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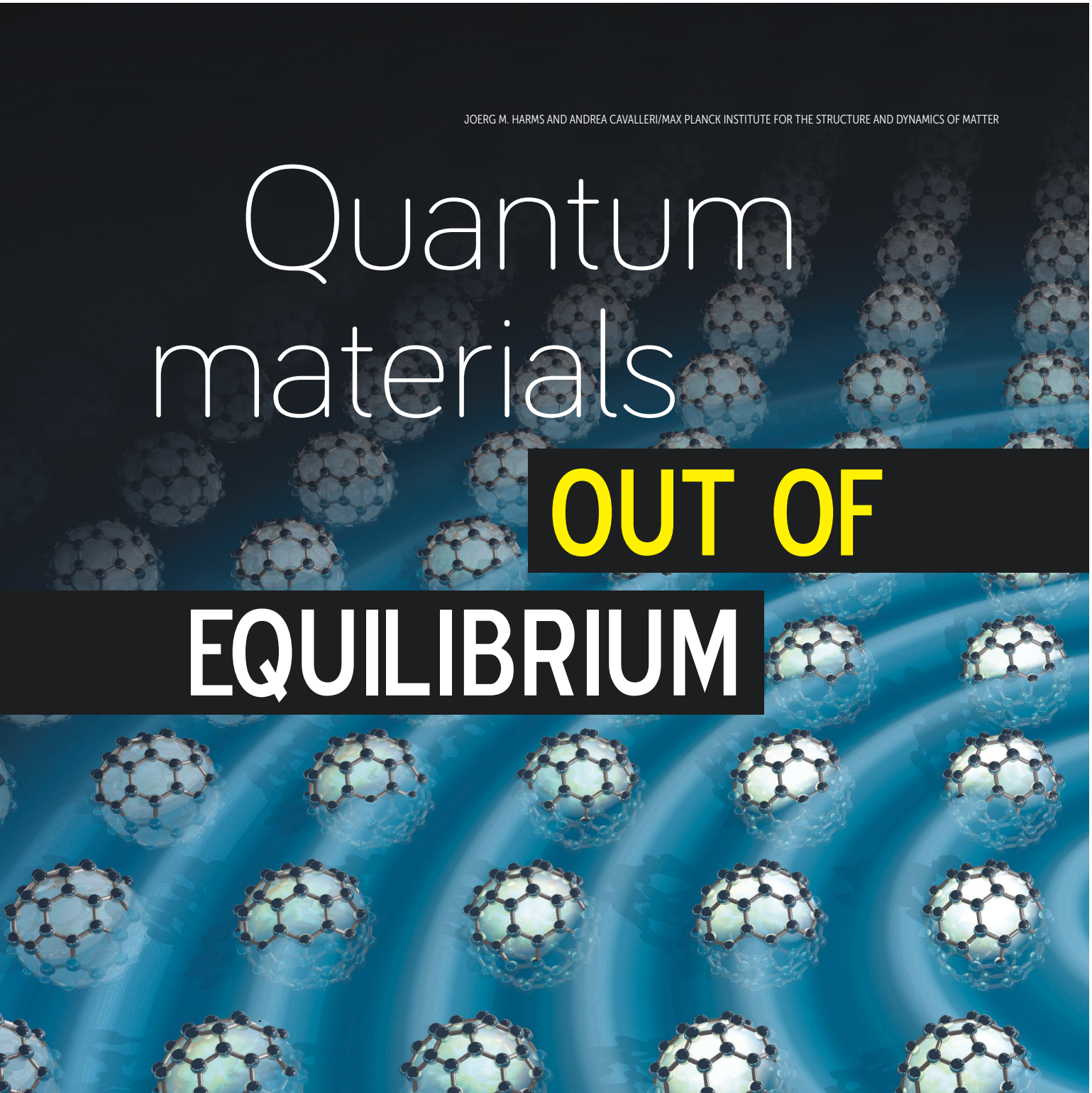

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Illuminating materials with lasers can create intriguing magnetic and topological states of matter.

Martin Rodriguez-Vega, Maia G. Vergniory, and Gregory A. Fiete

To appreciate some of the most interesting quantum phenomena associated with electrons in a material, imagine a flock of birds. Although each individual bird has its own flight pattern, the flock as a whole moves in sync and avoids any collisions. Such collective motion is ubiquitous in nature and is behind a central concept in condensed-matter physics: emergence. It describes how assemblies of objects behave in ways fundamentally different from their individual constituents. Examples include social networks and superconducting materials, whose collective properties come from pairs of electrons. As Nobel laureate Philip Anderson explains in his 1972 article "More is different," understanding that collective behavior is a fundamentally different task than understanding isolated components.[1]

Most theories developed to understand collective behaviors in materials hold only in the presence of equilibrium conditions, such as when energy is conserved. But releasing a physical system from that equilibrium constraint—for example, by introducing a behaviors. In the laboratory, laser light can supply the amount of "shaking" needed for the job.

