Crystals under Control

To alter properties of materials with light as if with the wave of a magic wand: that is Andrea Cavalleri's mission. The Director at the Max Planck Institute for the Structure and **Dynamics of Matter** in Hamburg uses lasers to change the behavior of crystals, fleetingly producing superconductors that conduct electricity without loss at room temperature.

TEXT ROLAND WENGENMAYR

use light to freeze water," says Andrea Cavalleri, explaining - albeit light-heartedly - the nature of his research to his six-year-old daughter. She's familiar with Princess Elsa from the animated film "Frozen," who uses magic to turn water into ice, so this gives her an idea of what Daddy does as a scientist when he uses light to manipulate matter.

And anyway, the Director at the Max Planck Institute for the Structure and Dynamics of Matter in Hamburg talks about his work so animatedly that it's easy to imagine him explaining it to a rapt group of children. Somewhat older listeners might tend to think of Harry Potter, who can transform things with a wave of his magic wand. As in Harry Potter's magical world, the materials the physicists in Hamburg manipulate retain their new properties for only a limited time - and it is still extremely short.

Cavalleri's magic wand consists of specially prepared laser light. It can briefly shift atoms around in such a

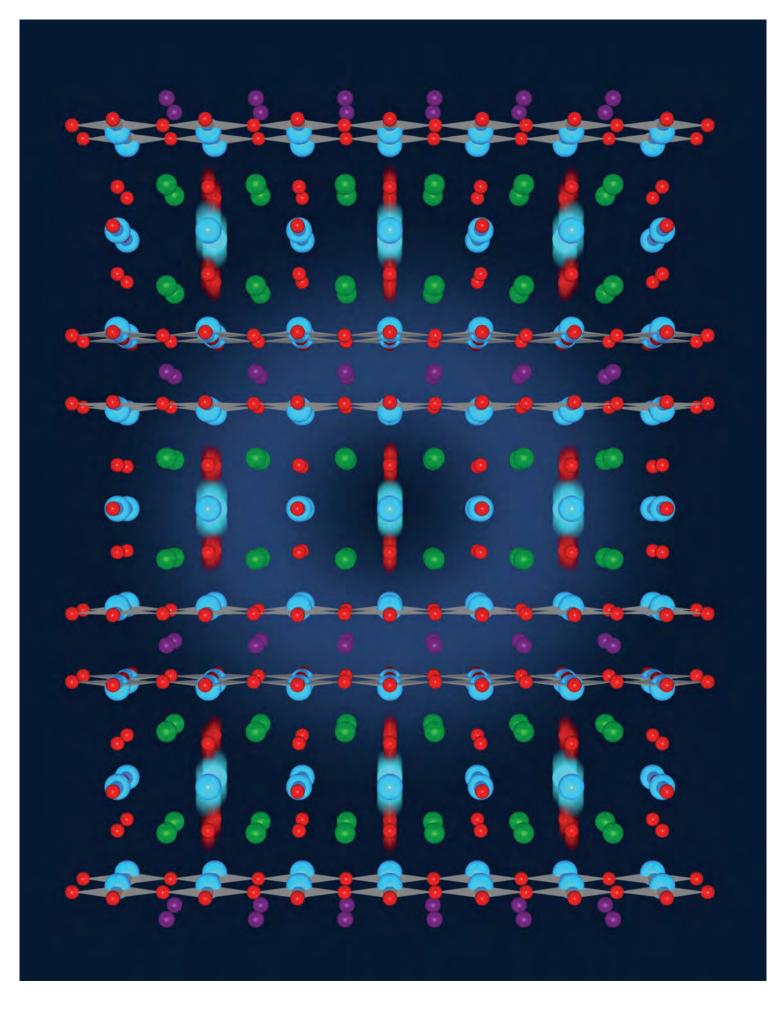
way that it completely alters the physical properties of a material for a brief moment. But water isn't yet on the list of research objects - he's concerned with other substances. In 2014, Cavalleri's team, in cooperation with other groups, achieved a major scientific breakthrough: for several picoseconds, the researchers were able to produce superconductivity - conduction of electricity without resistance - at room temperature. A picosecond is a billionth of a second, so this exotic state lasted for only an ultra-brief period.

