind of systems by some kind of displacement field  $\xi(\vec{r}, t)$  [12]. Clearly, inside of the frozen phase we may define deformations  $\vec{\nabla}\xi(\vec{r}, t)$ . To describe the phenomena of segregation more precisely, the order parameter field could be also introduced as follows [13]. From experimental data, let us define the region of a drum statistically occupied by the cluster of small particles at the final segregated state. When the process becomes stationary, one can take pictures, and the segregated zone will be defined after developing. The kinetic study, afterwards, will be reduced to counting the number of particles which belong to this segregated zone. From these operations, one can deduce the area  $\Omega(t)$  occupied by small particles in the segregated

zone defined above. The connected mass created this way is called reference mass [14] (a mass is said to be "connected" when its particles actually touch each other). Its volume or surface area reached in principle after an infinite time is denoted by  $\Omega(\infty)$ . Clearly  $\Omega(t) \leq \Omega(\infty)$ . At this point, it is natural to introduce an average ordering parameter  $P(t)$  that can vary between 0 (for completely random and homogeneous mixtures) and 1 (for fully developed reference mass). This parameter is defined in terms of  $\Omega(t)$  as

$$P(t) = \frac{\frac{\Omega(t)}{\Omega(\infty)} - \frac{\Omega(0)}{\Omega(\infty)}}{1 - \frac{\Omega(0)}{\Omega(\infty)}}. \quad (1)$$

As follows from experimental data [1–11,13], the relevant order parameter  $P(t)$  defined by Eq.(1) shows a global trend of increasing in time and saturating asymptotically (on the long run after the cylinder or 2D drum rotation has been started) (see Fig. 1). A typical time evolution of the average order parameter  $P(t)$  normally is well enough approximated by an exponential law of the form

$$P(t) = P(\infty) \left[ 1 - \exp\left(-\frac{t}{\tau_c}\right) \right] \quad (2)$$

with a characteristic segregation time  $\tau_c$  and a final amount of segregation  $P(\infty)$ . Note that, from the point of view of the general theory, it is not possible to generate an exact steady state trajectory in the phase space. This is because the measure of any dissipative non-equilibrium steady state within the phase space is zero [15–17]. We can use an equilibrium (relaxation) method of approaching the steady state. This picture can be expressed mathematically in the  $(\vec{r}, t)$  space by definition of the relevant order parameter which demonstrates the system evolution starting from the initial state passing the intermediate heterogeneous state and finishing with the asymptotic non-equilibrium stationary state (local equilibrium or steady state). Nevertheless, it seems clear that the segregation (as well as compaction) phenomena are purely dynamical effects unrelated to the Boltzmann–Gibbs measure. The approach proposed above could be the step to make a bridge between quasi-static properties and the corresponding relaxation behavior (and in general non-equilibrium dynamical properties of such complex dynamic dissipative systems as driven dry granular materials).

The purpose of this article is to consider the kinetic theory of non-equilibrium phase transitions (which is based on the Landau theory of phase transitions [18]) as an instrument for the investigation of fragmentation

within open dissipative systems (with the example of a driven dry granular system in the segregation state) close to the critical region of a metastable stationary state (steady state). The existence of those definitively follows from the experiments. On that way, considering the final non-equilibrium asymptotic state, when segregation is reached, as a steady state, one can describe the phenomenon of fragmentation (segregation) as a relaxation with a relevantly determined order parameter field. In other words, one can consider the phenomenon of fragmentation in open dynamic dissipative systems like, for instance, segregation in driven dry granular systems, as a non-equilibrium phase transition to steady state. This phase transition could be imagined as a critical slowing down on the dynamics of a multistable (bistable in the case of radial segregation) diffusion limited aggregated system in the neighbourhood of its steady state, where two mechanisms are active: diffusion due to the energy input from a driving force and thermalization due to (for instance) inelastic collisions (dissipation) which are modeled in the case of radial segregation in driven dry granular system as a relaxation of the relevant scalar order parameter to a non-equilibrium stationary state (steady state). In the vicinity of the steady state the respective model field equation of motion for the parameter of ordering will be constructed and solved both in the cases of non-conservative and conservative fields. Exact solutions of the equation of motion in the quasi-linear approximation have been found (neglecting the fluctuations). The asymptotic limit of the obtained solutions is in a remarkable qualitative agreement with experimental results interpolated by Eq.(2). We elucidate law (2) as an essentially nonlinear effect which occurs only if we consider the diffusion of an order parameter field in the presence of a nonlinear non-equilibrium (bistable) potential. We will show that this decay law has a universal character both in the cases of conservative and non-conservative order parameter fields. Thus, on that level of description, the question about the qualification of the observed phenomenon of segregation as a kinetic process of the first or second order requires more investigation. It is shown that the conservative order parameter field relaxes inhomogeneously. Namely, there is a selected mode (which corresponds to the relevant cooperative length within the system) which passes through a heterogeneous metastable state characterized by a slow (non-exponential) relaxation law. Such a state could resist to be destroyed by the changing of energy input (due to the increasing velocity of driving) and

this feature qualitatively corresponds to what probably observed in the Oyama's rotating drum [13]. It is clear that, in the absence of driving, the equations used in our model are no longer valid, and indeed any result based on kinetic theory becomes meaningless as the particle eventually come to rest. Another complication is the inelastic collapse known to occur inside clusters [19], i.e. an infinity of collisions in a finite time. In the approach developed in our present paper, we adopt the non-equilibrium stationary state (steady state) as maintained by an external driving.

