A thermodynamic theory of granular material endures

Theorists have tested what seemed like an untestable conjecture: that all the possible arrangements of grains in a packing are equally probable.

In some ways, the physics of granular materials—powders, sand, and the like—could hardly be simpler. The individual grains are, by definition, too big to be jostled by thermal fluctuations. Once they settle—say, in a heap—they sit there, each grain locked in place by its neighbors.

But granular materials' athermal nature also makes them confounding. (See the article by Anita Mehta, Gary Barker, and Jean-Marc Luck, PHYSICS TODAY, May 2009, page 40.) Once formed, a granular material won't spontaneously explore new configurations, so its behavior depends intimately on how it's prepared. A vibration that triggers an avalanche in one sandpile might barely register in another.

So when Cambridge University's Sam Edwards endeavored during the 1980s to construct a universal theory of granular packings, he'd set himself a tall order. He and his student Robert Oakeshott approached the problem by drawing an analogy between a packing of grains and a thermodynamic ensemble of atoms: The volume of the packing is to the grains as the energy of the ensemble is to the atoms, and the number of ways the grains can stably fill the volume—more precisely, the logarithm of that number—constitutes an entropy.[1] From those properties followed others, including the granular equivalents of temperature, free energy, and specific heat. Underpinning it all was a seemingly innocent conjecture: that all allowable configurations occur with equal probability.

Edwards's model yielded valuable insights into granular behavior. It helped explain, for example, the density fluctuations of powder shaken in a container. But Edwards himself never bothered to check whether the equiprobability conjecture was valid.

Now researchers led by Daan Frenkel of Cambridge University and Bulbul Chakraborty of Brandeis University have tested Edwards's conjecture in simulated packings and found that it holds at only a single point in parameter space.[2] As it happens, that point—at the boundary between stability and flow—might be the most important in all of granular physics.

A numbers game

If you suspect someone has rigged a die by shaving it, you might roll the die and note how frequently the different faces come up. The shaved face and its opposite should appear more often than the others. But only after several rolls—necessarily, many more rolls than there are faces on the die—does the bias become apparent. Statisticians call that the law of large numbers.

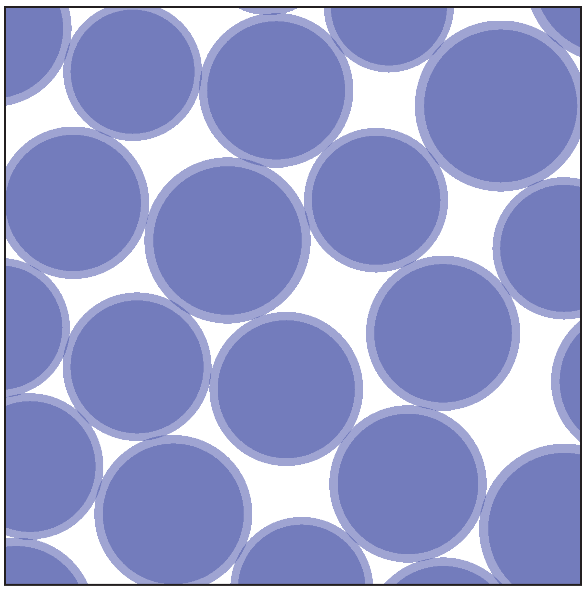

FIGURE 1. IN THIS GRANULAR PACKING, each grain is locked in place by its neighbors. Grains are modeled as partially compressible disks consisting of a hard core (dark purple) encircled by a soft outer ring (light purple). Hard cores are forbidden from overlapping, whereas outer rings repel each other with a spring-like force when they overlap. (Adapted from ref. 2.)

In theory, you could test Edwards's equiprobability conjecture in the same way—by either experimentally or numerically generating many different packings of the same grains. But the law of large numbers would work heavily against you. There are more ways to stably pack even 100 grains than there are atoms in the universe. A real-life granular packing is akin to an infinite-sided die.

