

# Frustrated magnets: New experiment reveals clues to their discontent

April 3 2015

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A crystal of frustrated magnet ( $\text{Tb}_2\text{Ti}_2\text{O}_7$ ). Credit: Jason Krizan

An experiment conducted by Princeton researchers has revealed an unlikely behavior in a class of materials called frustrated magnets, addressing a long-debated question about the nature of these discontented quantum materials.

The work represents a surprising discovery that down the road may

suggest new research directions for advanced electronics. Published this week in the journal *Science*, the study also someday may help clarify the mechanism of high-temperature superconductivity, the frictionless transmission of electricity.

The researchers tested the frustrated magnets—so-named because they should be magnetic at low temperatures but aren't—to see if they exhibit a behavior called the Hall Effect. When a [magnetic field](#) is applied to an [electric current](#) flowing in a conductor such as a copper ribbon, the current deflects to one side of the ribbon. This deflection, first observed in 1879 by E.H. Hall, is used today in sensors for devices such as computer printers and automobile anti-lock braking systems.

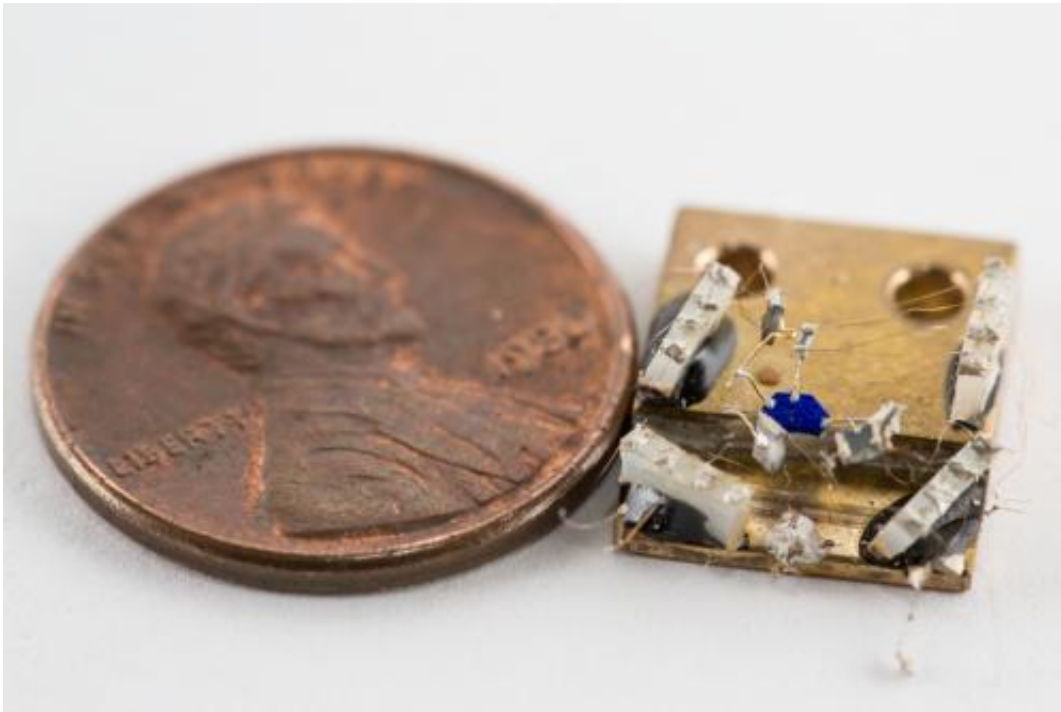
Because the Hall Effect happens in charge-carrying particles, most physicists thought it would be impossible to see such behavior in non-charged, or neutral, particles like those in frustrated magnets. "To talk about the Hall Effect for [neutral particles](#) is an oxymoron, a crazy idea," said N. Phuan Ong, Princeton's Eugene Higgins Professor of Physics.

Nevertheless, some theorists speculated that the neutral particles in frustrated magnets might bend to the Hall rule under extremely cold conditions, near absolute zero, where particles behave according to the laws of quantum mechanics rather than the classical physical laws we observe in our everyday world. Harnessing [quantum behavior](#) could enable game-changing innovations in computing and electronic devices.

Ong and colleague Robert Cava, Princeton's Russell Wellman Moore Professor of Chemistry, and their graduate students Max Hirschberger and Jason Krizan decided to see if they could settle the debate and demonstrate conclusively that the Hall Effect exists for frustrated magnets.