### 1. The Field Equation of Motion for the Order Parameter

Considering the radial segregation as a weakly non-equilibrium relaxation process in terms of a relevant parameter of ordering  $\varphi(\vec{r}, t)$ , we focus in this section on the evolution of  $\varphi(\vec{r}, t)$  in the vicinity of the steady state. We assume that evolution  $\varphi(\vec{r}, t)$  can be described with a master dynamic equation of the Landau—Ginsburg type in the case of a non-conservative field  $\varphi(\vec{r}, t)$  [18]:

$$\frac{\partial \varphi}{\partial t} = -\Gamma \frac{\delta H}{\delta \varphi} \quad (3)$$

or with an equation of the Cahn—Hilliard type in the case of a conservative field of ordering

$$\frac{\partial \varphi}{\partial t} = -\Delta \left\{ -\Gamma \frac{\delta H}{\delta \varphi} \right\}. \quad (4)$$

Here  $\Gamma$  is a kinetic coefficient, and  $H(\varphi)$  is the non-equilibrium potential functional which can be taken in the simplest bistable form,

$$H(\varphi) = \int \left[ \frac{c}{2} (\vec{\nabla} \varphi)^2 - \frac{a}{2} \varphi^2 - \frac{b}{4} \varphi^4 \right] d\vec{r}. \quad (5)$$

We choose the functional  $H(\varphi)$  which provides such a qualitative scenario: during the temporal evolution,  $H(\varphi)$  decreases through the steepest descent trajectory until it reaches one of its minima. The unstable structures are related to extrema of  $H(\varphi)$  of the saddle point type and define the magnitude of the barrier between different locally stable attractors. We will use this model with the aim of investigating the nature of critical slowing down in the dynamics (decay) in the critical region, where the system shows a structural instability. That kind of approach has been scarcely used for reaction-diffusion systems in [21], where it has been shown that the time evolution of the non-equilibrium potential  $H(\varphi)$  shows a critical slowing down in its

course towards stationary states: its time scale is given by the damped relaxation time, which depends on the system size and whose inverse is measured by the distance to the critical point in the parameter space.

Substituting the functional from (5) into (3),(4), we obtain (in dimensionless variables) the following equations of motion for  $\tilde{\varphi}(\vec{r}, t)$ :

$$\frac{\partial \tilde{\varphi}}{\partial \tau} = \Delta' \tilde{\varphi} + \tilde{\varphi} - \tilde{\varphi}^3 \tag{6}$$

and

$$\frac{\partial \tilde{\varphi}}{\partial \tau'} = -\Delta' \{ \Delta' \tilde{\varphi} + \tilde{\varphi} - \tilde{\varphi}^3 \}, \tag{7}$$

respectively. Here we define the dimensionless variables

$$\tau \equiv \Gamma a t ; \tau' \equiv \Gamma c t ; \vec{r}' \equiv \sqrt{\frac{a}{c}} \vec{r} \tag{8}$$

and measure the field  $\varphi(\vec{r}, t)$  in units of  $\sqrt{\frac{a}{b}}$ :

$$\tilde{\omega}(\vec{r}, t) \equiv \sqrt{\frac{a}{b}} \tilde{\varphi}(\vec{r}', \tau). \tag{9}$$

The nonlinear partial differential equations (6), (7) could not in general be integrated in quadratures. But in the vicinity of the imaginary point of the transition to a steady state, neglecting the fluctuations of  $\tilde{\varphi}(\vec{r}, t)$ , the quasi-linearization scheme [22, 23]

$$\varphi^3(\vec{r}, t) = \langle \varphi^2(t) \rangle \varphi(\vec{r}, t) \tag{10}$$

could be solved exactly, where the angular brackets denote the procedure of averaging over all initial states. It is possible to show that approximation (10) is going to be more adequate in the limit when the fluctuations of the order parameter field  $\tilde{\varphi}(\vec{r}, t)$  are negligible in comparison with the quasi-equilibrium value of the order parameter [18, 22, 23]. Consider first, in more details, the field equation of motion in the case of a non-conservative order parameter. Substituting (10) into (6), we obtain

$$\frac{\partial \tilde{\varphi}}{\partial \tau} = \Delta' \tilde{\varphi} + (1 - \langle \tilde{\varphi}^2(\tau) \rangle) \tilde{\varphi}. \tag{11}$$

Equation (11) seems open-circuited because of the presence of the second moment term  $\langle \tilde{\varphi}^2(\tau) \rangle$ . But the rigorous solution of Eq.(11), as we are going to show now, could be obtained in terms of  $\langle \tilde{\varphi}^2(\tau) \rangle$ , i.e. for a second moment of  $\tilde{\varphi}(\vec{r}, t)$ . In terms of Fourier transforms of the order parameter denoted as  $\tilde{\varphi}_{\vec{k}}(\tau)$ , Eq.(11) assumes the form

$$\frac{\partial \tilde{\varphi}_{\vec{k}}(\tau)}{\partial \tau} = (-k^2 + 1 - \langle \tilde{\varphi}^2(\tau) \rangle) \tilde{\varphi}_{\vec{k}}(\tau), \tag{12}$$

where

$$\tilde{\varphi}_{\vec{k}}(\tau) = \frac{1}{(2\pi)} \int e^{-i\vec{k}\vec{r}'} \tilde{\varphi}(\vec{r}', \tau) d\vec{r}'.$$

The solution of (12) is given by

$$\begin{aligned} \tilde{\varphi}_{\vec{k}}(\tau) &= \tilde{\varphi}_{\vec{k}}(0) \times \\ &\times \exp \left[ (-k^2 + 1) \tau - \int_0^\tau \langle \tilde{\varphi}^2(s) \rangle ds \right], \end{aligned} \tag{13}$$

where  $\tilde{\varphi}_{\vec{k}}(0)$  is the initial value for the Fourier component of the order parameter field. Taking the square modulus of (13) and integrating the results over the  $\vec{k}$  with the help of the Parseval theorem

$$\int |\tilde{\varphi}_{\vec{k}}(\tau)|^2 D\vec{k} = \int \tilde{\varphi}^2(\vec{r}', \tau) d\vec{r}',$$

we obtain the rigorous solution of Eq.(12) in the following closed form:

$$\begin{aligned} \langle \tilde{\varphi}^2(\tau) \rangle \exp \left[ 2 \int \langle \tilde{\varphi}^2(s) \rangle ds \right] &= \\ &= \exp(2\tau) \int g(\vec{k}) \exp(-2\tau k^2) d\vec{k}. \end{aligned} \tag{14}$$

After trivial manipulations with (14), we have

$$\langle \tilde{\varphi}(\tau) \rangle = \frac{J_1(\tau)}{1 + J_2(\tau)} \exp(2\tau), \tag{15}$$

where

$$J_1(\tau) \equiv \int g(\vec{k}) \exp(-2\tau k^2) d\vec{k}, \tag{16}$$

$$J_2(\tau) \equiv 2 \int_0^\tau \exp(2s) ds, \tag{17}$$

$$g(\vec{k}) \equiv \langle |\tilde{\varphi}_{\vec{k}}(0)|^2 \rangle. \tag{18}$$

Relations (15)–(18) give an exact solution of the considered problem in terms of the mean square of the parameter of ordering. It is natural that the solution

depends on initial conditions contributing through the static structure of initial states represented by  $g(\vec{k})$ , called also static structure factor. We will show further, that solution (15)–(18) obtained for a system of infinite size (in coordinate space) satisfies the natural time boundary conditions  $\langle \tilde{\varphi}(\tau) \rangle \rightarrow \text{const}$  when  $\tau \rightarrow 0$ ; and  $\langle \tilde{\varphi}(\tau) \rangle \rightarrow 1$  when  $\tau \rightarrow \infty$ . We are not going to study here the role played by boundary conditions in the coordinate space, to avoid mathematical complications. This problem will be discussed in our next paper, where we focus on 1D dissipative systems [20] and where this question will be rigorously considered. Note only that the qualitative character of the results presented here still remains valid in the case where the system has finite size.

