



# Cooperative length approach for granular media

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## Abstract

We analyze the non-equilibrium relaxation properties of granular materials in the perspective of a cooperative length approach of Adam and Gibbs. The existence of complex geometrical interactions between the grains induces a rough landscape in the structure of the allowable phase space and in the configurational entropy. At their turn these effects induce the need of complex cooperative rearrangements which account for the very slow relaxation dynamics of these systems. On this basis we relate quantitatively the characteristic dynamical properties to quasistatic quantities, e.g. free-volume and configurational entropy. © 1999 Elsevier Science B.V. All rights reserved.

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Granular matter represents a very rich research field which just recently started to interest people from statistical mechanics and physics in general [1–3]. In spite of our habit to handle them in our daily experience, granular materials show a very rich variety of features and unexpected behaviors, most of which are just barely understood in terms of basic physics principles. Compaction phenomena under vibration [4–12] segregation of different species [13], non-trivial properties of piling, very irregular stress distributions [14] are just few examples of this rich phenomenology.

In this paper we focus on the non-equilibrium dynamical properties of these non-thermal systems trying to make a bridge between some quasi-static properties and the corresponding relaxation behaviors. From this point of view it is worth to recall

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how much emphasis has been put on the close similarity of the relaxation properties of granular materials with the corresponding features of *glassy* systems [5–7,14–17]: slow-relaxations, divergence of the characteristic times, hierarchical structure of the accessible phase space, etc., are terms related to the phenomenology of granular media.

We thus try to identify the main ingredients which could account for these non-equilibrium properties. In this perspective we stress how *geometry* plays a crucial role in these non-thermal systems, many interactions between the grains being of geometrical origin (either like repulsive hard-core forces or entropically, always due to geometrical reasons, attractive forces). Generally speaking the introduction of whatever degree of frustration (quenched or annealed), say the difficulty for the particles to find the close packing configuration, makes some configurations forbidden, so inducing a drastic reduction of the volume of the allowable phase space. These modifications of the entropic properties of the system induce a hierarchy in the dynamics of the system which implies the need of cooperative rearrangements of the particles in order to allow a single grain displacement.

When the density increases the length associated to the cooperative region diverges and the system exhibits a glass transition which can be related to the so-called Reynolds-dilatancy transition.

Starting from this point it is possible to relate many non-equilibrium properties to configurational entropy and to excluded-volume (or equivalently free-volume [11]) effects.

The outline of the paper is as follows. We first develop a cooperative length approach inspired by the Adam–Gibbs theory for Glass-Forming liquids [18]. This approach allows us to estimate the relaxation time of a system whose configurational space reflects the presence of a certain degree of geometrical frustration. After having pointed out the relationship with the free-volume based theories we show in detail how complex relaxational behaviors arise as a consequence of the Vogel–Fulcher structure of the relaxation time. This relaxation obeys a logarithmic decay followed by an exponential one. Finally, we remark how in the case of the so-called *Tetris*-like models one observes the aforementioned phenomenology.

It is well known that by applying a sequence of taps to a granular system, the density relaxes logarithmically to its equilibrium value. This is found experimentally and in models [4–12]. A tap usually consists of a vibration applied to the system characterized by an energy  $E = A^2\omega^2$  where  $A$  and  $\omega/2\pi$  are the amplitude and the frequency of the applied vibration, respectively.  $E$  can be related to an effective *temperature*. In experiments usually the quantity  $\Gamma = \frac{A\omega^2}{g}$ , is implied to parametrize the intensity of the applied vibration, where  $g$  is the gravitational acceleration. After many taps, the system will reach a stationary state characterized by a density  $\rho_{\text{eq}}$  which, for a fixed frequency, will depend on  $E$  (or  $\Gamma$ ).

More generally, we consider the case in which the system is subjected to vibrations applied in a suitable way, and it reaches, after a long time  $t$ , an equilibrium density  $\rho_{\text{eq}}$ . The question that we address is how does the system relax to its stationary state as

function of  $\rho_{\text{eq}}$ . To answer this question we will adapt the cooperative length approach to the glass transition of Adam–Gibbs to granular materials. Free-volume theories and cooperative rearrangement theories are well known from the 1960s where they were proposed as phenomenological approaches to the relaxation behavior of glass-forming liquids (see for instance [18–20]). In the case of granular matter the complex entanglement of the grains, induces very strong correlations into the system: one cannot rearrange single elements of the system without rearranging at the same time an entire region. The cooperative length approach provides us with a quantification of how much important are these cooperative effects.

