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Granular matter: A wonderful world of clusters in far-from-equilibrium systems

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Abstract

We review several theoretical and experimental methods of modeling and investigating granular matter far from equilibrium. The theoretical methods include an extension of the classical Boltzmann equation to inelastic gases, scalar internal degrees of freedom, and Hamiltonian-like grain–grain interactions; the experimental technique is concerned with thermal properties of electrically conducting clusters. We discuss the results, focusing on phenomena nonexistent in physics of gases, fluids or solids, e.g. anomalous temperature gradients or electric resistance. One of the models is used to study the interplay between classical and self-organized criticality.

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1. Introduction

Granular matter is made of a large number of macroscopic solid entities, whose size ranges from that of dust (in powders) to that of rocks (in planetary rings). They

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are ubiquitous in nature and play an important role in a large number of industrial processes. Moreover, the theory of such systems can be applied to many phenomena that are traditionally not associated with granular matter, e.g. the motion of flux lines in superconductors [1], traffic flow, film growth, galaxy clustering, crystal agglomeration and sintering, company merging, and slow relaxation dynamics in glasses and flux lattices [2].

Granular systems have a very rich phenomenology, and their behavior often differs qualitatively from that observed in solids, liquids or gases. As an example, consider storage of cereals in silos. One might naively assume that to calculate the pressure at the silo walls, the hydrostatic approximation can be safely used. However, in reality, an irregular stress net is formed through the grains, and the local pressure at the walls can assume such a huge value that the silo may even break. Another example is the problem of controlling the mixing and segregation of powdered chemicals in pharmaceutical industry. Attempts toward understanding and controlling both static and dynamic properties of granular materials are thus of highest interest to many fields of physics, applied sciences and engineering.

For a physicist, a natural way to investigate granular matter is to construct relatively simple models and, whenever possible, compare their predictions with experimental data from mechanical, thermal, electric or magnetic measurements. Here we present a short review of several such models.

One way of constructing a physical model is to start from a well-known, much studied one and introduce to it some interactions that can be regarded as a perturbation. An example of this approach, consisting in generalization of the Boltzmann equation to the case of inelastic collisions, is briefly presented in Section 2 (see Refs. [3,4] for more comprehensive texts). An open problem in physics of granular matter is how to take into account interactions between individual grains. In Section 3 we introduce the magnetic ballistic deposition (MBD) model [5] of granular piles with the Ising-like interactions between grains, and we use it to discuss the impact of contact energy, grain anisotropy and an external field on the stress net and the cluster formation. Another concept related to granular matter is the so called self-organized criticality (SOC), first introduced by Bak, Tang, and Wiesenfeld in their famous sandpile model [6,7]. In Section 4 we discuss how the SOC phenomena are related to the standard criticality of statistical physics. Then, in Section 5, we will report an anomalous behavior of the electrical resistivity and the thermoelectric power (TEP) in densely packed conducting grains. Finally, Section 6 is devoted to conclusions.

2. Granular gases

In this section we focus on a dilute assembly of grains that interact through inelastic collisions (the so-called inelastic gas) in the limit of very low grain concentration. In this limit inelasticity can be treated as a small perturbation to the classical kinetic theory. Following this idea, one usually starts from writing down the *inelastic* Boltzmann equation [8], which is the classical Boltzmann equation with

some corrections reflecting inelasticity of collisions. Two new phenomena appear in this approach: irreversible dynamics and energy dissipation (Fig. 1). These effects are taken into account through an inelasticity parameter $\alpha \in]0, 1[$:

$$(\boldsymbol{\varepsilon} \cdot \mathbf{v}_{ij})^* = -\alpha(\boldsymbol{\varepsilon} \cdot \mathbf{v}_{ij}), \tag{1}$$

where $\mathbf{v}_{ij} \equiv \mathbf{v}_i - \mathbf{v}_j$, $\mathbf{r}_{ij} \equiv \mathbf{r}_i - \mathbf{r}_j$, $\boldsymbol{\varepsilon} \equiv \mathbf{r}_{ij}/|\mathbf{r}_{ij}|$; the asterisk marks the post-collision quantities, while the unleashed symbols correspond to the pre-collisional quantities. The elastic limit is recovered for $\alpha = 1$.

