X-ray laser glimpses how electrons dance with atomic nuclei in materials

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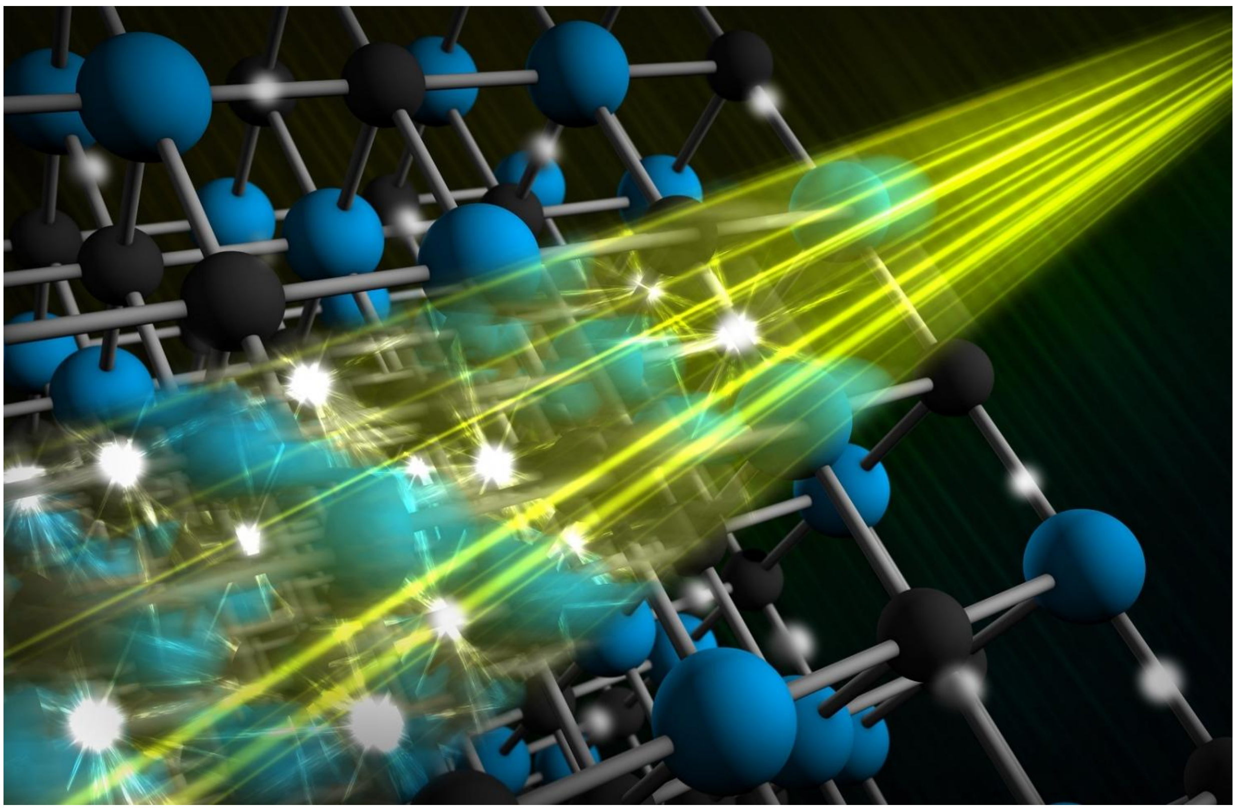


An illustration shows how laser light excites electrons (white spheres) in a solid material, creating vibrations in its lattice of atomic nuclei (black and blue spheres). SLAC's LCLS X-ray laser reveals the ultrafast "dance" between electrons and vibrations that accounts for many important properties of materials. Credit: SLAC National Accelerator Laboratory

From hard to malleable, from transparent to opaque, from channeling electricity to blocking it: Materials come in all types. A number of their intriguing properties originate in the way a material's electrons "dance" with its lattice of atomic nuclei, which is also in constant motion due to vibrations known as phonons.

This coupling between electrons and phonons determines how efficiently solar cells convert sunlight into electricity. It also plays key roles in superconductors that transfer electricity without losses, topological insulators that conduct electricity only on their surfaces, materials that drastically change their electrical resistance when exposed to a magnetic field, and more.

At the Department of Energy's SLAC National Accelerator Laboratory, scientists can study these coupled motions in unprecedented detail with the world's most powerful X-ray laser, the Linac Coherent Light Source (LCLS). LCLS is a DOE Office of Science User Facility.

"It has been a long-standing goal to understand, initiate and control these unusual behaviors," says LCLS Director Mike Dunne. "With LCLS we are now able to see what happens in these materials and to model complex electron-phonon interactions. This ability is central to the lab's mission of developing new materials for next-generation electronics and energy solutions."

LCLS works like an extraordinary strobe light: Its ultrabright X-rays take snapshots of materials with atomic resolution and capture motions as fast as a few femtoseconds, or millionths of a billionth of a second. For comparison, one femtosecond is to a second what seven minutes is to the age of the universe.