The further analysis of solution (15)–(18) requires the incorporation of the static picture of the order parameter field.

## 2. Static Structure Factor of the Order Parameter Field

In many ways, the problem of the static structure of dynamic dissipative systems in granular phases seems even more complicated than studying their dynamics in flow regimes. For example, the granular two-point correlation function shows the presence of pronounced long-range forces between particles [24,25]. In general, the definition of an initial state, both in the theory and in an experiment on granular matter, requires great care at least to avoid suffering from a lack of precise definitions. In the theory of classical fluids, the static statistical structure is described by the set of many-particle correlation functions [26]. Freely evolving granular fluids are linearly unstable (onset of clustering instability with respect to spatial fluctuations in density) [19]. In both driven and undriven granular fluids, there is only a weak separation of micro-scales and macro-scales which makes them behave very differently from molecular fluids. An important role is also played by the different intermediate intrinsic scales, related to viscosity, heat conductivity, and compressibility and controlled by the inelasticity parameters. For instance, in [9], the corresponding spatial correlation function of an inelastic hard disc system has been calculated by the method of molecular-dynamics simulations using the simplest way of incorporation of inelastic effects. It has been shown that the local structure of the colling granular medium remains essentially almost identical to that of an equilibrium field. Note also that our interest

in theory in most cases is focused on the calculation of average values of the dynamical variables weighted with the relevant correlation function. Furthermore, certain details in the behavior of the correlation function are not so important and, in the first approximation, could be even omitted from consideration. Along this line, one can go to a direct modelling of a static correlation function taking into account only its coarse-grained character, which follows from general definitions and empirical sources [23]. On that way, a certain constant which will be included in the relevant model, could be considered as a given physical parameter or determined by comparison of the respective final statistical characteristics, like structure factors, with the experiments. Let  $g(r)$  be approximated as

$$g(r) = C\theta(r_0 - r) + \theta(r - r_0) + A(r_0)\delta(r - r_0), \quad (19)$$

where  $\theta(z)$  and  $\delta(z)$  are the Heaviside (step) and Dirac delta functions, respectively;  $\frac{1}{2}r_0$  is the radius of a particle;  $C$  and  $A(r_0)$  are a certain constant and a function of parameters of the medium. The second and third terms in (19) could be considered as elements of the expansion of the initial step function  $\theta[r - (r_0 - A)]$  into a Taylor series in the small value of  $A$ . Note that  $A$  can be considered, for instance, as a function of the penetration area between two deformed particles and can be defined with the help of zero moment for  $g$  [20, 23]. If so,  $A$  should also naturally depend on the particle radius, and this we mean with writing  $A(r_0)$ . The first term in (19) is responsible for the nonzero probability for deformed particles to penetrate each other. After the Fourier transforming (19), we obtain for the Fourier component of  $g(r)$  denoted as  $\tilde{g}(\vec{k})$ :

$$\begin{aligned} \tilde{g}(\vec{k}) = & \delta(\vec{k}) - (1 - C) \sqrt{\frac{2}{\pi}} r_0^3 \frac{j_1(kr_0)}{kr_0} + \\ & + \sqrt{\frac{2}{\pi}} \frac{A(r_0)}{r_0} j_0(kr_0), \end{aligned} \quad (20)$$

where:  $j_0(z)$  and  $j_1(z)$  are Bessel spherical functions. Form (20) represents all the main properties of the behavior of a typical distribution function [26]: the presence of coordinate spheres, correlation decay, etc., but also includes certain parameters of the media ( $r_0, A(r_0)$ ). In principle, with the help of (20), one can construct also the relevant model equation of state [23].

### 3. The Relaxation of the Non-conservative Order Parameter Field

In this section, we study the time evolution of the order parameter field according to the exact solution defined in Sec.2, using the initial conditions modelled in terms of the static structure function  $g(\vec{k})$  (see the previous Section). The static structure factors have been calculated in Eq.(20). Substituting (20) into (16), (17), we obtain after some manipulations

$$\begin{aligned}
 J_1(\tau) \equiv J_1(\tau, \alpha) &= 1 - 2\pi\sqrt{2} \{ (1 - C) - \\
 &- \left[ \sqrt{\pi} \operatorname{erf} \left( \frac{\alpha}{\sqrt{\tau}} \right) - \alpha \exp \left( -\frac{\alpha^2}{\tau} \right) \right] - \\
 &- \frac{1}{4} \frac{A(r_0)}{r_0} \alpha^3 \frac{1}{\tau\sqrt{\tau}} \exp \left( -\frac{\alpha^2}{\tau} \right) \} \quad (21)
 \end{aligned}$$

where  $\operatorname{erf}(z)$  is the error function,  $\alpha \equiv \frac{r_0}{2\sqrt{2}}$ , and

$$\begin{aligned}
 J_2(\tau) \equiv J_2(\tau, \alpha) &= \\
 &= \exp(2\tau) - 1 + 4\pi\sqrt{2} \left\{ (1 - C) \left[ \sqrt{\frac{\sqrt{\pi}}{2}} \times \right. \right. \\
 &\times \left. \left. \left( 1 - \exp(2\tau) \operatorname{erf} \left( \frac{\alpha}{\sqrt{\tau}} \right) \right) + 4\alpha S(\tau, \alpha) \right] + \right. \\
 &\left. + \left[ (1 - C) \frac{\alpha}{2\sqrt{2}} - \alpha\sqrt{2}A(r_0) \right] \frac{\partial}{\partial\alpha} S(\tau, \alpha) \right\}, \quad (22)
 \end{aligned}$$

