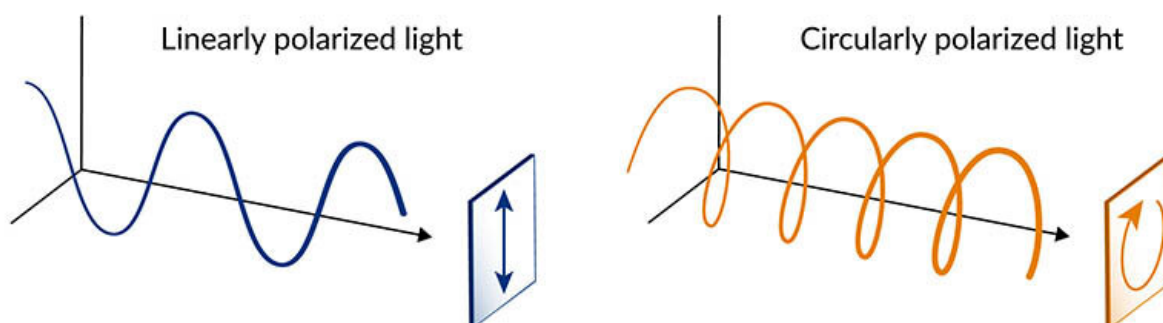


Experiments confirm a quantum material's unique response to circularly polarized laser light

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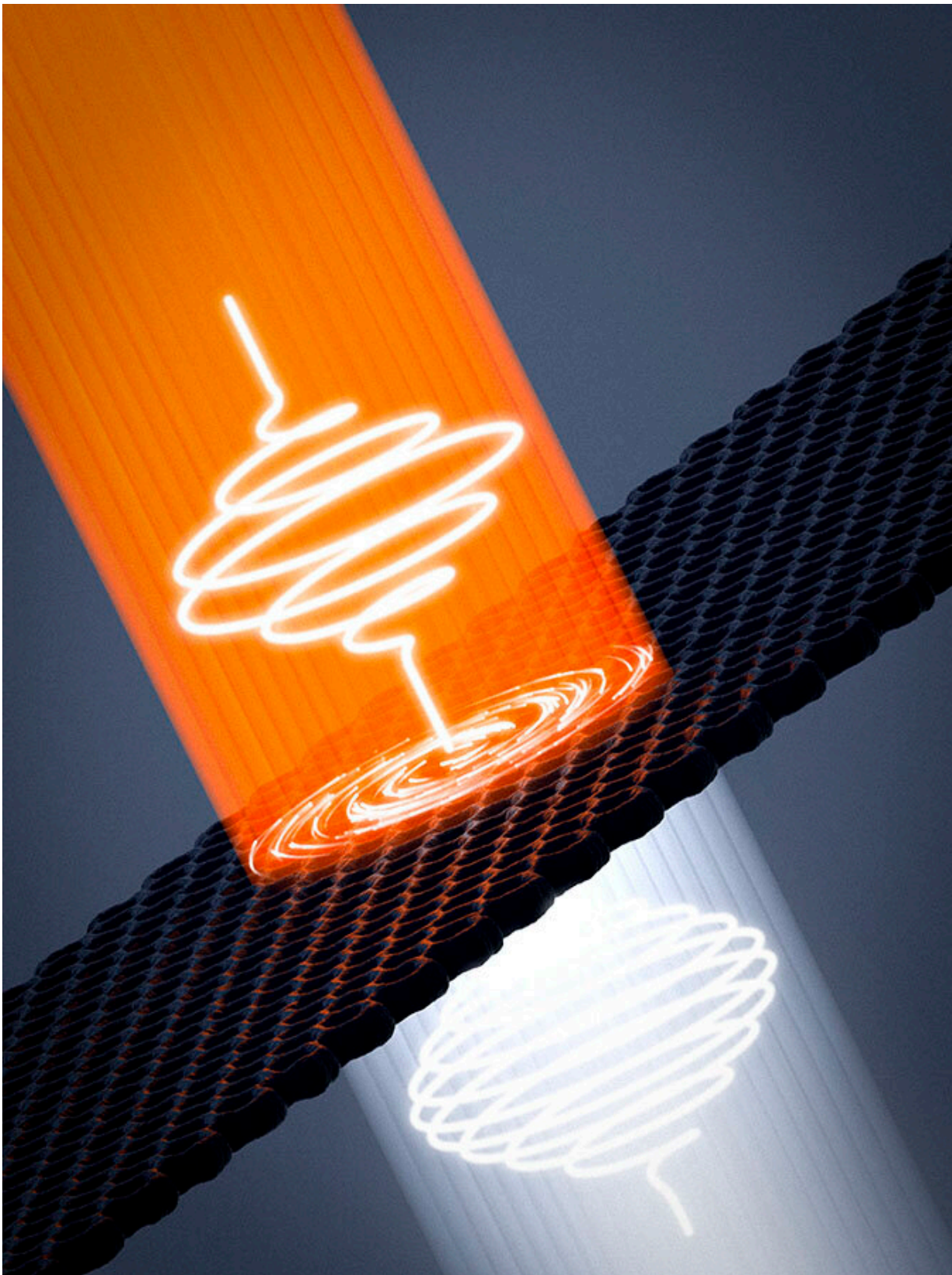
Laser light is usually linearly polarized, meaning that its waves oscillate in only one direction – up and down, in the example at left. But it can also be circularly polarized, at right, so its waves spiral like a corkscrew around the direction the light is traveling. A new study from SLAC and Stanford predicts that this circularly polarized light can be used to explore quantum materials in ways that were not possible before. Credit: Greg Stewart/SLAC National Accelerator Laboratory

