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Transient phenomena, self-diffusion, and orientational effects in vibrated powders

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We explore the rich behavior of vibrated powders, focusing on the relaxation of transients between vibrational steady states, on the diffusion of independent particles, and on "global" and "local" angular distributions of contacting particles. When examined as a function of vibrational intensity, these are shown to be explicable in terms of a model of independent-particle and collective excitations. Our data provide a wealth of information about topics as diverse as consolidation under packing, the roughness of granular piles, and the local structure of bridges and arches in shaken packings.

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Although granular materials have been studied by engineers [1] for a long time, it is only recently that they have become a fashionable and exciting area of theoretical and experimental [2–4] physics. There have been a number of theoretical approaches to flow in powders, of which one of the most important consists of treating the powder as a macroscopic continuum [5]; however, there are many circumstances in which it is crucial to consider the essential discreteness of the grains, for instance in the study of size segregation, aggregation and arching. Hence a fully microscopic description of powders is fundamental to the understanding of the many fascinating features that are associated with powders at rest and in motion.

Recently a combination of theoretical [6] and computational [7–9] techniques has led to a comprehensive description of relaxational mechanisms in a vibrated powder. The fundamental reason for studying vibrated powders is that in the real world, powders are frequently "shaken" for a variety of applications in science and technology. In this context, "shaking" refers to the application of any incoherent mechanical driving force and the details of the driving force, for instance the strength of its harmonic components and their coupling to individual particle motions, are frequently poorly defined and may vary between applications. Thus the observed behavior of granular materials in practical situations includes an element that is the response of a dense, disordered many-component system to ill-defined, complex driving forces.

However, we maintain that this response, although complicated, can be understood in terms of the competition and cooperation between the relaxation processes due to the motions of independent particles and of clusters, and that the overall relaxation of a powder during vibration is thus a combination of independent-particle and collective excitations [6–9]. Further, we suggest that the balance between independent-particle and collective relaxation, for a given vibration intensity, is only weakly affected by details of particulate shape and texture or of the nature of the driving force and is to a far greater extent generic to disordered, nonsequential, close packings of powders.

In this paper, we present computer-simulation results to support this model, and we also extend its scope to include the transient response (as distinct from the steady-state response which we have studied in earlier work [7–9]) of the bulk properties (e.g., the volume fraction) of the powder. This is done with a view to understanding the everyday phenomenon of "tapping", of which more will be said later. Secondly, we probe the self-diffusion of independent particles, which has been the subject of a recent experiment [4], and we provide a comprehensive interpretation of this phenomenon which extends across the regime of fast flows(studied in Ref. [4]) and that of slow flows. Finally, we verify earlier predictions [8,9] concerning the roughness of vibrated piles and the jaggedness of local structures within them, by studying "global" and "local" distributions of the contacts between neighbouring particles.

Our simulation method is hybrid in nature, with an algorithm involving Monte Carlo dynamics as well as nonsequential random close packing, which overcomes some of the difficulties associated with computer simulations of shaking (see, e.g., Ref. [10]). In our model, shaking is a periodic process. Each cycle of the shaking is represented by an instantaneous, homogeneous introduction of free volume followed by a nonsequential reorganization in a strong, uniaxial external field. Details of the method are presented elsewhere [8,9]. The powder is modeled as a collection of frictionless, monodisperse hard spheres; our results are, however, not changed significantly by the introduction of a small amount ($<10\%$) of polydispersity. (We note that the introduction of particles with different sizes into a shaken packing leads to size segregation occurring concurrently with structural relaxation, which is an unnecessary complication for the interpretation of this study.) The first part of the shake cycle is a uniform vertical expansion of the sphere packing, accompanied by random, horizontal shifts of the sphere positions. A sphere at height z is raised to a new height $z'=(1+\epsilon)z$. For each sphere new lateral coordinates are assigned according to the transformation $x'=x+\xi_x$, $y'=y+\xi_y$, provided they do not lead to an overlapping-sphere configuration. Here ξ_x and ξ_y are Gaussian random variables with zero mean and variance ϵ^2. The parameter ϵ is a dimensionless measure of the intensity of the vibration. Following this expansion, the spheres are nonsequentially redeposited by a combination of Monte Carlo moves and mechanistic random close packing. The simulation method allows the powder to experience a wide range of volume fractions during shaking in which the grains are, however, returned to a stable, close-packed configuration in each cycle: ours is thus a model of a finite-amplitude, low-frequency shaking process. Our model reduces the process of shaking to its essential components, which our previous results [7–9] suggest are those that lead to a wide variety of observed granular behavior.

