GRANULAR CRYSTALS: Nonlinear dynamics meets

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The freedom to choose the size, stiffness, and spatial distribution of macroscopic particles in a lattice makes granular crystals easily tailored building blocks for shock-absorbing materials, sound-focusing devices, acoustic switches, and other exotica.

Innovation in materials and the emergence of new engineering applications are closely intertwined. The ability to shape metals gave rise to the plow and the sword; the development of semiconductors led to the invention of transistors; and, more recently, the synthesis of composite materials such as reinforced plastics, polymers, and metal foams has produced lighter, more fuel-efficient cars and aircraft. In each case, the meaning of "material" can be interpreted in an expansive sense as more than chemical composition. The geometric arrangement of atoms also matters, and together both geometry and chemistry help determine a material's mechanical or electrical properties. Indeed, innovation in materials can occur not only by discovering new molecular compounds or doping existing ones but also by exerting precise control over the compounds' microstructure. Metal foams, for instance, are just one example of a material whose density and stiffness can be tailored by controlling the composition and porous microstructure during the fabrication process. (See the article by John Banhart and Denis Weaire, Physics Today, July 2002, page 37.)

Granular crystals similarly blur the definition of a material. As their name suggests, they are fabricated not from atoms or molecules but from macroscopic grains. More specifically, they are tightly packed lattices—or more disordered arrangements—of solid particles that deform on contact with each other. Prototypes can be assembled and studied using ingredients as generic as ball bearings, as shown above and in figure 1. Think of the balls as macroscopic analogues of atoms in a crystalline solid. Like atoms in a crystal, the particles in a granular crystal can be arranged in one-, two-, or three-dimensional lattices. However, unlike atoms, which interact through chemical bonds, macroscopic particles exchange forces and momenta through their geometric contact interactions.

The macroscopic particles can themselves be composed of different materials, such as metals, polymers, and ceramics. The choice of each particle's composition and shape affects the interactions and, ultimately, the mechanical response of the bulk granular crystal. Defects, such as dislocations, vacancies, and the presence of particles with a different composition, are akin to extended or point defects in atomic crystals; and they can similarly affect the bulk crystal behavior. In stark contrast to the random presence of defects in atomic crystals, however, defects in granular crystals can be placed in specific locations. That ability to precisely engineer defects into a material provides a knob for controlling its behavior.

As in other artificial materials, such as optical and acoustic metamaterials (see the article by Martin Wegener and Stefan Linden, Physics Today, October 2010, page 32), wave propagation in granular crystals can be controlled through the crystals' structural periodicity and local resonances. However, their dynamic behavior is governed by the elastic defor-

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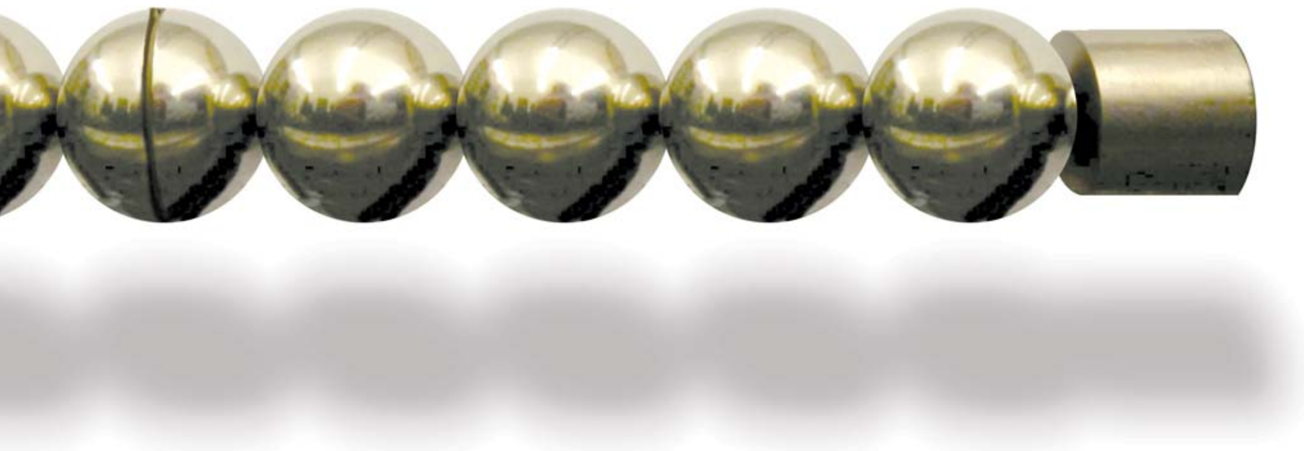
materials engineering



