Entropy for Relaxation Dynamics in Granular Media

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We investigate the role of entropic concepts for the relaxation dynamics in granular systems. In particular, we show how, in the framework of a mean-field model introduced for compaction phenomenon, there exists a *free-energy-like* functional which decreases along the trajectories of the dynamics and which allows one to account for the asymptotic behavior: e.g., density profile, segregation phenomena. Also we are able to perform the continuous limit of the above mentioned model which turns out to be a diffusive limit. In this framework one can single out two separate physical ingredients: the *free-energy-like* functional that defines the phase space and the asymptotic states and a diffusion coefficient $D(\rho)$ accounting for the velocity of approach to the asymptotic stationary states.

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Granular media enter only partially into the framework of equilibrium statistical mechanics and hydrodynamics. Their dynamics constitutes a very complex problem of nonequilibrium which poses novel questions and challenges to theorists and experimentalists [1,2].

Generally speaking granular materials cannot be described as equilibrium systems neither from the configurational point of view nor from the dynamical point of view. It is known, in fact, that these systems remain easily trapped in some metastable configurations which can last for long time intervals unless they are shaken or perturbed. A granular system may be in a number of different microscopic states at fixed macroscopic densities, and, more in general, for a given ensemble of macroscopic parameters. Many unusual properties are linked to this nontrivial packing [1]. The configurational space of these systems is very complex and presents a structure with several local minima. This structure induces a dynamic behavior characterized by hierarchical relaxation phenomena with several associated time scales. A general mechanism bringing to the existence of such a structure is based on the concept of *frustration* that, for instance, in granular media has a geometrical origin. 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. In their turn these effects induce the need of complex cooperative rearrangements which account for the very slow relaxation dynamics of these systems. At high densities (or very low temperatures for thermal systems) the system remains trapped in a local minimum and exhibits a nonergodic behavior as well as very slow relaxations: the logarithmic compaction in granular media [3–7] or the Kohlrausch-Williams-Watts (KWW) relaxations in glassy systems [8].

In this paper we try to elucidate the role that the concept of entropy can play in the dynamics of these systems. In particular, we show how, in the framework of a mean-field model introduced for the compaction phenomenon, there exists a *free-energy-like* functional which decreases along the trajectories of the dynamics and which allows one to account for the asymptotic behavior: e.g., density profile, segregation phenomena. Furthermore the continuous limit of the above mentioned models allows us to comment on the relationship between entropic and dynamical effects in explaining the relaxation phenomena in granular media [2,9,10].

We consider a simple model which describes the evolution of a system of particles which hop on a lattice of $k = 0, \ldots, N$ stacked planes, as introduced in [6]. In particular, the system represents an ensemble of particles which can move up or down in a system of N layers in such a way that their total number is conserved. We ignore the correlations among particle rearrangements and problems related to the mechanical stability of the system. The master equation for the density on a generic plane k, except for the $k = 0$ plane, is given by

$$\partial_t \rho_k = (1 - \rho_k) D(\rho_k) [\rho_{k-1} p_{\mathrm{up}} + \rho_{k+1} p_{\mathrm{down}})] - \rho_k [(1 - \rho_{k-1}) D(\rho_{k-1}) p_{\mathrm{down}} + (1 - \rho_{k+1}) D(\rho_{k+1}) p_{\mathrm{up}}], \quad (1)$$

where p_{down} and p_{up} (with $p_{\mathrm{up}} + p_{\mathrm{down}} = 1$) represents the probability for the particles to move downwards or upwards, respectively, among the different planes. With p_{up} and p_{down} we can define the quantity $x = p_{\mathrm{up}}/p_{\mathrm{down}}$ which quantifies the importance of gravity in the system. We can also associate with x a sort of temperature for the system given by $T \sim 1/\log(1/x)$. We shall return to this point later on.

$D(\rho_k)$ represents a sort of mobility for the particles given by the probability that the particle could find enough space to move. Apart from other effects it mainly takes into account the geometrical effects of frustration, i.e., the fact that the packing prevents the free movement of the particles.

