Experimental report

08/02/2024

Proposal:	**CRG-2998**	**Council:**	4/2023

Title: Magnetic Structure of the spin-1/2antiferromagnetic XXZ chain PbCo2V2O8 in a transverse (1,1,0) magnetic field

Research area: Physics

This proposal is a resubmission of 5-41-1211

Main proposer: **Cintli AGUILAR MALDONADO**

Experimental team: Cintli AGUILAR MALDONADO
Konrad PUZNIAK
Bella LAKE

Local contacts: Karin SCHMALZL
Wolfgang F SCHMIDT

Samples: PbCo2V2O8

Instrument	Requested days	Allocated days	From	To
D23	6	5	24/10/2023	29/10/2023

Abstract:

The one-dimensional (1D) spin-1/2 antiferromagnetic with Heisenberg-Ising (XXZ) anisotropy, is a fascinating model system to explore fundamental physics such as quantum phase transitions and exotic ground states and excitations. Although no long-range magnetic order is expected in the pure 1D case, real materials which have interchain coupling develop long-range magnetic orders that reflect the nature of these ground states. The ACo2V2O8 compounds (A=Sr, Ba) are great physical realizations of the Heisenberg-Ising spin-1/2 chains. They show exotic physics in zero field as well as [1,0,0] and [0,0,1] applied fields with complex magnetic phases and excitations that can be used to test physics concepts. We have discovered a previously unreported phase for the [1,1,0] transverse field direction that is accessible in only the newly synthesized PbCo2V2O8 member of this family due to its lower energy scale. The aim of this experiment is to determine the magnetic structure of this field-induced phase using single crystal neutron diffraction.

EXPERIMENTAL REPORT

ON EXPERIMENT PERFORMED AT JCNS INSTRUMENT AT ILL OR SNS

Proposal number:	CRG-2998		
Experiment title:	**Magnetic Structure of the spin-1/2 antiferromagnetic XXZ chain PbCo2V2O8 in a transverse (1,1,0) magnetic field**		
Instrument	D23		
Dates of experiment:	24/10/23 – 29/10/23	**Date of report:**	07/02/24
Experimental team: **Names**	**Addresses**		
	Cintli Aguilar Maldonado, Konrad Puzniak, Ralf Feyerherm, Nazmul Islam, Bella Lake Helmholtz-Zentrum Berlin f ̈ur Materialien und Energie GmbH, Hahn-Meitner Platz 1, D-14109 Berlin, Germany		
Local Contact:	Karin Schmalzl		

Abstract
The one-dimensional (1D) spin-1/2 antiferromagnetic with Heisenberg-Ising (XXZ) anisotropy, is a fascinating model system to explore fundamental physics such as quantum phase transitions and exotic ground states and excitations. Although no long-range magnetic order is expected in the pure 1D case, real materials which have interchain coupling develop long-range magnetic orders that reflect the nature of these ground states. The $ACo_2V_2O_8$ compounds (A=Sr, Ba) are great physical realizations of the Heisenberg-Ising spin-1/2 chains. They show exotic physics in zero field as well as [1,0,0] and [0,0,1] applied fields with complex magnetic phases and excitations that can be used to test physics concepts. We have discovered a previously unreported phase for the [1,1,0] transverse field direction that is accessible in only the newly synthesized $PbCo_2V_2O_8$ member of this family due to its lower energy scale. The aim of this experiment is to determine the magnetic structure of this field-induced phase using single crystal neutron diffraction.

Introduction
The $ACo_2V_2O_8$ family are model materials to explore fundamental physics due to their behavior as one-dimensional (1D) spin-1/2 antiferromagnets with Heisenberg-Ising (XXZ) anisotropy. The structure of this compounds is found to have 4-fold screw chains of magnetic ions (Co^{2+}) along the tetragonal c-axis with effective spin-1/2 where the intrachain coupling is strong and antiferromagnet meanwhile the interchain coupling is weak, giving rise to long-range, antiferromagnetic Neel order at low temperatures. In zero magnetic field these compounds have a spinon continuum above the Neel temperature T_N~5K, and they were used to demonstrate spinon confinement on cooling below T_N where the continuum is replaced by sharp bound-spinon modes [1,2]. A longitudinal magnetic field applied parallel to the easy axis which is the c-axis ([0,0,1]) also shows exotic physics. Above a critical field, the antiferromagnetic order is suppressed to much lower temperatures (T<1K) and the system undergoes a transition to a longitudinal spin density wave and then a transverse canted antiferromagnet with increasing magnetic field [3,4]. In the excitation spectrum, bound states of magnons known as Bethe strings which were predicted by Hans Bethe in 1931 [5], were observed for the first time in $SrCo_2V_2O_8$ using terahertz spectroscopy [6] and inelastic neutron scattering [4]. The behavior of the XXZ chain in a transverse magnetic field along the a-axis ([1,0,0]) is equally fascinating. Recent neutron diffraction and inelastic neutron measurements in applied field perpendicular to the easy axis of $BaCo_2V_2O_8$ revealed a quantum phase transition at 10T between two different types of solitonic topological objects [7]. At a lower magnetic field of 4.7T within the ordered phase, a hidden 1D quantum phase transition was identified by NMR [8]. It has a class of universality described by the exceptional E8 Lie algebra which is characterized by 8 gapped excitations whose gaps are theoretically predicted to have precise values [9]. These excitations were measured by inelastic neutron scattering [8] and terahertz spectroscopy [10] and compared successfully to this theory [11]. A magnetic field applied along the other transverse direction [1,1,0] was shown to give a different phase diagram than the [1,0,0] transverse field, with the Neel antiferromagnetic order that was found in zero field maintained to the highest [1,1,0]-fields measured, with only a gradual reduction in transition temperature observed [12,13]. The reason why the transverse [1,1,0] and [1,0,0] field directions are different, is due the complex g-tensor for the Co^{2+} ions as investigated in Ref [14].