When the COVID-19 pandemic shut down experiments at the Department of Energy's SLAC National Accelerator Laboratory early last year, Shambhu Ghimire's research group was forced to find another way to study an intriguing research target: quantum materials known as topological insulators, or TIs, which conduct electric current on their

surfaces but not through their interiors.

Denitsa Baykusheva, a Swiss National Science Foundation Fellow, had joined his group at the Stanford PULSE Institute two years earlier with the goal of finding a way to generate high harmonic generation, or HHG, in these materials as a tool for probing their behavior. In HHG, [laser light](#) shining through a material shifts to higher energies and higher frequencies, called harmonics, much like pressing on a guitar string produces higher notes. If this could be done in TIs, which are promising building blocks for technologies like spintronics, quantum sensing and quantum computing, it would give scientists a new tool for investigating these and other quantum materials.

With the experiment shut down midway, she and her colleagues turned to theory and computer simulations to [come up with a new recipe](#) for generating HHG in topological insulators. The results suggested that circularly polarized light, which spirals along the direction of the laser beam, would produce clear, unique signals from both the conductive surfaces and the interior of the TI they were studying, bismuth selenide—and would in fact enhance the signal coming from the surfaces.

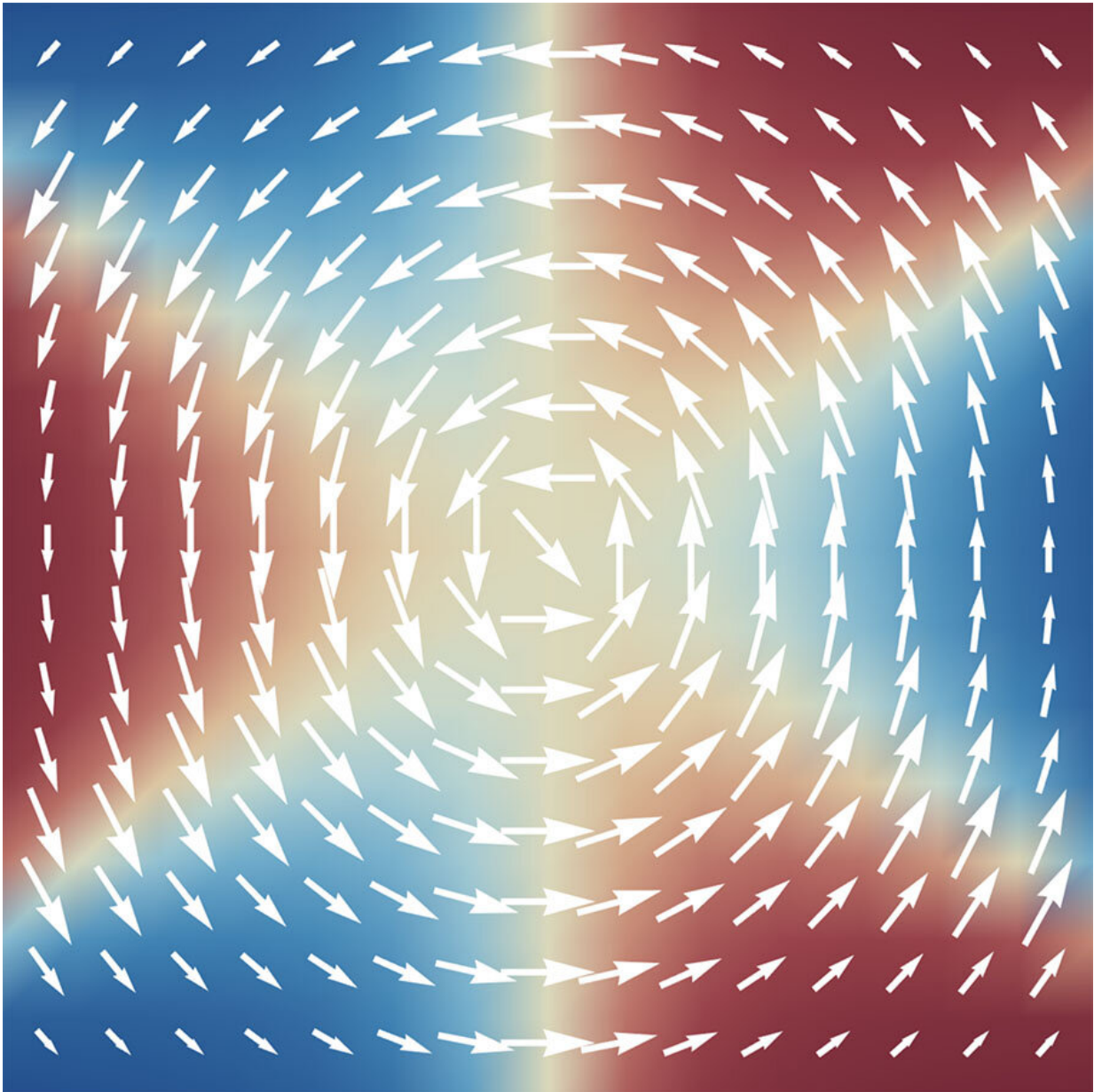


An illustration shows how circularly polarized laser light (top) is used to probe a topological insulator (black), a quantum material that conducts electric current on its surface but not through its interior. The light causes electrons in the material to fly apart, recombine and emit light (white) of higher energies and frequencies through a process called high harmonic generation. By analyzing this emitted light, scientists can measure the spin and momentum of electrons in the material. Experiments at SLAC confirm that these signals are a unique signature of topological surfaces. Credit: Greg Stewart/SLAC National Accelerator Laboratory