Later on we shall show how the analysis reported here is very general and does not depend on the exact functional form chosen for $D(\rho)$. For the sake of clarity before discussing the problem in its generality we shall consider a possible functional form for $D(\rho_k)$ suitable for the compaction problem. In a naive way one could imagine a functional form like $D(\rho_k) = \rho_k(1 - \rho_{k'})$ obtained by considering only the interactions with the nearest neighbor planes whose density is designed by $\rho_{k'}$. It is easy to realize that such an approach does not account for the complexity of the problem where the packing at high densities creates long range correlations in the system, and, using this functional form, the equations show a trivial relaxation. A possible general form of $D(\rho_k)$, which can be seen as the outcome of a theory based on the existence of regions of cooperativity [11] (as well as of a free-volume theory [5,7]) for granular media, includes a term like

$$D(\rho_k) = D_0 \exp[-\alpha/(1 - \rho_k)]. \tag{2}$$

The parameter α quantifies how much the shape and the dimensions of the particle frustrate its motion. The higher is α the higher will be the geometrical frustration felt by the corresponding particle. For instance, in the problems of parking of r-mers (i.e., segments of length r) on a line, under the hypothesis of an exponential distribution for the lengths of the empty (or filled) intervals, the probability for each r-mer to find a sufficient space to land is $\exp[-r/(1 - \rho)]$, where ρ is the occupation density on the line [5].

The question we want to address is whether there exists a variational principle driving the relaxation phenomena in this system and in general in granular media. In other words, one could ask if, in analogy with what happens for a liquid system, there exists some free-energy-like functional minimized (Lyapunov functional) [12] by the dynamical evolution. In a very general way it is possible to write explicitly a Lyapunov functional for the system of equations (1), say, the functional which decreases monotonically along the trajectories of the motion. This functional can be cast in the form of a free-energy-like function:

$$F = \sum_{k=0}^{\infty} [\gamma(x) k \rho_k - S(\rho_k)], \tag{3}$$

where the $S(\rho_k)$, the *entropiclike* contribution, and $\gamma(x)$ have to be determined in a self-consistent way by imposing that F decreases for any exchange of particles between two generic planes k and $k + 1$. Writing explicitly the expression for dF for a generic particle exchange and imposing $dF \leq 0$, for the particular choice of $D(\rho_k)$ we made, after some manipulations one gets for the functional (3) the expressions

$$S(\rho_k) = \rho_k \log\left(\frac{1 - \rho_k}{\rho_k}\right), \qquad \gamma(x) = \log\left(\frac{1}{x}\right). \tag{4}$$

Let us stress how $S(\rho_k)$ has exactly the same (critical) behavior, as $\rho_k \to 1$, of the entropy of a one-dimensional continuous system, filled with bars of unitary length at a certain density ρ. The functional F is a concave function because $\partial^2 F/\partial \rho_k^2 \geq 0 \ \forall \ k$ [13].

As a consequence there exists a unique minimum for this functional, and this minimum should correspond to the stationary state of the system. In the case of a monodisperse system we expect that the stationary state (i.e., the density profile for the system) is the one obtained, in a continuous process of shaking, after a very long transient.

In the general case with arbitrary N, and for $N \to +\infty$, it is possible to get the exact asymptotic stationary solution for the density on each plane. We denote with M the total "mass" of the system, i.e., the maximal number of planes which can be completely filled. Using a standard Lagrange multiplier method, where one tries to find the extremum of F subject to the constraint $\sum_k \rho_k = M$, the solution is given by the following implicit expression:

$$f(\rho_k) = f(\rho_0) x^k, \tag{5}$$

where

$$f(s) = \frac{s}{1 - s} e^{1/(1-s)}, \tag{6}$$

and ρ_0 is the density on the zeroth plane which is a complex function of the total mass of the system. In order to visualize the solution it is possible to extract the approximate explicit behaviors. In particular, one gets

$$\rho_k^\infty \simeq 1 - 1/[(M - k) \log(1/x)] \quad \text{for } k \ll M, \tag{7}$$

$$\rho_k^\infty \simeq e^{(M-k)\log(1/x)} \quad \text{for } k \gg M. \tag{8}$$

The stationary solution tends thus to a step function $\theta(k - M)$ in the limit $x \to 0$. This behavior is very well verified in the experiments [14].