Consider a  $d$ -dimensional granular system, whose volume is  $L^d$ . Imagine to divide the system in cooperative regions of volume  $\xi^d$  which are defined as the smallest regions that can be rearranged in a new configuration without the need of rearranging the particles outside their boundaries. These regions are then defined in such a way that the number of allowed configurations increases at most in a sub-exponential way with their size  $\xi$ . The probability of a rearrangement inside such a cell or, what is the same, the rate for an effective configurational change (measured in units of the corresponding quantity for a non-frustrated system) can be easily written as

$$\tau^{-1} \sim \frac{n_f}{n_u}, \quad (1)$$

where  $n_f$  and  $n_u$  are the number of configurations in which can be found the frustrated and the non-frustrated  $\xi$ -cell, respectively.  $n_f$  and  $n_u$  are thus measures of the phase space volume of the systems of linear dimension  $\xi$  (frustrated and non-frustrated). By indicating with  $\sigma_f$  and  $\sigma_u$  the configurational entropies per particle relative to  $n_f$  and  $n_u$ ,<sup>1</sup>

$$\sigma_{f/u} = \frac{1}{\xi^d} \log n_{f/u}, \quad (2)$$

we define with  $S_f$  and  $S_u$  the analogous quantities for the whole systems (of volume  $L^d$ ). Using the property of the entropy per particle to be an intensive quantity we have  $\sigma_f = S_f$  and  $\sigma_u = S_u$ . From (1) and (2) it follows:

$$\tau^{-1} \simeq e^{\xi^d(S_f - S_u)}. \quad (3)$$

Usually, the number of configurations increases exponentially with the volume, however, in above defined the cooperative regions, where only global rearrangements are allowed due to the entanglement of the grains,  $n_f$  increases in a sub-exponential way with  $\xi$  giving then  $\xi^{-d} \sim S_f$ . For large values of the density,  $\xi$  can grow and can even diverge at some value  $\rho^*$ , which implies that  $S_f \rightarrow 0$  for  $\rho \rightarrow \rho^*$ . Assuming that  $S_f$  vanishes linearly as  $S_f \sim \rho^* - \rho$ , from Eq. (3) then the relaxation time diverges according to the law

$$\tau \sim e^{c\rho/(\rho^* - \rho)}, \quad (4)$$

<sup>1</sup> We suppose that the  $\xi$ -cells are large enough to allow a statistics and in particular to allow for the definition of the configurational entropy.

where  $c$  is a constant. This is the Vogel–Tamman–Fulcher behavior, predicted by Adam and Gibbs for glass-forming liquids [18], which describes the characteristic dependence of relaxational time scales on density (or temperature) in real experiments [21,22].

In a granular medium when the density becomes high enough the cooperative regions extend up to the dimension of the system (eventually infinite) and a local rearrangement of particles can only occur through a global rearrangement of a large cooperative region. Eqs. (3) and (4) tell us then that when the density increases cooperative regions extend, the corresponding characteristic time for a configurational change increasing exponentially. In granular materials an exponentially diverging relaxation time could be checked experimentally, signalling the presence of a glass transition. We shall go back to this point later on.

Our picture can provide a mechanism for a better understanding of the Reynolds-dilatancy transition. It is well known that granular materials at high densities undergo a transition in density at  $\rho_c$ . For densities higher than  $\rho_c$ , the material must dilate (i.e. expand) if it is deformed. This transition has never been studied seriously from the theoretical point of view. Our cooperative length seems to be a good measure for a correlation length of that transition, since it describes the size of a frustrated blocked cluster. The divergence of this length, accompanied by the divergence of the viscosity, should describe the approach to this transition. If the density is increased even further, this length is infinite which means that there exists a spanning immobile cluster which can only be destroyed if the system dilates. In our picture the regime of high density would correspond to the glassy state and we expect therefore that  $\rho^* = \rho_c$ .

Our picture points out the importance of local dynamical heterogeneity. Beyond the simple picture of extended shear planes as macroscopically observed in deformed packings for  $\rho > \rho_c$ , one would expect for  $\rho < \rho_c$  a multitude of dilated regions of the characteristic size of the cooperative length and it would be interesting to experimentally verify the presence of these dynamical heterogeneities as also found in glasses [23].

The existence of a relaxation time with the general functional form (4), with  $\rho^* = \rho_{\max}$ , can be seen as the outcome of a free-volume theory for granular media. Although the Adam–Gibbs approach is in principle more general the two phenomenological approaches reproduce the same qualitative results. Following Bouteux and De Gennes reasoning [11] one can define the free volume as the excess of volume per particle which can be redistributed in the system at a certain density. In a box of total volume  $V$  a set of  $N$  monodisperse particles each one of volume  $w$  occupies a volume  $V_{\text{occ}} = Nw$ . If  $N_{\max}$  is the maximal number of particles the box can contain, the maximal volume  $V_{\max} = N_{\max}w$ , then the free-volume per particle, at each given volume fraction,  $\rho = Nw/V$ , is

$$v_{\text{free}} = \frac{V_{\max} - V_{\text{occ}}}{N} = w \frac{\rho_{\max} - \rho}{\rho}. \quad (5)$$

