Thermodynamic approach to dense granular matter: a numerical realization of a decisive experiment.

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Years ago Edwards proposed a thermodynamic description of dense granular matter, in which the grains (the ‘atoms’ of the system) interact with inelastic forces. The approach is intriguing but is not justified from first principles, and hence, in the absence of conclusive tests of its validity, it has not been widely accepted. We perform a numerical experiment with a realistic granular matter model specially conceived to be reproducible in the laboratory. The results strongly support the thermodynamic picture.

The similarities between a driven granular system and a system of molecules in a fluid have prompted the use of statistical ideas to describe granular matter \cite{2}, such as for example kinetic theories of gases to study rapid flows of low-density systems of inelastic particles \cite{3}. In this context, it is customary to associate the mean kinetic energy of the particles with a ‘granular temperature’, having no real thermodynamic meaning.

For the opposite limit of dense, slowly flowing granular matter, a more radical train of ideas was initiated more than a decade ago by Edwards and coworkers: the proposal of a statistical ensemble \cite{4,5}, and through it, thermodynamic notions such as entropy and temperature (the latter unrelated to the ‘granular temperature’ above). Although the idea of a thermodynamic description of granular matter was recognized as attractive, it was not universally accepted because there is no known first principle justification of Edwards’ statistical ensemble, such as there is for ordinary statistical mechanics of liquids or gases (Liouville’s theorem).

Recently, analytical developments and numerical work on schematic models originally devised for glasses have given a new perspective for the dynamics of dense granular matter \cite{6,7}. The theoretical support provided by this framework for a statistical approach has spurred a renewed interest in Edwards’ thermodynamics.

On the experimental side, years ago Nowak et al. \cite{13} studied in detail the density fluctuations in a vibrated granular material, and proposed that these fluctuations should reflect an underlying thermodynamics, much as they do in an ordinary thermal system. However, as far as the actual verification of Edwards’ approach, the evidence they found was if anything rather negative: we shall discuss below some possible reasons for this. On the other hand, Edwards’ approach has never been tested with simulations of realistic models of granular materials, characterized by the fact that energy is supplied by external driving (via shear or tapping) and dissipated by inelastic collisions and slippage between the grains.

In this work we describe a numerical experiment specially conceived to be reproducible in the laboratory, using a realistic model of sheared granular matter. Considering particles of different sizes inside the medium, we compute a dynamical temperature from an Einstein relation relating random diffusion and mobility (transverse to the shear direction) at long time scales. If there is an underlying thermodynamics, it will impose that the dynamical temperature be independent of the tracer’s shape: a very strong condition which we show to hold. We then perform an explicit computation to show that the temperature arising from Edwards’ thermodynamic description and the dynamic one measured from Einstein’s relation coincide. This last step cannot be performed in the laboratory, so the numerical simulation provides the missing link between thermodynamic ideas and diffusion-mobility checks.

Consider a ‘tracer’ body of arbitrary shape immersed in a liquid in equilibrium at temperature $T$. As a consequence of the irregular bombardment by the particles of the surrounding liquid, the tracer performs a diffusive, fluctuating ‘Brownian’ motion. The motion is unbiased, and for large times the average square of the displacement goes as $\langle |x(t) - x(0)|^2 \rangle = 2Dt$, where $D$ is the diffusivity. On the other hand, if we pull gently from the tracer with a constant force $f$, the liquid responds with a viscous, dissipative force. The averaged displacement after a large time is $\langle |x(t) - x(0)| \rangle = f\chi t$, where $\chi$ is the mobility. Clearly, the same liquid molecules are responsible both for the fluctuations and for the dissipation. Although both $D$ and $\chi$ strongly depend on the shape and size of the tracer, they turn out to be always related by the Einstein relation $D/\chi = T$, where $T$ is the temperature of the liquid.

Conversely, if in a fluid of unknown properties we find that several tracers having different diffusivities and mobilities yield the same ratio $D/\chi = T$, we may take this as a strong evidence for thermalisation at temperature $T$. Indeed, recent analytic schemes for out of equilibrium glassy dynamics have suggested the existence of such a temperature $T$ governing the slow components of fluctuations and responses of all observables \cite{6,10}.