Let us now ask what happens considering a bidisperse system, i.e., by considering two kind of particles defined by two different values of α ($\alpha_s = 1, \alpha_b = \alpha = 2$ in the specific case). In the case the master equations for the densities ρ_k^b and ρ_k^s on a generic plane k, except for the plane $k = 0$, are given by

$$\begin{aligned} \partial_t \rho_k^{b,s} = {} & D^{b,s}(\rho_k)[\rho_{k-1}^{b,s} p_{\mathrm{up}} + \rho_{k+1}^{b,s} p_{\mathrm{down}})] \\ & - \rho_k^{b,s}[D^{b,s}(\rho_{k-1}) p_{\mathrm{down}} + D^{b,s}(\rho_{k+1}) p_{\mathrm{up}}], \end{aligned} \tag{9}$$

where $\rho_k = \rho_k^s + \alpha \rho_k^b$ is the total density on the kth plane and $D^{b,s}(\rho_k)$ are the probabilities for a particle s or b, respectively, landing on the plane k, to fit the local geometrical environment. In this sense $D^{b,s}(\rho)$ are the analog of the mobilities for the two kinds of particles, and they take into account the cooperative effects on the dynamics generated by the frustration. A possible functional form for $D^{b,s}(\rho)$ (see for a similar approach [5–7,11]) is given by $D^s(\rho) \sim (1 - \rho) \exp[-1/(1 - \rho)]$ and $D^b(\rho) \sim (1 - \rho)^\alpha \exp[-\alpha/(1 - \rho)]$ with $\alpha \geq 1$.

Let us now look for a Lyapunov functional equivalent to Eq. (3) in this polydisperse case which now will have the form

$$F = \sum_{k=0}^{\infty} [\gamma(x) n \rho_k - S(\rho_k^b, \rho_k^s)], \tag{10}$$

where the $S(\rho_k^b, \rho_k^s)$ is again an *entropiclike* contribution. By repeating the calculation along the same lines as before one gets

$$S(\rho_k^b, \rho_k^s) = -\rho_k^s[\log(\rho_k^s) - 1] - \rho_k^b[\log(\rho_k^b) - 1] - (1 - \rho_k)[\log(1 - \rho_k) - 1] + \log(1 - \rho_k) \quad (11)$$

and $\gamma(x) = \log(1/x)$.

Even in this case the functional F is concave and there exists a unique solution corresponding to the asymptotic stationary state for the system. Using a standard Lagrange multiplier method one gets the result

$$\rho_k^b = \rho_k \frac{h_0(1 - \rho_k)^{\alpha-1} e^{-(\alpha-1)/(1-\rho_k)}}{1 + \alpha h_0(1 - \rho_k)^{\alpha-1} e^{-(\alpha-1)/(1-\rho_k)}},$$
$$\rho_k^s = \rho_k - \alpha \rho_k^b, \quad (12)$$

where $h_0 = h_0(\rho_0^b, \rho_0^s)$ is a constant which can be estimated by using the total mass M of the system:

$$h_0(\rho_0^b, \rho_0^s) = (1 - \rho_0)^{1-\alpha} e^{(\alpha-1)/(1-\rho_0)} \frac{\rho_0^b}{\rho_0^s}. \quad (13)$$

ρ_k is given by a nontrivial function of k that can be obtained exactly but in an implicit form. ρ_k is a monotonically decreasing function with the maximum for $k = 0$. Figure 1 shows an example of the asymptotic stationary solution obtained from (12) with $h_0 \simeq 10^3$ corresponding to $x \simeq 0.82$, $\rho_0 = 1 - \frac{c}{M \log(1/x)} \simeq 0.9$, and $\frac{\rho_0^b}{\rho_0^s} \simeq 1.5 \times 10^{-3}$. In particular, it shows ρ_k^b and ρ_k^s as a function of ρ_k. In this way, the effect of segregation [15] of the "big" particles (with $\alpha = 2$) on top of the "small" ones (with $\alpha = 1$) is clearly visible. The introduction of two different weights for the two species, say $p_{\text{up}}^{b,s}$ and $p_{\text{down}}^{b,s}$, gives rise to a rich phase space $(p_{\text{up}}^b, p_{\text{up}}^s, \alpha)$ with different regions corresponding to different behaviors for segregation.

