



	Experiment title: High Resolution Synchrotron Diffraction on the Spin Driven Jahn-Teller Effect Candidate Compound MnSc₂S₄	Experiment number: HE-2338
Beamline: ID31	Date of experiment: from: 26. Jan. 2007 to: 30. Jan. 2007	Date of report: 13. Aug. 2008
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Names and affiliations of applicants (* indicates experimentalists):

A. Krimmel, M. Mücksch, V. Tsurkan, A. Loidl, S. Horn

Institute of Physics, Centre for Electronic Correlations and Magnetism,

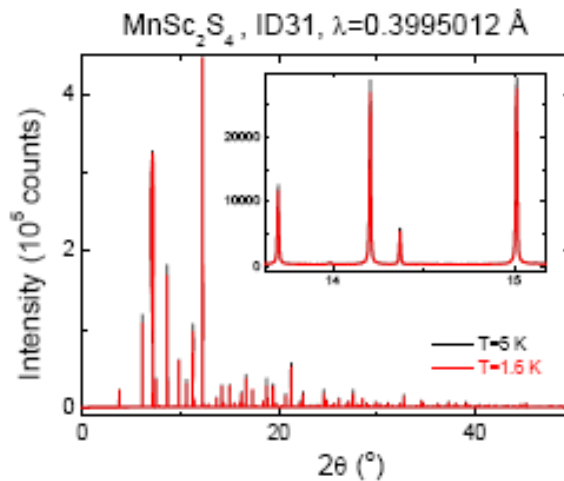
University of Augsburg, D-86159 Augsburg, Germany

P. Thompson, ESRF, BP 220, F-38043 Grenoble Cedex 9, France

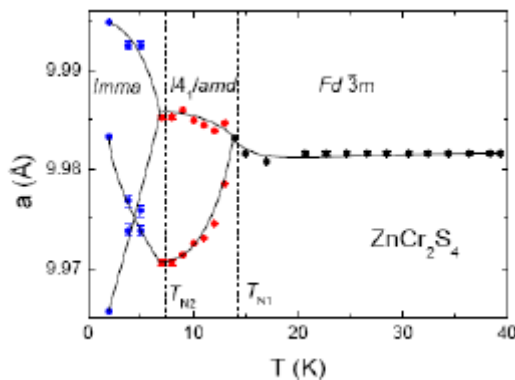
Report: Geometric frustration of spin and orbital degrees of freedom leads to highly degenerate ground states. Orbital degeneracy is usually lifted by a coupling of orbital to lattice degrees of freedom inducing orbital order with a lowering of the crystal symmetry, i.e. the orbital Jahn-Teller effect. The lifting of the spin degeneracy leading to spin order has been discussed in similarly via the spin-driven Jahn-Teller effect [1,2]. This effect may be purely dynamic in nature without involving static lattice distortions. It has been discussed for magnetically frustrated spinel compounds, in particular for Cr based ACr_2X_4 ($A=Zn, Cd, Hg$, $X=O, S, Se$) [3,4] and for MSc_2S_4 ($M=Mn, Fe$) [5,6]. In case of $MnSc_2S_4$, the magnetic spin-only ions Mn^{2+} form a diamond lattice with inherent geometric frustration. A Curie-Weiss temperature $\Theta_{CW}=-22$ K indicated antiferromagnetic (AFM) exchange. However, the compound shows a complex ordering process around 2 K into a non-collinear magnetic structure [6]. This magnetic structure is described by a propagation vector $\mathbf{k}=(0.75, 0.75, 0)$ indicating 3 spin rotations in a fourfold supercell within the a - b plane. High resolution neutron powder diffraction experiments gave no indications for any structural changes. However, the anisotropic cycloidal spin structure is incompatible with the cubic $Fd\bar{3}m$ symmetry of the spinel structure. We therefore performed high-resolution synchrotron powder diffraction experiments on ID31 to look for corresponding lattice distortions. The main experimental difficulty was the low ordering temperature of 2 K. The sample was placed in an aluminum container and mounted in a liquid-helium-cooled cryostat. Measurements could be performed in a temperature range 1.6 K - 40 K. An incident wavelength of $\lambda=0.3995$ Å in combination with a large range of the scattering angle $-5.964^\circ < 2\theta < 67.968^\circ$ allowed

measurements up to 17.8 \AA^{-1} of the scattering vector $Q=4 \pi \sin(\theta) / \lambda$. A first indication for the high quality of the sample was given by the observation of sharp Bragg reflections up to the highest available Q-values. However, no difference in the diffraction patterns of MnSc_2S_4 at 1.6 K ($T < T_N$) and 5 K ($T > T_N$) could be observed, as illustrated in Fig. 1. There is no peak splitting nor additional superlattice reflection. Even a peak broadening could not be detected. This zero result strongly points to a symmetry breaking on a local scale only.

We therefore decided to continue the measurements on ZnCr_2S_4 , a further magnetically frustrated spinel compound with strong spin-lattice coupling, as mentioned above. ZnCr_2S_4 shows two first order magnetic phase transitions at $T_{N1}=15 \text{ K}$ and $T_{N2}=8 \text{ K}$. Both magnetic phase transitions are accompanied by structural distortions evidenced by a corresponding peak splitting in the diffractograms. The resulting phase diagram is shown in Fig. 2 where the temperature dependent lattice constant (referred to the cubic one) is shown. At T_{N1} , a cubic to tetragonal phase transition takes place, followed by a further transition from tetragonal to orthorhombic symmetry. These results, combined with high resolution neutron powder diffraction studies, are currently being published [7].



X-ray powder diffraction pattern of MnSc_2S_4 at $T=1.6 \text{ K}$ (red line) and $T=5 \text{ K}$ (black line). No difference could be observed.



Temperature dependent normalised lattice constant of ZnCr_2S_4 . The magnetic Transition temperatures and the space groups of the different structures are indicated. The solid lines are to guide the eye

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