In real-world granular matter, α depends on \mathbf{v}_{ij} [9]. However, it is natural to start building a theory for such systems from disregarding this dependence [4]. However, this simplification has a dramatic consequence in MD simulations, the so-called collapse phenomenon [10], which is an extreme expression of the clustering instability specific to inelastic fluids and consists in the occurrence of an infinite number of collisions in a finite time.

Dissipativity of collisions contributes to an anomalous exploration of the phase space. The system kinetic energy is transferred to the internal degrees of freedom of

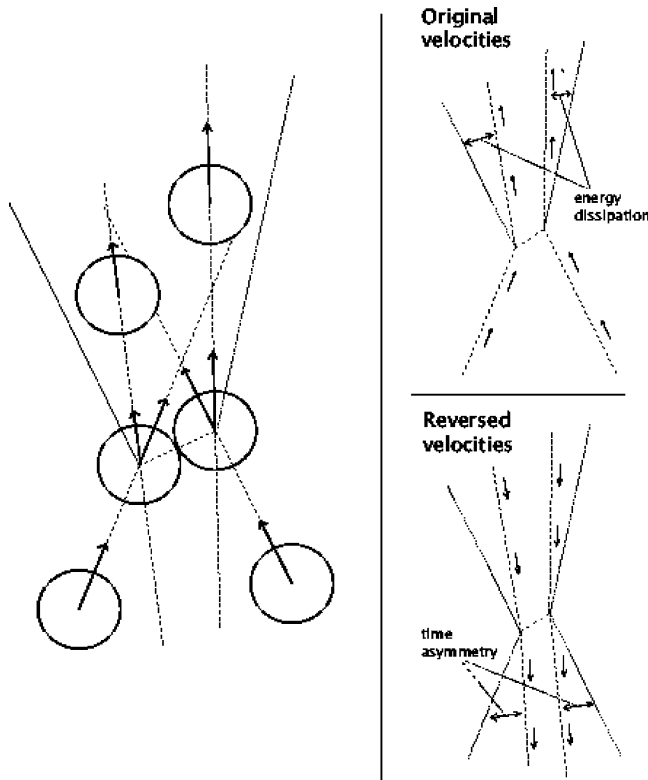


Fig. 1. Collision between inelastic hard spheres. Inelasticity makes the post-collisional velocities more parallel as compared to an elastic collision. Figures on the right illustrate the irreversibility of the microscopic dynamics: the reversed trajectories are no longer the same as the original ones.

the grains, and this renders the dynamics irreversible. As a consequence, some regions in the phase space are favored by the dynamics. This can lead to clustering of trajectories and heterogeneity of their density (Fig. 2). Moreover, if no external energy is transferred to the system, its evolution tends toward a perfectly resting state, in which the total kinetic energy vanishes. The system becomes non-ergodic and before it reaches the state of a complete rest, it passes through different intermediate states that usually depend on initial conditions. Let us stress that this expression of metastability does not rest on equilibrium-like mechanisms. The study of inelastic gases made it also possible to identify the main inelastic effects that alter the macroscopic dynamics: an anomalous coupling between the local energy and the local density, and a new time scale associated with the dissipative cooling.