Repeated application of the shaking algorithm leads to a steady state in the presence of vibration; by this we mean that the properties of the powder at each point of the shake cycle are statistically invariant. This steady state is independent of the sphere configurations chosen for the initial condition, and depends only on the shaking intensity. Details of the steady-state shaking regime (including the variation of the mean volume fraction ϕ in the stable close-packed phase of the shake cycle) as a function of intensity have been examined thoroughly elsewhere [8,9]. These results support the existence of two relaxation mechanisms for vibrated powders, where the faster of the two involves the motions of independent particles while the slower involves collective particle motions [6].

In the present work we develop this picture by considering the transitions between these steady states. We have chosen our reference, steady-state configurations to be those appropriate to a shaking intensity of $\epsilon=2.0$. In this reference state, the spheres are relatively loosely packed, with $\phi\approx0.55$, while all steady states with $\epsilon<2.0$ have denser packings. If one regards tapping as a small-intensity shaking process and pouring as a single large-intensity shake, this is in accord with the familiar observation that dry granular material, poured into a container, can be consolidated by "tapping" [11]. As mentioned before, it is one of the aims of this paper to reproduce the physics of this process, and to see how the mechanisms involved in the relaxation of transients produce this consolidation.

Having chosen our reference configurations as mentioned above, we have applied shaking vibrations with $0.05\leq\epsilon\leq0.75$ to them. In our granular bed, which consists of a periodic arrangement of cells, the primary cell has a square cross section with an area of (64 sphere diameters)^2 and an average depth of approximately 20 sphere diameters in a direction parallel to the external field; this primary cell is then repeated in the two directions orthogonal to the external field. Volume fractions in the close-packed phase of the shaking cycle are measured form the central portion of the bed in order to minimize surface effects, and time is measured in units of the shaking cycle. The transient response of the volume fraction, for shaking at five different intensities, is shown in Fig. 1. Each data set is an average over at least eight independent simulations. Also plotted, in each case, are two nonlinear least-squares-fitted functions. Dotted curves show the best single exponential fit (three parameters) and the solid lines show the best fit with a sum of two exponents (five parameters).

The simulation results show smooth, monotonic variations of volume fraction from the poured steady-state value to the shaken steady-state value. We note that the transient response of the volume fraction reflects a transition between steady states that have different statistics for nonsequential reorganizations driven by vibration and contain different densities of structural features such as bridges and large voids. If bed compaction were the result of a single vibration-drive process, then (since our driving force is coupled homogeneously to the powder) one would expect the excess unoccupied volume to decay with a single relaxation time—however, the poor fit achieved using the best single exponential relaxation is very noticeable. The improved fit, using a sum of two exponentials, indicates that the above expectation is unrealistic, and that the powder has a more complex response. (We note that similar improvements can be achieved by fitting a single stretched exponential.) Further, for each value of ϵ, the two time constants obtained from the double exponential fit are very different. For $\epsilon=0.05$, 0.1, 0.25, and 0.5 the two relaxation times are approximately 3 and 20 cycles, but for $\epsilon=0.75$ these times are approximately 1 and 50 cycles; also, the relevant fitted coefficients in each case for the fast and the slow relaxation times are of comparable magnitudes. We infer thus

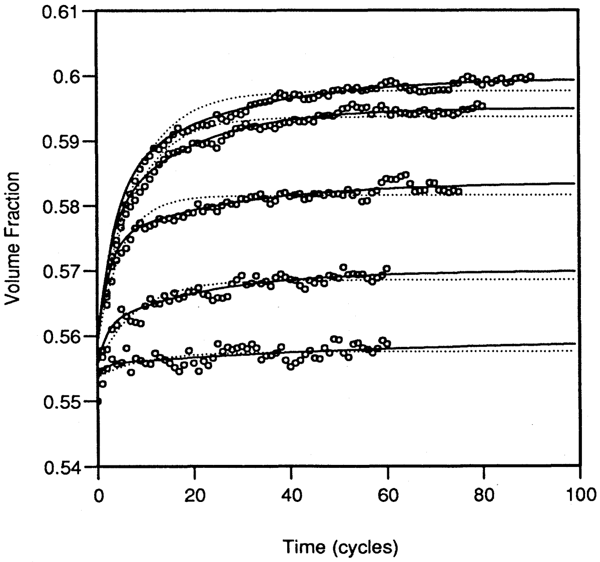

FIG. 1. The variation of the volume fraction (ϕ) with time in computer simulations of shaken granular deposits. The five data sets correspond, from top to bottom, to shaking intensity $\epsilon=0.05$, 0.1, 0.25, 0.5, and 0.75. Dotted curves show the best single exponential fits and solid curves show the best two exponential fits.

that the structural rearrangements that cause this densification are driven by two relaxation mechanisms and that these are the same as those which drive the structural relaxations in the steady state [8,9]—the faster relaxation corresponding to the motion of independent particles, and the slower to the motion of clusters.