Equation (11) makes evident how in the dynamics of granular media entropic effects are far from being negligible and the global behavior is given by a complex interplay between gravitational and entropiclike effects.

Let us now continue to push forward this point of view and ask whether the asymptotic stationary state corresponds to some sort of equilibrium state. At the equilibrium, i.e., asymptotically, one would expect that for any exchange of particles among different planes $dF = 0$ for the free-energy-like functional (10), or equivalently $dS/dE = 1/T =$ const. By defining the total energy $E = \sum_{k=0}^{\infty} k\rho_k$ and the total entropy $S_{\text{tot}} = \sum_{k=0}^{\infty} S(\rho_k)$, for any exchange of particles of type b or s between the planes k and $k + 1$ one has

$$\frac{dS_{\text{tot}}^{b,s}}{dE} = \frac{\partial S(\rho_{k+1})}{\partial \rho_{k+1}^{b,s}} - \frac{\partial S(\rho_k)}{\partial \rho_k^{b,s}}. \quad (14)$$

It is easy to realize how in the asymptotic stationary state one has

$$\frac{dS_{\text{tot}}^s}{dE} = \frac{dS_{\text{tot}}^b}{dE} = \log\left(\frac{1}{x}\right) = \text{const}, \quad (15)$$

or, what is the same, $\frac{dS_{\text{tot}}^{b,s}}{dE} =$ const everywhere in the system no matter which kind of particles we use for its definition. It is then tempting to associate $\log(\frac{1}{x})$ with the inverse of a temperature for the system. The analogy is made stronger by recalling that $1/\log(\frac{1}{x})$ is the usual quantity associated with a temperature in Monte Carlo dynamics. From this point of view the relaxation process of this system would correspond to an equilibration procedure in which $\frac{dS_{\text{tot}}^{b,s}}{dE}$ is made uniform everywhere. In the specific case of segregation one is forced to think that the system evolves in such a way that asymptotically all the particles have the same mobility, measured in this case in terms of the entropic change for every given displacement.

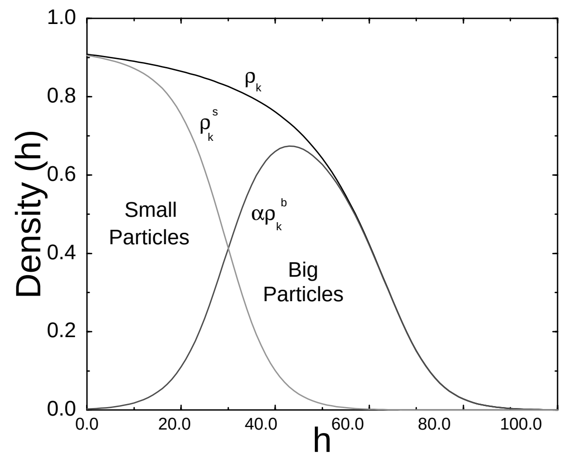

FIG. 1. Asymptotic density profile for the two species of particles in the N-plane model described in the text. Clearly visible is the effect of segregation of the more frustrated (big) particles ρ_b^k on top of the less frustrated (small) ones ρ_b^k.