In this article we discuss the properties of quantum materials in out-of-equilibrium conditions induced by laser irradiation. Remarkably, light pulses from a laser

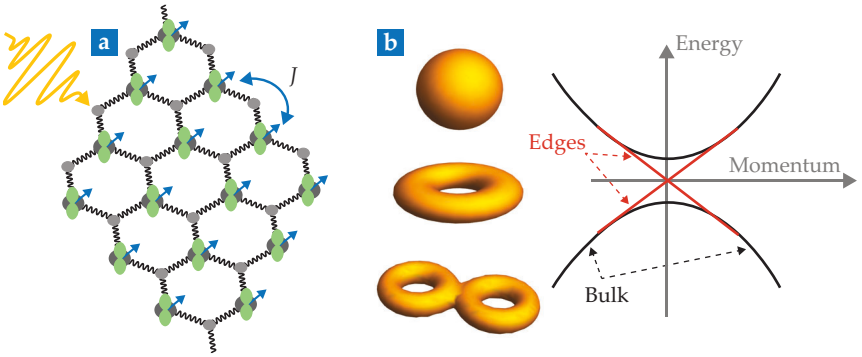


FIGURE 1. A REAL-SPACE illustration of a quantum material **(a)** displays interacting degrees of freedom: electron spins (blue arrows), electron orbitals (green lobes), phonons (black springs), and an incoming laser beam (yellow). The magnetic exchange parameter J reflects the energy of different orientations of two magnetic spins. **(b)** A material's topological aspects are distinguishable by its number of "holes." In an insulator, topology in the electronic states can appear as a protected surface or edge state (shown in red) in the material's energy–momentum diagram.

among electrons, and what kinds of topological properties can emerge? We then discuss how laser irradiation can modify those properties.

Topology, energy bands, and collective modes

Think of a quantum material as a collection of atoms that have interacting electrons and nuclei and that exhibit emergent behavior and topological properties—that is, properties that are impervious to local defects. Figure 1 illustrates that mathematically: A sphere and a torus are topologically distinct. The two objects have different numbers of "holes" in their structures. (A hole can be understood as an obstruction to contracting a loop on the surface.) And only a defect that changes the number of holes in a material's structure can change its topology. (See the article by Arthur Ramirez and Brian Skinner, PHYSICS TODAY, September 2020, page 30.)

Ever since the discovery in 1980 of the integer quantum Hall effect in two-dimensional systems, topological phases have played an important role in the field. With their extension to 3D systems in the late 2000s, topological phases have come to dominate the landscape of solid-state physics. Topology is an intrinsic property of electronic band structure, which describes the energy–momentum relations of electrons in a crystal lattice. Different topological materials exhibit previously unknown phases of matter. For example, many topological materials are insulating in the bulk but conduct an electric current on the

called magnons—and their interactions are fundamental to understanding the material behavior. Instead, one must consider such strongly correlated materials on a case-by-case basis. For example, the Coulomb interactions between electrons can produce insulating materials that even electronic band theory would predict to be metallic.[4] Such interaction-induced insulators are called Mott insulators. They are often described in terms of local moment (spin) models with collective spin excitations and a Hamiltonian,

$$H = \sum_{ij} J\mathbf{S}_i \cdot \mathbf{S}_j,$$

in which J signifies how strongly interacting spins $\mathbf{S}$ are coupled (the magnetic exchange energy), and i and j are space indices that label the positions of the local magnetic moments. Remarkably, in Mott insulators, certain circumstances may favor "fractionalized" topological states in which the electron, an otherwise fundamental particle, is split apart.[5] Such exotic states that combine topology and strong Coulomb interactions between electrons are good examples of emergent collective quantum behavior. (See the article by Philip Anderson, PHYSICS TODAY, October 1997, page 42.)

In the context of nonequilibrium quantum materials, collective modes play an outsized role because they are often the most interesting to selectively excite with a laser. Each collective mode—induced by correlations that result from interactions—possesses different excitation energy scales. Across materials, however, one can discern a range of distinct energies. For example, in insulating materials, the excitation energy of electrons can be measured in electron volts. On the other hand, lattice and spin collective modes are typically less energetic: Phonon energies are in the few tens to low hundreds of millielectron volts, and magnons are in the few millielectron volts or even lower. That energy hierarchy, combined with constraints imposed by the symmetries of the material, opens the door to selective mode excitation in quantum materials. See the box on page 47 for a discussion of crystal symmetries and their influence on phonons.