To do so, the research team turned to a class of the magnets called

pyrochlores. They contain magnetic moments that, at very low temperatures near absolute zero, should line up in an orderly manner so that all of their "spins," a quantum-mechanical property, point in the same direction. Instead, experiments have found that the spins point in random directions. These frustrated materials are also referred to as "quantum spin ice."



Graduate student Max Hirschberger lowers the assembled experimental setup into a high-field magnet system, capable of creating fields as strong as 250,000 times the earth's magnetic field. Credit: Jason Krizan

"These materials are very interesting because theorists think the tendency for spins to align is still there, but, due to a concept called geometric frustration, the spins are entangled but not ordered," Ong said. Entanglement is a key property of quantum systems that researchers hope to harness for building a quantum computer, which could solve

problems that today's computers cannot handle.

A chance conversation in a hallway between Cava and Ong revealed that Cava had the know-how and experimental infrastructure to make such materials. He tasked chemistry graduate student Krizan with growing the crystals while Hirschberger, a graduate student in physics, set up the experiments needed to look for the Hall Effect.

"The main challenge was how to measure the Hall Effect at an extremely low temperature where the quantum nature of these materials comes out," Hirschberger said. The experiments were performed at temperatures of 0.5 degrees Kelvin, and required Hirschberger to resolve temperature differences as small as a thousandth of a degree between opposite edges of a crystal.

To grow the crystals, Krizan first synthesized the material from terbium oxide and titanium oxide in a furnace similar to a kiln. After forming the pyrochlore powder into a cylinder suitable for feeding the crystal growth, Krizan suspended it in a chamber filled with pure oxygen and blasted it with enough focused light from four 1000-Watt halogen light bulbs to heat a small region to 1800 degrees Celsius. The final products were thin, flat transparent or orange slabs about the size of a sesame seed.

To test each crystal, Hirschberger attached tiny gold electrodes to either end of the slab, using microheaters to drive a heat current through the crystal. At such low temperatures, this heat current is analogous to the electric current in the ordinary Hall Effect experiment.

At the same time, he applied a magnetic field in the direction perpendicular to the heat current. To his surprise, he saw that the heat current was deflected to one side of the crystal. He had observed the Hall Effect in a non-magnetic material.

Surprised by the results, Ong suggested that Hirschberger repeat the experiment, this time by reversing the direction of the heat current. If Hirschberger was really seeing the Hall Effect, the current should deflect to the opposite side of the crystal. Reconfiguring the experiment at such [low temperatures](#) was not easy, but eventually he demonstrated that the signal did indeed reverse in a manner consistent with the Hall Effect.

"All of us were very surprised because we work and play in the classical, non-quantum world," Ong said. "Quantum behavior can seem very strange, and this is one example where something that shouldn't happen is really there. It really exists."

The use of experiments to probe the quantum behavior of materials is essential for broadening our understanding of fundamental physical properties and the eventual exploitation of this understanding in new technologies, according to Cava. "Every technological advance has a basis in fundamental science through our curiosity about how the world works," he said.

Further experiments on these materials may provide insights into how superconductivity occurs in certain copper-containing materials called cuprates, also known as "high-temperature" superconductors because they work well above the frigid temperatures required for today's superconductors, such as those used in MRI machines.

One of the ideas for how [high-temperature superconductivity](#) could occur is based on the possible existence of a particle called the spinon. Theorists, including the Nobel laureate Philip Anderson, Princeton's Joseph Henry Professor of Physics, Emeritus and a senior physicist, and others have speculated that spinons could be the carrier of a heat current in a quantum system such as the one explored in the present study.

Although the team does not claim to have observed the spinon, Ong said

that the work could lead in such a direction in the future. "This work sets the stage for hunting the spinon," Ong said. "We have seen its tracks, so to speak."

**More information:** Max Hirschberger, Jason W. Krizan, R. J. Cava, N. P. Ong. Large thermal Hall conductivity of neutral spin excitations in a frustrated quantum magnet. *Science*. 10.1126/science.1257340

Provided by Princeton University

Citation: Frustrated magnets: New experiment reveals clues to their discontent (2015, April 3)  
retrieved 19 April 2024 from

<https://phys.org/news/2015-04-frustrated-magnets-reveals-clues-discontent.html>

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