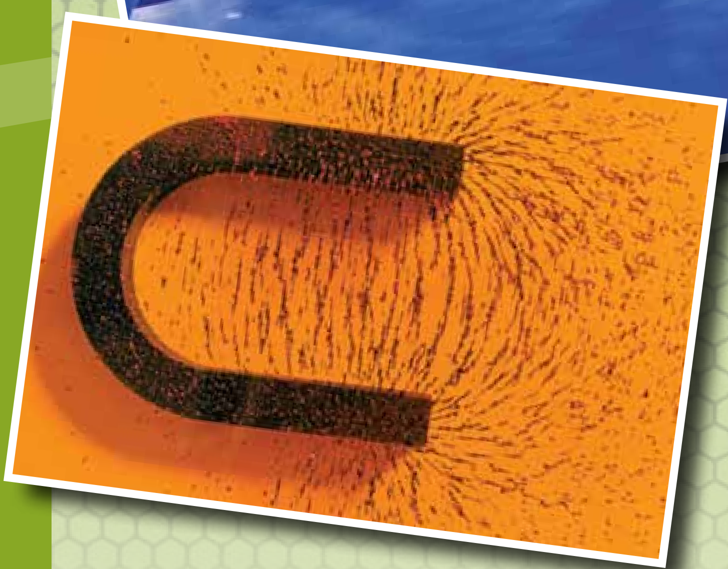


Studies of spin ice have increased our knowledge of the magnetic relationships among atoms, helping us better understand the other properties of materials.

Magnetism and Molecular Structure

Georg Ehlers and his colleagues are a bit like detectives on an atomic scale. They're looking for clues to explain how changes in molecular structure affect the magnetic relationships among atoms in a material called "spin ice." Because magnetic relationships affect other properties of materials, this research is applicable to a variety of fields.

The term spin ice is applied to materials whose magnetic structure is comparable to the crystal structure of water ice. As one might expect, water ice has been studied in depth, so the similarities between the two materials provide researchers with a model, of sorts, for their investigations. The spin ice Ehlers and his colleagues are studying is basically $Ho_2Ti_2O_7$, where some of the holmium has been replaced with nonmagnetic lanthanum. The reasons for doing this may be a little more obvious if we define a few key terms.

What is a magnet? Everyone has held two magnets together and felt them attract and repel each other. On the atomic scale, the definition of a magnet becomes a little broader—any substance containing atoms that exert a magnetic force on other atoms is considered to be a magnet. The biggest difference between these two situations is that, on the macroscopic scale, a magnet is made up of atoms whose magnetic moments all work in the same direction, acting together. On the atomic scale, however, a magnet can be composed of atoms whose moments point in many different directions—working together or potentially cancelling each other out.

What is a "frustrated" magnet? In a solid material, individual magnetic moments can point in a number of different ways, usually in the arrangement that requires the least energy to maintain. "This is the arrangement of magnetic moments that you would most often find," says Ehlers. "However, a frustrated magnet is a material where the spatial arrangement of the neighboring magnetic atoms prevents the magnetic moments from easily finding this sort of minimum energy configuration."

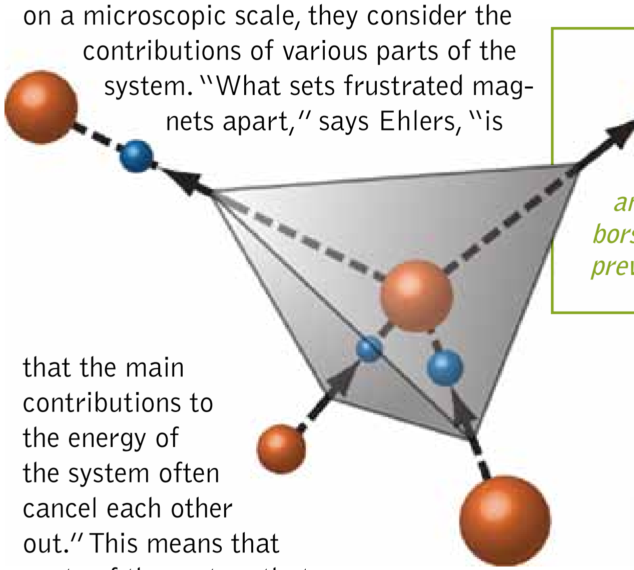
What is spin ice? Spin ice is a type of frustrated magnet. Similar to water, magnetic moments in spin ice can assume any of several different tetrahedral configurations, all with the same ground state energy. But when the ice freezes, the possible varied configurations prevent it from easily finding its "least energy" configuration, or long-range order.

In this project, Ehlers' team (which includes Eugene Mamontov, Michaela Zamponi, Jason Gardner, and K.C. Kam) was trying to determine what happens on the atomic scale when spin ice freezes—and in particular, how impurities in the spin ice, like the added lanthanum, change the freezing process. Ehlers notes that, in water ice, the frustration in the ice is in the hydrogen bonds. However, if you add certain chemical impurities to the water you can eliminate that frustration and make long-range-ordered ice crystals.