Among all the possible collective modes, phonons draw special attention because their excitation by laser radiation can modify a crystal's lattice structure and profoundly affect its

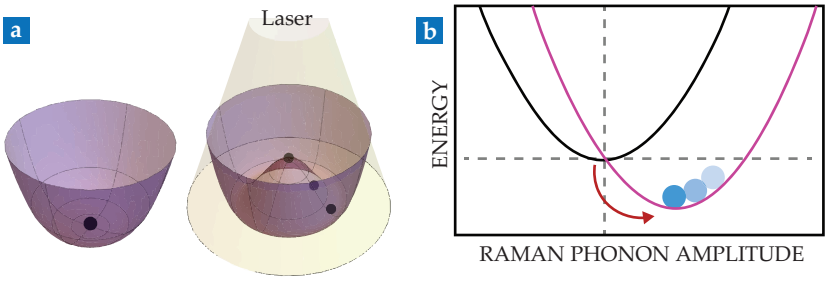


FIGURE 2. ENERGY CONFIGURATION in a material. **(a)** The black dots represent the ground state. The presence of laser light modifies the material's energy configuration and effectively produces a new ground state. **(b)** When a Raman phonon is excited by a laser beam, it shifts the energy minimum, produces a distorted lattice, and elicits new behavior. The blue dots illustrate the relaxation of the material (in progress) to a new energy minimum.

for certain types of phonons as determined by the symmetries of the material. For example, in materials with inversion symmetry—those whose structure is the same for an atom at position r as for an atom at position $-r$—the lattice excitations can be divided into so-called Raman phonons that are even under inversion and IR phonons that are odd under inversion. Only IR phonons directly interact with laser light because they have a finite dipole moment. Raman phonons are "optically silent" at their resonant frequencies. If an IR phonon is excited with strong enough laser light, however, nonlinear interactions between the phonons can excite Raman phonons with fascinating consequences. The mechanism is known as nonlinear phononics.

In their groundbreaking work, Matteo Rini, Andrea Cavalleri, and coauthors showed in 2007 that exciting an IR phonon in the insulating material $Pr_{0.7}Ca_{0.3}MnO_3$ can temporarily distort its orthorhombic lattice.[6] Such lattice modification, in turn, drives the electronic states into a high-conductivity phase that survives for several nanoseconds. The concept of nonlinear phononics was introduced by Cavalleri and coauthors four years later.[7] They showed that one could leverage nonlinear phonon–phonon interactions in $La_{0.7}Sr_{0.3}MnO_3$—an insulating perovskite with a rhombohedral distortion—to manipulate the crystal lattice and electronic structure. They demonstrated the electronic structure change, induced by phonon pumping with laser light, by using time-resolved reflectivity. The nonlinear mechanism prompts phonons to oscillate around new equilibrium positions, as shown in figure 2. From that early study, examples of the types of order that researchers can control in materials have grown rapidly.

Ferroelectrics compose an important class of quantum ma-

a new ultrafast switching route for ferroelectrics.

In the search for a longer-lived effect in ferroelectric order, Cavalleri's team also considered strontium titanate, an incipient ferroelectric.[9] As a function of applied strain, the compound transforms into a ferroelectric state. The lower the temperature, the lower the strain required for the transition. In their experiment, the researchers irradiated the sample with 20 THz femtosecond laser pulses in resonance with an IR phonon and tracked the time-dependent second-harmonic-generation signal. Their measurements revealed that the light-induced ferroelectric phase lasted for hours.

Irradiating magnets

Magnetism is perhaps the correlated electronic state people encounter most in everyday life. Storage and manipulation of information is one of the most relevant applications of magnetism in modern society. To that end, controlling the magnetic order by means other than magnetic fields has intrinsic appeal. It can increase information processing speeds and decrease device sizes. Nonlinear phononics could provide such a path, and recent experiments have already shown encouraging results.

Ankit Disa and colleagues considered the antiferromagnet cobalt fluoride.[10] The crystal's piezomagnetism is what makes it so intriguing. Under applied stress, the crystal acquires a net magnetic moment. Figure 3 shows a representation of the individual magnetic moments. In their experiments, the researchers identified a suitable phonon that could mimic the effect of the applied stress. That phonon, however, could not be excited directly by a laser beam. The researchers excited it indirectly using a nonlinear phonon process that simultaneously excites two other degenerate IR phonons. From Faraday rotation and circular dichroism measurements, they extracted the magnetic moment as a function of time. And that moment turned out to reach values two orders of magnitude larger than what can be achieved in equilibrium.