One of macroscopic consequences of inelasticity in granular gases is anomalous transport. By applying the Chapman–Enskog procedure to the inelastic Boltzmann equation [11], one can derive a generalized Fourier law for the heat flux of a granular fluid

$$\mathbf{q} = -\mu \hat{\partial}_r T - \kappa \hat{\partial}_r n, \quad (2)$$

and identify the mechanisms that make $\kappa \neq 0$. On the one hand, $\kappa \neq 0$ is a manifestation of the coupling between density and temperature which occurs in granular media: the local cooling rate depends on the local temperature. This non-local behavior discriminates the temperature dependence of neighboring hydrodynamic cells and couples the density field to the heat transport process, leading to an additional term $\sim \hat{\partial}_r n$, and to a positive contribution to κ . On the other hand, additional contributions to the heat flux come from the shape of the reference velocity distribution. This zeroth-order state in the Chapman–Enskog procedure (the homogeneous cooling state) is a homogeneous state which remains homogeneous, and whose time dependence occurs only through the granular temperature. It usually plays the role of the equilibrium state in the statistical description of granular gases. This contribution can be either negative or positive, depending on the variation to the Maxwellian.

A striking consequence of the generalized Fourier law is the phenomenon of temperature inversion in vibrated granular systems. It occurs in systems subject to gravitation and constant supply of energy through a vibrating bottom wall. Because

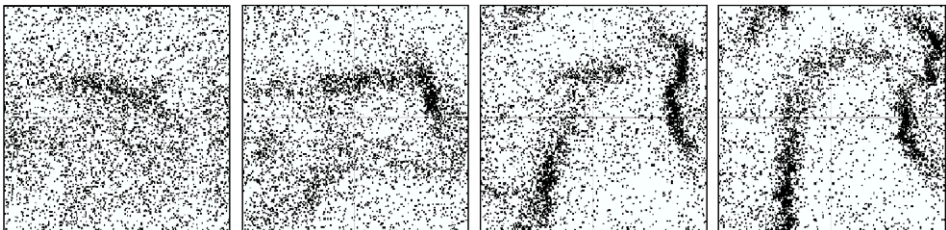


Fig. 2. Aggregation tendency and emergence of clustering in a freely cooling granular gas. The system is composed of 5000 inelastic discs and $\alpha = 0.9$ in Eq. (1).

of the energy injection, the inelastic fluid tends to an asymptotically stationary distribution with non-vanishing, *non-uniform* granular temperature [12,13] which develops a minimum at some distance from the bottom. Hence, the granular temperature can increase with height. This phenomenon has been predicted using (2) and verified both experimentally and numerically [4]. An open question remains whether the Fick and Ohm equations also have to be appropriately modified in their own context. Another question pertains to the validity of the Onsager relations.

Non-Maxwellian velocity distributions are another interesting property of inelastic gases. Their tendency toward overpopulated, “fat” high-energy tails is non-trivial, and their specific shape depends on the details of the model. However, overpopulation seems to be a generic feature of inelastic gases [8]. This property has been predicted theoretically and numerically, and observed experimentally in a large number of situations, see Ref. [4]. Maxwell models, which are simple kinetic models with a simplified collision operator in the inelastic Boltzmann equation, have been introduced to study the formation of the fat tails. Usually, two kinds of asymptotic velocity distributions are considered: (i) scaling solutions, which occur when the grains evolve freely, without external forcing, and correspond to a homogeneous cooling state, and (ii) heated stationary solutions, which are obtained by injecting energy into the system to counterbalance the energy loss. The energy is usually introduced by stochastic forces, often of Langevin type, to mimic the vibro-fluidized granular media. Another class of asymptotic solutions—stationary solutions of the unforced case—has been studied in Ref. [4]. By using similarities between the Maxwell model and a random walk in velocity space, it was shown theoretically and numerically that stationary solutions do exist. This means that the core of these distributions is stationary for an arbitrary time interval, while the system total energy decreases exponentially. These solutions correspond to truncated Lévy distributions.