$$\begin{aligned}
 S(\tau, \alpha) &= \frac{\sqrt{\pi}}{i4\sqrt{2}} \times \\
 &\times \left\{ \exp(i2\sqrt{2}\alpha) \operatorname{erf} \left( i\sqrt{2\tau} + \frac{\alpha}{\sqrt{2\tau}} \right) + \right. \\
 &+ \exp(-i2\sqrt{2}\alpha) \operatorname{erf} \left( i\sqrt{2\tau} - \frac{\alpha}{\sqrt{2\tau}} \right) - \\
 &\left. - \exp(i2\sqrt{2}\alpha) + \exp(-i2\sqrt{2}\alpha) \right\}, \quad (23)
 \end{aligned}$$

$$\frac{\partial}{\partial\alpha} S(\tau, \alpha) = \frac{\sqrt{\pi}}{i4\sqrt{2}} \left\{ i2\sqrt{2} \times
 \right.$$

$$\begin{aligned}
 &\times \left[ \exp(i2\sqrt{2}\alpha) \operatorname{erf} \left( i\sqrt{2\tau} + \frac{\alpha}{\sqrt{2\tau}} \right) - \right. \\
 &- \exp(-i2\sqrt{2}\alpha) \operatorname{erf} \left( i\sqrt{2\tau} - \frac{\alpha}{\sqrt{2\tau}} \right) - \\
 &- \exp(i2\sqrt{2}\alpha) - \exp(-i2\sqrt{2}\alpha) \left. \right] + \\
 &+ \exp(i2\sqrt{2}\alpha) \frac{\partial}{\partial x} \operatorname{erf} \left( i\sqrt{2\tau} \right) + \\
 &+ \frac{\alpha}{\sqrt{2\tau}} + \exp(-i2\sqrt{2}\alpha) \operatorname{erf} \left( i\sqrt{2\tau} - \frac{\alpha}{2\sqrt{2\tau}} \right) \left. \right\}. \quad (24)
 \end{aligned}$$

Eqs.(21)–(24) represent an exact solution of Eq.(11). The asymptotics of  $\langle \tilde{\varphi}^2(\tau) \rangle$ , given by Eq.(15) are thus determined by the asymptotic law for the behaviour of the functions  $J_1(\tau, \alpha)$  and  $J_2(\tau, \alpha)$ . Taking the relevant limit  $\tau \rightarrow \infty$  in Eq.(21)–(24), we obtain

$$\begin{aligned}
 \langle \tilde{\varphi}(\tau) \rangle &\simeq \left[ 1 + \frac{\pi}{2} \frac{A(r_0)}{r_0} \frac{r_0^3}{\tau\sqrt{\tau}} \exp \left( -\frac{r_0^2}{8\tau} \right) \right] \times \\
 &\times \left[ 1 + \hat{\Omega}(A, \alpha, C) \frac{1}{\sqrt{\tau}} - B(\alpha, C) \frac{1}{\sqrt{\tau}} \exp(-2\tau) - \right. \\
 &\left. - \tilde{\Omega}(A, \alpha, C) \exp(-2\tau) \right]^{-1}, \quad (25)
 \end{aligned}$$

where

$$\begin{aligned}
 \hat{\Omega}(A, \alpha, C) &\equiv 2\pi r_0 \left[ (1 - C) \cos 2\sqrt{2}\alpha - \right. \\
 &- \frac{A(r_0)}{r_0} r_0^2 \sin 2\sqrt{2}\alpha \left. \right], \\
 B(\alpha, C) &\equiv 2\pi\sqrt{2\pi} (1 - C) \alpha, \\
 \tilde{\Omega}(A, \alpha, C) &\equiv 2\pi\sqrt{\pi} \left[ (1 - C) \sin 2\sqrt{2}\alpha + \right. \\
 &+ \frac{A(r_0)}{r_0} r_0^2 \cos 2\sqrt{2}\alpha \left. \right]. \quad (26)
 \end{aligned}$$

In the limit  $\tau \rightarrow \infty$ ,  $\langle \tilde{\varphi}^2(\tau) \rangle$  tends to 1. As one can see from (25), quantitatively a different scenario of relaxation for  $\langle \tilde{\varphi}^2(\tau) \rangle$  could be realized asymptotically as dependent on certain conditions. Namely, when

$$\hat{\Omega}(A, \alpha, C) \frac{1}{\sqrt{\tau}} > \tilde{\Omega}(A, \alpha, C) \exp(-2\tau), \quad (27a)$$

we have

$$\langle \tilde{\varphi}^2(\tau) \rangle \simeq \frac{1}{1 + \frac{\hat{\Omega}}{\sqrt{\tau}}} \approx 1 - \frac{\hat{\Omega}}{\sqrt{\tau}}. \quad (27)$$

Respectively, in the case of the inequality opposite to (27a), one has

$$\langle \tilde{\varphi}^2(\tau) \rangle \simeq \frac{1}{1 - \tilde{\Omega} \exp(-2\tau)} \approx 1 + \tilde{\Omega} \exp(-2\tau). \quad (28)$$

Thus, within the presented model, the initial conditions influence the character of the asymptotic behavior of the order parameter field, providing long-time memory effects. As will be shown in the next Section, this feature of the presented model makes the rigorous qualification of the type of phase transition by examining the asymptotic behavior of the order parameter field only, problematic. This question requires more theoretical and experimental investigation.

#### 4. The Relaxation of the Conservative Ordering Field

Consider now the behaviour of the parameter of ordering in the case of conservative fields given by Eq.(7). Using the quasi-linearization procedure defined by Eq.(10) and repeating the scheme developed for the non-conservative field (see relation (13)), we obtain

$$\begin{aligned} \langle \tilde{\varphi}^2(\tau') \rangle &= \int d\vec{k} \tilde{g}(\vec{k}) \exp[-2\tau' k^4 + \\ &+ 2k^2 \int_0^{\tau'} \langle \tilde{\varphi}^2(s) \rangle ds], \end{aligned} \quad (29)$$

where  $\tilde{g}(\vec{k})$  is given by (20), and  $\tau'$  is the dimensionless time of (8). After some calculations, from (29), we get

$$\langle \tilde{\varphi}^2(\tau') \rangle = \frac{2\pi}{\sqrt{2\tau'}} \int_{-\frac{\Xi(\tau')}{\sqrt{2\tau'}}}^{\sigma} d\sigma \exp(\sigma^2) \times$$

$$\times \sqrt{\frac{2\sqrt{2\tau'}\sigma + 2\Xi(\tau')}{4\tau'}} \tilde{g} \left[ \sqrt{\frac{2\sqrt{2\tau'}\sigma + 2\Xi(\tau')}{4\tau'}} \right], \quad (30)$$

where  $\Xi(\tau') \equiv \tau' - \int_0^{\tau'} \langle \tilde{\varphi}^2(s) \rangle ds$ .