Dmytro Afanasiev and colleagues examined dysprosium ferrite ($DyFeO_3$) as a function of temperature.[11] The material is a complex magnetic system with multiple magnetic phases. Driving an IR phonon in its lattice to large amplitude can nonlinearly excite a Raman mode that modifies the magnetic interactions between the transition metal and rare-earth ions. Consequently, Afanasiev and coworkers achieved an antiferromagnetic-to-weakly-ferromagnetic phase transition.

Andrzej Stupakiewicz and colleagues achieved an elusive

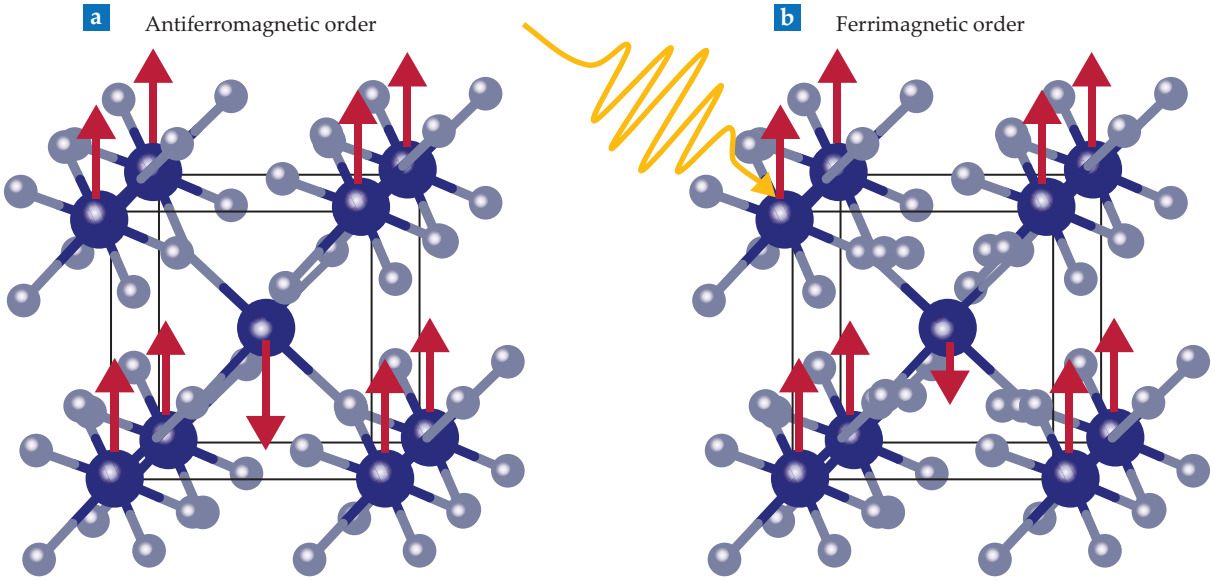


FIGURE 3. AN ANTIFERROMAGNET becomes ferrimagnetic under laser light. **(a)** Cobalt fluoride is normally antiferromagnetic at equilibrium, with magnetic moments (red arrows) that are spin up at corner sites and spin down in the center of its unit cell. The net magnetic moment is zero. **(b)** In its laser-induced distorted lattice, CoF_2 becomes ferrimagnetic, with magnetic moments that are both spin up and spin down, but whose net magnetic moment is now finite. The displaced atoms (relative to the antiferromagnetic lattice) produce that net moment.

phonon control of topological states. James McIver and coauthors used ultrafast laser pulses to induce a more general phenomenon, an anomalous quantum Hall state in graphene[13]—an exotic state of matter in which conducting states appear on the edge of the material and the bulk is insulating.

Unlike the examples we just discussed, graphene is a gapless semiconductor (see the article by Andrey Geim and Allan MacDonald, PHYSICS TODAY, August 2007, page 35). As such, electronic excitations are unavoidable when phonons are targeted, which complicates the picture of photon absorption. Phonon control of topological states, however, could be available in the insulating antiferromagnet $MnBi_2Te_4$ and related materials, in which magnetic order and topology are intertwined.[14] Thus it may only be a short time before we have experimental examples of phonon-induced electronic topological states.

