Experimental report

05/11/2019

Proposal: **5-41-984**

Council: 10/2018

Title: Magnetic properties of the new polymorphic phase of Co3TeO6

Research area: Materials

This proposal is a new proposal

Main proposer: **Nicolas BARRIER**

Experimental team:

Local contacts: Oscar Ramon FABELO ROSA

Samples: Co3TeO6

Instrument	Requested days	Allocated days	From	To
D9	12	6	23/07/2019	29/07/2019

Abstract:

In recent years, the transition metal orthotellurates, M3TeO6, with corundum-related structures, where M = Mn, Co, Ni, Cu, have been widely studied as multiferroic. Some of them have remarkable properties: Ni3TeO6, a gigantic magnetoelectric effect, and Co3TeO6, a rare inversion of ferromagnetic and ferroelectric domain patterns. The search for new orthotellurates, M3TeO6, with original crystalline structures is interesting for the research of new multiferroic compounds. Recently, we have synthesized a new polymorphic phase of Co3TeO6 as single crystals by transport method. The structure is based on different cobalt coordination polyhedra. Magnetic measurements showed an inclined ferroelectric or antiferromagnetic ground state and the presence of multiple phase transitions depending on the applied magnetic field. For this study, we need 12 days on D9 to correctly solve the complex magnetic structures expected at low temperature and to establish the magnetic phase diagram of this new polymorph under applied magnetic field using the normal beam geometry also available on D9.

Experiment 5-41-984
Magnetic properties of the new polymorphic phase of Co_3TeO_6

N. Barrier, S. Podchezertsev, J. Rodriguez-Carvajal, E. Suard, A. Pautrat.

Introduction

α-Co_3TeO_6 is a well-known representative of transition metal orthotellurates family. Being a type-II multiferroic this compound exhibits a rare inversion of ferromagnetic and ferroelectric domain patterns. A complex cryolite-related nuclear structure (sp. group *C2/c*) together with several successive magnetic phase transitions make explaining mechanisms of ferroelectricity within this system a challenging task. However recently we have synthesised a new polymorph β-Co_3TeO_6 with non-centrosymmetric, orthorhombic $Pna2_1$ structure. In this structure Co^{2+} ions have three different local environments: tetrahedral, octahedral and square based pyramid which gives rise to a complex magnetic behaviour. Temperature variation of magnetic susceptibility has two sharp features and shows ferri- or canted-antiferromagnetic ground state (figure 1). The aim of a current work was to study temperature evolution of β-Co_3TeO_6 for understanding a microscopic nature of a complex magnetic behaviour.

Results

Diffraction patterns were collected at 2 K, 32 K and 36 K with the use of a hot neutron single crystal diffractometer D9. Data refinement revealed complex magnetic structure which can be described with *Pn'a2₁'* (#33.146) Shubnikov space group, values of magnetic moments μ_{pyr} = 2.636(68) μ_B, μ_{oct} = 3.452(20) μ_B, μ_{tet} = 3.322(20) μ_B for Co^{2+} ions at pyramidal, octahedral and tetrahedral positions respectively (figure 2).

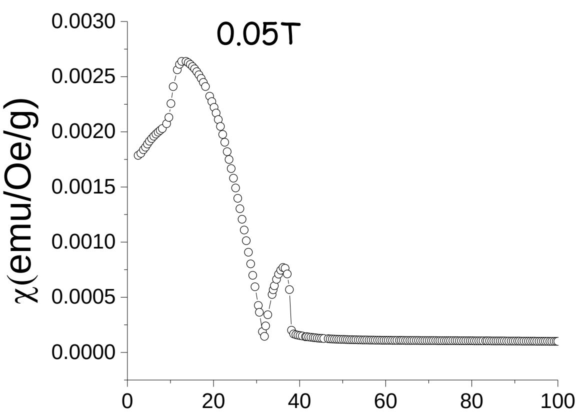

Figure 1. Temperature evolution of β-Co_3TeO_6 magnetic susceptibility

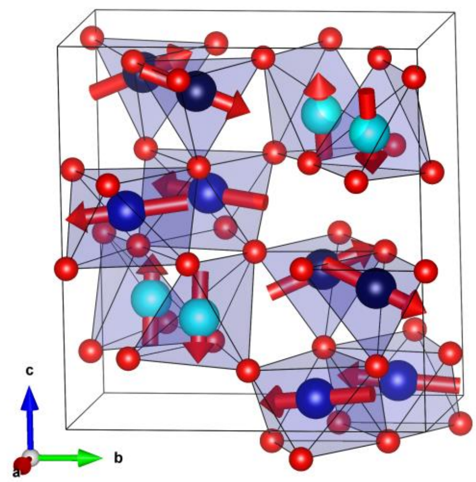

Figure 2. Magnetic structure of β-Co_3TeO_6 at 2 K

To detect possible changes within the magnetic subsystem we followed temperature evolution of four reflections with strong magnetic contribution: (0 2 0), (-1 0 -2), (1 0 0) and (1 -2 0). However no significant feature was observed in these measurements (figure 3). Nevertheless single crystal refinement of the data obtained at 32 K and 36 K provided a significant insight on macroscopic behaviour of β-Co_3TeO_6. It was revealed that for all three studied temperatures the magnetic space group remains the same. Spins at pyramidal sites compensate each other giving a zero value of total magnetization along *c*-axis whereas there's a non-zero magnetization along *b*-axis for all studied temperatures: M_b = -1.395 μ_B/u.c. at 2 K, M_b = 0.549 μ_B/u.c. at 32 K and M = 0.523 μ_B/u.c. at 36 K. Total magnetization obtained with neutron diffraction doesn't drop down to zero at 32K as it was expected from Macromagnetic measurements, nonetheless it changes sign between 32K and 2K with a change of ratios between Co^{2+} magnetic moments at octahedral and tetrahedral sites (figure 4)

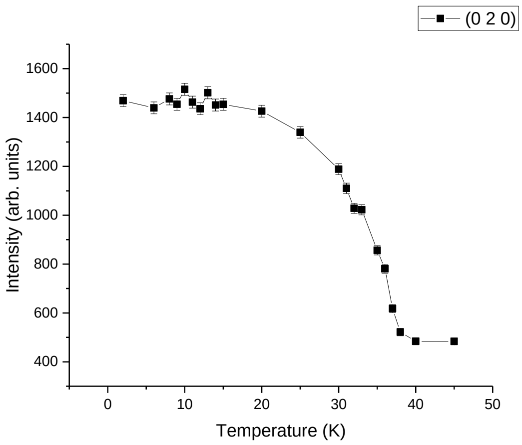

Figure 3. Temperature evolution (0 2 0) reflection

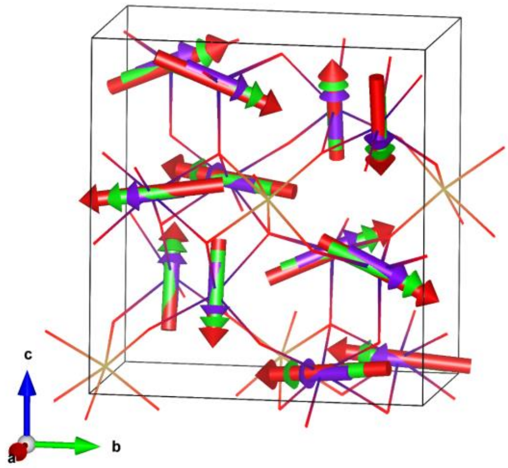

Figure 2. Magnetic structure of β-Co_3TeO_6 Red arrows - 2 K, green – 32 K and magenta – 36 K.