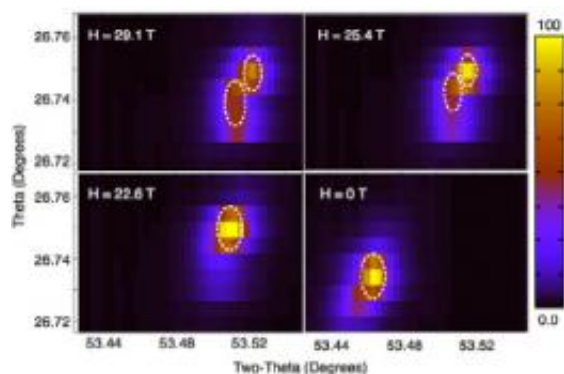


Probing spin liquids with a new pulsed-magnet system

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The (008) intensity color map on a θ vs. 2θ mesh. With increasing magnetic field the peak splits at a critical field of $H \sim 29$ T, which is a hallmark of a structural phase transition with a reduction from cubic to tetragonal or orthorhombic symmetry. Above the critical field, the peak broadens and two subtly distinct lattice parameters are observed.

Entirely new experimental vistas could be opened by a device called a precursor pulsed-magnet system developed by an international team of scientists. This system can generate magnetic fields as high as 30 Tesla for synchrotron x-ray scattering experiments. The researchers recently completed the first practical work using the system at the U.S.

Department of Energy's Advanced Photon Source (APS) at Argonne National Laboratory to study magnetoelastic effects in the rare-earth pyrochlore terbium titanate ($\text{Tb}_2\text{Ti}_2\text{O}_7$). Their findings were published in *Physical Review Letters*.

Terbium titanate is a member of a class of materials called “frustrated magnets” because of their lack of long-range magnetic order. Tetrahedral coordination of magnetic moments of Tb ions in the lattice structure prevents them from settling into a predictably ordered ground state. Such materials may exhibit magnetostrictive effects (a property of ferromagnetic materials that causes them to change their shape or dimensions when subjected to a [magnetic field](#)) and other exotic ferromagnetic and antiferromagnetic properties, which may be relevant to electronic transducers and switching applications.

Terbium titanate remains in a highly disordered spin-liquid ground state even at the lowest measurable temperature, while exhibiting magnetostriction exceeding that in commercial compounds such as Terfenol-D. Attempts to explain the material’s behavior by Hamiltonian models have been inconclusive, awaiting experimental data on its lattice properties in the spin-liquid state. “The problem in general of magneto-elastic effects, in particular in systems which don’t really order very well, hasn’t been well studied using structural probes such as x-rays,” said Argonne’s Zahirul Islam of the X-ray Science Division (XSD), lead scientist on the pulsed-magnet project.

The new pulsed-magnet system at the APS, which was described in an article in *Reviews of Scientific Instruments*, proved to be an ideal tool for direct structural observation of the $Tb_2Ti_2O_7$ compound at very low temperatures using single-crystal x-ray diffraction at the XSD 4-ID-D beamline. “There was a lot of incidental evidence that the way these little magnetic moments pointed was really strongly coupled to the actual positions of the atoms inside the crystal lattice,” explains Jacob Ruff, lead author of the [Physical Review Letters](#) article, who recently joined Argonne as a Director’s Fellow from Bruce Gaulin's group at McMaster University to advance x-ray studies of materials in pulsed magnetic fields. The new APS pulsed magnet allowed the experimenters to “carefully constrain and measure that directly with [x-rays](#),” said Ruff.

“We can apply high magnetic fields and we can actually see the lattice shift and bend and twist around.”

Pulsed-magnet experiments provide some unique advantages for materials science work. Islam explains, “Materials often display exotic phases under extreme conditions, which are fundamental to understanding their functionality, and the pulsed magnet is a way to generate this type of extreme condition in a contact-free way. Because the pulsed magnetic field is generated fast for a short duration, one may also combine it with the natural timing structure and high brilliance of synchrotron radiation for precision studies of structural relaxations and metastable phenomena on a micro-second level.”

The researchers subjected $\text{Tb}_2\text{Ti}_2\text{O}_7$ crystals to pulsed magnetic fields of up to 30 Tesla and temperatures as low as 4.4K, measuring the transverse magnetostriction in different conditions and observing the deformations of the crystal lattice via diffraction studies. They found anisotropic magnetoelastic effects that have not been previously observed or predicted in the rare-earth titanates. At high temperatures, the $\text{Tb}_2\text{Ti}_2\text{O}_7$ system behaves much like a conventional paramagnet, but crosses over to display a collective response in the spin-liquid regime. Under moderate magnetic fields, some cubic pyrochlore symmetry is restored, but this gives way to a structural phase transition when the magnetic field is increased.

The results indicate a strong, very large coupling between spin and lattice degrees of freedom in the spin-liquid state of $\text{Tb}_2\text{Ti}_2\text{O}_7$. Ruff observes, “The coupling between spin and lattice is very strong in this system. Most likely, the reason it doesn’t order at low temperature is that any ordered state would have internal magnetic fields and would actually have to distort or bend the lattice around.”

The split-pair coil used in the APS pulsed-magnet system was designed

and fabricated by Hiroyuki Nojiri and Yasuhiro Matsuda at the Institute for Materials Research at Tohoku. The experimental team is enthusiastic about the promise of the pulsed-magnet system and plans two complementary instruments that will allow different experimental orientations of the x-ray beam and magnetic fields, permitting researchers to design experiments utilizing various x-ray techniques.

While the pulsed magnets are certainly not the answer to every problem requiring magnetic fields, Islam notes that “it is the only practical approach that is available in the foreseeable future for studying many materials at a synchrotron in high magnetic fields.” The current work not only provides some unique insights into the $\text{Tb}_2\text{Ti}_2\text{O}_7$ system, but serves as the first major step in developing this exciting new capability at the APS to address high-field science at the frontiers of contemporary condensed matter physics.

More information:

-- Zahirul Islam, Jacob P.C. Ruff, Hiroyuki Nojiri, Yasuhiro H. Matsuda, Kathryn A. Ross, Bruce D. Gaulin, Zhe Qu, and Jonathan C. Lang, “A portable high-field pulsed-magnet system for single-crystal X-ray scattering studies,” *Rev. Sci. Instrum.* 80, 113902 (2009).

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