THE GOAL: ROOM-TEMPERATURE **SUPERCONDUCTIVITY**

Nevertheless, the discovery caused a sensation. After all, superconductivity at room temperature is the holy grail of materials research. Superconductors that lose their electrical resistance at temperatures well below zero degrees Celsius are already being wound into coils that generate extremely powerful magnetic fields, for example in medical MRI scanners. Zero-resistance power lines that don't require cooling could one day enable electricity to be transported without loss, for example from large-scale wind farms out at sea to remote areas on the mainland. The discoveries and insights of Andrea Cavalleri's team may well lay the foundations for such practical applications.

Before we turn our attention to the Hamburg-based researchers' projects, the two of us delve into the young Institute's history in Cavalleri's small meeting room. The Institute was officially founded on January 1, 2014, but Cavalleri, a young physicist who was already known internationally at that time, had been building it up since early 2008. Before that, the Italian scientist had been conducting research in the US. In 2004 he relocated to England, to the University of Oxford, where he was offered a professorship with tenure in 2006. "I was happy and thought I would never leave Oxford." he says. But an offer from the Max Planck Society to set up a brand-new in-

Light switch for superconductors: In a cuprate of yttrium (purple), barium (green), copper (blue) and oxygen (red), Cooper pairs carry current without loss in closely packed double layers even at relatively high temperatures. When a laser causes the copper atoms to vibrate between the double layers, the layers move closer together, and Cooper pairs are then able to flow perpendicular to them.







A racetrack for electrons: In the XFEL free-electron laser, particles whiz through a 3.4-kilometer-long tunnel between the DESY research center in Hamburg-Bahrenfeld and Schenefeld (top), producing X-ray flashes in the process. Employees travel through the X-ray laser tunnel on bicycles (bottom).

stitute in Hamburg was just too tempting. "I was the first employee," Cavalleri says. "I connected my phone myself at the time."

The Max Planck Institute for the Structure and Dynamics of Matter has grown rapidly since then and has now moved to the premises of the Center for Free-Electron Laser Science (CFEL). The striking three-story flat cylindrical building stands on the grounds of the German Electron Synchrotron (DESY), a research center that was originally dedicated solely to particle physics.

But accelerator technology has also produced brilliant light sources for materials research, and right next door, a very special light source went

into operation in September 2017: the European XFEL is the strongest X-ray laser in the world and is set to play a significant role in Andrea Cavalleri's research. X-ray light is required because its extremely short waves match the distances between atoms in materials. It is the only source that can precisely probe the structure of materials at the atomic level.

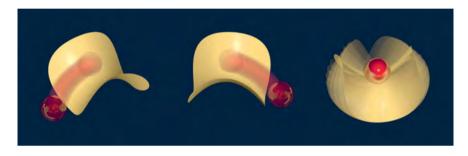
This is the perfect setting for the new Max Planck Institute, which currently consists of three departments in which 110 scientists conduct their research, with Cavalleri's team contributing about 30 people. While we talk, muted construction noise can be heard outside. The Institute's new building is currently being built in the neighborhood and is due to be completed in summer 2019. "I had to deal with a lot of new questions, such as what, exactly, a Max Planck Institute is," Cavalleri says about the founding period. "The Max Planck Society is a very good, highly flexible organization," he says, then adds: "Here you're limited only by your skills. If you fail, you have no one to blame but yourself."

CRYSTAL LATTICE AS A RACETRACK FOR ELECTRONS

Our conversation then turns to Cavalleri's favorite topic: his research. For a layperson, it's important to realize that much of the matter surrounding us consists of crystals. We usually associate crystals with table salt or precious diamonds, but many other materials metals, for instance – are also made up of tiny crystals. In the atomic world, crystals are characterized by a regular spatial order. Table salt, for example, consists of a lattice of myriad tiny cubes with sodium and chlorine atoms alternating at their eight corners.

Electrons are responsible for the quantum connections between the atoms in all crystals. In the case of an electrically conductive metal, the crystal lattice formed by the atoms becomes a three-dimensional racetrack for electrons. Those electrons can flow virtually freely through the crystal, carrying electric current and heat energy. Individual electrons in the crystals of other materials can also act like tiny, rotatable magnetic needles. They are usually then bound to specific atoms in the lattice. When these collectively turn in one direction, the crystal acts as a magnet. Highly complex crystals also exist; they include a group of substances known as cuprates. These copper oxides are known to be high-temperature superconductors and number among Cavalleri's research objects.

