Experimental observations of structural relaxation in granular matter

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The relaxational changes of the electrical capacitance of a system of grains poured abruptly into a vessel capacitor have been observed. Two power-law decays have been found. The faster relaxation at the beginning has been interpreted as being driven by independent-grain motion. The later slower process has been ascribed to the collective reorganization of the granular system. The observations seem to be an experimental illustration of the computer simulation predictions of Mehta and Barker [Rep. Prog. Phys. **57**, 383 (1994); Phys. Rev. A **45**, 3435 (1992); Phys. Rev. Lett. **67**, 394 (1991); Nature **364**, 486 (1993); Phys. Rev. E **47**, 184 (1993)]. [S1063-651X(96)09812-1]

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I. INTRODUCTION

Recently physicists dealing with theory, computer simulations, and experiments have become interested in problems concerning granular matter at rest and in motion. Powders reveal various fascinating features that seem to make these systems the paradigm of complexity in physics [1–3]. In spite of the fact that small grains stick together only due to gravitational and frictional forces, the free-standing sandpile stops being mechanically stable only when the packing (volume) fraction k is less than 0.52 [4]. The parameter k is defined as the ratio of the volume of grains themselves to the volume of the whole sample, i.e., the volume of the grains and the voids between them. Thus, in general, the volume occupied by grains is not much larger than the volume of empty spaces; in fact, the maximum packing fraction is equal to 0.64 [5], which means a lack of order in the spatial distribution of grains. A unique feature of the granular materials is that they can sustain voids. The distribution of the various-size empty spaces between grains is sensitive to the detailed history of the sample preparation. The contacts between locally disordered grains can be divided into close ones that are ''active'' [3], i.e., take part in the transmission of the disturbance through the system, and loose ones protected by the active ones from the external stress, current (for conducting grains), etc. Due to the lack of Brownian motion (present in many-body systems such as gases and liquids) there is no thermal averaging of the spatial distribution of the grains. Thus complicated structures of random close-packed grains, once created, are relatively stable and long lasting, a phenomenon called arching [6]. A large diversity of metastable configurations of grains with various bridges and arches is the basis of hysteresis effects. Moreover, the properties of the granular materials seem to be universal. Generally they do not depend on the type and size of the grains, but are attributed to the granular structure of the system.

The unusual features of the granular materials have influence on their dynamics, which includes also the relaxational behavior. The computer simulations [7] and the phenomenological considerations [8] indicate the complex evolution of the granular system towards a steady state in response to the mechanical driving force, for example, shaking. Mehta and Baker [1,7–10] postulate that two mechanisms, an independent-particle one and a collective one, take place in a relaxational process in a system of equal-size spheres exposed to operations that simulate vibrational motions. Relaxation is allowed by reorganization of the grains following the creation of the free volume in the sample by shaking cycles. Fast relaxation corresponding to the motion of independent grains dominates in the initial period of observation and then is followed by much slower relaxation, which is the result of the cooperational rearrangement of many grains, i.e., the motion of clusters of grains. The aim of this paper is to present the results of a very simple experiment undertaken for checking the correctness of the interpretation of relaxation in the granular material in the form of two processes.

II. EXPERIMENT AND RESULTS

In our experiment, dielectric grains of size of about 0.5 ± 0.1 mm were used. A vessel capacitor with two conducting plates of size 5×7.5 cm^2 along two parallel walls with a 3-cm gap between them was used. So that many grains could be accommodated above the level of the plates we made the plates smaller in height than the vessel containing the grains. To observe the relaxation phenomenon for the system of grains the perturbation applied had the form of an abrupt pouring of the grains into the vessel at time $t=0$. The pouring procedure is regarded as a single large-intensity shake [10]. In the system created by abrupt pouring, the distribution of grains is metastable. The contacting grains enclosed relatively large voids and the packing fraction is relatively small. Thus the grains start the gravitationally driven motion into more stable positions. Through the changes of their mutual positions, the system relaxes towards the steady state with higher packing fraction. Some of the grains above the plates fall into the capacitor as the empty spaces between the grains diminish. The growth of the number of the grains in between the plates leads to higher global capacitance, which reflects the enlargement of the packing fraction k. In the measuring system the capacitor is supplied by the generator with a sinusoidal voltage of 10 kHz frequency. The voltage drop on the capacitor filled by grains is amplified, rectified, and measured by the digital voltmeter. The sketch of the measuring system is presented in Fig. 1. In our experiment the relaxational enlargement of the packing fraction k