mations experienced by each particle. When two component spheres are adjacent, contact initially occurs at a single point. When the spheres are slowly pressed together, they overlap by a length δ, and the contact point becomes a 2D region. The compressive force is proportional to $\delta^{3/2}$, as outlined in the box below. The strongly nonlinear response sets granular crystals apart from other materials—for instance, microtruss lattices and microlaminate composites—that have a custom-made microstructure (see PHYSICS TODAY, January 2012, page 13).

The behavior of granular crystals has inspired numerous studies of the interplay between nonlinearity and discreteness.[1] The nearly three decades of work on such crystals since the pioneering studies of Vitali F. Nesterenko in the 1980s and early 1990s has produced exciting advances in experiment, computation, and theory. For example, new theoretical developments capture the effects of dissipation on propagating waves, and new experiments and numerical simulations include the effect of plasticity on contact between spheres.[1]

As with granular crystals, amorphous granular materials such as soils, sand, and cereals are also tunable, but their physics is very different and beyond the scope of this article. See, for example, reference 2 and the article by Anita Mehta, Gary Barker, and Jean-Marc Luck, PHYSICS TODAY, May 2009, page 40.

In this article, we focus on recent developments of granular crystals—in particular, the major advances toward such potential engineering applications as the creation of shock-absorbing materials, acoustic lenses, switches, logic elements, and energy-harvesting systems.

Hertzian interactions

In 1882, Heinrich Hertz published a description of the contact interaction between two elastic bodies.[18] When the particles are compressed, they deform slightly and exert repulsive forces. Such forces, which are nonlinear because the contact area increases with the deformation, govern the interactions between each pair of particles in a granular crystal.

Consider two identical spheres that, when pressed together, squish each other by an "overlap" length δ, which is on the order of microns for materials such as stainless steel, aluminum, bronze, brass, and Teflon. The force F between the two particles is given by Hertz's law: $F = A\delta^{3/2}$, where $A = E\sqrt{2R}/[3(1-\nu^2)]$, the elastic modulus E is a measure of stiffness, the Poisson ratio ν is a measure of a material's tendency to expand in lateral directions when compressed, and R is the spheres' radius. A version of Hertz's law also applies when considering nonidentical spheres. For stiff materials, it is typical (and often reasonable) to ignore dissipative effects in the model equations for granular materials.

Ball bearings, commonly with radii of 1–10 mm, are a convenient type of stainless steel sphere used in experiments. The elastic modulus of steel is about 193 GPa, and its Poisson ratio is about 0.3. Remarkably, granular crystals assembled from such spheres have a dramatically low bulk wave speed—below 100 m/s—due to the large inertia from each particle's mass. The speed of sound in bulk stainless steel, by contrast, is about 6000 m/s.

Particles with different geometries can yield interaction exponents that differ from $3/2$. For example, cylindrical particles have a variable interaction potential that depends on axis orientation, and hollow spheres have interaction laws that vary as a function of shell thickness.