The scientist studies the physical properties of crystals and how they can be manipulated with light. The crux of the matter here is that, in every crystal lattice, the properties of the material are determined by the exact arrangement of atoms. "Imagine the atoms in the crystal lattice as spheres that are connected by coil springs," he explains. "You can pull the spheres



A balancing act with atoms: The method by which the Max Planck researchers in Hamburg hope to stabilize a superconducting non-equilibrium state of crystals can be compared to an attempt to hold a ball on a saddle. Normally it would roll off. However, if the saddle is rotating very quickly, it acts like a bowl that traps the ball.

apart at one point, but as soon as you let go, they spring back to their original positions."

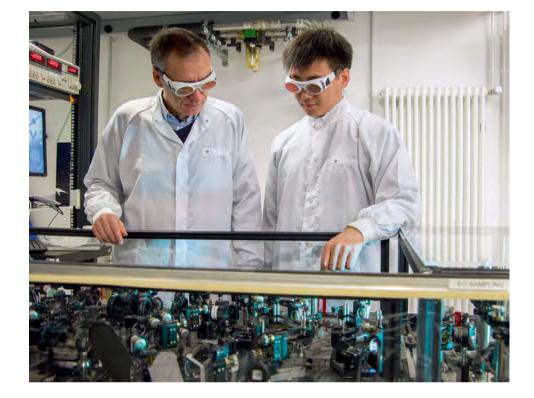
ATOMS IN A **NON-EQUILIBRIUM STATE**

When the spheres are in this equilibrium state, the spring forces all cancel each other out. In a true crystal, as well, the atoms strive to take up equilibrium positions - like marbles rolling into a trough. The equilibrium state therefore determines the crystal's permanent physical properties.

Cavalleri's team is working on methods that can artificially nudge selected atoms in crystals out of their comfortable state of equilibrium. "Our aim is to selectively tug on the springs between the spheres, distort the interatomic bonds and then see what happens," Cavalleri explains. "It's about maneuvering the atoms, at least momentarily, into positions in the crystal where they wouldn't normally remain of their own accord. It's a balancing act." Cavalleri illustrates this on the computer with an animation of a ball on a saddle. When the saddle is at rest, the ball immediately rolls to the side. But if the saddle rotates fast enough, it forms a virtual trough in which the ball remains trapped.

This is precisely what the scientists hope to one day achieve with specially prepared laser light: like the rotating-saddle model, they want to hold atoms within a crystal in a non-equilibrium state, thus changing the crystal's properties. This is Cavalleri's dream, because it would make it possible to alter material properties as long as the material is exposed to laser light.

The trick is for suitable laser light to induce oscillations - physicists call them phonons – within crystal lattices. The method can be visualized by considering what happens when children jump up and down on a mattress. In this analogy, the metal springs in the mattress correspond to the crystal lattice. For example, jumping could result in the springs always squeaking at a



This page When Michael Först and doctoral student Biaolong Liu induce properties in crystals with light, they work at a bench on which an array of optical instruments has been set up. They first use a laser pulse to produce oscillations that alter the material's behavior. With a second pulse, they then probe the momentary state of the crystal.

Right page Using a pump-probe experient, the Hamburg-based researchers produced superconductivity in a crystal consisting of soccer-ball-like fullerene molecules.

specific point during compression before rebounding. The squeak would then reveal the sought-after non-equilibrium state.

Continuing with this analogy, the researchers want to know precisely how the mattress springs are deformed in that state. This information can provide a snapshot of the mattress that captures it at just the right moment to yield a sharp spatial image of the squeak-inducing state.

That is, in principle, exactly what the researchers are doing with crystals. In scientific terminology, the jumping is known as "pumping," while the squeaking corresponds to "probing." Both pumping and probing of the momentary state are achieved using strong laser systems developed by Andrea Cavalleri and his team in the Institute's laboratories. The researchers also carry out the probing measurements, which, like the squeak of a spring, tell them indirectly whether they are on the track of the sought-after non-equilibrium state. Once they're certain they are, it's time for the intricate X-ray snapshots.