A more direct way to tell if a die has been shaved is to measure the areas of its faces. In essence, that's how Frenkel and his colleagues approached the testing of Edwards's conjecture. They noted that the possible arrangements of grains in a packing can be described with a many-dimensional energy landscape. Each local minimum represents a stable configuration, to which nearby unstable configurations equilibrate. Those nearby states form what's known as a basin of attraction; much like the area of a die face, the volume of a stable configuration's basin of attraction reflects the probability of that configuration arising by chance.

For systems larger than a few grains, however, the energy landscape is too complicated to explicitly calculate. How, then, to measure the basins?

To start, the researchers randomly select a stable configuration in the landscape. That is, they simulate the compression of loose grains to some predetermined packing density. For computational simplicity, they consider a two-dimensional system of frictionless grains: Each grain is modeled as an incompressible disk surrounded by a soft outer ring, as illustrated in figure 1; the packing density is the fraction of the total area covered by disks.

Once the packing has stabilized, the group initiates a Monte Carlo algorithm that displaces disks at random, in tiny increments. Before finalizing a displacement, the code checks that the perturbed packing remains in the original basin of attraction; displacements that would tip the system to a new basin are rejected. In that way, the algorithm performs a statistical random walk that's confined to the original configuration's basin, as illustrated schematically by the arrows in figure 2. From the properties of the walk, the team can estimate the basin's volume, despite incomplete knowledge of its boundaries. Crucially, one needn't repeat the process for every basin to know whether Edwards's conjecture holds up; a representative sampling will do.

Wrong 'til it wasn't

In 2011 Frenkel teamed with Ning Xu (University of Science and Technology of China) and Andrea Liu (University of Pennsylvania) to apply the Monte Carlo method to packings of 16 disks, the most the code could handle at the time.[3] Although the researchers considered only a single packing density, they found that the basin volumes differed wildly, by up to several orders of magnitude. That result jibed with a 2009 study by Corey O'Hern (Yale University), Mark Shattuck (City College of New York), and coworkers, who enumerated all the possible configurations of a seven-disk packing and found that some configurations arose in experiments more frequently than others.[4]

So when Frenkel's new team of collaborators revisited the packing configuration problem, recalls group member Stefano Martiniani, "We didn't go in with the aim of checking whether the conjecture was right or wrong. We already felt it was wrong, full stop."

Instead, the researchers were mainly interested in studying larger systems and seeing how the basin-volume distribution would vary with packing density. Martiniani and postdoc Julian Schrenk had spent three and a half years rewriting and fine-tuning the code. It was now fast enough to compute the basin volume of a 64-disk configuration in just a week's time.

The team found that the basin-volume distributions in those packings narrowed as packing density decreased. At a density known as the unjamming point—the lowest density at which a stable packing can form—the basin volumes were nearly identical. Suspecting that the residual differences might have been an artifact of the finite system size, the group used a scaling technique to extrapolate their results to the limit of infinitely many grains. In that limit, the variation in basin volumes disappeared altogether at the unjamming point. At that density, and seemingly nowhere else, Edwards's equiprobability conjecture held.

"Lovely if true"

The new results don't close the case on the Edwards conjecture. For one, it's unclear whether simulations of frictionless disks will generalize to real 3D packings of frictional, irregularly shaped grains. Plus, the simulated packings were formed through a specific protocol of isotropic compression; real granular packings are more likely to form under the directed forces of gravity or shear.

But if the conjecture does hold up, even if only at the unjamming point, the implications could be far reaching. The unjamming point is where static packings destabilize and begin to flow. If Edwards's model works there—and especially if it can be expanded to nearby densities—it might reveal universal precursors to avalanching, shearing, and other types of mechanical failure.

Even where the simulation results are at odds with the equiprobability conjecture, the discrepancies could be instructive. It's relatively straightforward to amend Edwards's definition of entropy to permit a distribution of configuration probabilities. The math becomes more difficult, but the equations don't substantially change.