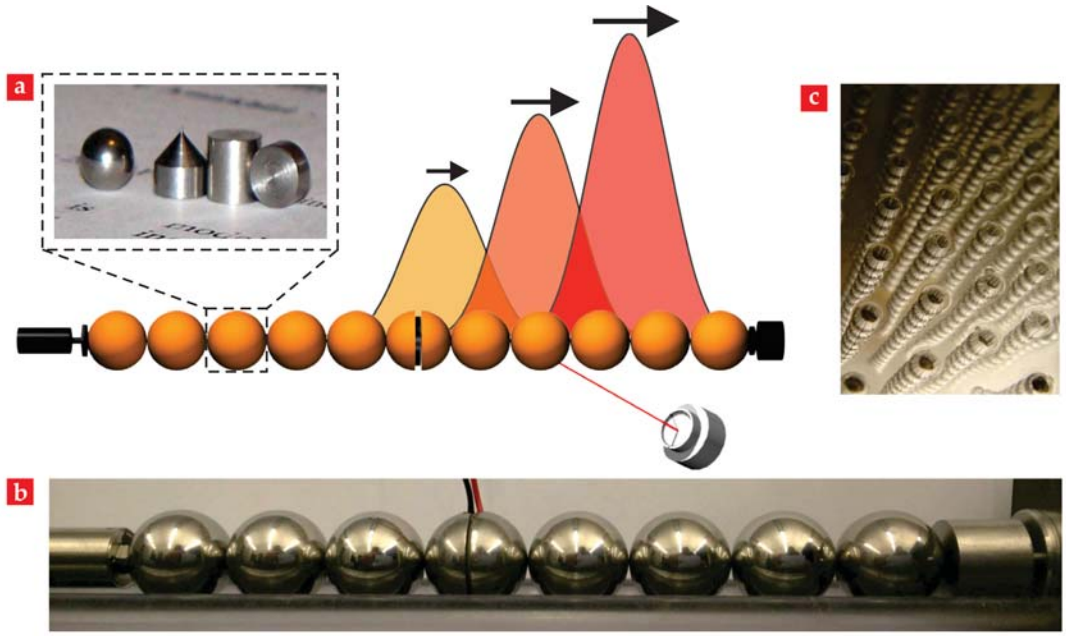


Figure 1. A granular crystal is a lattice of macroscopic particles. **(a)** In the chain sketched here, the particles are stainless steel ball bearings; they could also have some other geometry (inset) or composition. The chain supports traveling waves that can be excited by a piezoelectric actuator attached to one end; the greater the wave amplitude, which can be measured by a static force sensor at the opposite end, the faster the wave propagates through the chain. A vibrometer uses a laser (red line) to measure the displacement and velocity of a selected particle, and a sensor embedded in another (central) particle measures the forces it experiences.[15] **(b)** In this experimental implementation of the chain, each stainless steel ball bearing is about 2 mm in diameter. **(c)** An array of such chains embedded in a polymer matrix forms a three-dimensional granular metamaterial. (Panels a and b courtesy of Joseph Lydon.)

Control knobs

An uncompressed, 1D crystal—that is, a granular chain with no external force applied to its boundaries—is sometimes described as a sonic vacuum, because the speed of sound is 0 in a system whose interparticle interactions are purely nonlinear. That is, the equations of motion contain no linear term. Such chains (see figure 1) support the propagation of so-called solitary waves.[1] Those elastic waves, which remain highly localized and coherent while traveling along the chain, have fascinating features. For example, the waves' speed depends on their amplitude—the larger their amplitude, the larger their speed—but their wavelength is independent of amplitude and roughly equal to five particle diameters.

However, if one applies a static external force to compress the ends of a chain, such that the particles' static deformations from the load are comparable to or exceed their dynamic deformations when struck by another object, the nonlinear behavior in granular crystals weakens and may even become almost linear. In that near-linear regime, granular crystals have features familiar from discrete periodic systems, including the presence of bandgaps in their dispersion relation.[3]

The precompression just described is only one convenient way to tune a granular crystal's mechanical behavior. Engineers also have access to a large variety of solid materials, whose different elastic properties produce different sound speeds through the crystal. Control over particle geometries—cylindrical, toroidal, ellipsoidal, conical, or others—offers further tunability of the nonlinearity governing local contact interactions. Moreover, when one increases the particles' complexity, such as by coating them with another material, nesting them inside larger particles, or embedding them inside a polymer matrix, it becomes feasible to study other dynamic effects triggered by internal degrees of freedom—including local potentials, whose presence modifies the contact interactions.[4]