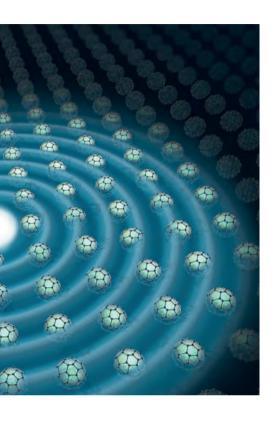
For this, the scientists in Hamburg need a free-electron laser that generates X-rays. In the past, they had to travel with their experimental setups to the Linac Coherent Light Source in Stanford, California up to three times a year. Until recently, the Linac facility was the world's most powerful X-ray source. Now, the even more powerful European XFEL is located right on the Institute's doorstep. Like a camera, it can capture extremely sharp images of atomic positions at just the right moment.

A PUMP-PROBE EXPERIMENT

"We want to bring the search for new materials and new material properties to the optical bench," Andrea Cavalleri stresses. However, it's very difficult to generate laser pulses (light flashes) that can induce the desired oscillations (phonons) in a crystal.

This is demonstrated by senior scientist Michael Först in one of Cavalleri's four laboratories on the ground floor. Before entering, we must don cleanroom clothing and laser goggles. "Dirt on the mirrors plus intensive laser beam equals broken mirrors," the physicist jokes. Many of the mirrors and lenses scattered around the heavy optical benches, creating elaborate labyrinths for laser light, are expensive, custom-made instruments. The experimental setup before us covers the area of an average apartment. It is a complete pump-probe experiment. The laser flash used for probing the crystal state – physicists call it the probe pulse - must be very short to provide a snapshot of the lattice structure. Without X-ray laser light, however, it would be impossible to capture an image of the atoms' positions. Moreover, the probe pulse must be a diverted copy of the pump pulse, because only if both pulses are synchronized can the probe pulse provide meaningful information about the excited crystal lattice.

A titanium sapphire laser generates strong laser flashes. Each laser pulse



lasts only a few femtoseconds, which is one-millionth of one-billionth of a second. A semi-transparent mirror splits the beam. The more powerful of the twin pulses is then used to excite the sample, corresponding to the children jumping on the mattress. The second, weaker pulse is sent via an optical delay circuit, so it arrives at the sample after the pump pulse and captures its momentary state. By varying the delay time, the researchers can find the precise moment at which the crystal takes on the state of interest the point at which the mattress emits a squeak.

However, the setup in front of us is even more complex. Like the optical equivalent of a reduction gearbox, it must convert the infrared light of the femtosecond laser to a wavelength that is up to 25 times longer. This terahertz radiation, as it is known, lies between infrared light and microwaves on the electromagnetic spectrum and can induce the desired oscillations in the crystal lattice. The atoms in crystals are electrically charged by the emission or absorption of electrons. The oscillating electric field of the terahertz waves thus pulls the atoms along, like buoys bobbing up and down on a passing water wave.

Recently, Cavalleri's group succeeded in using terahertz laser radiation to create a kind of artificial magnetism in a material called erbium iron oxide. In simple terms, the laser induced a precisely choreographed mix of oscillations in the crystal lattice. Collectively, the phonons generated a magnetic field in which the iron atoms in the crystal momentarily rotated in one direction like tiny magnetic needles. The result was laser-switchable magnetism. This effect could be of interest for future materials and electronic components.

COOPER PAIRS FORM A QUANTUM FLUID

Having returned from the laser lab to Cavalleri's room, we talk about the discovery of short-term superconductivity at room temperature in cuprates, the best-known high-temperature superconductors. "They are actually copper rust," Cavalleri says of copper oxides. Superconductivity is based on the fact that certain quantum effects in crystal lattices marry two electrons to form Cooper pairs, which behave quite differently from individual electrons. They gather in a collective quantum state to form a sort of quantum liquid that can flow through the crystal lattice without any resistance whatsoever.

In the case of cuprates and other high-temperature superconductors, it is still unclear what mechanism binds Cooper pairs together, but it works relatively well. It forms Cooper pairs even at room temperature, whereas in other types of superconductors they fly apart at temperatures well below minus 200 degrees Celsius due to tiny thermal movements. Even before he came to Hamburg, Cavalleri had found indirect experimental evidence of relatively temperature-resistant Cooper pairs.