Says Karen Daniels, an experimentalist at North Carolina State University, "Away from jamming, there might not be equiprobability, but if we can put the correct probabilities into our models—and simulations like Frenkel's tell us how to do that—then we might be able to better understand our experimental data. It's lovely if the Edwards conjecture is true—and interesting if it isn't."

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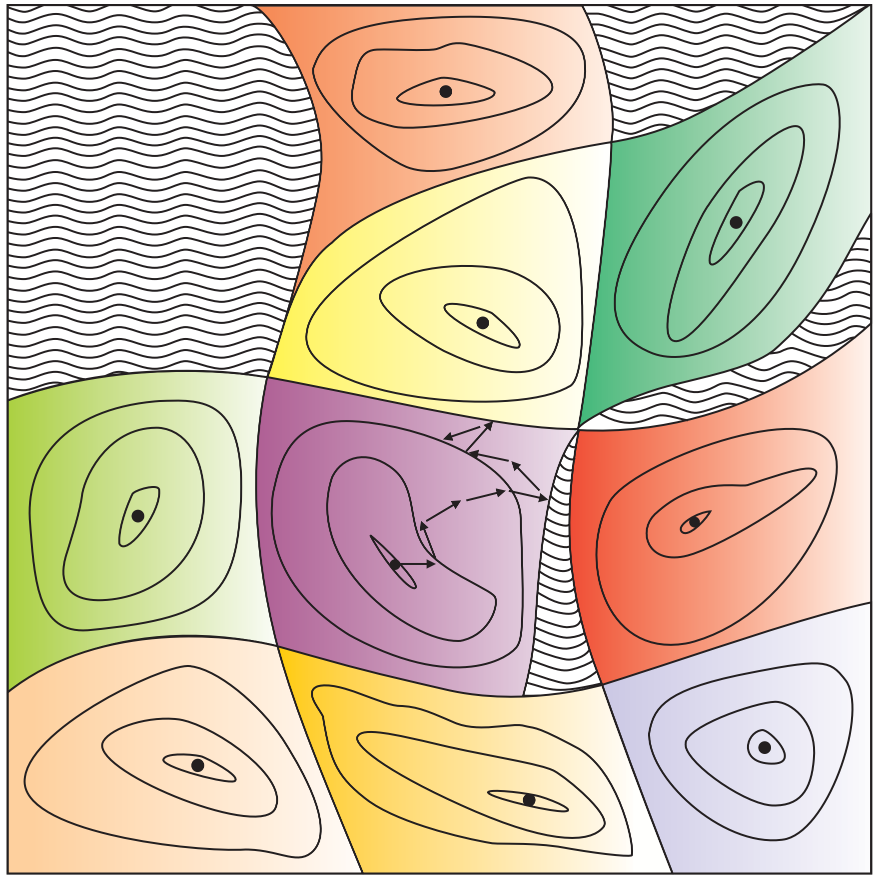

FIGURE 2. AN ENERGY LANDSCAPE depicts the possible arrangements of grains in a packing: Stable configurations (dots) correspond to local minima to which "basins" (colored areas) of nearby unstable configurations are attracted. (Contours are equal-energy curves.) Striped regions are inaccessible because they would require the partially compressible grains to be either unallowably close or too distant to touch one another. The many-dimensional energy landscape can't be calculated explicitly, but the volume of a configuration's basin of attraction can be estimated numerically from a random walk confined to that basin's configuration space, as illustrated by the arrows. (Adapted from ref. 2.)

References

1. S. F. Edwards, R. B. S. Oakeshott, *Phys. A* **157**, 1080 (1989).
2. S. Martiniani et al., *Nat. Phys.* (in press).
3. N. Xu, D. Frenkel, A. J. Liu, *Phys. Rev. Lett.* **106**, 245502 (2011).
4. G.-J. Gao et al., *Phys. Rev. E* **80**, 061304 (2009).