When the lab reopened for experiments with covid safety precautions in place, Baykusheva set out to test that recipe for the first time. In a paper published today in *Nano Letters*, the research team report that those tests went exactly as predicted, producing the first unique signature from the topological surface.

"This material looks very different than any other material we've tried," said Ghimire, who is a principal investigator at PULSE. "It's really exciting being able to find a new class of material that has a very different optical response than anything else."

Over the past dozen years, Ghimire had done a series of experiments with PULSE Director David Reis showing that HHG can be produced in ways that were previously thought unlikely or even impossible: by beaming laser light into a [crystal](#), a [frozen argon gas](#) or an [atomically thin semiconductor](#) material. Another study described how to use HHG to generate attosecond laser pulses, which can be used to observe and control the movements of electrons, by shining a laser through ordinary glass.



This pattern of arrows reflects the combined spin and momentum of electrons in the surface layer of a topological insulator, a quantum material that conducts electric current on its surface but not through its interior. Experiments at SLAC discovered that circularly polarized laser light couples to this spin polarization to produce a unique pattern of high harmonic generation that is a signature of topological surfaces. Credit: Denitsa Baykusheva/Stanford PULSE Institute

But [quantum materials](#) had steadfastly resisted being analyzed this way, and the split personalities of [topological insulators](#) presented a particular problem.

"When we shine laser light on a TI, both the surface and the interior produce harmonics. The challenge is to separate them," Ghimire said.

The team's key discovery, he explained, was that circularly polarized light interacts with the surface and the interior in profoundly different ways that boost high harmonic generation coming from the surface and also give it a distinctive signature. Those interactions, in turn, are shaped by two fundamental differences between the surface and the interior: the degree to which their electron spins are polarized – oriented in a clockwise or counterclockwise direction, for instance – and the types of symmetry found in their atomic lattices.

