feature article

Heterogeneities in granular materials

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Different parts of a sandpile can exhibit very different dynamical behaviors ranging from jammed to fluid. To understand them, one needs to look at the networks of contacts between individual grains.

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At first sight, granular materials such as sand on the beach, coffee in a jar, and broken rocks flowing along a chute at a construction site appear homogeneous on scales above the size of individual particles. However, closer inspection reveals that is not always true; beaches can exhibit sand ripples, coffee granules gradually segregate according to size, and delivery chutes sometimes jam. Far from being anomalies, such behaviors are generic consequences of physics at the scale of a grain, and their analysis represents a major challenge in modern statistical physics.

Heterogeneities in granular materials complicate even the most mundane predictions—the pressure distributions on silo walls or the maximum flow rate from a particular hopper configuration, say. As a result, industrial efforts to process granular materials efficiently remain heavily reliant on empirical methods or continuum approximations that are not always appropriate.

Since the late 1980s, much research in statistical physics has focused on the nature of spatial disorder and temporal unpredictability in complex systems. Complexity is manifest in emergent systems—those in which macroscopic properties result from the collective behavior of many individual particles in large assemblies. A classic example of such a system is a pile of sand. A typical sand grain lives in a spatially disordered world and is too large for thermal fluctuations to affect its dynamics. Those two simple facts have complex consequences: Granular systems are strongly hysteretic since they do not, at ambient temperature, manifest the diffusion characteristic of Brownian motion. The hysteresis shows up, first, in structures that are embedded in the sandpile at the time of its formation and, second, in the influence of the structural memory on ensuing dynamics. Such spatiotemporal characteristics are heterogeneous by definition, because homogenizing processes such as Brownian diffusion cannot act to average them away.

Sandpile systems, especially their computerized cellular-automaton representations, have become a paradigm of

