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Commentary on "An intermediate state between the kagome-ice and the fully polarized state in $Dy_2Ti_2O_7$"

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The authors of Ref. [1] provide experimental evidence on the existence of an intermediate state between the kagome-ice and the fully polarized state in $Dy_2Ti_2O_7$ single crystals and try to support this finding with theoretical calculations. After recalling the state-of-the-art knowledge of these systems, the authors present AC susceptibility measurements and Monte-Carlo simulations tentatively performed to reproduce and better characterize this intermediate state. The spin-ice systems are a hot topic in molecular magnetism because they represent a class of model systems to study frustration not only experimentally, but also theoretically. The magnetic moments of rare earths in the pyrochlore derivatives $Ln_2Ti_2O_7$ (Ln=Dy,Ho) sit on a lattice of corner-shared tetrahedra and have Ising character, as a result of the crystal field. In this geometry, the spin configuration "two in two out" minimizes the (partially free) magnetic charges, and thus the dipolar energy, within each tetrahedron. With this "ice rule", it is impossible to fulfill simultaneously all the six ferromagnetic nearest-neighbor couplings among spins. Therefore, frustration arises already at the level of a single tetrahedron, with a six-fold degenerate ground state. This degeneracy is enhanced at the bulk level, where it produces a macroscopic entropy at low temperature. For this reason, spin-ice systems possess a macroscopic number of quasi-degenerate ground states and a low-temperature entropy very similar to the one predicted by Pauling for protons in water. The study presented in this paper clearly evidences the occurrence of an intermediate state between the kagome-ice and the fully polarized state. The specific-heat measurements suggest that there is some disorder in this state. It would be interesting to investigate more in detail this spin-disorder by using a multi-technique approach, for example, using cantilever magnetometry on single crystals coupled with a sub-Kelvin cooling system and with proper experimental setup. Moreover, different degrees of disorder are expected to split differently the energy levels of rare earths: Electronic Paramagnetic Resonance measurements could, in principle, provide useful information about the new energy levels structure and, indirectly, about the newly discovered spin-ice phase. Alternatively, neutrons and muons could also be employed to extract useful information about the phonons and the local magnetic fields that are related to the spin structure of the system. The knowledge of the actual type of ordering of this novel intermediate state would be precious to deeply understand the role and the weight of the dipolar interaction in the Spin Hamiltonian used for the rationalization of the spin-ice behavior. Indeed, the treatment that is commonly used for modeling these systems uses Ewald summations to obtain an absolutely convergent effective dipole-dipole interaction between two spins,

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but this method does not provide any evidence of the intermediate state. For this reason, a refinement of the model is mandatory. Indeed, the authors attempt to reproduce the intermediate phase using the generalized dipolar spin-ice model, proposed in Refs. [2,3] that has successfully explained many other features of these materials. They perform Monte-Carlo simulations on a 2D Ising lattice in which the Ewald summations are used to handle dipole-dipole interactions among spin pairs. Unfortunately, the intermediate phase observed in experiments is not encountered in the Monte-Carlo simulations, which points out the need to develop more sophisticated theoretical models. The study conducted on this pyrochlore derivative could be easily extended to $Ho_2Ti_2O_7$ to investigate if this partially-disordered state is a common feature of the two systems. All the aforementioned suggestions could help achieve a complete scenario of the magnetic behavior of the pyrochlore spin-ice systems that is necessary for the development of a more accurate model.

Concluding, this study provides a further step in the understanding of the magnetic behavior of these systems. On one side, it reveals the presence of an intermediate state between the kagome-ice and the fully polarized state in $Dy_2Ti_2O_7$ that was not detected so far. On the other side, it points out that the theory commonly used to describe the magnetic behavior of this compound should be refined in order to account for all the states/phases through which the system passes while the temperature is varied.

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