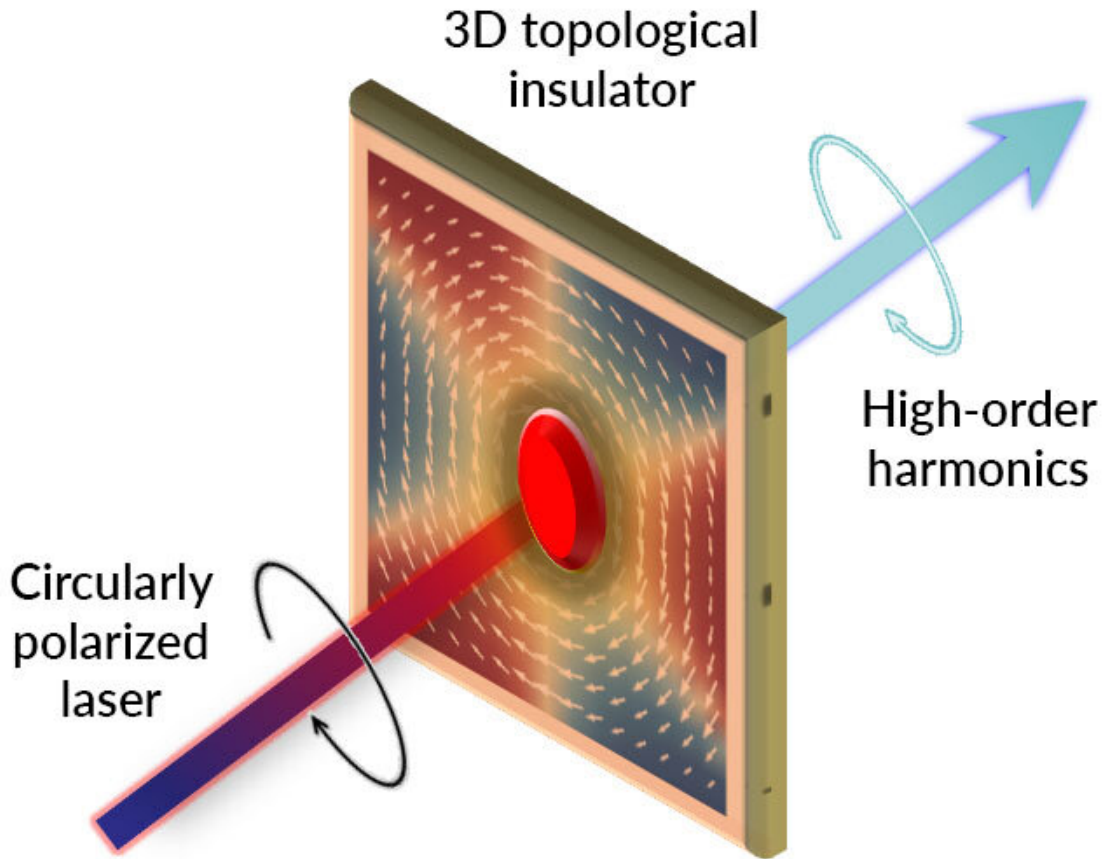


Diagram of an experimental setup at SLAC's high-power laser lab where scientists used circularly polarized laser light to probe a topological insulator – a type of quantum material that conducts electric current on its surfaces but not through its interior. A process called high harmonic generation shifts the laser light to higher energies and frequencies, or harmonics. This produces polarization patterns in a detector (arrows) that reveal the spin and momentum of electrons in the conductive surface layer – a unique signature of the topological surface. Credit: Shambhu Ghimire/Stanford PULSE Institute

Since the group published their recipe for achieving HHG in TIs earlier

this year, two other research groups in Germany and China have reported creating HHG in a topological insulator, Ghimire said. But both of those experiments were with linearly polarized light, so they did not see the enhanced signal generated by circularly polarized light. That signal, he said, is a unique feature of topological surface states.

Because intense laser light can turn electrons in a material into a soup of electrons—a plasma—the team had to find a way to shift the wavelength of their high-powered titanium sapphire laser so it was 10 times longer, and thus 10 times less energetic. They also used very short [laser](#) pulses to minimize damage to the sample, which had the added advantage of allowing them to capture the material's behavior with the equivalent of a shutter speed of millionths of a billionth of a second.

"The advantage of using HHG is that it's an ultrafast probe," Ghimire said. "Now that we've identified this novel approach to probing the topological surface states, we can use it to study other interesting materials, including topological states induced by strong lasers or by chemical means."

Researchers from the Stanford Institute for Materials and Energy Sciences (SIMES) at SLAC, the University of Michigan, Ann Arbor, and Pohang University of Science and Technology (POSTECH) in Korea contributed to this work.

More information: Denitsa Baykusheva et al, All-Optical Probe of Three-Dimensional Topological Insulators Based on High-Harmonic Generation by Circularly Polarized Laser Fields, *Nano Letters* (2021). [DOI: 10.1021/acs.nanolett.1c02145](https://doi.org/10.1021/acs.nanolett.1c02145)

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