Theoretical aspects

All the experiments described above have motivated the research community to redouble their efforts to understand quantum materials out of equilibrium and to devise theoretical tools to

energy potential, including the driving term introduced by the laser interaction with the IR phonons. The solutions describe the transient lattice distortions. First-principles calculations help to determine the new electronic ground state provided the electrons relax much faster than the lattice does and follow that relaxation adiabatically. Besides providing the theory for the experiments discussed earlier, the approach has produced predictions that await experimental verification.[15]

More accurate simulations of light-induced effects in quantum materials could be achieved by using frameworks that are based on time-dependent density functional theory[16] or extensions of it. Current theoretical efforts, such as the Octopus project at the Flatiron Institute in New York City and the SALMON project at the University of Tsukuba in Japan, are focused on the real-space computation of light-matter interactions. Those first-principles approaches could, at least in principle, capture the dynamics of all the relevant interacting degrees of freedom and provide accurate descriptions of light-induced effects with increased predictive power. Such schemes are computationally expensive, though, and limited by system sizes and simulation

PHONONS IN GROUP THEORY

Symmetry is one of the most fundamental concepts in physics. It is also quite useful. The symmetry of a crystal lattice in its equilibrium state determines the symmetries of the lattice's vibrational modes, even when the crystal is out of equilibrium. Selectively exciting specific modes with a laser can tailor the topology of a material's electronic band structure and manipulate its magnetic states. The figure illustrates the equilibrium lattice symmetries of chromium triiodide (CrI_3), a two-dimensional magnetic insulator at low temperatures. Blue dots represent chromium atoms, and purple dots iodine atoms. The CrI_3 lattice has twofold rotational symmetry about the center of a chromium hexagonal lattice unit because of the lower symmetry arrangement of the iodine atoms.

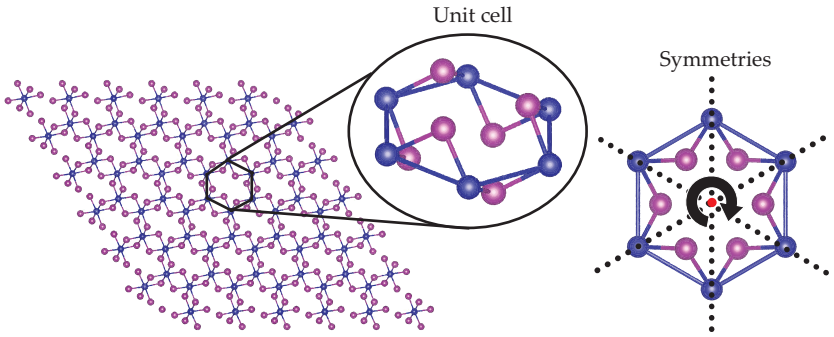
The application of symmetry to physics has far-reaching implications and is much broader than lattice vibrations. In her 1918 work "Invariant variation problems," Emmy Noether showed that symmetries are mathematically associated with conserved physical quantities. The conservation of energy, for example, is associated with the time-translational symmetry of physical laws derived from time-independent Hamiltonians.

Each quantum material is characterized by its own set of crystal symmetries, which are the mathematical operations that when performed on a material structure leave it unaltered. Translations and rotations are examples. (See the 2008 book *Group Theory: Application to the Physics of Condensed Matter*, by Mildred Dresselhaus, Gene Dresselhaus, and Ado Jorio, for a pedagogical introduction.)

The collection of crystal symmetries defines the space group of the material, which includes the translational symmetries. Each material is constructed by repeating its unit cell. Typically, the space group of a material is established through a combination of group-theory studies and x-ray or neutron scattering experiments.

Once the material symmetries are established, one can determine the phonons' symmetries. In the case of phonons excited by light, the discussion can be restricted to long-wavelength phonons. Mathematically, one need consider only point groups. For example, the point group of monolayer CrI_3 is D_{3d}, one of the 32 point groups in three spatial dimensions.

Group theory can also help block-diagonalize the dynamical matrix, which determines the real-space atom motions for each phonon type. That is done by constructing the projection operators for each phonon type. Those displacements do not correspond directly to the actual phonons but form a basis for them. To determine the energetics of the phonons, one must employ density functional theory.



by-case basis. For example, recent experiments by Stephen Wilson's group at the University of California, Santa Barbara, showed that CsV_3Sb_5 has topological electronic bands in its normal state. At low enough temperatures (around 2.5 K), however, it becomes a superconductor. The next step might be to induce dynamical transitions between the material's topological and superconducting states by using nonlinear phononics.

The integration of advanced, time-dependent density functional theory calculations with machine-learning approaches could provide a path to extend the time scales and system sizes that researchers can simulate. Molecular dynamics and spin control of a lattice by phonon manipulation promises to unlock new functionalities from materials in out-of-equilibrium contexts, in which new phases of matter could emerge. Shaken is indeed different.

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