Waves in chains

A granular chain of N spherical particles can be described as a set of coupled oscillators, whose internal interactions between each pair of particles are governed by Hertz's law, as discussed in the box. In the 1D setting of chains, the Newtonian equations of motion are

$$\frac{d^2u_n}{dt^2} = \frac{A_n}{m_n}\left[\Delta_n + u_{n-1} - u_n\right]_+^{3/2} - \frac{A_{n+1}}{m_n}\left[\Delta_{n+1} + u_n - u_{n+1}\right]_+^{3/2},$$

where the + subscript signifies that the term in brackets is kept when it is positive but set to 0 when negative, u_n is the displacement of the nth particle from its equilibrium position in the initially compressed chain, m_n is its mass, and $\Delta_n = (F_0/A_n)^{2/3}$ is a static displacement that arises from a constant precompression F_0. The interaction parameter A_n depends on the elastic and geometric properties of the nth and $(n-1)$th particles.

Granular chains can support three primary types of waves: traveling waves, shock waves, and intrinsic localized modes, which are also known as discrete breathers—the spatially localized, time-

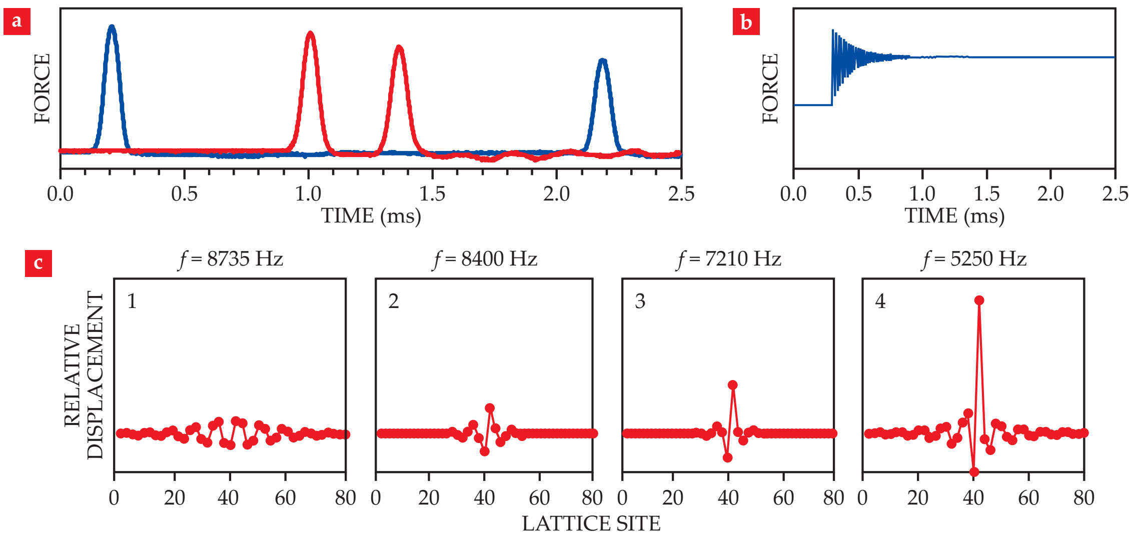


Figure 2. Granular chains support three primary types of waves: **(a)** dispersionless traveling waves; **(b)** shock waves, whose trailing amplitude profile decays slowly; and **(c)** discrete breathers, which are spacially localized, temporally periodic, and long-lived excitations. Panel a plots the force experienced by two particles—number 10 (blue) and 35 (red) on a 40-particle chain—when a single wave propagates along the chain. The wave passes through the two particles in succession and then subsequently reflects at the chain's end and propagates back toward the left. Panel b shows the temporal evolution of the compressive force experienced by a particle in a chain subject to a shock wave. Panel c shows the normalized relative displacement experienced by a family of discrete breathers. Each breather oscillates at a frequency *f* close to or within the bandgap of a numerically simulated chain of 81 particles, whose displacements are represented as red dots. Subpanels 1–4 depict breathers excited at ever-decreasing frequencies: (1) close to the lower edge of the optical band; (2 and 3) inside the bandgap; and (4) near the acoustic band edge of the compressed chain. (Panel a courtesy of Jinkyu Yang; panel b adapted from ref. 6; panel c adapted from ref. 16.)