In order to test the existence of this temperature for dense granular matter, we perform, with a realistic numerical model, a diffusion-mobility experiment in con-
ditions that can be reproduced in the laboratory. In the model, deformable spherical grains interact with one another via non-linear elastic Hertz normal forces, and non-linear elastic and path-dependent Mindlin transverse forces [4]. The normal force, \( F_n \), has the typical \( 3/2 \) power law dependence on the overlap between two spheres in contact, while the transverse force, \( F_t \), depends on both the shear and normal displacements between the spheres. For two spherical grains with radii \( R_1 \) and \( R_2 \): \( F_n = \frac{1}{4} k_n R_1^{1/2} w^{3/2} \), \( \Delta F_t = k_t (R w)^{1/2} \Delta s \). Here \( R = 2R_1 R_2/(R_1 + R_2) \), the normal overlap is \( w = (1/2) (R_1 + R_2) - |\vec{x}_1 - \vec{x}_2| > 0 \), and \( \vec{x}_1, \vec{x}_2 \) are the positions of the grain centers. The normal force acts only in compression, \( F_n = 0 \) when \( w < 0 \). The variable \( s \) is defined such that the relative shear displacement between the two grain centers is \( 2s \). The prefactors \( k_n = 4G/(1 - \nu) \) and \( k_t = 8G/(2 - \nu) \) are defined in terms of the shear modulus \( G \) and the Poisson’s ratio \( \nu \) of the material from which the grains are made (typically \( G = 29 \) GPa and \( \nu = 0.2 \), for spherical glass beads). We assume a distribution of grain radii in which \( R_1 = 0.105 \) mm for half the grains and \( R_2 = 0.075 \) mm for the other half. The observables are measured in reduced units: length in units of \( R \), force in units of \( GR^2 \), time in units of \( \sqrt{\rho R^2 / G} \), where \( \rho \) is the density of the particles. Internal dissipation is included in two ways: (1) via a viscous damping term proportional to the relative normal and tangential velocities, and (2) via sliding friction: when \( F_t \) exceeds the Coulomb threshold, \( \mu F_n \), the grains slide and \( F_t = \mu F_n \), where \( \mu \) is the friction coefficient between the spheres (typically \( \mu = 0.3 \)).

We perform molecular dynamics (MD) simulations for a binary system of 200 large and small spheres in a periodic 3D cell. Our calculation begins with a numerical protocol designed to mimic the experimental procedure used to prepare dense packed granular materials. The simulations start with a gas of spherical particles located at random positions in a periodically repeated cubic cell of side \( L \). The system is then compressed and extended slowly until a stationary situation with a specified value of the pressure and volume fraction—above the random close packing fraction—is achieved. We then apply a gentle shear in the \( y \)-direction at constant volume by moving the periodic images at the top and bottom of the cell with velocities \( \dot{\gamma} L/2 \), where \( \dot{\gamma} \) is the shear rate (Lees-Edwards boundary conditions). In this simple shear flow the gradient of the velocity along \( z \) is uniform (no shear bands). We apply a shear rate of \( \dot{\gamma} = 10^{-4} \), which allows the system to be in the quasi-static regime. We focus our study on the slow shear rate, quasi-static limit where the system is always close to jamming, but moves just barely enough to avoid stick-slip motion [15]. Here the external pressure is large enough (\( \sim 10 \) MPa in a typical simulation) and the average coordination number is high enough (typically \( \sim 7 \)) that deformation and elasticity of the particles play the dominant role, as opposed to the collision dominated rapid flow regime described by kinetic theories. We checked that shear induced segregation is absent at the times scales of our simulations. The contacts between particles are enduring and the internal stresses in the system are transmitted via a network of ‘force chains’ [6], independent of the shear rate.