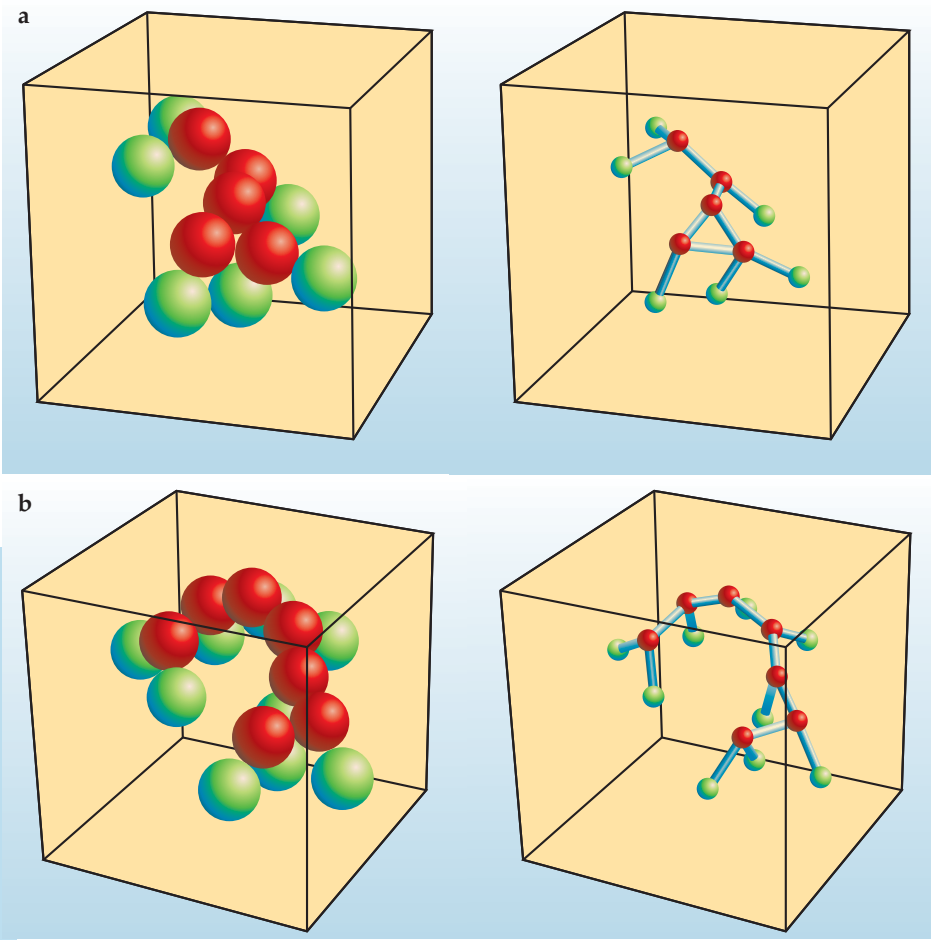

Figure 1. Bridges in granular assemblies. **(a)** A five-particle complex bridge (red) with six bridge-supporting base particles (green), with its corresponding contact network at right. **(b)** A seven-particle linear bridge (red) with nine base particles (green), with its corresponding contact network. (Adapted from ref. 3, A. Mehta, G. C. Barker, J. M. Luck.)

complex-systems research. Extensive analysis of them has led to the identification of avalanches and scaling behaviors in many social, economic, and biological systems. Examples range from electronic traffic jams on the internet to stock-market crashes. Somewhat surprisingly, however, the simplest variants of the sandpile models have not automatically led to results that are immediately applicable to granular materials. The interpretation of heterogeneities in real granular media, for example, requires a much deeper understanding of granular systems.[1]

Bridges

Arches—or bridges, as they are known in the granular community—are classic examples of spatial heterogeneity and were identified long before physicists entered the world of sand grains.[2] Bridges are collective structures in which grains rely on each other for mutual stability, much like their real-life analogues on roads and highways. Mutual stability means that cutting even one link of a bridge leads to its collapse. Cooperation among grains is thus intrinsic to the formation of bridges and is essential to their modeling. Therefore, computer algorithms that prescribe a purely sequential dynamics for grain deposition—each landing one after the other—produce granular packings that are entirely without bridges. Such "isostatic" packings can arise when there is no friction between grains.

In real granular packings, bridges form and are sustained precisely because of the presence of friction between grains. In a typical packing, up to 70% of the particles are part of bridge configurations, largely formed by processes such as shaking and pouring, in which cooperative effects emerge naturally when grains move together. Ensuing dynamics are also cooperative, which leads to the deformation of bridges rather than the destruction that sequential motion would have caused.[3]

Studies of the coordination number c, the average number of contacts for any particular grain, verify that picture: Frictional packings in d dimensions generated by cooperative dynamics contain bridges, and their coordination numbers, $c = d + 1$, are lower than those of frictionless packings, where sequential dynamics do not allow bridges to form and where $c = 2d$.

Materials science distinguishes at least two fundamental types of bridging, cohesive and interlocking. Cohesive bridges form in the presence of attractive intergrain interactions and have been analyzed by continuum approximations. Interlocking bridges, in contrast, can form when a group of hard particles becomes trapped even in the absence of attractive interactions. In that case, multiple direct, particle-to-particle contacts render further relative motion impossible. The essential discreteness of the scenario—without the bonding from attractive interactions—makes it unsuitable for analysis by continuum models. Although rarer in industrial applications than their cohesive counterparts, interlocking bridges may prove much more significant for the understanding of densely packed materials.

A fundamental unit of heterogeneity, an interlocking bridge can be further classified as linear or complex, depending on the topology of its backbone or contact network: Complex bridges have backbones with branches or loops; linear bridges do not.[3] As pictured in figure 1, a linear bridge made of n particles always rests on $n + 2$ base particles; what distinguishes bridge and base particles is the criterion of mutual stability. Cutting a link between two bridge particles causes them both to collapse; base particles stabilize bridge particles but are not stabilized by them in return.

Figure 2. A thin slice through a colloidal gel illustrates the heterogeneous dynamics of component particles. Arrows indicate the direction of motion for particles with displacements greater than 0.2 μm over a time interval of 600 seconds; lighter colors signify particles with larger displacements. The arrows are all the same length in three dimensions, so shortened arrows indicate motion into or out of the plane. (Adapted from ref. 10, E. R. Weeks, D. A. Weitz.)

