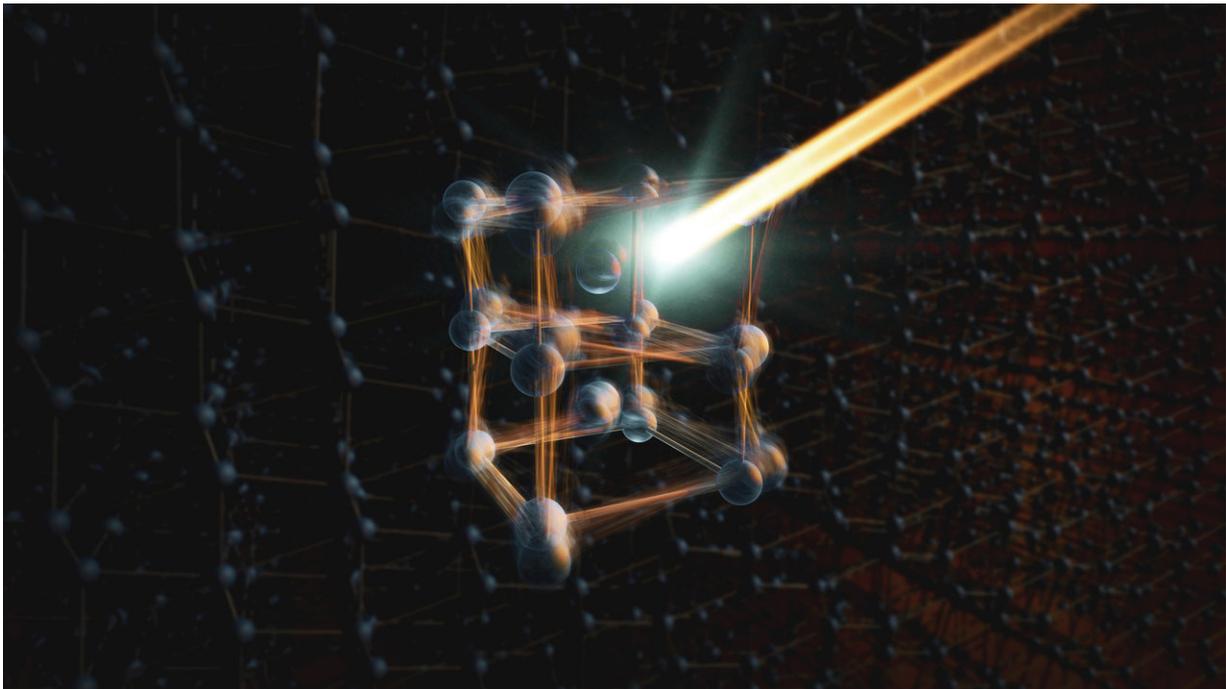


In materials hit with light, individual atoms and vibrations take disorderly paths

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Two studies with a new X-ray laser technique reveal for the first time how individual atoms and vibrations respond when a material is hit with light. Their surprisingly unpredictable behavior has profound implications for designing and controlling materials. Credit: Greg Stewart/SLAC National Accelerator Laboratory

Hitting a material with laser light sends vibrations rippling through its latticework of atoms, and at the same time can nudge the lattice into a

new configuration with potentially useful properties – turning an insulator into a metal, for instance.

Until now, scientists assumed this all happened in a smooth, coordinated way. But two new studies show it doesn't: When you look beyond the average response of atoms and vibrations to see what they do individually, the response, they found, is disorderly.

Atoms don't move smoothly into their new positions, like band members marching down a field; they stagger around like partiers leaving a bar at closing time.

And laser-triggered vibrations don't simply die out; they trigger smaller vibrations that trigger even smaller ones, spreading out their energy in the form of heat, like a river branching into a complex network of streams and rivulets.

This unpredictable behavior at a tiny scale, measured for the first time with a new X-ray laser technique at the Department of Energy's SLAC National Accelerator Laboratory, will have to be taken into account from now on when studying and designing new materials, the researchers said – especially quantum materials with potential applications in sensors, smart windows, energy storage and conversion and super-efficient electrical conductors.

Two separate international teams, including researchers at SLAC and Stanford University who developed the technique, reported the results of their experiments Sept. 20 in *Physical Review Letters* and today in *Science*.

"The disorder we found is very strong, which means we have to rethink how we study all of these materials that we thought were behaving in a uniform way," said Simon Wall, an associate professor at the Institute of

Photonic Sciences in Barcelona and one of three leaders of the study reported in *Science*. "If our ultimate goal is to control the behavior of these materials so we can switch them back and forth from one phase to another, it's much harder to control the drunken choir than the marching band."

Lifting the haze

The classic way to determine the atomic structure of a molecule, whether from a manmade material or a human cell, is to hit it with X-rays, which bounce off and scatter into a detector. This creates a pattern of bright dots, called Bragg peaks, that can be used to reconstruct how its atoms are arranged.