Integral (30) is rapidly (namely exponentially) convergent and furthermore can be well estimated by the following expression:

$$\begin{aligned} \langle \tilde{\varphi}^2(\tau') \rangle &= \frac{\pi\sqrt{\pi}}{2} \frac{\sqrt{\Xi(\tau')}}{\tau'} \exp\left[\frac{\Xi^2(\tau')}{2\tau'}\right] \times \\ &\times \operatorname{erfc}^2\left[-\frac{\Xi(\tau')}{\sqrt{2\tau'}}\right] \tilde{g}\left[\sqrt{\frac{\Xi(\tau')}{2\tau'}}\right]. \end{aligned} \quad (31)$$

We are now going to simplify the rigorous functional (31). Since the function  $\exp\left[\frac{\Xi^2(\tau')}{2\tau'}\right] \operatorname{erfc}^2\left[-\frac{\Xi(\tau')}{\sqrt{2\tau'}}\right]$  behaves exponentially and the function  $\tilde{g}\left[\sqrt{\frac{\Xi(\tau')}{2\tau'}}\right]$  shows limited variations within the interval  $\left[\tilde{g}[0], \tilde{g}\left[\frac{1}{\sqrt{2}}\right]\right]$ , without losing the general features, functional (31) can be represented as follows:

$$y(\tau') = \gamma \frac{\Xi(\tau')}{\tau'} \exp(\tau'), \quad (32)$$

where

$$y(\tau') \equiv \langle \tilde{\varphi}^2(\tau') \rangle, \quad \gamma \equiv \frac{\pi^3}{4} \tilde{g}^2\left(\frac{1}{\sqrt{2}}\right).$$

Expression (32) produces a differential equation for the definition of  $y(\tau')$  of the Abelian type, namely

$$\frac{du}{d\tau'} = f_3 u^3 + f_2 u^2 + f_1 u, \quad (33)$$

where

$$\begin{aligned} f_3 &\equiv -2\nu\gamma^2\tau'^2 \exp(\tau'), \quad f_2 \equiv \nu\gamma^2 \exp(\tau'), \\ f_1 &\equiv \beta, \quad u(\tau') \equiv 1/2\tau'^2 y(\tau'), \end{aligned} \quad (34)$$

$$\nu = \begin{cases} 4, & \tau' \rightarrow \infty \\ 1, & \tau' \rightarrow 0, \end{cases}$$

$$\beta = \begin{cases} -\frac{1}{\tau'}, & \tau' \rightarrow 0 \\ \frac{1}{2} - \frac{1}{\tau'}, & \tau' \rightarrow \infty. \end{cases} \quad (35)$$

Eq.(33) cannot be integrated in quadratures, except for a few particular cases [ 29–31]. But, to conclude the qualitative analysis in our case, it is enough to show that exponentially relaxing functions belong to the class of asymptotical solutions of Eq.(33). In fact, it is simple to show that functions of the type

$$u(\tau') = [1 + \varepsilon(\tau')] / 2\tau'^2, \tag{36}$$

where

$$\varepsilon(\tau') = \left| \begin{array}{l} \rightarrow 0 \\ \tau' \rightarrow \infty \end{array} \right. \lim_{\tau' \rightarrow \infty} \varepsilon(\tau') = 0,$$

satisfy Eq.(33) if  $\varepsilon(\tau')$  tends to zero more rapidly than any power law (in principle, this strong condition is even not required). Eq.(36) perfectly satisfies Eq.(33) for any  $\varepsilon(\tau') = \hat{\Delta}(\tau') e^{-\Gamma\tau'}$  (where  $\hat{\Delta}(\tau')$  belongs to the class of functions of limited variations). Thus, the exponential relaxation law  $\langle \tilde{\varphi}^2(\tau') \rangle = 1 - \exp(-\tau'/\tau'_0)$  definitively follows from the scenario given above. Note that, strictly speaking, observable experimental data for the behaviour of the order parameter in a segregation show a weakly non-monotonic behavior superposed on the general saturation tendency (see Fig. 1) [3, 6, 13]. This behaviour can be simply described with the help of the factor  $\hat{\Delta}(\tau')$  which could be imagined to behave also non-monotonically (within the limited variation in the given interval). Note that solutions of the type  $\hat{\Delta}(\tau') \exp(-\hat{\Gamma}\tau')$  also asymptotically satisfy (33). When the Abelian equation (33) with enough degree of accuracy is reduced to a differential equation of the Riccati type

$$\frac{du}{d\tau'} = \gamma^2 u^2 - \frac{u}{\tau'} \tag{37}$$

which has a simple integral [29–31]:

$$u(\tau') = \frac{1}{c\tau' - \gamma^2\tau' \ln \tau'}. \tag{38}$$

Expression (38) produces the relevant function

$$y(\tau') = \langle \tilde{\varphi}^2(\tau') \rangle = \frac{c_1}{2\tau'} - \frac{\gamma^2 \ln \tau'}{2\tau'}, \tag{39}$$

where  $c_1$  is a certain constant. As follows from (39) for the relevant dimensionless times  $\tau'$ , the order parameter can behave nonexponentially slowly (we call this behavior as a metastable “heterogeneous state”). Such a critical dynamic decay belongs, of course, to the specific character of the considered model. But,

at the same time, it is known from the experiments with driven granular materials [3, 13] that a typical relaxation time of a segregation cluster remains almost unchanged as the rotation velocity varies to a value larger by one order of magnitude. This behavior could reflect the existence of a critical slow dynamic regime (in terms of the order parameter field description — the slow relaxation metastable state regime). It is clear that, in the framework of the considered model, the relaxation picture cannot depend qualitatively on parameters like, e.g., the ratio of the diameters of particles in a (binary)mixture because, on a naked eye level, the definition of order parameter for growing segregated phases is not influenced by such parameters.