The enhanced stability of loops in complex bridges means that the number of base particles is reduced for a given n. That loop-induced stability also means that the longer the bridge, the more likely it is to be complex, since linear bridges become exponentially rare as their length increases.[3] Moreover, the results of a kinetic "tube model" of linear bridges suggest that small linear bridges are almost never flat.[4] In the rare event that they remain linear as they become longer, linear bridges get weighed down, arching over to form domes supported by the walls of the container.

Those theoretical results, supported by computer simulations of sphere packings,[3] suggest that linear bridges evolve in a manner characteristic of a planar "self-avoiding" walk: Their generic fate is eventual collapse into complex bridges and, in rare cases, the formation of system-spanning domes. The tube model of linear bridges also predicts, in agreement with experiments on 2D bridges,[5] that the vertical growth of bridges is diffusive—that is, they grow as $n^{1/2}$—and their horizontal growth is faster than diffusive. Analogous results for complex bridges show that their size distributions follow a power-law decay and suggest that they evolve in a manner characteristic of percolation clusters.[3]

Jamming of particles at the mouth of a container is an everyday occurrence for many large-scale granular-processing operations. Bridges are entirely responsible for the phenomenon.[5] All downward granular motion becomes inhibited when a dome, for example, stably forms at any point of a packing. By extension, one sees that the potential of a bridge of size n to jam the container opening is related to the projection of its full 3D structure onto the basal plane of the container.

Indeed, judging by computer simulations, the size distribution of those projections has a well-defined peak that indicates the presence of a fairly characteristic base projection for 3D bridges of a given length. That makes it relatively easy to predict whether such bridges would jam a particular

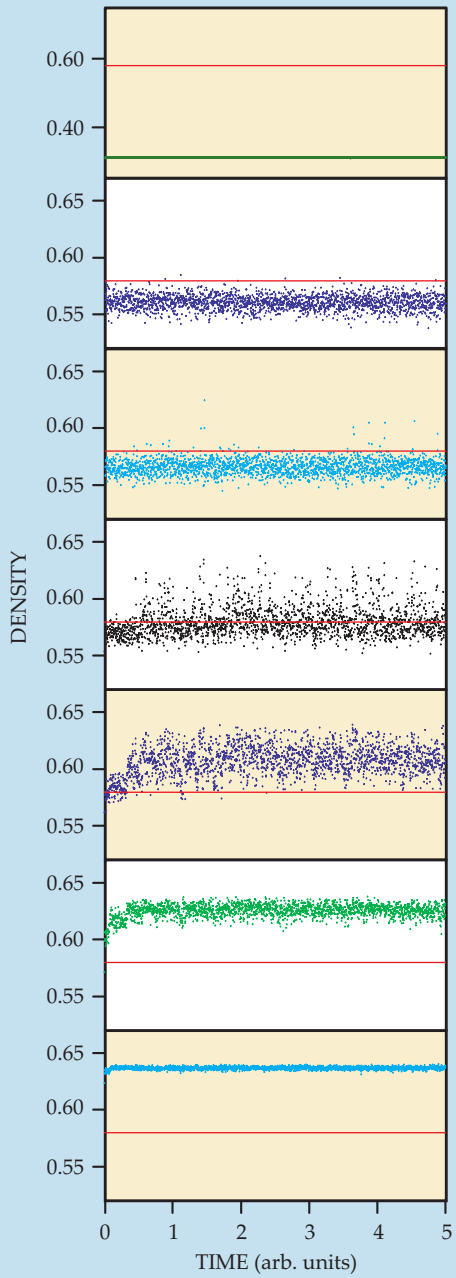

Figure 3. The densities of successive layers in a vibrating box of 1300 simulated grains as a function of time. The top panel represents the density of the uppermost layer and descending panels represent densities of progressively deeper layers; the red line, pegged at 0.58 in each panel, represents the packing density of grains laid sequentially. The densities of layers at the top, where particles are most free and mobile, experience few fluctuations about a consistently low density; those nearest the bottom, where particles are most likely to be jammed or slow moving, also experience few fluctuations, but around a consistently high density. The greatest density fluctuations occur in the middle of the box, where the widest range of particle environments are found—some particles are loosely packed, others jammed together. (Adapted from ref. 13.)

opening. The results are crucial for industries such as beer brewing, which suffers from the routine jamming of grain hoppers.