Finally, let us recall the granular Demon experiment, conceived to visualize the above-mentioned energy–density coupling. In this experiment, a box is divided into two equal compartments by a vertical wall starting from the bottom of the box. The wall has a hole that allows the grains to pass from one compartment to the other. The box is filled with inelastic identical particles submitted to gravitation. Energy is supplied by a vibrating bottom wall. This simple system exhibits an order–disorder transition. Indeed, for a high energy input, the system presents a homogeneous steady state, while for a lower energy input, a phase transition occurs and an asymmetric steady state prevails. This transition was explained by Eggers’ effusive model [14]. This system has been also studied in Ref. [4], where a stochastic urn model of Lipowski [3,15] was used. Several generalizations of the original Maxwell Demon experiment have been considered, and have led to a rich phenomenology: (i) systems with an arbitrary number of compartments (metastable states); (ii) systems where the energy is input asymmetrically (hysteresis and strong similarity to a ferromagnetic system in a external magnetic field); (iii) the original experiment applied to mixtures (horizontal segregation and emergence of non-stationary oscillations called *granular clocks*).

3. Ballistic magnetic deposition model

At higher densities, granular media lock into piles. In modeling them, it is crucial to remember that even if the grains can be thought of as made of hard matter [16], like rice or sand grains [17,18], hardly ever can they be assumed to be spherical and slide friction-free on each other. The contact forces between grains are responsible for specific angles of repose [19], jams and arches [20].

We have introduced the magnetically controlled ballistic rain-like deposition (MBD) model [5,21,22] of granular piles of interacting entities and numerically investigated its static properties in 2D. The grains are characterized by a two-state scalar degree of freedom, a “nip”, which can represent the grain anisotropy or position with respect to neighboring grains. The nip–nip interaction is introduced through an Ising-like Hamiltonian. An external field of arbitrary origin is assumed to forcefully make each grain rotate during its deposition. This effect is controlled through a parameter q .

The simulation algorithm creates a pile with a fixed probability q for any grain coming in the ballistic grain fall to choose the “up” direction (or +1 value). At each time step the local energy gain $E = -J \sum_{\langle i,j \rangle} n_i n_j$ is calculated, where $n_j = \pm 1$ is the nip value at j . If $\Delta E \leq 0$ or if the site just below is already occupied, the grain sticks to the cluster immediately in its current “nip” state. Otherwise the grain either sticks to the cluster with a probability $p_s = \exp(-\Delta E)$ or continues to fall down with the probability $1 - p_s$.

The density and the order parameter (“niptization”) have been measured, as well as fractal characteristics of clusters. Cluster size and specific size distributions were found to depend on q and J (Figs. 3 and 4). The stronger the nip–nip interaction, the larger the difference between the piles [21,22].

Apparently the pile growth dynamics and structure depend on whether $q < q_c$ or $q > q_c$ [22], where q_c is a function of J . The two regimes can be identified through analysis of the cluster-mass distribution, which can be either exponential or follow a power-law form. Most probably the regimes are distinguishable because of a percolation-like transition at finite q_c , with $q_c \simeq 0.85$ and $q_c \simeq 0.75$ for $J < 0$ and $J > 0$, respectively. There is no theory to account for this at the moment.

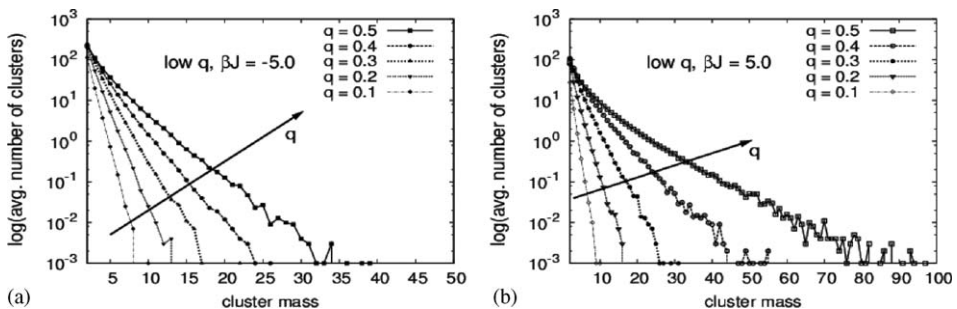


Fig. 3. Semilog plots of the cluster size distribution for low q values: (a) $\beta J = -5$, (b) $\beta J = 5$. Observe the exponential law-like behavior of the distribution.