periodic, and stable (or at least long-lived) excitations sometimes found in periodic systems[3,5] (see the article by David Campbell, Sergej Flach, and Yuri Kivshar, PHYSICS TODAY, January 2004, page 43). Figure 2 illustrates each type of wave.

A straightforward way to produce traveling waves is to strike one end of the chain. The traveling waves resemble conventional solitary waves,[6] but with tails that decay at a much faster rate (doubly exponentially) than that of a typical solitary wave[7] when the tunable parameter $\Delta_n = 0$. When $\Delta_n \neq 0$, the waves decay exponentially and can, at least for small amplitudes, be approximated by hyperbolic-secant-squared profiles. Chains of disparate particles, such as alternating steel and aluminum spheres in a diatomic chain, produce intriguing generalizations of the traveling waves. Just a few years ago, however, it was realized that the traveling waves do not survive long in such heterogeneous chains, except for isolated parameter values—particular mass ratios of adjacent spheres in the chain.[8]

The ability of granular crystals to support the propagation of stresses in the form of solitary waves makes them attractive as materials that can focus and guide coherent structures. For one thing, traveling waves in granular crystals are far less susceptible to

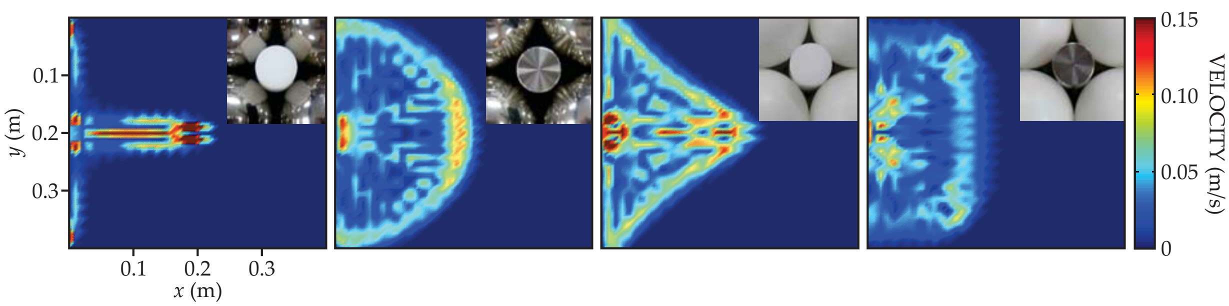


Figure 3. Wave propagation through a face-centered, square granular crystal. The crystal consists of a 20 × 20 square array of spheres containing a 19 × 19 square array of cylinders. Waves are produced by a localized impact on the left side of the crystal. Variations in the distribution of the material ingredients in the unit cell (inset) dramatically alter the shape of the wavefront. From left to right, the crystal is composed of polymer cylinders and steel spheres, steel cylinders and steel spheres, polymer cylinders and polymer spheres, and steel cylinders and polymer spheres. (For a larger picture of the last configuration, see page 88.) The cylindrical polymers are made of Teflon, and the spherical polymers are made of Delrin, which is about six times as stiff. (Adapted from A. Leonard, C. Daraio, *Phys. Rev. Lett.* **108**, 214301, 2012.)

dispersion than such waves in linear materials. For another, the propagation of such waves can be tuned to ultraslow speeds, a characteristic suitable for impact mitigation and other applications.