### 5. Intermediate Scattering Functions and Dynamic Structure Factors. Scaling Behaviour.

The dynamics of the order parameter field can be properly expressed in the behavior of an intermediate scattering function

$$S(\vec{k}, t) \equiv \langle \tilde{\varphi}_{\vec{k}}^2(t) \rangle$$

as well as in the dynamic structure factor

$$S(\vec{k}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} S(\vec{k}, t) dt. \tag{40}$$

Explicit relations for the definition of  $S(\vec{k}, \omega)$  can be obtained using Eq.(13) as well as its analogue in the case of conservative fields, namely

$$S^{(n)}(\vec{k}, \tau) = \tilde{g}(\vec{k}) \exp[-2k^2\tau + 2\Xi(\tau)]. \tag{41}$$

$$S^{(c)}(\vec{k}, \tau') = \tilde{g}(\vec{k}) \exp[-2k^4\tau' + 2k^2\Xi(\tau')], \tag{42}$$

where indices  $n$  and  $c$  denote that the functions belong to the case of a non-conservative and conservative field, respectively. Substituting Eq. (14) into (41) and taking into account Eq. (15)–(18), we easily obtain a rigorous expression for  $S^{(n)}(\vec{k}, \tau)$  in the following form:

$$S^{(n)}(\vec{k}, \tau) = \tilde{g}(\vec{k}) (1 + J_2(\tau)) \exp[-2k^2\tau + 2\tau], \tag{43}$$

where  $\tilde{g}(\vec{k})$  and  $J_2(\tau)$  are defined by Eq.(20) and Eqs.(22, 23). On that way, considering Eq.(42)



simultaneously with Eqs.(29–31), one has an exact functional form, which defines  $S^{(c)}(\vec{k}, \tau')$ . Considered as functions of  $\vec{k}$ , relations (41), (42) are peaked for  $k_n^{(n)} = 0$ ;  $k_{c1}^{(c)} = 0$ ;  $k_{c2}^{(c)} = \frac{1}{\sqrt{2\tau'}} \sqrt{\Xi(\tau')}$ . As expected, in the case of non-conservative fields, only a soft critical mode characterized by the critical value of the wave vector  $k_c^{(n)} = 0$  is present. This is reminiscent of the second order phase transition. In the case of a conservative order parameter, together with the soft critical mode with  $k_{c1}^{(c)} = 0$  which is also present, we have a hard critical mode with  $k_{c2}^{(c)} = \sqrt{\Xi(\tau')/2\tau'}$ , which normally characterizes the first order phase transition. It is easily seen that, in the asymptotic limit  $\tau' \rightarrow \infty$ , if we adopt simply  $\langle \tilde{\varphi}^2(\tau') \rangle \simeq 1 / [1 + \tilde{A}e^{-2\tau'}]$  (see Sec. 3, 4), the relaxation of the conserving order parameter field is characterized by a certain dominant length scale (the scale of inhomogeneity):

$$l_c = \frac{2\pi}{k_{c2}^{(c)}} = 2\sqrt{2}\pi \sqrt{\frac{\tau'}{\Xi(\tau')}} = 2\sqrt{2}\pi \times \sqrt{\frac{\tau'}{\left(1 - \frac{1}{\tilde{A}}\right)\tau' + \frac{1}{2\tilde{A}} \ln \left[\frac{1+\tilde{A}}{1+\tilde{A}\exp(-2\tau')}\right]}}. \quad (44)$$

In the limit  $\tau' \rightarrow \infty$ , Eq.(44) can be approximated as

$$l_c \simeq \frac{2\sqrt{2}\pi}{\sqrt{1 - \frac{1}{\tilde{A}}}} \times \left[ 1 - \frac{\ln(1 + \tilde{A})}{4\tilde{A}} \cdot \frac{1}{\tau'} + \frac{1}{4} \frac{\exp(-2\tau')}{\tau'} \right], \quad (45)$$

$$\lim_{\tau' \rightarrow \infty} l_c(\tau') = 2\sqrt{2}\pi / \sqrt{1 - \tilde{A}}.$$

Since  $l_c(\tau')$  evidently increases with time,  $k_{c2}^{(c)}(\tau')$  shifts to smaller wave vectors as time proceeds. This means that the segregation cluster, for instance, increases in size. This behaviour also implies the increasing of the amplitude of the intermediate scattering function as

$$\sim \exp \left\{ \left(1 - \frac{1}{\tilde{A}}\right)\tau' + \frac{1}{2\tilde{A}} \ln \left[ \frac{1 + \tilde{A}}{1 + \tilde{A}\exp(-2\tau')} \right] \right\}. \quad (46)$$

This restructurization (because of the conservation of the integral intensity) requires also the simultaneous narrowing of the maximum (peak) of  $S^{(c)}(\vec{k}, \tau')$ . It is interesting to note that if we assume that Eq.(45) is still valid for time intervals large enough to provide the competition between the inverse power-law and the exponential behavior of  $l_c$ , we have

$$l_c(\tau') = 2\sqrt{2}\pi \sqrt{\frac{\tilde{A}}{\tilde{A} - 1}} \times \begin{cases} 1 + \frac{1}{4} \cdot \frac{\tilde{A}}{\tilde{A} - 1} \frac{\exp(-2\tau')}{\tau'} & , \tau' < \tau'_0, \\ 1 & , \tau' = \tau'_0, \\ 1 - \frac{1}{4} \cdot \frac{\ln(1 + \tilde{A})}{\tilde{A}} \frac{1}{\tau'} & , \tau' > \tau'_0. \end{cases} \quad (47)$$

Therefore we obtain that, for a certain time,

$$\tau'_0 = \frac{1}{2} \ln \frac{\tilde{A}}{\ln(1 + \tilde{A})}, \quad (48)$$

$l_c(\tau')$  becomes time independent (that means also that  $l_c(\tau')$  is time independent in a close vicinity of  $\tau'_0$ ). For times smaller than  $\tau'_0$ , with growing  $\tau'$ ,  $l_c(\tau')$  decreases almost exponentially ( $\tilde{A} > 1$ ).