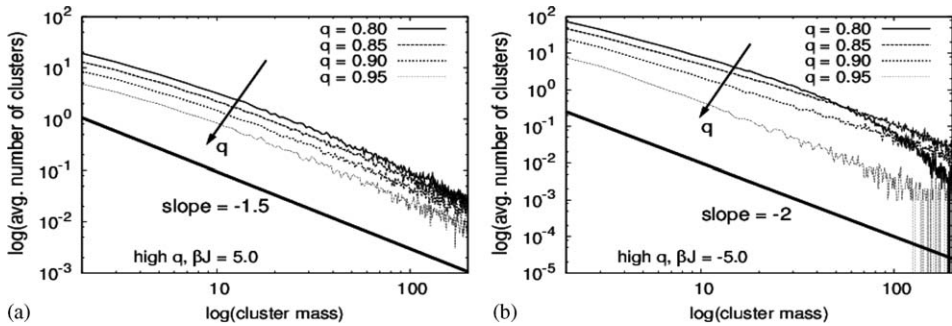


Fig. 4. Log–log plots of the cluster size distribution for high q values: (a) $\beta J = -5$, (b) $\beta J = 5$. Notice the power law-like behavior of the distribution.

It is of high practical interest to calculate the stress net in a pile. Although this is a hyperstatic problem [23], we found that for hard disks deposited as in the MBD model, the most frequent contact number is 4 [24]. In the case of elongated disks, the stress net might be thus more easily determined in two dimensions (2D) than in 3D, especially that it has been found to be unique in 2D. Moreover, the contact number value of 4 seems to remove rotation-frustration constraints.

Extensions toward binary, polydispersed or more complex objects have not received much attention, yet. However, recent studies on granular piles show importance of such investigations [25].

4. Hybrid sandpile model

One of the phenomena most commonly associated with granular matter is the formation of sandpiles, and one of the most interesting properties of sandpiles is their ability to “self-organize” into a critical state in which various quantities, e.g. avalanche size, mass and duration, are scale-free and exhibit power-law distribution tails [7]. Although the relation between the “classical” and “self-organized” criticality has been studied by many authors [26], the origin and precise definition of the SOC phenomena, as well as its relation to its classical counterpart, remains elusive.

We decided to tackle this problem by studying a hybrid model which, by construction, is expected to exhibit both classical and self-organized critical phenomena. In defining such a model we follow ideas of Ref. [27] and build it from two components. One of them is the Blume–Emery–Griffiths (BEG) [28] model, which is a standard Hamiltonian lattice spin model with a rich phase diagram. The other one is the Bak–Tang–Wiesenfeld (BTW) sandpile model [6], which, owing to its elegant mathematical structure [29], has practically become a paradigm for self-organized criticality studies. We combine these models by taking

(short-range) interactions from the BEG model and the (non-local) constraints from the BTW model.

The hybrid model is defined on a honeycomb lattice. Each of the lattice sites j is in one of three states that can be regarded either as “spins” $S_j \in \{-1, 0, 1\}$ or local pile heights $h_j \in \{0, 1, 2\}$. The entities located at the lattice sites interact with their neighbors through their spin representation and the BEG Hamiltonian

$$H = -J \sum_{\langle i,j \rangle} S_i S_j - K \sum_{\langle i,j \rangle} S_i^2 S_j^2 + D \sum_i S_i^2, \quad (3)$$