Let us now generalize the results obtained so far. Equation (1) represents a particular case of a general class of equations that can be written as

$$\partial_t \rho_k = g(\rho_k)[f(\rho_{k-1})p_{\text{up}} + f(\rho_{k+1})p_{\text{down}}] - f(\rho_k)[g(\rho_{k-1})p_{\text{down}} + g(\rho_{k+1})p_{\text{up}}], \quad (16)$$

where f and g are generic functions for which we require only $f \geq 0$, $g \geq 0$, $df/d\rho \geq 0$, and $dg/d\rho \leq 0$. All the results we have shown in the particular case (1) are valid under these general assumptions. In particular, we can prove that there does exist a functional that decreases along the trajectories of the motion and its expression is given by Eq. (3) with

$$S(\rho_k) = \int_{\rho_k} \log \frac{g(\rho)}{f(\rho)} d\rho, \qquad \gamma(x) = \log\left(\frac{1}{x}\right). \quad (17)$$

A deeper insight in the above mentioned phenomenology is obtained by considering the continuum limit for the model described by Eq. (16). More precisely we consider a diffusive limit that consists of scaling the space variable as $\frac{1}{\epsilon}$, the time variable as $\frac{1}{\epsilon^2}$, and the drift term $p_{\text{down}} - p_{\text{up}}$ as ϵ. Therefore $x = \epsilon k$, $\tau = \epsilon^2 k/2$, $p_{\text{down}} - p_{\text{up}} = \epsilon \beta/2$, and we consider the evolution of $u(x) \equiv \rho(k)$.

We get the continuum limit by taking the Taylor expansion of the right member of Eq. (16) around $x = k\epsilon$. For example, $\rho(k+1) \equiv u(x+\epsilon) = u(x) + \epsilon \partial_x u + \frac{1}{2}\epsilon^2 \partial_{xx} u + O(\epsilon^3)$.

We get, formally,

$$\partial_\tau u(x) = \beta \partial_x(fg) + (g\partial_{xx} f - f\partial_{xx} g) + O(\epsilon), \tag{18}$$

which, in the limit $\epsilon \to 0$, gives

$$\partial_\tau u(x) = \beta \partial_x(fg) + (g\partial_{xx} f - f\partial_{xx} g). \tag{19}$$

This is a nonlinear diffusion equation that may be conveniently written in the following form:

$$\partial_\tau u = \partial_x\left(D(u)\partial_x \frac{\partial F}{\partial_u}\right), \tag{20}$$

where $D = fg$, $\frac{\partial F}{\partial_u}$ denotes the functional derivative of F with respect to u, $F = \int_0^\infty [\beta u x - S(u)]\, dx$, and $S' = \log(\frac{g}{f})$. Notice that the functional F decreases with the dynamics induced by Eq. (19). One has, in fact,

$$\partial_\tau F = \int dx \frac{\partial F}{\partial u}\, \partial_\tau u = \int dx \frac{\partial F}{\partial u} \partial_x\left(D(u)\partial_x \frac{\partial F}{\partial u}\right) \tag{21}$$

that, after an integration by parts, gives

$$-D(u)\left(\frac{\partial F}{\partial_u}\right)^2 \leq 0. \tag{22}$$

Therefore there exists a "free energy"-like functional F for Eq. (20) which has exactly the same form of the functional defined for the discrete model [see Eq. (17)]. We can notice that while the functional form of S and the value of β determine in a unique way the asymptotic state they are not sufficient to determine the dynamical behavior of the system. In particular, in order to know it one should know the functional form of $D(\rho)$.

What we have discussed so far suggests the possibility of introducing, for nonthermal systems as granular media, equilibrium concepts as free-energy, entropy, and temperature. More precisely it is possible (in the case studied here) to predict the asymptotic state by means of the minimization of a suitable functional which can be constructed by entropic arguments. It is worth stressing how granular systems often exhibit memory and so the existence of a unique Lyapunov functional is not guaranteed in general. In general one could expect that several Lyapunov functionals are associated with different stationary states reached with different dynamical paths.

Finally, let us notice how Eq. (20) allows us to comment on the relationship between entropic and dynamical effects in explaining the relaxation phenomena in granular media. Once the Lyapunov functional is known (and thus also the entropic properties of the system) it is possible to predict the asymptotic state. However, one cannot specify whether the asymptotic state is reached in a finite time unless one knows the connectivity properties of the phase space, which in our case corresponds to know $D(\rho)$.

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