A related phenomenon of industrial importance concerns the compaction of particles following a bridge collapse.[3] When grains are packed to the point that individual grain motion cannot further increase the packing density—a point that occurs at a 3D packing fraction of 0.58 and corresponds to the maximum density achievable by sequential deposition of grains—only the coherent and collective collapse of bridges can squeeze out trapped space between grains and produce yet denser granular packings. The applications are relevant to a broad range of fields, wherever dense packing is desirable—from the manufacture of pills to the making of pillars.

Research on bridges has a long history in the literature, but recent work in granular physics has focused on force chains,[6] linear connections of stronger-than-average contact forces that stretch through dense granular structures but involve just a fraction of all particles. Researchers have identified force chains as routes through which stresses are preferentially—and thus heterogeneously—transmitted. That identification has prompted a chicken-and-egg discussion: Which is more fundamental, the granular structure or the force networks inside an already formed structure? That is, does the formation of bridges lead to force chains, or do interparticle forces that become organized into chains lead to the formation of bridges?

There are compelling arguments on both sides. On the one hand, granular structures form as a result of incipient forces during deposition. On the other, knowledge of the granular structure, along with some assumptions relating to material properties, allows researchers to compute a complete set of forces—at least in stable packings. With the imaging technology available today, 3D structures can be determined relatively easily; measuring the forces directly, even in two dimensions, is far more difficult.

Structural rather than force-related measurements might thus be more effective in probing static heterogeneity. From that perspective, the remarkable and detailed similarity of the distribution of bridge base projections to that of experimental measurements of normal-force distributions in granular packings is intriguing.[7] Indeed, because the normal forces are proportional to the area that the force chains subtend on the basal plane, it is tempting to speculate that long-lived linear bridges are the carriers of force in force chains.[1] Current experimental measurements to test such ideas could provide valuable clues about heterogeneity—for structures or forces—in granular packings at rest.

Fluctuations and correlations

Dynamical heterogeneities are equally significant keys to the behavior of complex multicomponent systems. When a large crowd leaves a stadium, some groups get stuck while others appear to move smoothly toward the exits. The presence of diverse dynamics in the same system has been observed in many granular materials and has led to intriguing concepts such as "rattlers in a cage," in which individual grains or small groups of them can move, albeit in a restricted space bordered by their stuck neighbors.

When a granular structure relaxes—in response to shaking, for example—particle bridges can give rise to those heterogeneous responses. Such structural relaxation invariably leads to bridge collapse, which can directly affect other grain reorganizations; the effect might remain local, or it might spread throughout the system in an avalanche. Computer simulations of model systems have confirmed both cases near jamming,[1] but the phenomena are difficult to observe in experiments unless they occur close to a free surface.

In simulations, the displacements of individual grains were found to be anticorrelated on average, and a subsequent theoretical model of grains near jamming corroborated the observation.[8] Analogous analyses in molecular and colloidal glasses revealed similar phenomena.[9,10] The experimental illustration in figure 2 shows dynamical heterogeneities in a 2.5-μm-thick slice through a 3D colloidal sample. Cooperative rearrangements between large groups of particles are

clearly visible. As the glass transition is approached, the cluster of less mobile particles grows larger and rearrangements become much less frequent.

A great deal of work in recent years has tried to unify the phenomenology of dynamical heterogeneities in dry granular media, on the one hand, and dispersed colloidal suspensions on the other. For a novel and illuminating example of such research, consider pastes, dense colloidal suspensions often used for foods and household preparations.[11] Those materials present the most difficult aspects of hydrodynamics and mechanics in granular media; they are, however, uniquely convenient to package and to handle in industrial applications. The slow heterogeneous dynamics that occurs in and around interlocking bridges in granular deposits may reveal details about the motions that occur in liquids as they approach the glass transition—details that otherwise are notoriously difficult to glean. From that point of view, a deep understanding of pastes is likely to be invaluable as much to pure science as to industry.

Particle bridges invisibly come and go as granular systems slowly change in response to their environment, just as crowds of people ebb and flow as they disperse. Structural and dynamic heterogeneity are inextricably linked to patterns on length and time scales beyond those relevant to individual particles. Experiments performed a decade ago in Chicago using nothing more complex than a glass tube filled with sand captured the essence of spatiotemporal heterogeneity in granular systems.[12] The experiments showed that different parts of a vibrated granular system behave differently depending on their location. More specifically, both the average density and the fluctuations in density varied strongly throughout the shaken column of grains as a function of the depth.