Two recent studies made use of these capabilities to study electron-phonon interactions in lead telluride, a material that excels at converting heat into electricity, and chromium, which at low temperatures has peculiar properties similar to those of high-temperature superconductors.

Turning Heat into Electricity and Vice Versa

Lead telluride, a compound of the chemical elements lead and tellurium, is of interest because it is a good thermoelectric: It generates an electrical voltage when two opposite sides of the material have different temperatures.

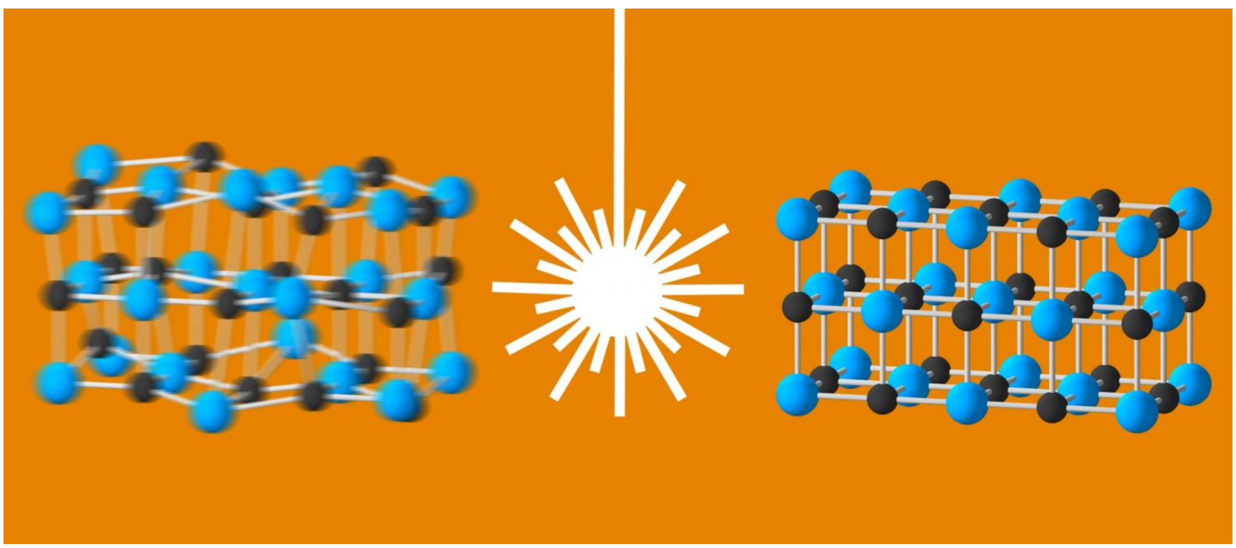
"This property is used to power NASA space missions like the Mars rover Curiosity and to convert waste heat into electricity in high-end cars," says Mariano Trigo, a staff scientist at the Stanford PULSE Institute and the Stanford Institute for Materials and Energy Sciences (SIMES), both joint institutes of Stanford University and SLAC. "The effect also works in the opposite direction: An electrical voltage applied across the material creates a temperature difference, which can be exploited in thermoelectric cooling devices."

Mason Jiang, a recent graduate student at Stanford, PULSE and SIMES, says, "Lead telluride is exceptionally good at this. It has two important qualities: It's a bad thermal conductor, so it keeps heat from flowing from one side to the other, and it's also a good electrical conductor, so it can turn the temperature difference into an electric current. The coupling between lattice vibrations, caused by heat, and electron motions is therefore very important in this system. With our study at LCLS, we wanted to understand what's naturally going on in this material."

In their experiment, the researchers excited electrons in a lead telluride sample with a brief pulse of infrared laser light, and then used LCLS's X-rays to determine how this burst of energy stimulated lattice vibrations.

"Lead telluride sits at the precipice of a coupled electronic and structural transformation," says principal investigator David Reis from PULSE, SIMES and Stanford. "It has a tendency to distort without fully transforming – an instability that is thought to play an important role in its thermoelectric behavior. With our method we can study the forces involved and literally watch them change in response to the infrared laser pulse."

The scientists found that the light pulse excites particular electronic states that are responsible for this instability through electron-phonon coupling. The excited electrons stabilize the material by weakening certain long-range forces that were previously associated with the material's low thermal conductivity.



This illustration shows the arrangement of lead and tellurium atoms in lead telluride, an excellent thermoelectric that efficiently converts heat into electricity and vice versa. In its normal state (left), lead telluride's structure is distorted and has a relatively large degree of lattice vibrations (blurring). When scientists hit the sample with a laser pulse, the structure became more ordered (right). The results elucidate how electrons couple with these distortions – an interaction that is crucial for lead telluride's thermoelectric properties. Credit: SLAC National

"The light pulse actually walks the material back from the brink of instability, making it a worse thermoelectric," Reis says. "This implies that the reverse is also true – that stronger long-range forces lead to better thermoelectric behavior."