As we have discussed, a short input signal can lead to a solitary wave, but a longer input—or, alternately, a force from a particle whose mass far exceeds that of the particles in the chain—can produce a shock wave. The spatial profile of a shock wave is that of a solitary wave but with a trailing wave train whose modulated amplitudes eventually decay to a constant. Theoretical and experimental methods have been used to study shock waves in both homogeneous and heterogeneous granular chains.[9] Additionally, to determine the merits of granular crystals as potential shock absorbers in armor and sports helmets, several groups are now analyzing numerical simulations and experiments to understand the materials' response to large-amplitude blasts and other threats.

Breathers and bands

Since the late 1990s, experimentalists have observed discrete breathers in such disparate physical systems as magnetic solids, Josephson junctions, and laser-induced photonic crystals. In 2010, we and several colleagues proposed and experimentally demonstrated that these spatially localized waves can be excited in compressed diatomic chains of alternating steel and aluminum spheres.[5] In this setting, the precompression is crucial for the breathers to exist. Small-amplitude Fourier modes associated with a linear dispersion relation can propagate through a granular crystal's acoustic and optical frequency bands. The signature of a discrete breather is the emergence of sustained oscillations at a frequency that lies inside the band gap of the system's Fourier spectrum, accompanied by exponential localization in space.

As illustrated in figure 2c, each particle in a chain oscillates, or breathes, by itself with an amplitude that decreases exponentially from the central particle. The panel shows (nearly) delocalized waveforms at the bandgap edges and localized waveforms inside the gap.

Granular-crystal breathers have earned the attention of the materials engineering community because of their potential for focusing and harvesting mechanical vibrations. The nonlinear interactions of a crystal, which allow it to confine noise or mechanical vibrations in a specific frequency range and in a specific location, make it an ideal material for converting signals into electrical current that could drive small sensors or transmitters with microwatt to milliwatt levels of power.

Although wave propagation through granular crystals has been studied most thoroughly in one dimension, coherent structures in 2D and 3D crystals have also been investigated. The higher-dimensional systems provide additional freedom for controlling the propagation directions of nonlinear waves. For instance, the wavefront of a wave in a 2D granular array can be changed radically by selecting different particle-packing geometries or material arrangements of the same packing geometry. For the case shown in figure 3, in which cylinders are embedded within the interstitial spaces of an array of spheres, the lateral stress propagation depends on the mass and elasticity of both types of particles.

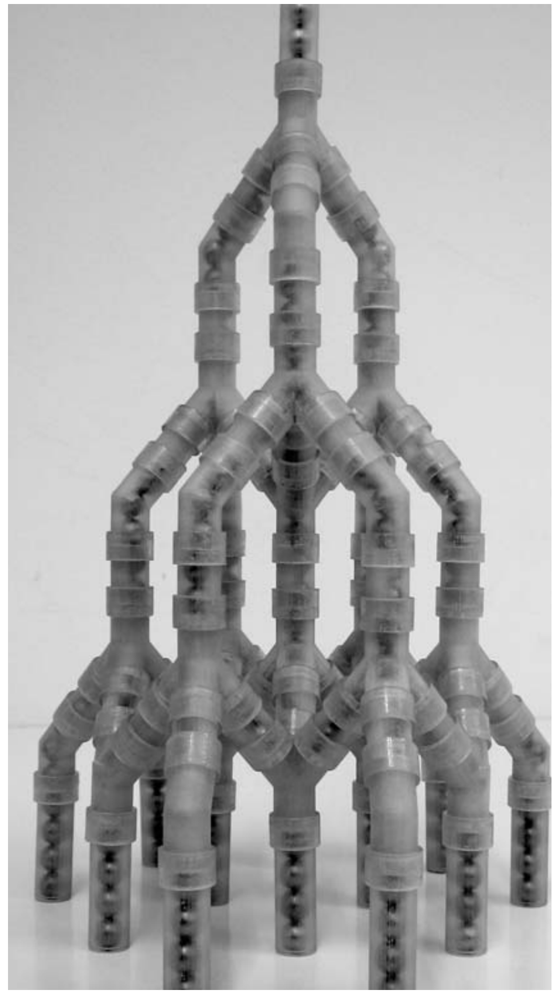


Figure 4. This impact-dispersing granular network consists of branching one-dimensional chains of stainless steel spheres, each 1 cm in diameter, contained in a surrounding polymer matrix.[12] The 1D chains in each branch support the formation and propagation of solitary waves, which are partially reflected and partially split at each branching point. The result is an exponential decay of the wave in both the lateral and vertical orientation of the photograph. (Courtesy of Andrea Leonard.)