We note that for computer-simulated powders, the particle reorganizations induced by shaking are only subject to geometrical constraints, whereas for real powders, consolidation is far more complicated. For real powders, other factors like cohesive forces, and particle fragmentations combine to constrain the structural collapse. Our simulation technique, by its very nature, is able to isolate the fundamental geometrical constraints from other extraneous effects; it is thus able to provide a valuable benchmark for evaluating the results of sophisticated consolidation schemes in industrial materials [11,12].

In each cycle of a finite-amplitude, low-frequency shaking process, the powder has periods of both quasistatic (low kinetic energy) and dynamic (high kinetic energy) behavior. In the quasistatic regime the particles are either static or move together as a rigid body, but in the dynamic regime, the particles are mobile and so lose information concerning their relative positions at a finite rate. This loss of information induced by shaking can be considered as a diffusive process in the following way—as the positions of a particle, measured at the same phase point of consecutive shake cycles, will be slightly displaced, we see readily that a sequence of these finite displacements forms a three-dimensional random walk that represents the diffusive motion of the particle due to shaking.

We have measured the average displacements of approximately 200 spheres over 30 cycles in the steady-state regime for $0.05 \leq \epsilon \leq 1.0$. In each case we observe a linear increase of the squared displacement with time. In Fig. 2 we have plotted, as a function of ϵ, the gradients D_T and D_Z which are obtained from least-squares fits of $\langle (\Delta x)^2 + (\Delta y)^2 \rangle$ against $2t$ and $\langle (\Delta z)^2 \rangle$ against t, respectively. Here Δx and Δy are two orthogonal displacements that are perpendicular to the uniaxial (gravitational) field and Δz is the displacement parallel to the field. For all intensities $D_Z > D_T$, as expected for vertical shaking under gravity, since the diffusive motion of a particle in the direction of shaking will always be greater than that in the other two orthogonal directions.

The results in Fig. 2 indicate the existence of two different diffusive regimes: there is a fast regime for $\epsilon > 0.2$, where D_T and D_Z are linearly dependent on ϵ, and a slower regime at smaller shaking intensities. This picture is in qualitative agreement with recent experimental observations of self-diffusion in vibrated beds of granular material [4], which the authors interpret via a hydrodynamic approach [5] considered to be appropriate for rapid flows and large voidage. Our simulations, however, span both slow and rapid flows, as well as large and small voidage, so that the following interpretation of the self-diffusion observed in our simulations is more comprehensive, encompassing both the so-called hydrodynamic and viscous regimes.

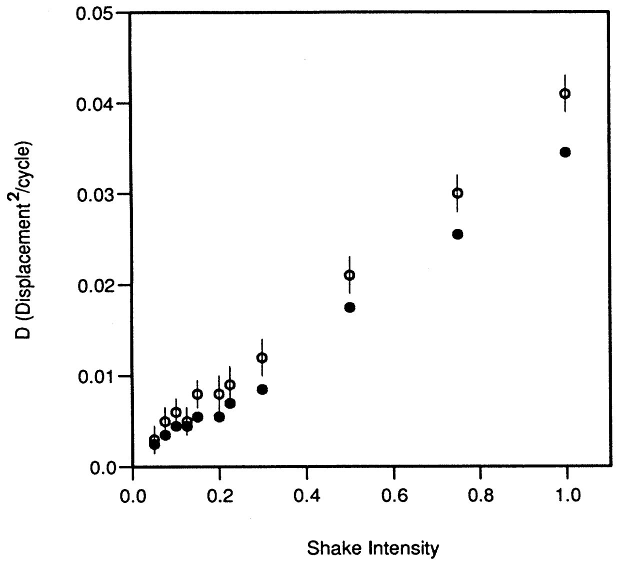

FIG. 2. The effective diffusion coefficients D_Z (open circles) and D_T (closed circles) against the shaking intensity ϵ. For clarity only one set of error bars is included.

The processes which underlie the diffusive motions of the spheres during vibration are not straightforward. In each shake cycle, the spheres spend some of the time subject to a direct fluctuating force that arises from effective collisions between pairs of moving particles; in addition, they also spend some time following deterministic trajectories, including rolling and falling, on a complicated potential-energy surface. However, this energy surface changes from one shake cycle to another, so that it, too, can be considered to fluctuate. Thus the random displacements of the spheres, during one shake cycle, result from a combination of different fluctuating processes. Phenomenologically, the random motion of the spheres corresponds to "hopping" between potential wells, and it is clear that the hopping times and the distribution of the wells are complex functions of the shaking intensity [6].

