ICFO



New X-ray imaging technique to study the transient phases of quantum materials

An international team of researchers has recently demonstrated for the first time the use of a new lensless ultrafast X-Ray method to image phase transitions. This new method described in Nature Physics enables the direct observation of the dynamics o quantum materials at the nanoscale.

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The use of light to produce transient phases in quantum materials is fast becoming a novel way to engineer new properties in them, such as the generation of superconductivity or nanoscale topological defects. However, visualizing the growth of a new phase in a solid is not easy, due in-part to the wide range of spatial and time scales involved in the process. Although in the last two decades scientists have explained light-induced phase transitions by invoking nanoscale dynamics, real space images have not yet been produced and, thus, no one has seen them.

In the new study published in Nature Physics, ICFO researchers Allan S. Johnson and Danie Perez-Salinas, led by former ICFO Prof. Simon Wall, in collaboration with colleagues fro Aarhus University, Sogang University, Vanderbilt University, the Max Born Institute, th Diamond Light Source, ALBA Synchrotron, Utrecht University, and the Pohang Accelerato Laboratory, have pioneered a new imaging method that allows the capture of he light-induced phase transition in vanadium oxide (VO_2) with high spatial and temporal resolution.?

The new technique implemented by the researchers is based on coherent X-ra hyperspectral imaging at a free electron laser, which has allowed them to visualize and bette understand, at the nanoscale, the insulator-to-metal phase transition in this very well-know quantum material

The crystal VO_2 has been widely used in to study light-induced phase transitions. It was the first material to have its solid-solid transition tracked by time-resolved X-ray diffraction and its electronic nature was studied by using for the first time ultrafast X-ray absorption techniques. At room temperature, VO2 is in the insulating phase. However, if light is applied to the material, it is possible to break the dimers of the vanadium ion pairs and drive the transition from an insulating to a metallic phase.

In their experiment, the authors of the study prepared thin samples of VO2 with a gold mask to define the field of view. Then, the samples were taken to the X-ray Free Electron Laser facility at the Pohang Accelerator Laboratory, where an optical laser pulse induced the transient phase, before being probed by an ultrafast X-ray laser pulse. A camera captured the scattered X-rays, and the coherent scattering patterns were converted into images by using two different approaches: Fourier Transform Holography (FTH) and Coherent Diffractiv Imaging (CDI). Images were taken at a range of time delays and X-ray wavelengths to build u a movie of the process with 150 femtosecond time resolution and 50 nm spatial resolution but also with full hyperspectral information

The surprising role of the pressure

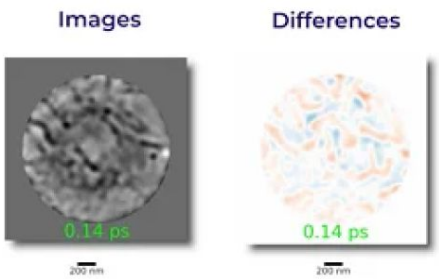
The new methodology allowed the researchers to better understand the dynamics of the phase transition in VO2. They found that pressure plays a much larger role in light-induced phase transitions than previously expected or assumed.

"We saw that the transient phases aren't nearly as exotic as people had believed! Instead of a truly non-equilibrium phase, what we saw was that we had been misled by the fact that the ultrafast transition intrinsically leads to giant internal pressures in the sample millions of times higher than atmospheric. This pressure changes the material properties and takes time to relax, making it seem like there was a transient phase" says Allan Johnson, postdoctoral researcher at ICFO. "Using our imaging method, we saw that, at least in this case, there was no link between the picosecond dynamics that we did see and any nanoscale changes or

exotics phases. So, it looks like some of those conclusions will have to be revisited".
To identify the role played by the pressure in the process, it was crucial to use the hyperspectral image. "By combining imaging and spectroscopy into one great image, we are able to retrieve much more information that permits us to actually see detailed features and decipher exactly where they come from," continues Johnson. "This was essential to look at each part of our crystal and determine whether it was a normal or an exotic out-of-equilibrium phase-and with this information we were able to determine that during the phase transitions all the regions of our crystal were the same, except for the pressure".

Challenging research

One of the main challenges the researchers faced during the experiment was to ensure that the crystal sample of VO2 returned to its original starting phase each time and after being illuminated by the laser. To guarantee that this would occur, they conducted preliminary experiments at synchrotrons where they took several crystal samples and repeatedly shone the laser on them to test their capacity to recover back to their original state.
The second challenge resided in having access to an X-Ray free electron laser, large research facilities where the time windows to conduct the experiments are very competitive and in-demand because there are only a few in the world. "We had to spend two weeks in quarantine in South Korea due to the COVID-19 restrictions before we got our one shot of just five days to make the experiment work, so that was an intense time" Johnson recalls.
Although the researchers describe the present work as fundamental research, the potential applications of this technique could be diverse, since they could i¿½look at polarons movi g inside catalytic materials, try imaging superconductivity itself, or even help us understa d novel nanotechnologies by viewing and imaging inside nanoscale devicesi¿½ concl des Johnson.



Link to the video