After a transient of the order of the inverse shear rate, we start measuring the spontaneous \( \langle |x(t) - x(0)|^2 \rangle \) and force-induced displacements \( \langle |x(t) - x(0)|/f \rangle \) along the \( x \)-direction for the two types of particles with different sizes. The results are shown in Fig. 1. We notice that the diffusivities and the mobilities are different for the two type of particles, since they have different sizes. However, when we draw the parametric plot of \( \langle |x(t) - x(0)|^2 \rangle \) versus \( \langle |x(t) - x(0)|/f \rangle \) (Fig. 2) we find parallel straight lines for large time scales, implying an extended Einstein relation:

\[
\langle |x(t) - x(t_w)|^2 \rangle = 2 T_{dyn} \frac{\langle |x(t) - x(t_w)| \rangle}{f},
\]

valid for both particles with the same \( T_{dyn} \) for widely separated time scales \( t \gg t_w \). This suggests that \( T_{dyn} \) can be considered to be the temperature of the slow modes.

For small time scales (fast rearrangements, near the origin in Fig. 1) the fluctuation-dissipation plot shows that there is not a well defined Einstein-temperature relation. We expect this result since the fast motion of the grains depends on the microscopic interactions dominated by inelastic collisions [17]. We also calculate the ‘granular kinetic temperature’ defined in terms of the velocity fluctuations in the \( x \)-direction. Unlike \( T_{dyn} \), we find that this temperature is different for the two types of particles, and their values are two orders of magnitude smaller than \( T_{dyn} \). We expect this result since the kinetic granular temperature is dominated by the fast (high frequency) modes, which are not thermalised to \( T_{dyn} \).

We also repeat the numerical experiment for a system of Hertz spheres without transverse forces (\( \mu = 0 \); experimental realizations of elastic spheres with viscous forces but without sliding friction are foams and compressed emulsions [18,19]) and find that \( T_{dyn} \) is well defined at long time scales for this case as well (Fig. 1). Thus, our results suggest that the validity of a structural temperature for long-scale displacements (larger than a fraction of the particle size) holds in the presence of viscous forces between grains or even of a sliding threshold (Coulomb’s law).

The existence of a single temperature is an instance of the zero-th law of thermodynamics, for which we find positive evidence here, at variance with the experimental result in Nowak et al. [14]. Three possible reasons for the apparent violation of the ‘zero-th law’ in their experiments are i) the effect of an unknown height-dependent pressure (as pointed out by authors), ii) a rather high tapping amplitude (Edwards ensemble in principle only valid in the high compaction limit [14]) and iii) the fact
that the density fluctuations considered are integrated over all frequencies, thus including also ‘fast’ relaxations.

Next, we treat the question whether it is possible to relate the dynamical temperature obtained above to the thermodynamic construction of slowly driven out of equilibrium systems proposed by Edwards. Whereas in the Gibbs construction for equilibrium statistical mechanics one assumes that the physical quantities are obtained as average over all possible configurations, Edwards ensemble consists of only the blocked configurations (static or jammed) at the appropriate energy and volume. The strong ‘ergodic’ hypothesis is that all blocked configurations of given volume and energy can be taken to have equal statistical weights. This formulation leads to an entropy \( S_{Edw}(E, V) \), and the corresponding temperature \( T_{Edw}^{-1} = \partial S_{Edw}/\partial E \) and compactivity \( X_{Edw}^{-1} = \partial S_{Edw}/\partial V \).

In order to calculate \( T_{Edw} \) and compare with the obtained \( T_{dyn} \) we need to count the number of blocked configurations at a given energy and volume. (For this calculation we concentrate in the case without tangential forces and sliding friction, in order to avoid path dependency which would lead to an ambiguity in the definition of blocked configurations—see below). To do this in practice, we extend the ‘auxiliary model’ method to the case of deformable grains. We define an auxiliary model composed of the true deformation energy \( E, (the \) Hertzian energy of deformation of the particles) and of a term that vanishes in (and only in) the blocked configurations, depending on how they are accessed. The ensemble of blocked configurations is then ill-defined. We have not tried to construct any ensemble, but content ourselves with the observation that \( T_{dyn} \) is also in this case independent of the particle size (Fig. 2)—we suspect that thermodynamic concepts apply, but that the relevant ensemble goes beyond Edwards’ construction as it stands.

