Study reports high-harmonic generation in an epsilon-near-zero material

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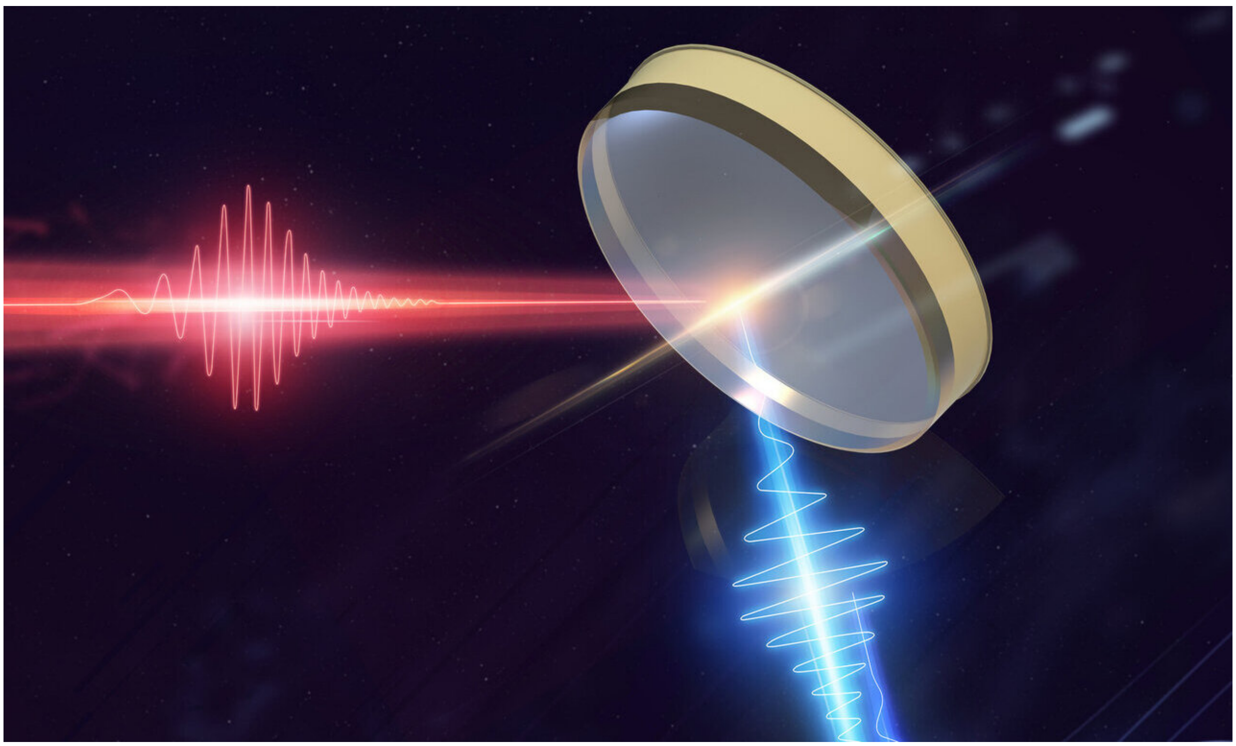

Credit: Yang et al.

High-harmonic generation (HHG) is a nonlinear optical phenomenon through which high harmonics of an intense laser beam are generated in a target material, typically a gas. Physicists have been studying HHG in atomic gases for decades, but more recently a team of researchers at SLAC National Accelerator Laboratory has started investigating this process in solids.

One of the advantages in using solid targets is more efficient generation, due to the high density at interaction. The first experiment was performed on a zinc oxide crystal, with the observation of harmonics up to 25th orders . Since then, HHG has been successfully observed in several dielectrics, including a magnesium oxide crystal quartz.

One of the latest examples of this stems from a collaborative effort involving a team of researchers at Sandia National Laboratories, Tsinghua University, SLAC National Accelerator Laboratory, the University of New Mexico and North Carolina State University. In their paper, published in *Nature Physics*, they reported HHG arising from a low-loss, indium-doped cadmium oxide thin film, which was attained by leveraging the material's epsilon-near-zero (ENZ) effect.