The researchers hope their results, published July 22 in *Nature Communications*, will help them find other thermoelectric materials that are more abundant and less toxic than lead telluride.

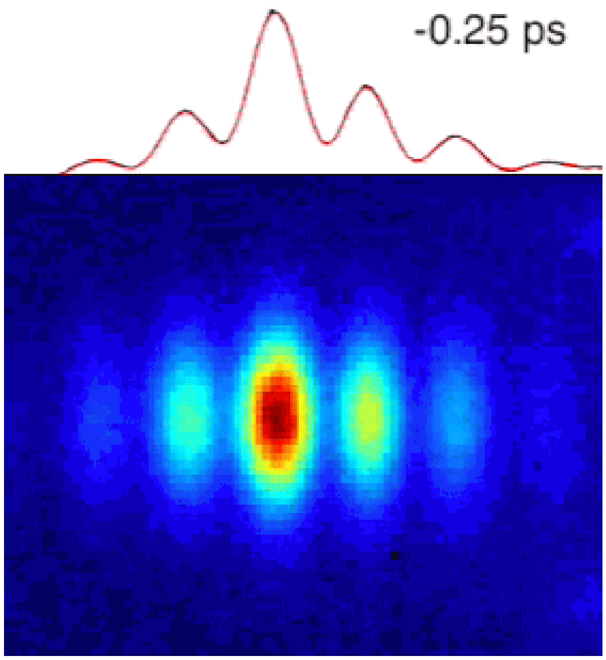
Controlling Materials by Stimulating Charged Waves

The second study looked at charge density waves – alternating areas of high and low electron density across the nuclear lattice – that occur in materials that abruptly change their behavior at a certain threshold. This includes transitions from insulator to conductor, normal conductor to superconductor, and from one magnetic state to another.

These waves don't actually travel through the material; they are stationary, like icy waves near the shoreline of a frozen lake.

"Charge density waves have been observed in a number of interesting materials, and establishing their connection to material properties is a very hot research topic," says Andrej Singer, a postdoctoral fellow in Oleg Shpyrko's lab at the University of California, San Diego. "We've now shown that there is a way to enhance charge density waves in crystals of chromium using laser light, and this method could potentially also be used to tweak the properties of other materials."

This could mean, for example, that scientists might be able to switch a material from a normal conductor to a superconductor with a single flash of light. Singer and his colleagues reported their results on July 25 in *Physical Review Letters*.



This movie shows how a laser pulse hitting a chromium crystal causes charge density waves – alternating areas of high and low electron density within the crystal – to oscillate in height, or amplitude. The signal seen here is made by X-ray laser pulses scattering off the crystal. The timescale of the oscillations is shown in picoseconds, or trillionths of a second. Credit: A. Singer/University of California, San Diego

The research team used the chemical element chromium as a simple model system to study charge density waves, which form when the crystal is cooled to about minus 280 degrees Fahrenheit. They stimulated the chilled crystal with pulses of optical laser light and then used LCLS X-ray pulses to observe how this stimulation changed the amplitude, or height, of the charge density waves.

"We found that the amplitude increased by up to 30 percent immediately after the laser pulse," Singer says. "The amplitude then oscillated, becoming smaller and larger over a period of 450 femtoseconds, and it kept going when we kept hitting the sample with laser pulses. LCLS provides unique opportunities to study such process because it allows us to take ultrafast movies of the related structural changes in the lattice."

Based on their results, the researchers suggested a mechanism for the amplitude enhancement: The light pulse interrupts the electron-phonon interactions in the material, causing the lattice to vibrate. Shortly after the pulse, these interactions form again, which boosts the amplitude of the vibrations, like a pendulum that swings farther out when it receives an extra push.

A Bright Future for Studies of the Electron-Phonon Dance

Studies like these have a high priority in solid-state physics and materials science because they could pave the way for new materials and provide new ways to control material properties.

With its 120 ultrabright X-ray pulses per second, LCLS reveals the electron-phonon dance with unprecedented detail. More breakthroughs in the field are on the horizon with LCLS-II – a next-generation X-ray laser under construction at SLAC that will fire up to a million X-ray flashes per second and will be 10,000 times brighter than LCLS.

"LCLS-II will drastically increase our chances of capturing these processes," Dunne says. "Since it will also reveal subtle electron-phonon signals with much higher resolution, we'll be able to study these interactions in much greater detail than we can now."

More information: M. P. Jiang et al. The origin of incipient ferroelectricity in lead telluride, *Nature Communications* (2016). DOI: 10.1038/ncomms12291

A. Singer et al. Photoinduced Enhancement of the Charge Density Wave Amplitude, *Physical Review Letters* (2016). DOI: 10.1103/PhysRevLett.117.056401

Provided by SLAC National Accelerator Laboratory