where J, K, D are some parameters. However, in contrast to the original BEG model, not all spin configurations are allowed in the hybrid model—we assume that the phase space is restricted to the so-called recurrent configurations of the BTW sandpile model on the lattice. What are the recurrent states of the BTW model then? The BTW model mimics the process of building a sandpile by adding to it grains one by one. As more grains are added to the pile, now and then it becomes unstable, which manifests itself through avalanches. In brief, the recurrent states of the BTW model correspond to the stable states of the sandpile at criticality, see Ref. [29]. Alternatively, the hybrid model can be seen as an extension of the “nip” model of Section 3: one continually adds grains to a pile, which may (but does not have to) become unstable and reconfigure through an avalanche; in a stable configuration grains interact through the BEG-like Hamiltonian and we are interested in the equilibrium properties of this system. An example of an allowed configuration is depicted in Fig. 5.

Here we report only a few major results for this model [30]. First, we have calculated the temperature dependence of the specific heat for different lattice sizes from energy fluctuations,

$$C = \frac{N}{T^2} (\langle u^2 \rangle - \langle u \rangle^2), \quad (4)$$

and found that $C \sim T^{-2}$ for large T , which is typical of Ising-like spin systems. We also found that for somewhat lower temperatures, $C(T)$ develops a clear peak, typical of a second-order phase transition. This finding was also confirmed by studying the finite-size effects. Consequently, the SOC constraints do not affect thermodynamics of the system.

Secondly, we investigated the impact of interactions on the self-organization of the system. One signature of “criticality” of the BTW sandpile model is a power-law decay of two-point correlation functions $P_{kl}(r)$, of nodes at a distance r apart, having heights k and l

$$P_{kl}(r) = P_k P_l + p_{kl} r^{-4} + \dots \quad (5)$$

for $k = l = 0$ in the bulk [31] and for all $0 \leq k, l \leq 3$ near the boundary of the system [32]. We have examined the case $k = l = 0$. The results for a rather high temperature $T = 10$, where the BEG interactions should not play a significant role, are presented in Fig. 6. As can be seen, an algebraic fit is quite good. However, it is based on only nine relevant points and an exponential fit cannot be ruled out. Since it is impossible

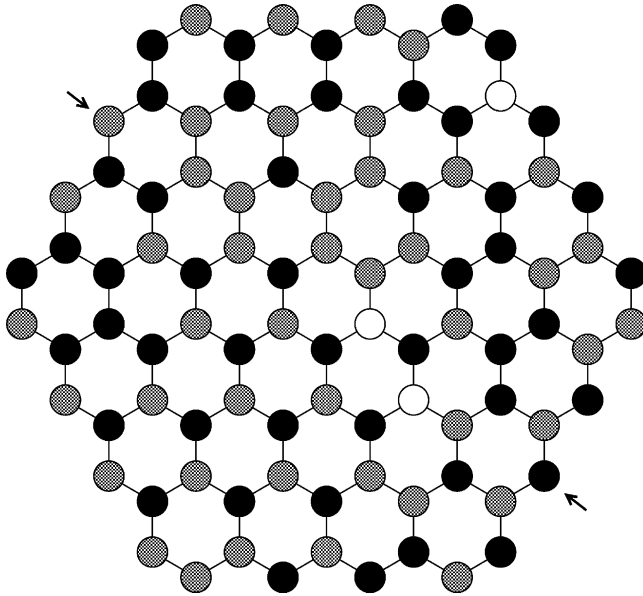


Fig. 5. An example of an allowed configuration on a honeycomb lattice of linear size $L = 4$. Empty, shadowed, and filled circles represent the nodes with heights $h_j = 0, 1, 2$ (or spins $s_j = 0, -1, +1$), respectively. The arrows show an example of a pair of boundary nodes that interact via Hamiltonian (3).