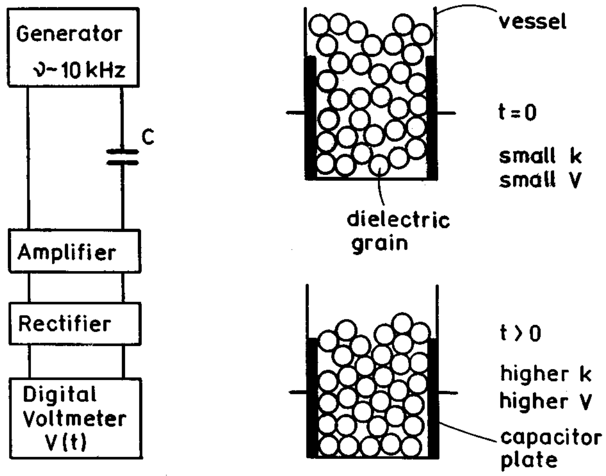

FIG. 1. Sketch of the experimental system. Changes of $V(t)$ reflect the growth of the packing fraction $k(t)$ after application, at $t=0$, of the perturbation to the vessel.

is registered as the growth of the voltage $V(t)$ from the initial value V_0 for the vessel just after it was filled with grains. It has been checked in several experiments that $V_0=410\pm 7$ mV only if the external conditions have been preserved. For the empty vessel the voltage was equal to 103 mV. Detailed quantitative observations of the changes of $V(t)$ were made for 3 h. During the first 2 min when the rapid growth of $V(t)$ occurred, the results were recorded every 5 s. The final registered value of $V(t)$ was 8% higher than V_0. The very small changes of the voltage $V(t)$ were observed during the whole day, reflecting a very slow relaxation process rather than a long-time drift of the electronic equipment.

Theoretical arguments [11] show that to describe the physical properties and behavior of the granular materials, the appropriate parameter is the so-called compactivity $X=1/k$ instead of the packing fraction k [8]. (For the granular matter, if only the energy is replaced by volume, the compactivity X corresponds to the temperature in the statistical mechanics description used for thermodynamical systems.) The time dependences of $V_0/V(t)$ observed in this experiment, which reflect the $X(t)$ changes, are presented in Fig. 2. In the relaxation process $V_0/V(T)$ [and thus also the $X(t)$ parameter] seems to be the response function of the disturbed system of grains.

III. DISCUSSION

The decay of $V_0/V(t)$ shows that the relaxation process observed is very slow and complex. The dependence of $\ln[V_0/V(t)]$ vs t is far from being the straight line corresponding to a single exponential decay.

After analysis of many dielectric and mechanical relaxation data, Jonscher [12] observed that the responses differ from the exponential decay $\exp(-t/\tau)$, which corresponds to independently relaxing entities characterized by the single relaxation time τ [13]. For various dipolar and nondipolar materials the universal response of two power-law temporal dependences has been found. The local and long-range interactions between each relaxing entity and its surroundings are understood as the mechanisms causing, respectively, the short- and long-time deviations of relaxation from the exponential decay. The power-law exponents describe the size of the correlation between relaxing entities in the system: α, for the short-time decay, and β, for the long-time decay. The experimental values have been found to cover the following limits: $-1<\alpha<0$ and $-2<\beta<0$, where the pair $(\alpha,\beta)=(0,-2)$ is the limit for noninteracting entities [14,13].

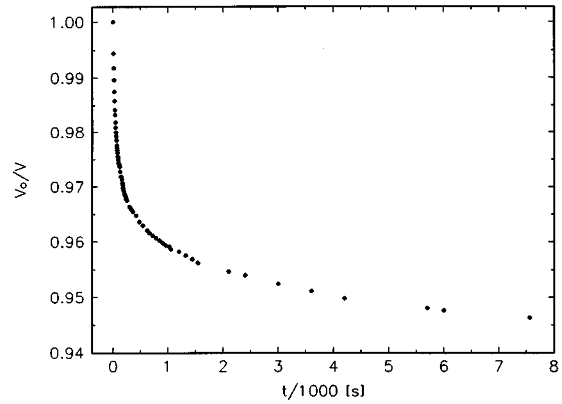

FIG. 2. Decay of the response function $V_0/V(t)$ after the grains have been poured abruptly into the vessel capacitor.

