DIMENSIONALITY OF MAGNETISM IN TRIRUTILE $CoTa_2O_6$ AND ITS DERIVATIVES

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Abstract

In this thesis, we addressed the question of low dimensionality of trirutile compound $CoTa_2O_6$ and studied how the low dimensionality evolved with doping of Mg on Co-site. In order to study low dimensionality in $CoTa_2O_6$ and its derivative compounds $Co_{1-x}Mg_xTa_2O_6$ (x = 0.1, 0.3, 0.5, 0.7, and 1), we used different techniques: X-ray diffraction, magnetic susceptibility, magnetization, specific heat and elastic neutron diffraction. We have addressed the question of low dimensional magnetism of $CoTa_2O_6$ by preparing phase-pure samples of the compound. In $CoTa_2O_6$ a broad feature is observed in magnetic susceptibility at 10 K and an antiferromagnetic phase transition is confirmed to occur at 6.2 K through magnetization, specific heat and neutron diffraction. It is noted that the transition peak at 6.2 K in the parent compound is robust up to 7 T and corresponds to an antiferromagnetic transition. With the addition of Mg, the peak at the magnetic transition is suppressed. Significant short-range spin fluctuations are present in $CoTa_2O_6$ as evidenced through specific heat analysis. The analysis of magnetic susceptibility combined with neutron diffraction data points towards a quasi-low-dimensional magnetic structure in $CoTa_2O_6$ which is suppressed with the addition of non-magnetic atom due to dilution of interaction between Co-atoms. $Co_{1-x}Mg_xTa_2O_6$ with $x > 0.5$ show slight enhancement in ferromagnetism. Structural analysis using X-ray and neutron diffraction data shows anomalies in the lattice parameters corresponding to $x = 0.5$. Future studies will address the role of structure in observed magnetic features and the magnetic excitations in this trirutile compound series.

Table of Contents

List of Tables

List of Figures

Chapter 1: Introduction

Research interest in low dimensional magnetic systems is mainly due to the fact that they provide unique possibilities to realize novel magnetic ground states, magnetic excitations, possible new phases of magnetic matter incorporating the interplay of quantum and thermal fluctuations. The magnetic properties of low-dimensional spin systems are strongly dependent on *S* (spin) of the electron as well as on the existence of single-ion anisotropy, along with the physics brought about by dimensionality of the crystal structure. Control of magnetic order in low-dimensional systems can be important milestone for the nanoelectronics and spintronics. It is difficult to realize long-range magnetic order in low dimensional bulk magnetic systems as it is suppressed by the thermal fluctuation. The Mermin Wagner theorem prohibits the formation of a long-range order in bulk spin systems of reduced dimensionality [1]. There are several 1D and 2D spin systems that are interesting from the perspective of low-dimensionality. In the present thesis, we discuss the physics of a class of compounds called trirutiles of general formula MTa_2O_6 (M = transition metal) where the lattice formed by M can be treated as a square net of spins. The M lattice could also be perceived as a chain of spins aligned along a certain crystallographic axis. The trirutile structure of MTa_2O_6 is derived from the rutile (TiO_2) structure. In rutile structure, each cation, Ti^+, is surrounded by 6 oxygen ions, O^{-2}, which forms an octahedral environment. The metal cation in a trirutile has the same basic structure as rutile, but the cations will order in such a way to triple the *c*-axis. Trirutile compounds crystallize in the space group $P4_2/mnm$, space group number 136 [2]. Since the present thesis deals with the trirutile $CoTa_2O_6$, a schematic of the crystal structure is presented in Figure 1.1. In the figure, the crystal structure composed of CoO_6 and TaO_6 octahedra are shown in blue and grey colored