Progress toward applications

Granular crystals assembled from particles with diameters on the scale of centimeters respond to input signals between 1 Hz and 20 000 Hz, the sonic range relevant for sound barriers, shock-protection layers, and underwater sonar devices. Using acoustic waves in other common engineering applications—including the nondestructive evaluation of solids, ultrasonic medical imaging, and surgery—calls for frequencies on the order of a megahertz. To achieve such frequencies, the sizes of component particles have to be reduced to the micron scale.

Preliminary studies on micron-scale crystals re-

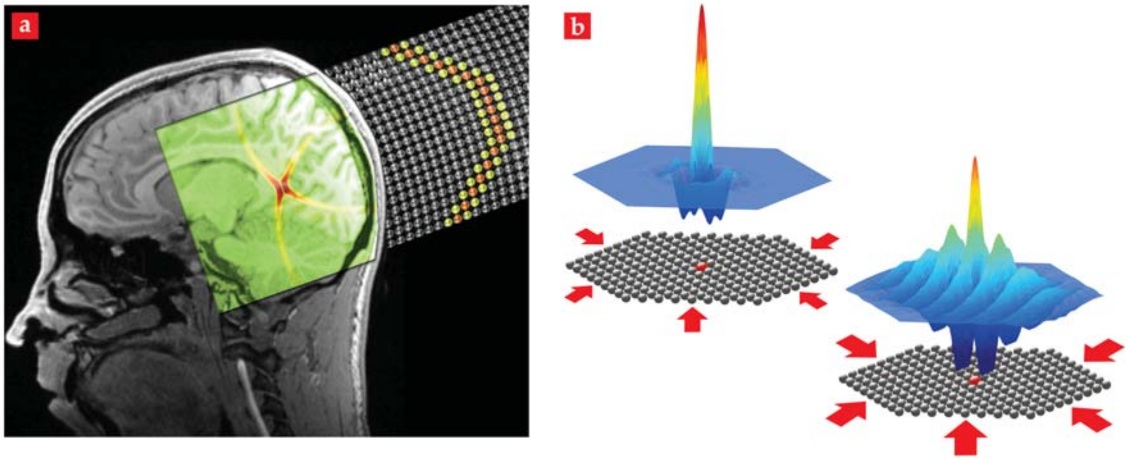


Figure 5. Tunable precompression. (a) An acoustic lens can be made from a granular crystal such as the two-dimensional array of spheres illustrated here. In this fanciful depiction, an incident acoustic signal generates traveling waves that, provided the crystal is properly clamped, experience phase delays and constructively interfere to focus the signal into a highly localized pulse known as a sound bullet. The focused pulse is superimposed on a brain MRI scan to suggest possible medical applications. **(b)** In a hexagonal crystal array containing a single, small-mass defect (red), one can tune the particles' oscillations near the defect from localized to delocalized by varying the extent of precompression (red arrows) at the crystal boundaries.[17] (Panel a courtesy of Alessandro Spadoni and Mike Tysazka; panel b adapted from ref. 17.)

veal that as the components are miniaturized, the contact dynamics become increasingly difficult to control and increasingly sensitive to particle imperfections and environmental conditions. Moreover, the interactions between microscopic particles are no longer adequately modeled as exclusively Hertzian. The particles can be sensitive to electrostatic forces, adhesive forces, asperities, hydrodynamic effects (or more specifically, changes to the particles' elasticity from fluid pressure), and surface charges.