If  $\tau' < \tau'_0$ , a rapid growing of the inhomogeneity takes place (i.e., the infrastructure of a segregated cluster is strongly developed). Then, for a certain  $\tau' \simeq \tau'_0$ , we obtain an inhomogeneous intermediate state which further relaxes non-exponentially slowly to the steady state. In this intermediate state, the development of the segregated cluster becomes a slow (non-exponential) process due to, for instance, the entropy of displacement [17]. Taking Fourier transforms of (41), (42) and again adopting the asymptotic form for  $\langle \tilde{\varphi}^2(\tau) \rangle$ , after integration ([27,28]), and selecting the real part of the resulting form, we obtain dynamic structure factors  $S^{(n)}(\vec{k}, \omega)$  and  $S^{(c)}(\vec{k}, \omega)$  in the following analytical form:

$$S^{(n)}(\vec{k}, \omega) = \frac{1}{2} \tilde{g}(\vec{k}) \exp\left(\frac{\ln(1 + \tilde{A})}{\tilde{A}}\right) \times \Gamma \left[ k^2 - \left(1 - \frac{1}{\tilde{A}}\right) \right] \cos \tilde{\theta}(\omega, k) \times \prod_{j=0}^{\infty} \frac{\left| j + k^2 - \left(1 - \frac{1}{\tilde{A}}\right) \right|^2}{\sqrt{\frac{\omega^2}{4} + \left[ j + k^2 - \left(1 - \frac{1}{\tilde{A}}\right) \right]^2}}, \quad (49)$$

$$\begin{aligned}
 S^{(c)}(\vec{k}, \omega) &= \frac{1}{2} \tilde{g}(\vec{k}) \exp \left[ \frac{\ln(1 + \tilde{A})}{\tilde{A}} k^2 - \right. \\
 &- 4 \left( k^2 - \left( 1 - \frac{1}{\tilde{A}} \right) \right) k^2 \ln k \left. \right] \times \\
 &\times \Gamma \left[ k^2 \left( k^2 - \left( 1 - \frac{1}{\tilde{A}} \right) \right) \right] \times \\
 &\times \cos(2\omega \ln k + \theta(\omega, k)) \times \\
 &\times \prod_{j=0}^{\infty} \frac{|j + k^4 - (1 - \frac{1}{\tilde{A}}) k^2|}{\sqrt{\frac{\omega^2}{4} + [j + k^4 - (1 - \frac{1}{\tilde{A}}) k^2]^2}},
 \end{aligned} \tag{50}$$

where

$$\begin{aligned}
 \tilde{\theta}(\omega, k) &= -\frac{\omega}{2} \psi \left[ k^2 - \left( 1 - \frac{1}{\tilde{A}} \right) \right] + \\
 &+ \sum_{j=0}^{\infty} \left[ \frac{-\frac{\omega}{2}}{j + k^2 \left( 1 - \frac{1}{\tilde{A}} \right)} - \right. \\
 &\left. - \operatorname{arctanh} \frac{-\frac{\omega}{2}}{j + k^2 - \left( 1 - \frac{1}{\tilde{A}} \right)} \right]
 \end{aligned} \tag{51}$$

$$\begin{aligned}
 \theta(\omega, k) &= -\frac{\omega}{2} \psi \left[ k^4 - \left( 1 - \frac{1}{\tilde{A}} \right) k^2 \right] + \\
 &+ \sum_{j=0}^{\infty} \left[ \frac{-\frac{\omega}{2}}{j + k^4 - \left( 1 - \frac{1}{\tilde{A}} \right) k^2} - \right. \\
 &\left. - \operatorname{arctanh} \frac{-\frac{\omega}{2}}{j + k^4 - \left( 1 - \frac{1}{\tilde{A}} \right) k^2} \right]
 \end{aligned} \tag{52}$$

and  $\Gamma(z)$  and  $\psi(z)$  are gamma and digamma functions, respectively. In the low frequency limit ( $\omega \rightarrow 0$ ), Eqs.(49–52) lead to

$$S^{(c)}(\vec{k}, \omega) \simeq \frac{1}{2} \tilde{g}(\vec{k}) \exp \left[ \frac{\ln(1 + \tilde{A})}{\tilde{A}} k^2 - \right.$$

$$\begin{aligned}
 &- 4 \left( k^2 - \left( 1 - \frac{1}{\tilde{A}} \right) \right) k^2 \ln k \left. \right] \times \\
 &\times \cos \left[ \omega \left( 2 \ln k - \frac{1}{2} \right) \psi \left[ k^4 - k^2 \left( 1 - \frac{1}{\tilde{A}} \right) \right] \right],
 \end{aligned} \tag{53}$$

$$\begin{aligned}
 S^{(n)}(\vec{k}, \omega) &\simeq \frac{1}{2} \tilde{g}(\vec{k}) \Gamma \left[ k^2 - \left( 1 - \frac{1}{\tilde{A}} \right) \right] \times \\
 &\times \cos \left[ \frac{\omega}{2} \psi \left[ k^2 - \left( 1 - \frac{1}{\tilde{A}} \right) \right] \right].
 \end{aligned} \tag{54}$$

Note that, in the limit when  $k \rightarrow 1$ , the ratio  $S^{(c)}(\vec{k}, \omega) / S^{(n)}(\vec{k}, \omega)$  becomes independent of the initial state and depends on the value of the parameter  $A$  only:

$$\left. \frac{S^{(c)}(\vec{k}, \omega)}{S^{(n)}(\vec{k}, \omega)} \right|_{\omega \rightarrow 0} \simeq \exp \left( \frac{\ln(1 + \tilde{A})}{\tilde{A}} \right). \tag{55}$$

