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The Magnetic and Structural Properties of Layered Materials

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Summary

Theoretical work has shown that magnetically frustrated centrosymmetric materials with trigonal symmetries are promising candidates for exhibiting phases that host magnetic skyrmions. The main aim of this thesis is to investigate the (magneto)structural and magnetic properties of layered frustrated magnetic compounds with trigonal symmetries.

A magnetic skyrmion is a two-dimensional swirling spin texture, akin to a magnetic vortex, which is topologically non-trivial. Skyrmions possess a topology-related energy barrier which provides additional stability. Skyrmion phases also exhibit emergent electromagnetic phenomena such as ultralow-current-driven motion and the topological Hall effect. These properties make magnetic skyrmions attractive candidates for magnetic data storage applications such as racetrack memories, information carriers in spin-logic devices and artificial neurons or synapses in neuromorphic applications. In this thesis several compounds are investigated for their potential to host skyrmions.

Chapter **3** describes the synthesis and magnetostructural properties of single crystals of FeCl$_3$. Previously performed studies leave several key questions open regarding its magnetic ordering and the evolution of the field-dependent magnetization. In this chapter, we show that an antiferromagnetic structure is obtained below $T_N = 8.57$ K which is best described by a propagation vector that differs from the previously reported magnetic structure. However, disordered crystallographic models including a large density of stacking faults are required to accurately reproduce the diffuse scattering observed in powder neutron diffraction patterns. This prevents a full determination of the magnetic structure. A field-induced spin reorientation process is observed at $H = 40$ kOe ($H \parallel$ *c*-axis) and at significantly lower fields of $H \approx 25$ kOe ($H \perp$ *c*-axis) where the spins lie predominantly in the basal plane.

By inserting a single layer of graphite (graphene) between the FeCl$_3$-layers, the effect of increasing interlayer spacing on the magnetic properties can be investigated. In Chapter **4**, the magnetic properties of stage-1 graphite intercalated FeCl$_3$ are investigated. In previous studies inconsistent and sample-dependent behavior of the magnetic properties is reported. Therefore, there are large uncertainties regarding the nature of the magnetic ground state and the evolution of the magnetic phase under applied magnetic fields. This chapter reveals that in small applied magnetic fields, two magnetic phase transitions occur sequentially. A cluster glass-like phase, which is characterized by slow magnetization dynamics, is observed at $T_{f1} \approx 4.2$ K. The cluster glass-like phase transitions into a long-range ordered phase at $T_{f2} \approx 2.7$ K. This investigation allows construction of a complex magnetic phase diagram that accounts for all the observed magnetic phase transitions as a function of temperature and magnetic field.

Compounds belonging to the transition metal/ethylenediamine/SO_4 system commonly adopt crystal structures that lead to geometric frustration. Chapter **5** describes the hydrothermal synthesis and magnetocaloric properties of single crystals of the novel layered organic-inorganic hybrid material $Co_4(OH)_6(SO_4)_2[enH_2]$. We show that in a low applied magnetic field of 1 T, easily accessible by permanent magnets, the values for the magnetic entropy and the adiabatic temperature change are among the highest reported for any material in the 15 – 25 K range. This temperature range is relevant for hydrogen liquefaction. This study shows the promise of transition metal-based hybrid materials as magnetocalorics, a field currently dominated by rare-earth containing intermetallics.

In Chapter **6**, the hydrothermal synthesis and structural properties of four novel Mn- and Fe-based amine-templated hybrid compounds belonging to the transition metal/ethylenediamine/SO_4 system are described. We show that $Mn_3(SO_4)_3(OH)_2(H_2O)_2(NH_4)_2$ undergoes a ferro/ferrimagnetic phase transition and that the magnetic behavior of $Fe_7(SO_4)_{15}(OH)_3H_2O[enH_2]_6$ is indicative of an antiferromagnetic phase transition.