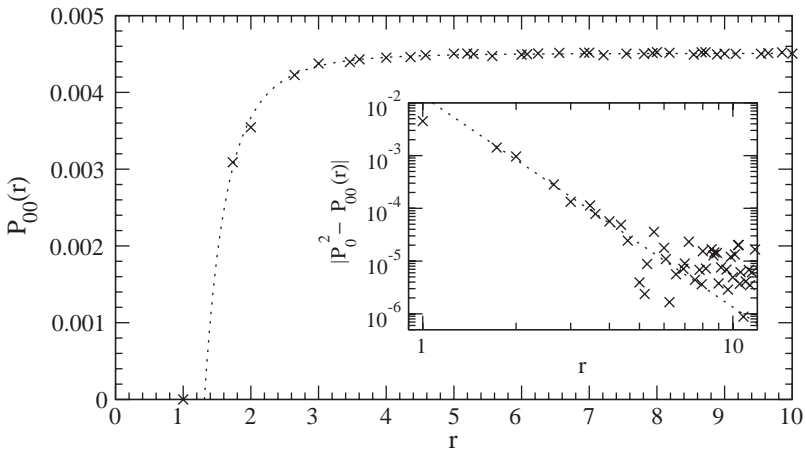


Fig. 6. Probability $P_{00}(r)$ of finding two empty lattice nodes r units apart for $L = 40$ and $T = 10$. The dotted line is a fit calculated from (5). The inset presents the log–log plot of $|(P_0)^2 - P_{00}(r)|$.

to increase the number of relevant points significantly, we resort to physical insight. If the decay of correlations is governed by interactions and thermal effects, it should be exponential with the T -dependent correlation length. If the correlations are

controlled by the SOC constraints, the ratio $(P_0)^2/p_{00}$ in (5) should be independent of T . On checking these hypothesis for a wide range of temperatures, we found that the system is in an SOC state at all $T > T_c$. Note, however, that this has absolutely no impact on the thermodynamic properties of the system! We draw the following conclusion: while it is true that thermodynamic criticality implies power-law decay of correlations, power-law decay of correlations does not imply thermodynamic criticality.

In the above study, the lattice basis is a regular structure. It has been shown that for fractal basis the avalanches present an extra feature, a log-periodicity, which depends on the fractal dimension and connectivity of the lattice [18]. Translated into a complex fractal dimension [33], the sandpile model can serve as an analogy to endogenous financial crashes [34].

5. Conducting densely packed matter

Some granular materials are electrically conductive. An electric current running through them may heat up and strongly modify the oxide layers that are often formed at the grain surfaces [35], even up to welding. It is therefore important to investigate the temperature dependence of electrically conductive packed materials.

Such systems are usually modeled as a disordered resistor network with the electronic conduction occurring in a strongly localized regime. Their transport properties depend on whether the system is above or below the percolation threshold. Since the resistance is dominated by carrier hopping between grains, electrically conducting GM displays features of the variable-range-hopping (VRH) phenomenon observed in doped semiconductors. In particular, their electrical resistivity obeys the fractional temperature dependence,

$$\rho(T) = \rho_0 \exp[(T_0/T)^p], \quad (6)$$

where T is the temperature, T_0 is a characteristic temperature, $p = 1/(d + 1)$, and d is the dimensionality of the system [36].

However, the electric current might avoid the oxide layer and penetrate the internal structure of the grain. As this would invalidate the VRH mechanism, we synthesized and compacted a crystalline granular metallic system, CaAl_2 [37], to obtain tiny crystals. Chemical analysis indicates a complex microstructure inherent to the phase diagram [38]. An EDX analysis shows that the system is made of Al-rich dendrites embedded in an Al-poor matrix. The dendrites are made of CaAl_2 , while the matrix is a mixture of $\text{Ca}_{13}\text{Al}_{14}$ and Ca_8Al_3 , according to the phase diagram. These phases have been found to be metallic with a very similar electrical resistance [39].