If one postulates [11] an exponential distribution,  $f(v)$ , of empty volumes into the system,  $v_{\text{free}}$  will represent the average empty volume in  $f(v) = (1/v_{\text{free}})e^{-v/v_{\text{free}}}$ . Each

particle will be able to make a move provided that it finds an empty volume greater or equal to its own volume. The distribution of voids larger than  $w$  is

$$P(v > w) = \int_w^\infty f(v) dv = e^{-w/V_{\text{free}}} = e^{-\rho/(\rho_{\text{max}} - \rho)}. \tag{6}$$

This behavior can lead to exponentially long relaxation times as the free-volume tends to vanish [24,25,8,9]. It is worth to stress how free-volume theories are, in principal, less general than the theory based on the existence of cooperative regions. Free-volume theories just take into account geometrical effects whereas a cooperative length approach, being very general, can take into account, in principle, any factors inducing a frustration in the system, e.g. energetic barriers.

Let us now push forward our approach *à la* Adam–Gibbs. In order to get more detailed informations on the relaxation process in the system, we consider its general master equation for the density  $\rho(x, t)$  at any spatial point  $x$  at time  $t$  which can be written

$$\partial_t \rho(x, t) = - \sum_{x'} W(x, x') \rho(x, t) + \sum_{x'} W(x', x) \rho(x, t), \tag{7}$$

where  $W(x, x')$  is the transition probability per unit time from  $x$  to  $x'$ . We assume that the density at site  $x$  is out of equilibrium and is surrounded by a reservoir which is at equilibrium with a density  $\rho = \rho_{\text{eq}}$ . Notice that  $\rho_{\text{eq}} \leq \rho^* \leq \rho_{\text{max}}$ . In this case Eq. (7) reads

$$\partial_t \rho(t) = - \frac{\rho(t)}{\tau(\rho_{\text{eq}})} + \frac{\rho_{\text{eq}}}{\tau(\rho(t))}, \tag{8}$$

where we have defined  $\tau^{-1}(\rho_{\text{eq}}) = \sum_{x'} W(x, x')$  and  $\tau^{-1}(\rho(t)) = \sum_{x'} W(x', x)$  taking into account that the characteristic time  $\tau$  only depends on the density at that point. In order to include the cooperative effects of the entire system the characteristic time is chosen, in the region where  $\rho$  is close to  $\rho^*$ , with the same form predicted by the Adam–Gibbs theory, i.e.  $\tau(\rho) = \exp[\rho/(\rho^* - \rho)]$ , where, for sake of simplicity, we choose  $c = 1$  and the prefactor equal to unity. Remember that  $\rho^*$  is the density where the relaxation time diverges,  $\rho^* = \rho_{\text{max}}$  being a particular case.

In the region where  $\rho$  is close to  $\rho_{\text{eq}}$ ,  $\delta\rho = (\rho_{\text{eq}} - \rho(t))/\rho_{\text{eq}} \ll 1$ , one can identify two typical regimes for the density relaxation. For  $\delta\rho > (\rho^* - \rho_{\text{eq}})^2/\rho_{\text{eq}}^2$  the second term in Eq. (8) dominates the dynamics and gives rise to a logarithmic behavior (similar to that of Refs. [5–10]):

$$\frac{\rho(t) - \rho^*}{\rho^* - \rho(0)} = - \frac{1}{1 + B \ln(t/\tau_0 + 1)}, \tag{9}$$

where  $B$  and  $\tau_0$  are dependent on  $\rho^*$  and  $\rho_{\text{eq}}$ . Very close to the equilibrium density, i.e. when  $\delta\rho \ll (\rho^* - \rho_{\text{eq}})^2/\rho_{\text{eq}}^2$ , one can linearize the equation and an exponential regime takes place with a characteristic time of the order of  $\tau(\rho_{\text{eq}}) = \exp(\rho_{\text{eq}}/(\rho^* - \rho_{\text{eq}}))$ . Let us notice how as  $\rho_{\text{eq}}$  approaches  $\rho^*$ , by changing, for instance, the vibration intensity, the relaxation time diverges, suggesting the existence of a glass transition at  $\rho = \rho^*$ . The relaxation time scale given by  $\tau_{\text{rel}} = \tau(\rho_{\text{eq}})$  gives the crossover, in the density

relaxation, from a non-trivial dynamical behavior at short times to a long time simply exponential decay.