"In the spin ice," he says, "we find that the changes resulting from the introduction of an impurity are much less significant than we would expect." Ehlers explains that researchers' expectations in this regard are guided by their experience with other frustrated systems and how the freezing process is altered in these systems when impurities are added. "We found that, in spin ice, the frustration effects are much more resistant to external changes," he says.

However, as it turns out, the frustration effects in the spin ice were not entirely unaffected by these changes. The addition of lanthanum atoms, which are slightly larger than the holmium atoms they replaced, created stress in the structure of the spin ice. "This didn't create long-range order," says Ehlers, "but it created slightly different effects than putting in an impurity which is exactly the same size as the holmium. So, although we were not successful in creating long-range order and relieving the frustration completely, we found that different impurities cause different effects."

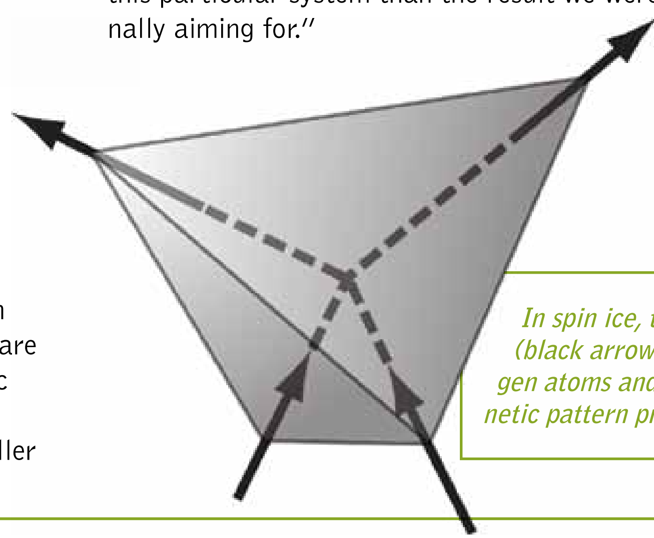
Another, more subtle, finding suggests that influence over the structure of spin ice can come from unexpected places. When physicists describe magnets on a microscopic scale, they consider the contributions of various parts of the system. "What sets frustrated magnets apart," says Ehlers, "is that the main contributions to the energy of the system often cancel each other out." This means that parts of the system that would normally have a negligible impact can sometimes play key roles in determining the magnetic configuration.



In water ice, the central water molecule can take six orientations by moving the locations of its hydrogen atoms (blue circles) among the four hydrogen bonds to its neighbors. This variation in hydrogen arrangement prevents long-range order in the ice crystals.

A system's main magnetic moments are created in the electron shells of atoms, but nuclei also can have magnetic moments. These nuclear moments are about a thousand times smaller than the magnetic moments of the electrons. "In the spin ice system we have looked at, we identified one of these smaller contributors that apparently has a major impact," Ehlers says. "That is the magnetic moments of the holmium nuclei."

The influence of these nuclei appears under fairly specialized circumstances. When spin ice is cooled, it freezes—as does ice—in an irregular, frustrated way until nothing moves. However, at the temperature where the moments of the electrons are frozen, the moments of the nuclei are still dynamic because the nuclei freeze at a much lower temperature. "It appears that the continued presence of dynamics in the nuclear moment system introduces residual dynamics in the system of the electronic moments," says Ehlers. "As a result, the sample has difficulty getting into the truly frozen ice state. We came across something unexpected and new, and it took us some time to realize what it was," he says. "It turns out that this new result should have more impact on the knowledge of this particular system than the result we were originally aiming for."



In spin ice, the direction of its magnetic moments (black arrows) is similar to the position of the hydrogen atoms and the oxygen ions in water ice. This magnetic pattern prevents long-range order in the spin ice.

So what's the next step? "It all depends on what your ultimate goal is," says Ehlers. He mentions several potential avenues of inquiry: continue his group's studies but with other parameters such as external pressure or an electric field, compare the material used in this study with other frustrated materials, and conduct theoretical studies of the importance of other energy terms and their interaction energies.

Regardless of how the research proceeds, Ehlers stresses that much of the data generated by this study could not have been gathered at any other facility. "We were studying something that we thought we already understood fairly well, but the SNS Backscattering Spectrometer allows us to see details ten times more closely than we could before."

For more information see:

G. Ehlers, E. Mamontov, M. Zamponi, A. Faraone, Y. Qiu, A. L. Cornelius, C. H. Booth, K. C. Kam, R. Le Toquin, A. K. Cheetham, and J. S. Gardner, "Frustrated spin correlations in diluted spin ice $Ho_{2-x}La_xTi_2O_7$," *J. Phys.: Condens. Matter* **20**(23), 235206 (April 2008).

G. Ehlers, E. Mamontov, M. Zamponi, K. C. Kam, and J. S. Gardner, "Direct observation of a nuclear spin excitation in $Ho_2Ti_2O_7$," *Phys. Rev. Lett.* **102**, 016405 (2009).