The results in Fig. 2 can be interpreted in terms of two distinct hopping processes. The major contribution to the particle displacements for steady-stae shaking with $\epsilon > 0.2$ occurs during the expanded, dynamic regime. For these shaking intensities the volume expansion, at the beginning of each shake cycle, is sufficient to destroy a large number of particle clusters [6], and therefore many particles spend some time in random motion before new clusters are formed, the duration of this period being proportional to the shaking intensity. For steady-state shaking with $\epsilon \leq 0.2$, local clusters remain largely intact for the whole of the shake cycle, and the major component of particle displacements arises from their deterministic motion inside their slightly deformed local environments. The size of the cluster deformations is not strongly dependent on ϵ for $\epsilon \leq 0.2$. This interpretation is consistent with earlier theoretical and computational work which relies on the same picture of inter- and intracluster particle motion [6–9].

Our shaking model does not allow a clear interpretation of vibration frequency, and hence we cannot classify our results in terms of a dimensionless combination of frequency, amplitude, and gravitational acceleration. However, we note that for a packing with a random close-packed volume fraction $\phi \approx 0.6$, the shaking intensity $\epsilon = 0.2$ leads to an expanded packing in which the volume occupied by the particles is the same as the volume occupied by voids; this is the so-called hydrodynamic regime studied by the authors of Ref. [4]. In this case, the expanded configuration contains spaces which are approximately equal in size to the particle size; it is reasonable to expect, therefore, that a change of hopping behavior occurs for $\epsilon < 0.2$, when the voidage is smaller and the flow is slower. We look forward to experimental verification of this, which is in progress [4].

The volume fraction and the mean coordination number z are the simplest structural descriptors for random close-packed solids. In our simulations both these quantities vary monotonically with the shaking intensity, but in our view, this is *not* an indication of a simple relationship $z(\phi)$ (see, e.g., the empirical relationship proposed in Ref. [13]) between ϕ and z. We consider the following counterexample. The range of volume fractions for monodisperse spherical particles that can be achieved by nonsequential reorganizations of the form discussed above includes the sequential random close packing fraction $\phi_{\mathrm{RCP}} = 0.581$; in contrast, the mean coordination number associated with nonsequential packing is substantially below that for sequential close packing, $z_{\mathrm{RCP}} = 6.00$. Thus $z(\phi)$ is clearly a multivalued function of ϕ that depends on the detailed history of the powder. This shows that a specification of both ϕ and z as independent quantities is insufficient to enumerate adequately many of the structurally dependent properties of random packings; in this respect, the distribution of the orientations for particle-particle contacts contains valuable additional information about granular structures. This second-order descriptor underlies most of the tensor properties of powdered materials [14].

We study this orientational distribution in terms of the angle θ between the z axis and a line drawn between the centers of contacting particles, with $0 \le \theta \le \pi/2$; we have plotted in Fig. 3(a) the distribution of $\cos(\theta)$ for configurations taken from the stable close-packed phase of the shaking steady states with $\epsilon = 0.05$ and 1.0. For comparison we have also plotted this distribution function for sequential random close packings. The contact angle distribution is flatter for the shaken deposits, which indicates that the nonsequential structures are more isotropic; this is to be expected, since the concurrent rearrangement of particles possible with our cooperative reorganizations will lead to less "sharpness" in configurational shapes which is the inevitable (and unrealistic) result when particles are forced to order one after the other. This distribution is in a sense a global indicator of the shapes of particle clusters, and thus of the roughness of the pile—the structures which are wasteful of space are those where there is a sharp distribution of contact angles because particles are not able, once deposited, to reorganize collectively to minimize voids. The sequential deposits and the nonsequential deposits at high intensity ($\epsilon = 1.0$) are in this category, while those at low intensity ($\epsilon = 0.05$) have a far more homogeneous distribution of contact angles, indicating that particles have organized themselves into flatter structures by slipping slowly into available voids to form a smoother pile—this behavior is in accord with earlier predictions [6–9].

