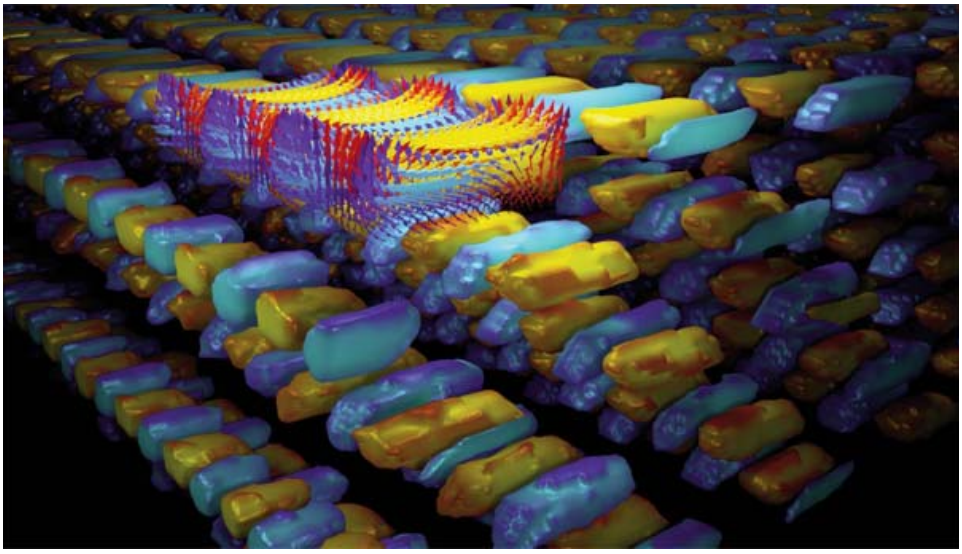
The Advanced Photon Source

Supercrystal:

A Hidden Phase of Matter Created by a Burst of Light

"Frustration" plus a pulse of laser light resulted in a stable "supercrystal," one of the first examples of a new state of matter with long-term stability transfigured by the energy from a sub-picosecond laser pulse. The research focused on states of matter with unusual properties (e.g., new forms of magnetic or electric states) that do not exist in equilibrium in nature.



A three-dimensional image of a supercrystal from phase-field simulations using the software μ-PRO. Image: L-Q Chen Group / Penn State

The challenge was to find a way to maintain the intermediate state of matter, since the state may exist for only some tiny fraction of a second and then disappear. The team accomplished this by "frustrating the system" — not allowing the material to do what it wants to do, which is to allow it to minimize its energy fully without constraints.

Frustration was achieved by using single atomic layers of two compounds, lead titanate and strontium titanate, stacked in alternating layers on top of each other to build up a three-dimensional structure. Lead titanate is a ferroelectric material, while strontium titanate is not. This mismatch forced the electric polarization vectors to take an unnatural path, curving back on themselves to make vortices, like water swirling down a drain.

The layers were grown on top of a crystal substrate whose lattice spacing was intermediate in size between the two layered materials. This provided a second level of frustration, as the strontium titanate layer tried to stretch to conform with the crystal structure of the substrate, and the lead titanate had to compress to conform to it. This put the whole system into a frustrated state with multiple phases randomly distributed in the volume.

The laser pulse produced free charges in the material, adding extra electrical energy to the system and driving it into a new state of matter: a supercrystal. These supercrystals have a unit cell much larger than any ordinary inorganic crystal, with a volume one million times larger than the unit cells of the original two materials.

Unlike transient states, this supercrystal state stays around potentially forever at room temperature — at least a year in this study — unless it is heated to about 350° Fahrenheit where it is erased. The process can be repeated by hitting the material with a light pulse and erased using heat. This state can only be created by ultrashort laser pulses with a threshold energy, and not by spreading out that energy over longer pulses.

The team used high-energy x-ray diffraction at the Advanced Photon Source to examine the supercrystal before and after it formed, clearly showing the transformation from disordered matter into a supercrystal.

By virtue of its short pulse duration, an ultrafast laser imprints excitations in materials faster than their intrinsic response time. While such dynamical transformations were already explored for decades to stimulate the ordering of materials, a strategy for their steady state stabilization seemed out of reach until now.

The high-resolution x-ray diffraction, combined with imaging at the nanoscale level, was used to observe the evolution of irreversible structural reordering. For the first time, it was seen that a single ultrafast laser pulse irradiation of artificially layered polar material can induce long-range structural perfection when starting from relative disorder.

See: V.A. Stoica, N. Laanait, C. Dai, Z. Hong, Y. Yuan, Z. Zhang, S. Lei, M.R. McCarter, A. Yadav, A.R. Damodaran, S. Das, G.A. Stone, J. Karapetrova, D.A. Walko, X. Zhang, L.W. Martin, R. Ramesh, L.-Q. Chen, H. Wen, V. Gopalan, J.W. Freeland, "Optical creation of a supercrystal with three-dimensional nanoscale periodicity," Nat. Mater. **18**, 377 (18 March 2019). DOI: 10.1038/s41563-019-0311-x

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Call for APS General-User Proposals

The Advanced Photon Source is open to experimenters who can benefit from the facility's high-brightness hard x-ray beams.

General-user proposals for beam time during Run 2019-3 are due by Friday, July 5, 2019.

Information on access to beam time at the APS is at http://www.aps.anl.gov/Users/apply_for_beamtime.html or contact Dr. Dennis Mills, DMM@aps.anl.gov, 630/252-5680.

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