Computer simulations of those experiments reproduced the spatiotemporal effects evident in the density fluctuations of different sections of the column. More specifically, they verified that the time-averaged value of density, about which temporal fluctuations occur, is an increasing function of depth.[13] One of the most surprising features of those results, at least at first glance, is that the range of density fluctuations is greatest near the middle of the column, as seen in figure 3. However, that feature can be simply explained. Sand grains at the top of the column are fluidlike, with complete freedom of motion in response to vibrational excitations, so they experience few fluctuations about consistently low-density packing. The region near the bottom, consisting of mostly jammed particles, has little chance to fluctuate about consistently high-density packing. The middle of a column, though, experiences a quickly changing particle environment, which can vary from jammed to loosely packed as a function of time.

Tracking movement

Modern experimental and software developments provide powerful representations of motion inside a dense, complex material such as a colloidal gel. In confocal microscopy, for example, index-of-refraction differences between densely packed regions and loosely packed ones offer enough imaging contrast that researchers can follow the cooperative motion of several thousand colloidal particles.[10] Computer simulations have paralleled the achievement and offer a glimpse of the spatiotemporal phenomena by following the tracks of an individual particle inside granular material.

The results confirm the existence of heterogeneity: The particle's behavior depends on whether it is traversing a region closer to the top or the bottom of a granular pack.[13] Figure 4 tracks the history of a particle that begins in the middle of the box and subsequently traverses the whole system.

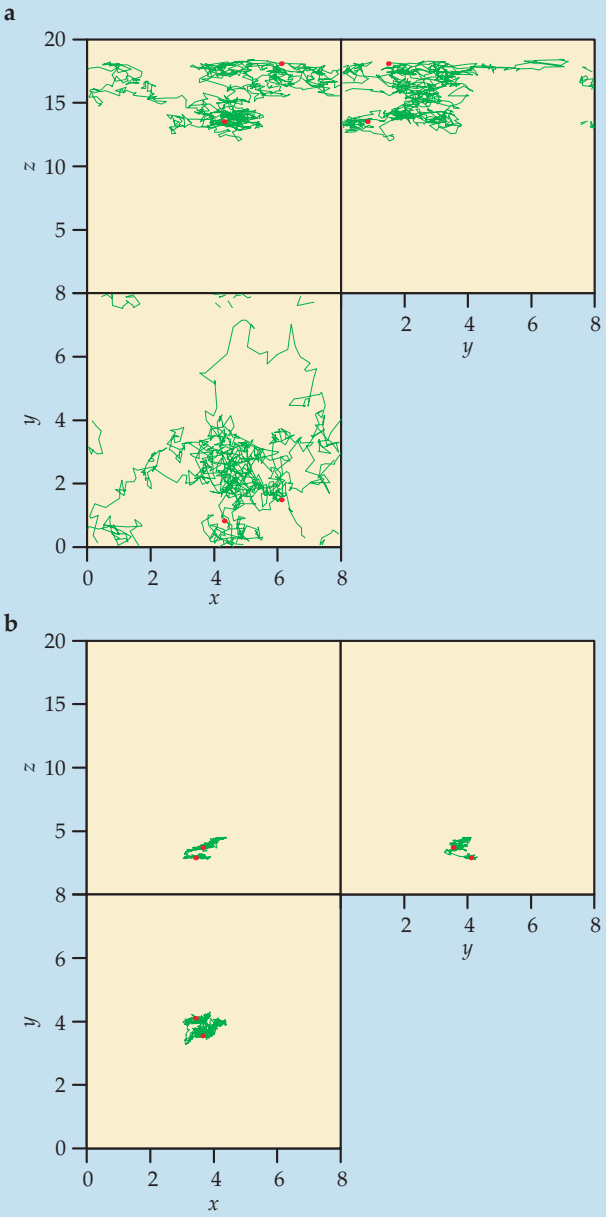

Figure 4. Simulation results for the trajectories of a single grain (green), initially located in the middle of a vibrating box of 1300 grains. The trajectories are projected onto the *xy*, *yz*, and *xz* planes, with start points and endpoints in red. **(a)** When the particle explores the near-surface region, its movement is fluidlike and relatively unimpeded. **(b)** When it travels through lower regions, it becomes localized, constrained by its neighbors. The spatial units are arbitrary, but on the order of the grain size. (Adapted from ref. 13.)