"The history of this research traces back to our extensive activities in epsilon-near-zero materials and phenomena," Igal Brener and Yuanmu Yang, two of the researchers who are co-authors in the study, told Phys.org via email.

ENZ materials, such as the film used by the researchers, are a new class of materials with a vanishing permittivity (i.e. equal to zero) at some wavelength (i.e. frequency). Recent studies suggest that they also exhibit ultrafast nonlinear efficiencies within their sub-wavelength propagation lengths.

One of the consequences of these material's permittivity going to zero at a predetermined wavelength is that when shining ENZ thin films under the right conditions (i.e. angle, polarization), the optical fields inside these films are significantly enhanced (with a ratio of 10 to 100X). This means that any phenomenon that relies on the intensity of these fields, such as optical nonlinearities, should be vastly enhanced.

"We had done some prior nonlinear optical experiments (i.e. harmonic generation) in other ENZ materials (ITO) and saw some indications of enhanced efficiencies; so did a couple of other research groups," Brener and Yang said. "Highly doped CdO (material grown by co-author Jon-Paul Maria) is a much superior ENZ material (higher electron mobility which translates into lower optical losses and higher optical field enhancements). Therefore, we wanted to study HHG in these films."

In recent years, there has been [a growing interest] in finding new ways to produce attosecond pulses, especially in a compact experimental setup, i.e., by replacing the big gas tubes and expensive high-intensity laser systems in which these pulses are generated today. In their study, Brener, Yang and their colleagues set out to explore this possibility further, using a low-loss, indium-doped cadmium oxide thin film.

The sample used in their experiments consists of a thin (75nm) film of highly doped CdO with a plasma frequency that lies at the equivalent wavelength of ~2um, which is the ENZ wavelength. This sample is grown on MgO and has a metal top layer introduced to create what is known as "perfect absorption."

The researchers illuminated their sample with short pulses at 2.08um from the substrate at oblique incidence and p-polarization. They then measured the harmonics generated on the reflected optical path using standard UV-Vis spectrometers and detectors.

"Because of the absorption of the substrate, in this reflection configuration, we were only able to measure up to the ninth harmonic; that is the shortest wavelength we could measure," Brener and Yang explained. "In the future, samples without the gold top layer could be tried in the transmission geometry so this issue can be mitigated.

In their study, the researchers observed that ENZ-assisted harmonics exhibited a pronounced spectral redshift and a line-width broadening. This was the result of photo induced electron heating and the consequential time-dependent ENZ wavelength of the material they used.

The attosecond science community is interested in materials that exhibit this behavior, as it could potentially improve how these specialized pulses are generated. Replacing the gas used in typical systems with a solid material, such as a thin film of cadmium oxide, would allow researchers to observe some of the fastest events in nature in an easier, less expensive and possibly more detailed way.

Compared to the observations gathered in other experiments with solid state materials, the harmonics achieved by the researchers required about two order of magnitude less of pump optical power. Therefore, the material and process they used greatly simplifies the hardware required for HHG and attosecond spectroscopy.

A further interesting finding of their study is that the optical nonlinearity arises from the electrons that are present in the highly doped CdO and the nature of the band structure of CdO. The combination of optical pumping at the ENZ wavelength and the nature of the nonlinearity giving rise to HHG could offer guidance for further improvements, while also informing the search for other materials that exhibit similar behavior.

For the members of the collaboration who are working at Sandia, this research stemmed from a broader interest in nonlinear optics, which they plan to continue studying in other ways. For instance, Sandia has already explored a related phenomenon in which light passing through cadmium oxide becomes more than 10 times brighter inside the material. In their work, they used this effect to build a high-contrast optical switch that could eventually help to speed up optical communications.

More information: Yuanmu Yang et al. High-harmonic generation from an epsilon-near-zero material, *Nature Physics* (2019). DOI: 10.1038/s41567-019-0584-7

Shambhu Ghimire et al. High-harmonic generation from solids, *Nature Physics* (2018). DOI: 10.1038/s41567-018-0315-5

Orad Reshef et al. Nonlinear optical effects in epsilon-near-zero media, *Nature Reviews Materials* (2019). DOI: 10.1038/s41578-019-0120-5