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Magnetostructural Phase Diagram of Multiferroic $(ND_4)_2FeCl_5{\cdot}H_2O$

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Introduction

Spin and polarization flop transitions are fascinating, especially when controlled by external stimuli like magnetic and electric field and accompanied by large material responses involving multiple degrees of freedom. Multiferroics like $MnWO_4$, $TbMnO_3$, and Ni_3TeO_6 are flagship examples and owe their remarkable properties, for instance field control of polarization and polarization flops combined with spin helix reorientation, to the anisotropy and heavy centers that bring in spin-orbit coupling. The family of $A_2FeX_5{\cdot}H_2O$ erythrosiderites (A = K, Rb, NH_4; B = Fe, Mn, Co; X = Cl, Br, H_2O) drew our attention due to the rich chemical tuning possibilities, complex phase diagrams, and topological similarities to oxide multiferroics.[1] $(NH_4)_2FeCl_5{\cdot}H_2O$ is the flagship example (Fig. 1(a)). It displays a high temperature order-disorder transition involving long-range hydrogen bonding of the NH_4^+ group and two successive low temperature magnetic transitions below which non-collinear magnetic order and ferroelectricity are established.[1] In addition to the magnetically-induced electric polarization that arises below 6.9 K (P = 3 $\mu C/m^2$ along a and a smaller component along b), applied field reveals a peculiar hysteretic spin flop transition near 4.5 T above which polarization flops from the a- to the c-axis.[1] There are elastic components as well. Taken together, these findings raise questions about the interactions that induce this behavior and whether additional non-equilibrium phases might be accessed under even higher magnetic fields.

Results and Discussion

In order to deepen the understanding of spin-charge mixing in magnetically-driven multiferroics, we recently carried out high field magnetization measurements of $(NH_4)_2FeCl_5{\cdot}H_2O$ and the deuterated analog at 0.6 K and 65 T at the National High Magnetic Field Laboratory in Los Alamos (Fig. 1(b)). In addition to revealing the two-component nature of the spin-flop transition from the derivative of $M(B)$ (dovetailing perfectly with on-going inelastic neutron scattering measurements (Fig. 1(c)), these experiments uncover a magnetic quantum phase transition near 30 T. The latter is anisotropic. A preliminary magnetic field-temperature phase diagram for $B \parallel a$ is shown in Fig. 1(d). An analysis of the critical exponents is on-going.

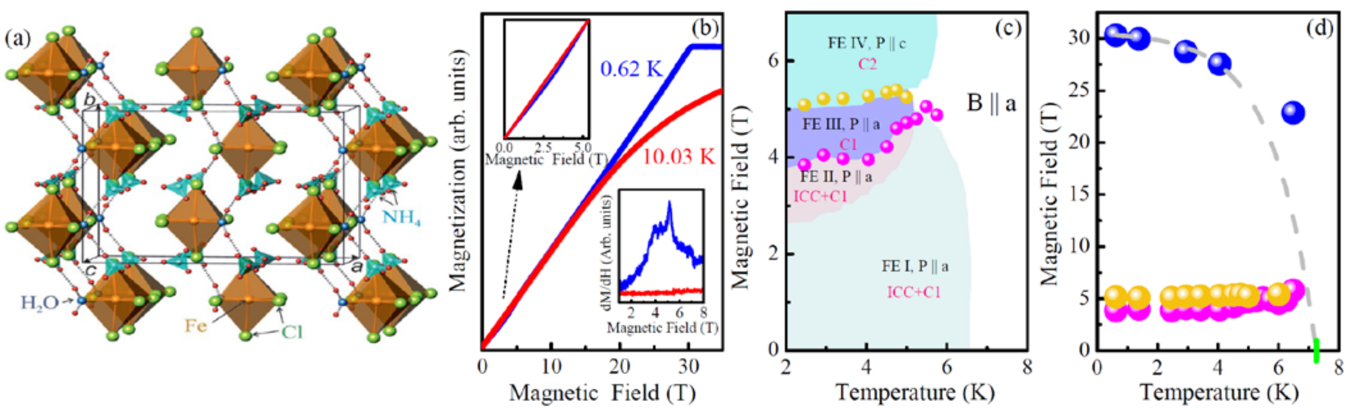

Fig. 1: (a) Crystal structure of $(ND_4)_2FeCl_5{\cdot}D_2O$. (b) Pulsed field magnetization at temperatures above and below the ordering transition. (c) $B - T$ phase diagram of $(ND_4)_2FeCl_5{\cdot}D_2O$ drawing together the low field magnetization data and inelastic neutron scattering to reveal the magnetoelectric response. (d) Developing $B - T$ phase diagram of $(ND_4)_2FeCl_5{\cdot}D_2O$ at higher fields revealing the 30 T magnetic quantum phase transition.[2]

Acknowledgements

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References

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