The fluidlike trajectory of the particle near the top of the box (figure 4a) is in sharp contrast to the nearly jammed one when the particle explores regions near the base (figure 4b) after a long period of time measured in cycles of the vibrational excitation. Both types of trajectory look remarkably similar to ones observed in particle-tracking experiments in colloidal suspensions, where the presence of Brownian motion throughout the system prohibits spatial segregation of

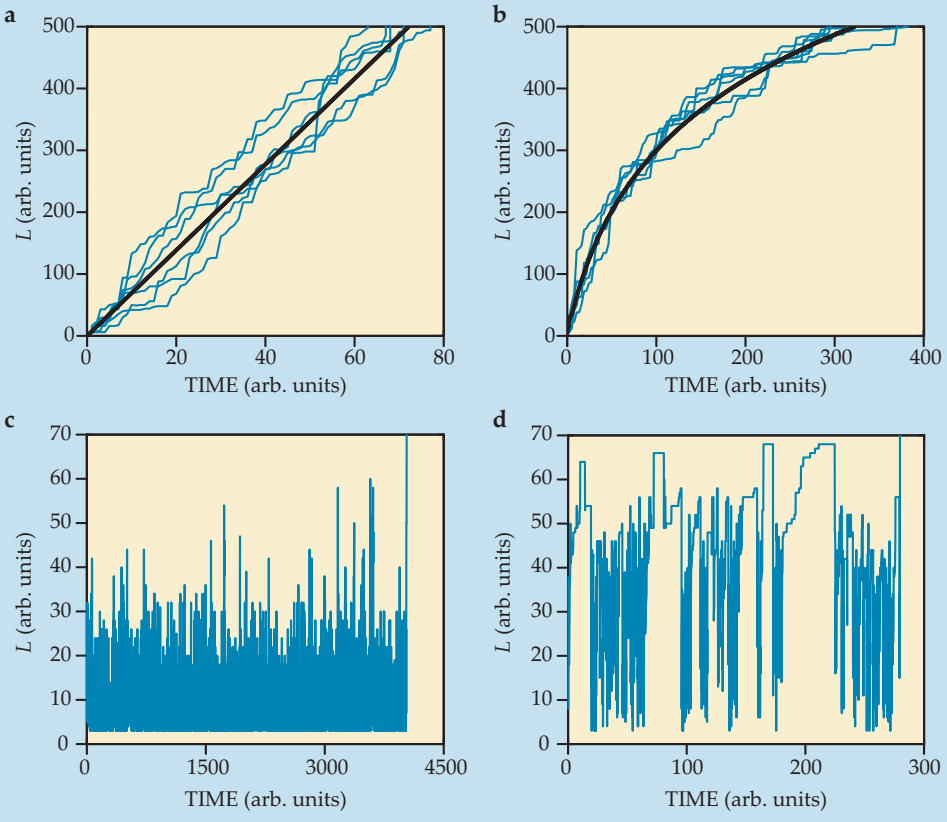

Figure 5. Zero-temperature dynamics. Lightly tapping a system can guide particles into their ground-state, ordered configuration. In these numerical plots, which illustrate the four dynamical phases of a granular column, L is the length of an ordered layer as a function of time. **(a)** The "ballistic" phase denotes the dynamics of particles near the top of the column. Nearly free, these particles become ordered linearly in time. **(b)** The "logarithmic" phase denotes the depth-dependent slowing of the dynamics in time; each track here and in the ballistic case represents a separate simulation of the system's evolution, with the bold line a best-fit average. **(c)** The "activated" phase, here represented by a single track, emerges from rapid density fluctuations experienced by middle-layer grains and reveals the seemingly random variation in ordered lengths as a function of time. **(d)** The "glassy" phase denotes dynamics in which particles are frequently jammed and immobile. The system remains unaltered for large periods of time separated by sudden fluctuations in the length of the ordered state. At the end of the trajectory, it jams into one of two intrinsic states. (Adapted from ref. 13.)

dynamical phenomena. The similarities between particle tracks in granular media and in colloidal gels raise intriguing but unresolved questions about the origin and effect of heterogeneities in the different media.