Eq.(55) shows that, in the considered low-frequency limit,  $S^{(c)}(\vec{k}, \omega)$  and  $S^{(n)}(\vec{k}, \omega)$  are simply proportional to each other. Thus, for a certain value of size of the growing segregated cluster  $l_c \sim \frac{1}{k} \sim 1$ , the dynamical picture of relaxation of the order parameter field has the same character in both cases of conservative or non-conservative fields. It should be noted that the function  $S^{(c)}(\vec{k}, \omega)$  could show dynamic scaling behavior. Namely, it follows from Eq.(42) that:

$$\frac{S^{(c)}(\vec{k}, \tau')}{S^{(c)}(\vec{k}_0, \tau')} = \exp \left\{ \Xi(\tau') k_0^2 \left[ -\frac{k^4}{k_0^4} + 2 \frac{k^2}{k_0^2} - 1 \right] \right\}, \tag{56}$$

where

$$k_0^2 = \Xi(\tau') / 2\tau' \tag{57}$$

is the coordinate of the peak of the function  $S^{(c)}(\vec{k}, \tau')$ . Of course, generally speaking, scaling is always approximate since there is never rigorously a single length scale, but only the dominance of a single length scale. But it is possible to show that scaling becomes more accurate as time proceeds, and especially for

some particular types of relaxation of  $\langle \tilde{\varphi}^2(\tau') \rangle$  in the asymptotic region, namely if  $\langle \tilde{\varphi}^2(\tau') \rangle$  follows Eq.(27) (this law,  $\langle \tilde{\varphi}^2(\tau') \rangle = 1 - \frac{\hat{\Omega}}{\sqrt{\tau'}}$ , asymptotically satisfies Eq.(33) as a matter of fact and thus belongs to the class of solutions of the general equation of motion (7)).

The dynamic scaling represented by Eq.(56) becomes explicit ( $\Xi(\tau') k_0^2 \rightarrow 1$  when  $\tau' \rightarrow \infty$ ). It should be stressed that, in general, the dynamic structure factor scaling functions depend basically on the initial stage, and possibly on the particular manner in which the quenching is realized. However, we found that there is a remarkably little variation of the scaling functions as functions of the initial parameters. This property of the equation of motion for the order parameter makes the dynamic scaling functions universal in the sense that they are independent of the initial conditions and quenching characteristics. The corresponding analysis of the sum rules and different asymptotic forms for explicit expressions of the dynamic structure factors is performed in [20].

## Conclusions

The proposed approach, based on an analysis of experiments with segregation in driven dry granular systems which include systems (mixtures) of beads incorporated in driven containers (for instance in 2D rotating drums or 3D cylinders), shows the possibility (at least from the outlook) of qualitatively describing this phenomenon with a partial nonlinear differential equation coupling the two phases and reducing it to a simple reminiscence of the Landau—Ginsburg (for non-conservative fields of the relevant parameter of ordering) or the Kahn—Hilliard (for conservative ordering fields) pictures of phase transitions. Qualitatively, our model theoretical results agree well with experiments and also with molecular dynamics simulations. The outlined analogies between segregated granular systems and phase transitions suggest a link between visual mechanics (dynamics) and statistical physics which remains to be constructed. New results are obtained in the presented models on the basis of rigorous solutions and estimation methods, in terms of order parameter relaxation field behavior, amenable to experimental checks. We suggest that the segregation in driven granular systems (as well as in general — diffusion-limited aggregation [20]) is a kinetic critical phenomenon.

The obtained results, which have a rigorous character within the purposed quasi-linearization procedure, could

be applied also without any transformations to the investigation of the kinetics of the first order and second order phase transitions near the critical points, where the investigated metastable relaxational state could play an important role [16]. After the analysis given in previous Sections, one may expect that the phenomenon of segregation can be pictured in the form of the formation of metastable, even locally frozen, clusters. We believe that the specific features of this model are not too crucial in the argument, and a similar reasoning can be carried over to other models like, for instance, percolational segregation or some others types of partial ordering. We require that spatial variations be small in comparison with mean free paths and with the size of any of the constituent particles. And one of the sources of the possible slow motion is the granular flow of the considered quantities from one part of the system to another (for instance, from the periphery to the axis of symmetry in a driven container). These notions may be described by stating that each part of the system is in a local thermodynamic equilibrium and then using conservation laws as flow equations, as was done in our case. Leaving the detailed description apart, note here that if we will interpret the average order parameter, for instance as a probability for a certain quantity to belong to a segregated cluster state (steady state), then the Ruelle—Sinai—Bowen—Cohen theorem for dynamic dissipative systems is valid [15, 16]. This fact seems a demonstration also of the Sinai theorem for adiabatic piston-type behavior in terms of simply determined parameters of systems with a complex behavior [15, 16]. Note that additional slow motions may arise when there is a broken symmetry in the system. We shall not worry much about the broken symmetries here. The next less well understood (and experimentally unexplored) source of slow relaxation is glassy behavior, with its partial freezing of degrees of freedom and its concomitant slow relaxation to full equilibrium (this behavior was predicted and described in Section 5 as a slow relaxational heterogeneous state of our proposed model in the case of conservative order parameter field). Here we should say that one may expect a considerable glassiness in driven granular systems. In our opinion, a detailed analysis of experimentally observed (and theoretically explained, see Sections 1—5) data reveals the existence of damped oscillations superimposed on the exponential relaxation of the order parameter. Our results demonstrate that while the proposed model qualitatively behaves in agreement with observed scenarios, the phenomena of segregation in terms of order parameter fields still offer surprises.

A full and detailed description of this effect (among many other intriguing properties of granular materials) continues to be an interesting challenge in theoretical and experimental physics.

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#### КІНЕТИКА СЕГРЕГАЦІЇ В ГРАНУЛЬОВАНИХ СИСТЕМАХ

O.I. Герасимов, П.П.Дж.М. Шрам, К. Китахара

#### Резюме

Запропоновано розв'язувану кінетичну модель, яка описує процес сегрегації у гранульованій суміші як фазовий перехід до асимптотичного квазірівноважного стану. Нелінійне рівняння руху для відповідно визначеного параметра порядку розв'язано точно у квазілінійному наближенні. Отримані теоретичні результати якісно пояснюють прямий фізичний експеримент з радіальної сегрегації сухої гранульованої суміші в полі віброприскорювань в термінах релаксації поля параметра порядку.

#### КІНЕТИКА СЕГРЕГАЦИИ В ГРАНУЛИРОВАННЫХ СИСТЕМАХ

O.I. Герасимов, П.П.Дж.М. Шрам, К. Китахара

#### Резюме

Предложена решаемая модель, которая интерпретирует процесс сегрегации в гранулированной смеси как фазовый переход в асимптотическое квазиравновесное состояние. Нелинейное уравнение движения для соответствующим образом определенного параметра порядка проинтегрировано в квазилинейном приближении. Полученные результаты качественно описывают данные прямого физического эксперимента по наблюдению радиальной сегрегации в смеси сухих гранулированных материалов, помещенных в поле виброускорений.