But the cohesion of Cooper pairs in cuprates evidently still isn't sufficient for them to become superconducting at room temperature and thus conduct a current without resistance. The main obstacle is the sandwich-like layer structure of the complex cuprate crystals. At certain levels, which can be likened to the buttery layers of a sandwich, the Cooper pairs are able to slide around smoothly even at high temperatures. Above and below those, however, are layers of copper oxide that pose an insurmountable obstacle at room temperature. Those layers could be compared to layers of thick bread in a multi-layer sandwich. Only when this layer is compressed, so the buttery layer spreads out somewhat, do they approach each other close enough for Cooper pairs to slip through the layers vertically, as well. That is the precise moment when three-dimensional superconductivity occurs. Cavalleri's team has succeeded in inducing this state in a cuprate for ultra-short periods of time. At the X-ray laser in Stanford, the researchers also determined the exact positions of the atoms in a superconducting crystal.

The Max Planck researchers have now studied high-temperature superconductivity on a completely different



Magic with light: Andrea Cavalleri explores how material properties can be altered with laser pulses. His insights could find applications in sensors and optoelectronic devices.

material. "It's actually a superconducting plastic," Cavalleri explains, halfjokingly. This superconductor has no crystal lattice of atoms, but rather a cube-shaped lattice of molecular spheres. The soccer-ball-shaped molecules are buckminsterfullerenes consisting of 60 carbon atoms each.

It has long been known that the fulleride crystals become superconducting at temperatures below minus 253 degrees Celsius. However, Cavalleri's team recently produced superconductivity with terahertz laser radiation at a comparatively warm minus 170 degrees Celsius. The discovery that superconductivity is possible in a wide variety of materials at relatively high temperatures could provide insights into the universal properties of this quantum phenomenon.

Andrea Cavalleri's idea of altering the properties of materials with laser light could stimulate the invention of innovative sensors, for instance for electromagnetic radiation. It could also lead to the development of optoelectronic devices in which electrons are controlled by light or vice versa, or the development of tiny mechanical drives for nanotechnology. It is also conceivable to use light to rapidly turn window glass opaque, Cavalleri says - so his experiments in the microcosm could also find applications in our macroworld. ◀

TO THE POINT

- Andrea Cavalleri and his team at the Max Planck Institute for the Structure and Dynamics of Matter manipulate the properties of materials with laser light.
- · Using special laser pulses, the researchers briefly induce the crystals of a cuprate high-temperature superconductor to conduct current without loss at room temperature. In the same way, they also produce superconductivity in crystals of spherical buckminsterfullerenes at relatively high temperatures.
- With the help of X-ray lasers such as the XFEL in Hamburg, the scientists are investigating what changes in the arrangement of the atoms are responsible for these effects.
- · The findings could lead to the development of innovative sensors, for instance for electromagnetic radiation or optoelectronic components.

GLOSSARY

Cooper pairs: In superconductors, two electrons join together to form Cooper pairs, causing the electrical resistance of the material to drop to zero. The mechanism by which the electrons pair up is understood only for conventional superconductors (see below): lattice oscillations in a metal force conducting electrons together at low temperatures.

Superconductivity: Among the materials that conduct electricity without loss, physicists distinguish between conventional and unconventional superconductors. In conventional superconductors, superconductivity under normal pressure usually begins below minus 196 degrees Celsius, so that the materials must be cooled using very expensive liquid helium. Unconventional superconductors, in which the formation of Cooper pairs isn't caused by lattice oscillations, include high-temperature superconductors. The most prominent examples are cuprate ceramics, which, in addition to copper oxide, contain other metals such as barium and yttrium. In these materials, superconductivity occurs above minus 196 degrees Celsius, and in extreme cases at minus 140 degrees Celsius or higher. They can therefore be cooled with relatively inexpensive liquid nitrogen.

Terahertz radiation: That portion of the electromagnetic spectrum with a wavelength of between 100 micrometers and 1 millimeter, placing it between infrared and microwave radiation. In addition to many research applications, it is used in body scanners for security screening, and may also be useful in medical diagnostics.

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