COMMENTARY

On the way to understanding $Yb_2Ti_2O_7$

Sylvain Petit[a,1]

As the history of science shows, research is rarely a linear process. Rather, it is built more like the roots of a tree, which branches out, retraces its steps, hesitates, and stammers, even if, on a larger scale, progress is palpable. Often, research is slow to provide answers, but, over time, it refines the "right" way to ask the question. The paper we are dealing with here is particularly interesting for what it brings, but also from this point of view.

Let me first briefly summarize the main results of this research. This paper by Allen Scheie et al. (1) is a very nice investigation of the low-energy properties of the pyrochlore material $Yb_2Ti_2O_7$, a compound which belongs to the family of geometrically frustrated systems, which has aroused tremendous interest in the two last decades. The authors combine neutron scattering experiments (elastic, inelastic, small-angle scattering), theoretical calculations, and detailed analysis, to bring light to this issue. The main point of the paper is to show the existence of multiphase magnetism in this material, corroborating the fact that the physics of $Yb_2Ti_2O_7$ is not that of a quantum spin ice but of a material precisely, and perhaps accidentally, located on a transition line between competing phases. One of the strong lessons that can be drawn from this article is that $Yb_2Ti_2O_7$ continues to be a particularly interesting and enigmatic compound that advances our knowledge of physics, but that its study was approached for "the wrong reasons," from a probably inappropriate point of view. To show the significance of these results, a long digression is necessary.

Search for New States of Matter

The last decades in solid-state research have seen the rise of rich and novel physics, beyond the Néel paradigm and transcending conventional descriptions based on Landau theory. In the very context of magnetism, "correlated paramagnets"—or "spin liquids"—have thus stimulated a lot of research activity; indeed, these systems are characterized by the emergence of fractionalized excitations, and can be accompanied by topological properties (5). The minimal "recipe" to obtain a spin liquid is to enhance the conditions that can suppress magnetic order. These include small spins (typically S = 1/2), low-dimensional systems (one-dimensional [1D], 2D), but also frustration. This concept is actually quite common, and plenty of illustrations can be found in areas as different as protein folding, stellar nuclear matter, or social dynamics. Frustrated magnetism has contributed to these developments in major ways, through new concepts like the "Coulomb phase," a highly degenerate state of matter brought to light by the discovery of spin ice in rare-earth pyrochlore networks (2–4).

Frustration, Spin Ice, and Quantum Spin Ice

Here, frustration is ensured by the low connectivity of the underlying substructure, as is the case here for tetrahedra connected by their vertices building the pyrochlore network. In a situation where it is combined, for instance, with ferromagnetic (FM) interactions and Ising spins (lying along the axes that connect a vertex to the center of its tetrahedron), frustration prevents the existence of a unique classical magnetic ground state, and yields a large (macroscopic) landscape of energetically equivalent ones instead. This does not mean that the assembly of spins forms a featureless paramagnet nevertheless: As a matter of fact, the spins are strongly correlated, with correlations governed by a "local constraint," or "organizing principle," which is satisfied within the manifold of degenerate states (6). For instance, in the case mentioned above, the case of spin ice, this organizing principle states that each tetrahedron must have two spins pointing in and two pointing out (7) (in close analogy with the two-close and two-far hydrogen atoms surrounding an oxygen in water ice; hence the name ice rule). Even more fascinating, the idea that this local constraint can be considered as the conservation law of an emergent magnetic flux (div B = 0) was quickly imposed (8–10). Quantum fluctuations can cause this flux to change with time, giving rise to an

[a]Laboratoire Léon Brillouin, Université Paris-Saclay, CNRS, Commissariat à l'Energie Atomique et aux Énergies Alternatives Centre d'études de Saclay, F-91191 Gif sur Yvette, France

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[1]Email: sylvain.petit@cea.fr.

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emergent electric field, and, eventually, to an emergent quantum electromagnetism. The quantum spin ice state hosts spinon (monopoles in the spin ice language) and photon-like excitations (11–15). Early research on $Yb_2Ti_2O_7$ proposed that this material could be a realization of this physics (16, 17).

The key point of Scheie et al.'s (1) paper is to show that researchers were on the wrong track. Rather, $Yb_2Ti_2O_7$ lives at the edge of competing FM and antiferromagnetic (AFM) phases and should not be understood as a quantum spin ice. To be quite honest, voices had already been raised in the literature to push this idea forward (18, 19). Scheie et al.'s paper quotes those studies and makes it clear that the Hamiltonian already proposed is essentially identical to the one the authors have determined. However, it is based on a decisive and comprehensive argumentation: The small-angle neuron scattering (SANS) measurements, especially, provide direct evidence for the coexistence of FM and AFM, while it was only inferred from the analyses presented earlier. Furthermore, the amazing sample quality allows observation of spin waves, an experimental feat that had never been done before. The paper also proposes an interpretation of their width, in terms of admixtures of a large number of individual different responses arising from static magnetic configurations extremely close to each other. The argumentation is built on the following points:

1) A description of the experimental conditions of the various techniques and of the preparation of the sample is provided. It is worth mentioning that growing single crystals of this quality is a tour de force.
2) A further section details inelastic neutron scattering data. It confirms previous reports, yet the exceptional quality of the sample allows deeper insight, as shown by the comparison with the still-remarkable work of other research groups. The present analysis also shows that the interpretation is compatible with the averaged superimposition of different spin wave spectra, arising from AFM and FM static configurations.
3) A section is devoted to SANS measurements, a relatively new experimental technique in the community of researchers on frustrated magnetism. The data show rod-like features along (111) directions, attributed to lamellar domains walls extending perpendicular to the $\langle 111 \rangle$ crystallographic axis. The authors then propose a plausible interpretation in terms of walls switching from FM to AFM and then back to FM again.
4) Finally, convincing Monte Carlo simulations probing the stability of the AFM and FM phases are also reported. The spin wave spectra calculated based on the average of each response tend to confirm the results about the apparent broadening.

In their final discussion, the authors (1) are concerned about the first-order character of the transition to the ordered state, while their result rather pleads for a continuous transition. At the same time, this first-order character, in itself, suggests the coexistence of phases, which a continuous transition would likely not do. Finally, even if the idea of interpreting the width of the excitations as the superimposition of several individual spectra is interesting and allows progress, it is not certain whether it might correctly account for the spin dynamics in the immediate area of the stability lines of three phases, of which two, moreover, are degenerated. It is difficult to imagine that the fluctuations in the so close vicinity of three different phases do not play any role, leading to some kind of unconventional excitations.

Beyond these considerations, this article perfectly illustrates two seemingly fundamental points in condensed matter physics. This is only sensical, but it is important to insist. The first one concerns the sample quality. A good part of success is there. It is essential that solid-state chemists and physicists work together to move forward. Secondly, it is clear that neutron scattering is a technique of impressive strength and formidable efficiency to investigate quantum materials, and makes it possible to bring to light microscopic details, to see complex magnetic structures, correlated disorder via diffuse scattering, collective excitations (sometimes exotic), and features of magnetic domains.

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