In practice, even simple models of granular behavior can be used to explore the complex spatiotemporal patterns. One theoretical model for a column of grains near jamming starts with grains that can orient themselves in one of two states.[8] Compaction occurs in the column when all excess voids—trapped space between grains—are squeezed out. The model assumes that the column is sufficiently jammed that particles can no longer diffuse; they can only change orientations. Every grain has to choose the orientation that is most favorable to minimizing voids with respect to those above it as well as to those below it. The choices are frequently not the same, which leads to geometrical frustration. A further complication is that in the presence of gravity, the lower, more weight-bearing grains move more slowly than less-burdened grains higher in the packing.

The effect can be included in the model as a simple depth-dependent local frequency at which grains can reorganize. That minimal model, whose two main ingredients are frustration and the effect of gravity, predicts the existence of density fluctuations and suggests that their magnitude increases from the top to the middle of the column, in agreement with numerical simulations and the Chicago experiments.

Tops, middles, and bottoms

A variant of zero-temperature dynamics, traditionally used to find the ground states of a system, can also be used to probe dynamical behavior. That theoretical tool monitors the propagation of order in a system at minimal perturbation; here, the dynamics are reflected in the behavior of the ordering length—that is, the length over which the column is ordered—at zero vibrational intensity (see figure 5).[13]

A complete picture of spatiotemporal heterogeneity emerges from particle behavior in different regions. At the top of the column, the dynamics are ballistic: Grains move fast and freely in response to vibrational excitations. Away from the top, the motion slows logarithmically as a function of depth because of weight-related effects. Near the middle of the column, a region that "feels" both upper and lower parts of the packing equally, grains experience frustration. Their dynamics are referred to as activated; in that region, increases in density take place by chance, when local entropic barriers are crossed.

Deeper still, the effects of frustration are twinned with the extremely slow motions of grains weighed down by higher layers. Near the base of the column, the model predicts glassy dynamics: The dynamics of the density profile vary between rapidly fluctuating periods and slowly fluctuating ones close to a ground state. Moreover, not all the ground states are equal; the lowest-lying intrinsic, or "crystalline," states are impossible to reach at finite vibrational intensities when the system typically jams into a higher band of metastable ground states.

Intuitively, the heterogeneities reveal a simple and compelling picture: Granular assemblies have tops, middles, and bottoms, which separately respond to vibration by giving rise to ballistic, activated, and glassy dynamics. Among many other things, the diverse dynamics caution us to be careful when assigning effective temperatures to shaken granular media. In contrast to the effect of heating or cooling on the phase behavior of a system, vibration can often lead to ordering.[14] In fact, the existence of dynamical and spatial heterogeneities should properly be regarded as a clear consequence of the athermal nature of granular media.

Understanding the nature of the heterogeneities has a potentially enormous bearing on industrially relevant problems—from the efficient transport of particles to the avoidance of jamming of materials as disparate as rocks, grains, and pharmaceuticals. Equally important is the insight that understanding would bring to a range of geological phenomena, such as snow avalanches and earthquakes, in which the granular picture is fundamental.

Unexpected benefits may follow from comparisons with other systems such as polymer and molecular glasses. Recent investigations have shown that the melting of polymer glasses in response to external stress manifests strong spatial heterogeneity in a way that seems consistent with the emerging picture of granular relaxations.[15] No less intriguing, the way that polymer glasses flow, in terms of local structural relaxations, appears to depend on whether the driving force is shear stress or temperature change.

Many questions on spatiotemporal heterogeneities in granular media remain unsolved. How, for example, do distinct dynamic regimes couple? What are the effects of particle shape? What are the effects of adhesion, cohesion, and other intergrain forces? Can we link static and dynamic heterogeneity by looking at the dynamics of heterogeneous structures like bridges? The field is wide open to theorists and experimentalists, as a rich variety of complex behaviors in deceptively simple systems remain unexplored. As William Blake reminds us, there truly is a world in a grain of sand.

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