SLAC's Linac Coherent Light Source (LCLS), with its super-bright and ultrafast X-ray [laser pulses](#), has allowed scientists to determine atomic structures in ever more detail. They can even take rapid-fire snapshots of chemical bonds breaking, for instance, and string them together to make "molecular movies."

About a dozen years ago, David Reis, a professor at SLAC and Stanford and investigator at the Stanford Institute for Materials and Energy Sciences (SIMES), wondered if a faint haze between the bright spots in the detector – 10,000 times weaker than those bright spots, and considered just background noise – could also contain important information about rapid changes in materials induced by laser pulses.

He and SIMES scientist Mariano Trigo went on to develop a technique called "ultrafast diffuse scattering" that extracts information from the haze to get a more complete picture of what's going on and when.

The two new studies represent the first time the technique has been used to observe details of how energy dissipates in materials and how light

triggers a transition from one phase, or state, of a material to another, said Reis, who along with Trigo is a co-author of both papers. These responses are interesting both for understanding the basic physics of materials and for developing applications that use light to switch the properties of materials on and off or convert heat to electricity, for instance.

"It's sort of like astronomers studying the night sky," said Olivier Delaire, an associate professor at Duke University who helped lead one of the studies. "Previous studies could only see the brightest stars visible to the naked eye. But with the ultrabright and ultrafast X-ray pulses, we were able to see the faint and diffuse signals of the Milky Way galaxy between them."

Tiny bells and piano strings

In the study published in *Physical Review Letters*, Reis and Trigo led a team that investigated vibrations called phonons that rattle the atomic lattice and spread heat through a material.

The researchers knew going in that phonons triggered by laser pulses decay, releasing their energy throughout the atomic lattice. But where does all that energy go? Theorists proposed that each phonon must trigger other, smaller phonons, which vibrate at higher frequencies and are harder to detect and measure, but these had never been seen in an experiment.

To study this process at LCLS, the team hit a thin film of bismuth with a pulse of optical laser light to set off phonons, followed by an X-ray laser pulse about 50 quadrillionths of a second later to record how the phonons evolved. The experiments were led by graduate student Tom Henighan and postdoctoral researcher Samuel Teitelbaum of the Stanford PULSE Institute.

For the first time, Trigo said, they were able to observe and measure how the initial phonons distributed their energy over a wider area by triggering smaller vibrations. Each of those small vibrations emanated from a distinct patch of atoms, and the size of the patch – whether it contained 7 atoms, or 9, or 20 – determined the frequency of the vibration. It was much like how ringing a big bell sets smaller bells tinkling nearby, or how plucking a piano string sets other strings humming.

"This is something we've been waiting years to be able to do, so we were excited," Reis said. "It's a measurement of something absolutely fundamental to modern solid-state physics, for everything from how heat flows in materials to even, in principle, how light-induced superconductivity emerges, and it could not have been done without an X-ray free-electron laser like LCLS."

A disorderly march

The paper in *Science* describes LCLS experiments with vanadium dioxide, a well-studied material that can flip from being an insulator to an electrical conductor in just 100 quadrillionths of a second.

Researchers already knew how to trigger this switch with very short, ultrafast pulses of laser light. But until now they could only observe the average response of the atoms, which seemed to shuffle into their new positions in an orderly way, said Delaire, who led the study with Wall and Trigo.

The new round of diffuse scattering experiments at LCLS showed otherwise. By hitting the vanadium dioxide with an optical laser of just the right energy, the researchers were able to trigger a substantial rearrangement of the vanadium atoms. They did this more than 100 times per second while recording the movements of individual atoms

with the LCLS X-ray laser. They discovered that each atom followed an independent, seemingly random path to its new lattice position. Computer simulations by Duke graduate student Shan Yang backed up that conclusion.

"Our findings suggest that disorder may play an important role in some materials," the team wrote in the *Science* paper. While this may complicate efforts to control the way [materials](#) shift from one phase to another, they added, "it could ultimately provide a new perspective on how to control matter," and even suggest a new way to induce superconductivity with light.

In a commentary accompanying the report in *Science*, Andrea Cavalleri of Oxford University and the Max Planck Institute for the Structure and Dynamics of Matter said the results imply that molecular movies of [atoms](#) changing position over time don't paint a complete picture of the microscopic physics involved.

He added, "More generally, it is clear from this work that X-ray free electron lasers are opening up far more than what was envisaged when these machines were being planned, forcing us to reevaluate many old notions taken for granted up to now."

More information: Samuel W. Teitelbaum et al. Direct Measurement of Anharmonic Decay Channels of a Coherent Phonon, *Physical Review Letters* (2018). [DOI: 10.1103/PhysRevLett.121.125901](https://doi.org/10.1103/PhysRevLett.121.125901)

Simon Wall et al. Ultrafast disordering of vanadium dimers in photoexcited VO₂, *Science* (2018). [DOI: 10.1126/science.aau3873](https://doi.org/10.1126/science.aau3873)

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