Let us notice how this treatment shares some similarities with other proposed approaches [8,9,11]. In particular, Boutreux and De Gennes [11] considered an equation with just the second term in the right-hand side of Eq. (8) with  $\rho_{\text{eq}} = \rho^*$ . Nowak et al. [7–9] considered an absorption–desorption equation for a plane exchanging particle with a reservoir such that it can receive particle with a characteristic time independent of its density. In our approach we stress how the equilibrium density  $\rho_{\text{eq}}$  could be different from  $\rho^*$ , the density where the relaxation time diverges. That allows for an exponential regime after the logarithmic one. Furthermore, we consider the interaction with a real environment which is not perturbed by the arrival of particles but which poses problems to their packing via the term  $\tau(\rho_{\text{eq}})$  dependent on  $\rho_{\text{eq}}$ . This is the simplest way to introduce the cooperative effect given by the interaction with other parts of the system.

The general features in the above approach can be relevant to other systems where the relaxation time follows the Vogel–Fulcher behavior. For instance, in glass forming liquids we may expect, near the ideal glass transition and in the long-time regime, a logarithmic behavior eventually followed by an exponential tail.

Our theoretical analogies between glasses and granular systems do of course have their limitations. While the individual constituents of a glass are made of molecules, granular systems are made of particles with mass of many orders of magnitude larger than that of molecules, therefore there is an important difference concerning the concept of temperature. Moreover typically a granular packing is shaken, giving to the internal movements a periodic time dependence, eventually an anisotropic direction and often also shock-like waves. In a glass, temperature is a scalar, the motions are isotropic and random in time. Therefore in order to mimic our concept of temperature in a granular experiment, one would have to put vibrators at many locations, several frequencies and several directions. In fact, it is known that granular packings can have internal texture as described by the fabric tensor primarily due to the direction of the applied excitations. Considering all this, in the ideal case, the proposed experiments should be made to measure the relaxation time and check the proposed Vogel–Fulcher law. For instance one can consider a system lying on the reversible curve  $(\Gamma, \rho_{\text{eq}})$ , i.e. in the curve where at each given vibration *energy* corresponds a specific equilibrium density  $\rho_{\text{eq}}$  [8,9]. Starting at the density corresponding to the highest  $\Gamma$ , one can reduce progressively  $\Gamma$  and measure the characteristic time to reach the new equilibrium density  $\rho_{\text{eq}}(\Gamma)$ ,  $\tau(\rho_{\text{eq}})$ . According to our approach one would expect for this characteristic time a form given by Eq. (4). When  $\rho_{\text{eq}}$  gets closer and closer to  $\rho^*$  the characteristic time starts to diverge signaling the existence of a glass-like transition at  $\rho^*$ .

It is worth to stress, as a specific example of a system which is supposed to possess all the properties of the systems we mentioned up to now, the case of the so-called “Tetris” model [10,26] that exhibits a very slow relaxation dynamics due to high entropic barriers (originated by geometrical constraints) which have to be passed by the particles to improve the global packing. In this case one observes how the introduction

of frustration induces a dramatic effect on the entropy which vanishes as the density  $\rho \rightarrow 1$ . We do not present the long expression for  $S_F(\rho)$  but it is worth to note that near  $\rho = 1$  one has  $S_F(\rho)|_{\rho \rightarrow 1} \simeq (1 - \rho)\log(1 - \rho)$ . Let us notice how this represents an explicit example of a system for which it holds the hypothesis that  $S_F(\rho) \rightarrow 0$  for  $\rho \rightarrow \rho^*$  with  $\rho^* = \rho_{\max} = 1$ .

Furthermore one can calculate  $\xi$ , the size of the cooperative regions, for this particular system. If  $\rho$  is the coverage density, one has  $\xi \simeq \rho/(1 - \rho)$  and again we find a cooperative length which diverges (at  $\rho^* = 1$ ) and which brings, once inserted in (3), to a diverging characteristic time  $\tau \sim e^{\rho/(1-\rho)}$ . This expression has been checked by direct simulations on the *Tetris*-like system [26].

In summary in this paper we have proposed a simple phenomenological approach to explain the slow relaxation behaviors observed in granular media. It turns out that a crucial role for these systems is played by the so-called free volume that gives a measure of the extension of the cooperative regions into the system. It is the geometrical frustration felt by the grains that induces at its turn drastic modifications of the allowable phase space, i.e. of the configurational entropy, which corresponds to the existence of the cited cooperative regions. We have then related quantitatively the relaxation dynamical properties of these systems to quasistatic and geometrical properties. These concepts turn out to be relevant in many phenomena involving granular matter. We just mention the compaction under vibration, which exhibits a very slow inverse logarithmic behavior as a consequence of the high entropic barriers the system has to overcome to reach the close-packing density configuration, and the interesting phenomena related to the segregation of species in a medium subjected to shaking. We have proposed a set of possible experiments to check the predicted Vogel–Fulcher law for the behavior of the characteristic relaxation time. Finally, we have suggested how a similar approach could be relevant to describe the long-time behavior in glass forming liquids.

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