We measured the electrical resistivity and thermoelectric power of a packed crystal under standard conditions. As depicted in Fig. 7, their temperature dependence shows three regimes. The resistivity was found to depend on T as $\rho(T) \simeq T^{-3/4}$ at low temperatures (Fig. 7a). This can be interpreted as a *thermal* effect taking place on geometrically disordered backbone that does not change with the temperature. The 60–70 K break (or crossover) indicates the energy range at which the thermal

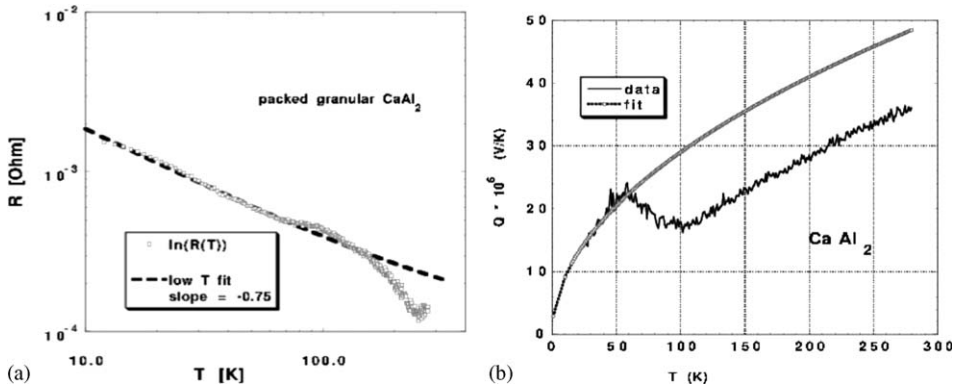


Fig. 7. (a) Electrical resistance R vs. temperature T of a densely packed granular CaAl_2 on a log–log plot, with a low-temperature fit to $R(T) \propto T^{-3/4}$. (b) Thermoelectric power Q vs. temperature T of a densely packed granular CaAl_2 with a square-root fit at low temperatures.

process takes over on the geometric disorder. At higher T , the smooth decay of $R(T)$ can be attributed to further charge carrier delocalization resulting from the high-temperature form of the Fermi–Dirac distribution.

The (positive) thermoelectric power [37] shows a bump near 60 K, after an unusual, square-root dependence on T at low temperature (Fig. 7b). Above 100 K, a log–log plot (not shown) reveals that $Q(T)$ grows as $T^{3/4}$ rather than linearly, as might be expected for metallic systems at high T . Since a TEP measurement implies no external electrical current, it is unlikely that some “barrier ageing” or “hot spots” occur in the investigated temperature range. Therefore both effects, an increase of charge and heat conductivity with growing temperature and a large thermoelectric effect at room temperatures, can be understood as a consequence of a delocalization process that takes place on the intricate barrier network, with competing characteristic mean free paths and weakening of the contact TEP due to the Fermi surface widening. The three thermal regimes are thus a manifestation of competition between geometric and thermal processes in weakly conducting clusters of densely packed granular matter.

6. Conclusions

Except for some notable attempts by Faraday and Coulomb, it was not until quite recently that granular systems started to attract attention of the physics community, and now studies on their properties are considered as an important part of applied physics and technology. There are several reasons for this new interest. One of them arises from the fact that despite its apparent simplicity, granular matter exhibits surprisingly rich and often counterintuitive behavior. For example, entropy effects are often out-weighted by dynamics. Because of dissipative nature of collisions,

exploration of the phase space is restricted, the system is non-ergodic and the dynamics irreversible.

In this short review we have discussed several methods of introducing grain–grain interactions to models of granular matter. We first focused on dilute systems, and used this approximation to extend the classical kinetic theory to dilute assemblies of inelastic grains (“inelastic gases”). One of the most peculiar predictions of this theory is a new term in the Fourier equation of heat conduction. Then we outlined a model of gravitational deposition of interacting grains. We also showed how sandpile models with interacting grains can be used to investigate similarities and differences between self-organized and classical criticality. Finally, we have discussed anomalous thermal properties of dense granular electrically conducting clusters. These are only a few examples of a rapidly growing field of modeling granular matter.

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