The insights gained from prototype granular systems are starting to affect practical applications beyond the laboratory. Recent studies have demonstrated the feasibility of nonlinear acoustic devices that are analogous to electronic devices. To make an acoustic rectifier, for example, consider a granular chain that is homogeneous except for one less-massive particle close to its left boundary.[10] The oddball particle's presence produces a localized wave at frequency f_{defect} that decays exponentially around the light-mass defect. A driving force applied to the right end of the chain with a frequency f_{drive} near f_{defect} in the gap above the acoustic passband produces a wave that cannot propagate leftward. The same force applied to the left end, however, produces a wave that interferes with the defect mode and produces a linear combination of the respective frequencies, $c_1 f_{\text{drive}} \pm c_2 f_{\text{defect}}$, where c_1 and c_2 are integers. Some of those frequencies lie in the passband and are thus transmitted through the chain.

Last year, two of us (Kevrekidis and Daraio) and colleagues proposed that acoustic switches and logic elements could be developed from a similar approach.[11] Unlike the rectifier, a switch works on the basis of two input signals: the first, at a frequency in the chain's bandgap, acts as the input; and the second, at a frequency in the passband, acts as a control. Combinations of the two signals transmit the input; however, when the control signal is turned off, the input signal is not transmitted. Acoustic analogues of AND and OR gates work on the same principle using dual control signals. On and off excitation states correspond to Boolean states 1 and 0, respectively.

Quasi-1D systems that consist of interconnected chains of particles can serve as efficient load bearers to mitigate both vibrations and impacts. The granular network in figure 4, in which a 1D chain branches into a root-like configuration of several limbs, is a good example. The branches fragment stress waves spatially and temporally. An initial pulse is first broken into a train of solitary waves that travel at different velocities. The wave train is then partially reflected and partially split at the branches. As a result, the waves' amplitudes decay exponentially in both propagation and lateral directions.[12]

Ordered 2D and 3D arrangements of granular crystals can also be used as acoustic lenses to focus energy for applications like noninvasive surgery, outlined schematically in figure 5a; damage detection in materials; and underwater imaging. Very recent demonstrations of underwater focusing and imaging have moved the technology a step closer to practical use.[13] The idea in an acoustic lens is to precompress a granular array differentially, such that the phase velocity of incoming acoustic waves is delayed in some parts of the array. Compact acoustic pulses, known as sound bullets, form from the constructive interference of incident waves at the desired focal point in an object of interest. The dramatic focusing effect from the coalescence of waves in a granular crystal can deliver pulses with energies that are orders of magnitude larger than lenses made from other materials.

Recent work on hexagonally packed 2D crystals has revealed another advantage of tunable compression—the ability to switch between localized and delocalized energy states near a defect,[14] as illustrated in figure 5b.

On the horizon

Research in the fundamental physics and emerging applications of granular crystals is flourishing, and many developments are in progress. Disordered configurations of particles, for instance, enable one to study strongly nonlinear generalizations of phenomena like Anderson localization. (For a primer on Anderson localization, see the article by Ad Lagendijk, Bart van Tiggelen, and Diederik Wiersma, PHYSICS TODAY, August 2009, page 24.) Another major challenge is the study of metamaterial lattices, whose particles have their own structure—containing internal or external masses—which dramatically alters a granular crystal's band structure and resonance frequencies. Still others include miniaturization—the behavior of micron-scale crystals are slowly becoming experimentally accessible—and the need to model the effects of intermolecular adhesive forces and surface roughness on the behavior of granular crystals.

An increasing amount of work is being devoted to 2D and 3D granular crystals, which produce a much richer set of phenomena than 1D crystals. To understand these higher-dimensional phenomena, it is important to consider additional physical effects, such as rotational forces between particles. On the practical side, researchers continue to make progress in developing novel, tunable, and even programmable acoustic materials that might, for example, find their way into industrial production lines as quality-control sensors. Our article just scratches the surface of applications that may mature with time and ingenuity.

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