Such a general picture of a two part relaxation response function seems to correspond well with Mehta and Barker's results of computer simulation experiments for relaxation in granular matter [7–10]. Having that in mind, we have presented the observed decay $V_0/V(t)$ vs t in the double logarithmic scales (Fig. 3) to check if the experimental points compose themselves into two straight lines. It can be seen that the response $V_0/V(t)$ of the system to the perturbation in the form of the abrupt pouring procedure can be regarded as consisting of two parts. The first part lasts about 15 s and is documented by four experimental points only. The slopes of two straight lines used to describe the short- and long-time dependences of the $\ln[V_0/V(t)]$ vs $\ln(t)$ are $\alpha=-0.0038\pm 0.0002$ and $\beta=-0.0069\pm 0.0001$, respectively. On the other hand, when there is only a single power-law decay in the plot of $\ln[V_0/V(t)]$ vs $\ln t$, then the fitting

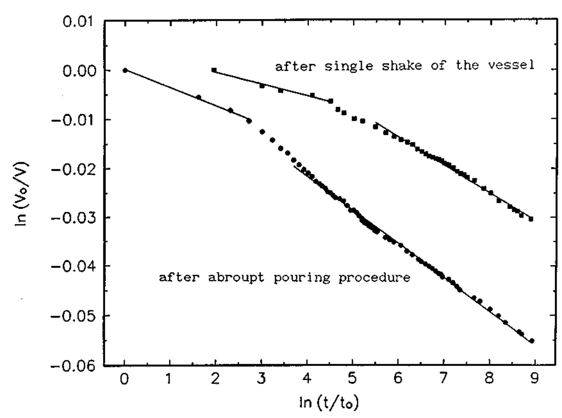

FIG. 3. Dependence of $\ln[V_0/V(t)]$ vs $\ln t$ for the abrupt pouring of the grains into the vessel (dots) and for a single shake of the vessel (squares). $t_0=1$ s.

procedure of the single straight line to all the data gives the slope $\gamma=-0.007\pm0.007$, where the large error is caused by the relatively large deviations of the first four points. The conclusion is that the process can be much better described by the two-power-law relaxational decay than by the single-power-law one.

The structure of contacting grains that appears after abruptly pouring them into the vessel is relatively wasteful of space, i.e., it is characterized by a large void. Thus, at the beginning it is easy to minimize voids by gravitationally driven motion of single grains. In this period the displacement of one grain can be independent of the displacement of its neighbors. In the course of time the structure becomes compact rather quickly. The motion of the grains to more stable positions becomes more and more difficult. Further densification needs structural rearrangement, which can be by cooperational movements of many grain clusters. Their motion starts to dominate in relaxation. As an effect of the collective mechanism, the decrease of the compactivity measured by $V_0/V(t)$ changes is slower and slower. This scenario seems to be confirmed well by the values of the α and β exponents calculated for our granular system: $\alpha\sim 0$ points to a nearly single-grain mechanism at the beginning of relaxation, while $\beta\geqslant -2$ points to definitely non-single-grain mechanism later on. It can be said than in the first few seconds of the experiments the relaxation is very similar to the exponential decay, while later on the deviation from such behavior is significant and the process in which the system reaches an equilibrium is extremely slow and lasts a very long time. After the perturbation the granular system is evidently a metastable state with a macroscopic lifetime.

In the second part of our experiment the different perturbation in the form of a single strong shake of the vessel was applied. The result is similar to $\alpha=-0.002\pm0.0007$ and $\beta=-0.0057\pm0.000\,01$ (see the boxes in Fig. 3). The voltage drop on the resting vessel capacitor filled with grains before it has been shaken was about 434 mV. The duration of the single-particle relaxation was found to be longer (about 100 s) for this perturbation than in the response observed after the abrupt pouring. Thus, in the second experiment the disturbed sample seems to have more empty spaces than the disturbed sample in the first one: Much longer single-grain displacements will fill the voids before the cluster's rearrangements become necessary for that. In order to investigate more precisely the physics of relaxation in granular matter, detailed studies of the response $V_0/V(t)$ to well-defined shaking procedure with several frequencies and amplitudes of vibration applied are being prepared.

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