We conclude with some remarks: i) Since the blocked configurations are the same whatever the inter-grain dissipation coefficient, Edwards’ ensemble (and hence its temperature) are insensitive to viscous dissipation.

ii) On the contrary, tangential forces and sliding friction block certain configurations, depending on how they are accessed. The ensemble of blocked configurations is then ill-defined. We have not tried to construct any ensemble, but content ourselves with the observation that \( T_{dyn} \) is also in this case independent of the particle size (Fig. 2)—we suspect that thermodynamic concepts apply, but that the relevant ensemble goes beyond Edwards’ construction as it stands.

iii) We have tested the validity of the thermodynamics in an ideal homogeneous system with periodic boundary conditions by explicitly avoiding structural features of dense granular flows such as shear bands and segregation of the species. Even though it remains to be seen whether the thermodynamic picture can account for these inhomogeneous effects, our ideal system may prove to be useful in deriving constitutive relations to be used in macroscopic theories of slow granular flows.

To summarize: we have performed numerically an experiment with dense granular systems specially conceived to be a ‘dress rehearsal’ for the real laboratory one. The independence of the Einstein-relation temperature on the tracer provides a strong test for the thermodynamic ideas. We have also showed that this temperature, obtained from measurable quantities, indeed matches Edwards’ one. If, as now seems likely, this result is confirmed in the laboratory, this will give an experimental foundation for the use of the powerful tools of statistical mechanics as the framework to study this kind of far-from-equilibrium dissipative dynamical system.

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Fig. 2. Parametric plot of diffusion vs response function for small and large grains and for spheres interacting with tangential forces and without tangential forces (Coulomb frictionless). The fitting at long time scales shows the existence of a well defined temperature which is the same for small and large grains: $T_{dyn} = 2.8 \times 10^{-5}$ for grains without transverse forces and $T_{dyn} = 1.2 \times 10^{-4}$ for grains with Mindlin transverse forces and Coulomb friction. We calculate the response function for several small external fields and find the same temperature indicating that we are in the linear response regime. Plotted are results for a system without transverse forces using: $f = 1.7 \times 10^{-5}$ (small grains $\square$, large grains $\bigcirc$) and $f = 2.6 \times 10^{-5}$ (small grains $\blacksquare$, large grains $\bullet$). For a system with tangential forces and Coulomb friction we show the case $f = 6 \times 10^{-5}$ (small grains $\triangle$, large grains $\times$).

Fig. 1. (A) Diffusion and (B) response function for the large and small particles in a sheared granular material measured perpendicular to the shear plane as a function of time in MD steps. Both quantities depend linearly on time at the late stages of the evolution. The response function is measured by applying a constant force $f$ in the $x$-direction to each type of particle. Averages are taken over 30 temporal realizations. The obtained diffusivities and mobilities depend on the particles size as expected.
FIG. 3. Annealing procedure to calculate $T_{Edw}$ at different true energies. We plot the true deformation energy versus $T_{aux}$ together with the distribution of deformation energies obtained during shear (dashed curve, mean value $\langle E \rangle = 8.4 \times 10^{-4}$). We equilibrate the system for 40 million iterations at A: ($T^* = 3.4 \times 10^{-2}, T_{aux} = 3 \times 10^{-8}$). We then anneal slowly both temperatures until B: ($T^* = 3.4 \times 10^{-4}, T_{aux} = 3 \times 10^{-10}$), where we split the trajectory in three paths in the ($T^*, T_{aux}$) plane. Path 1: we anneal $T_{aux} \to 0$ and $T^* \to 2.8 \times 10^{-5}$ which corresponds to $T_{dyn}$ obtained during shear (Fig. 2). Path 2: we anneal $T_{aux} \to 0$ and $T^* \to 3.4 \times 10^{-6}$. Path 3: we anneal $T_{aux} \to 0$ but keep $T^* = 3.4 \times 10^{-4}$ constant. When we set $T^* = T_{dyn}$ (Path 1), the final true energy value when $T_{aux} \to 0$ falls inside the distribution of energies obtained during shear and it is very close to $\langle E \rangle$. This proves that $T_{dyn} = T_{Edw}$ under the numerical accuracy of the simulations. For other values of $T^* \neq T_{dyn}$ the final $E$ falls out of the distribution obtained during shear (Paths 2 and 3). We also follow different trajectories (not shown in the figure) to $T^* \to 2.8 \times 10^{-5}, T_{aux} \to 0$ and find the same results indicating that our procedure is independent of the annealing path.