We turn finally to a much more local indicator of cluster shapes. In a stable close packing, each particle has three special neighbors which form the bases for its sta-

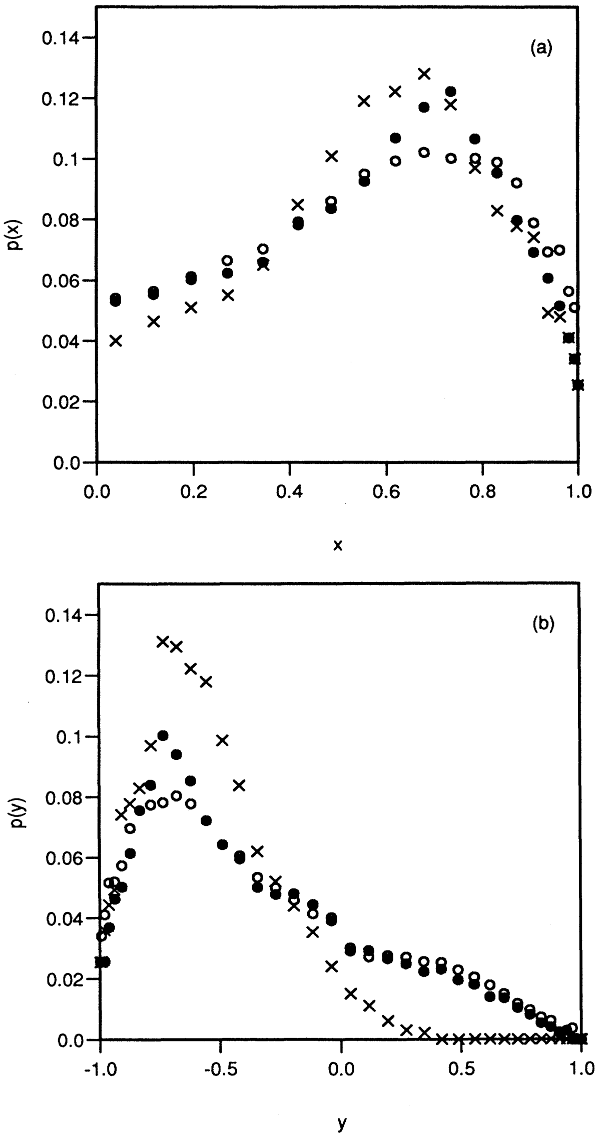

FIG. 3. The distribution of orientations for particle-particle contacts in random close packings of monosize spheres. Crosses correspond to sequentially constructed packings and open (closed) circles correspond to vibrated packings with $\epsilon = 0.05(1.0)$. In (a) $x = \cos(\theta)$ is the cosine of the angle between a particle-particle contact vector and the z axis. In (b), $y = \cos(\psi)$ is the cosine of the angle between the z axis and the contact vectors which form the stabilizing contacts of each particle.

bilization; that is, each particle has a subset of three of its contacting neighbors which define the local potential-energy minimum in which it rests. The distribution for $\cos(\psi)$, where $0 \leq \psi \leq \pi$ is the angle between the z axis and a line from the center of each particle to the center of each one of its three stabilizing contacts, is shown in Fig. 3(b). Again distribution functions are plotted for sequential random close packing and for configurations from the close-packed phase of shaking steady states with $\epsilon = 0.05$ and 1.0. The results in Fig. 3(b) show that a large number of the particles in the shaken packings are stabilized by particles whose centers are above [higher z, $\cos(\psi) > 0$] their own; while the number of these upward stabilizations observed in shaken packings is not strongly dependent on ϵ, this number is significantly higher than that found in sequential deposits. Since upward stabilizations are strong indicators of bridges, we are able to make two observations, the first and more obvious one being that nonsequential reorganizations are demonstrably essential for realistic packings of powders which contain bridges and arches. Secondly, although the observed dependence of $\cos(\psi)$ on ϵ is not strong, there does appear to be a trend suggesting that higher intensities lead to sharper distributions of $\cos(\psi)$ (i.e., more jagged local structures such as bridges) while lower intensities lead to flatter distributions (e.g., bridges that are less wasteful of space). In other words, this distribution is the "local" equivalent of the one presented in Fig. 3(a); we can argue, therefore, that high ϵ produces more jagged cluster shapes [sharper distributions of $\cos(\psi)$] and hence rougher piles [sharper distributions for $\cos(\theta)$] while the reverse holds for small ϵ, in accord with earlier predictions [6–9].

In conclusion, we have explained the physics behind the consolidation of a powder under "tapping," presented a theory of self-diffusion in granular flows which extends across both the relevant regimes of flow and voidage, and demonstrated the effects of vibrational intensity on cluster shapes and the roughness of the pile, within the framework of the theory of independent-particle and collective relaxation presented in earlier work [6–9]. In addition, we have highlighted the role of nonsequential algorithms in these studies as having the necessary realism to incorporate the physics of independent- and cooperative-particle motions. It is our hope that these issues will form the subject of inquiry by serious experimentalists, to aid in the development of the integrated approach to the microscopic physics of vibrated powders, which we regard as essential.

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