Objectives
We have discovered a previously unreported phase for the [1,1,0] transverse field direction that is accessible only for $PbCo_2V_2O_8$. The aim of this experiment is to determine the magnetic structure of this field-induced phase using single crystal neutron diffraction as well as corroborate the borders of the phase diagram for this novel phase.

Experimental Results
A single crystal of $PbCo_2V_2O_8$ (5g) was prepared and cut to appropriate dimensions for the experiment. The sample was aligned with the [1,1,0] direction vertical and parallel to the magnetic field direction and glued with x60 glue into a copper sample holder with the pertinent size for the 15T vertical magnet with ^{3}He insert experimental setup. A copper monochromator was required to provide a neutron wavelength of 1.28 Å, the ^{3}He insert was also required to achieve temperatures down to 0.5K since the novel [1,1,0] phase is evident below 1.8K at 9T and at lower temperatures as the field increases. Bragg peaks within the (H,H,L) plane were measured. The measurements at zero field were performed to obtain the intensities of the nuclear peaks which is especially important for the magnetic structure resolution in the case the magnetic peaks coincide with the nuclear peaks in the high field phase. The specific goal of this experiment was the corroboration of the borders

for the novel phase, for this reason variations on temperature and field were measured, 4 reflections (two structural and two magnetic) were followed along the variations.

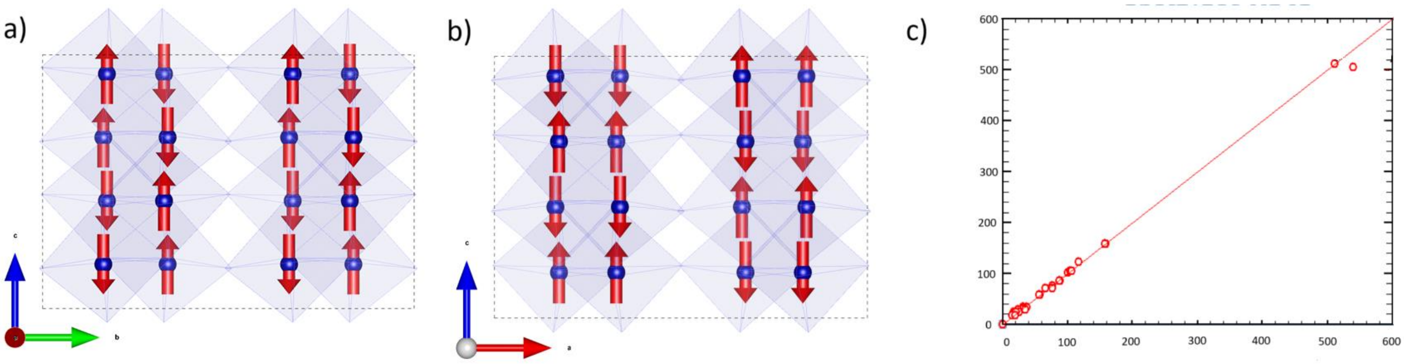
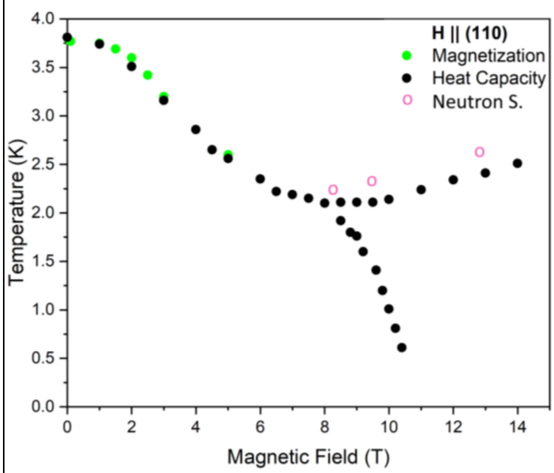


*Figure 1. Projection on the **bc**-plane (a) showing antiferromagnetic ordering along the **b**-axis, whereas the chains present a ferromagnetic coupling along the **a**-axis. (c) F_{calc} vs F_{obs} plot of the least squares fitting for the magnetic structure model for $PbCo_2V_2O_8$ at T=0.5K and μ_0H=0T.*

Preliminary refinement results confirms that $PbCo_2V_2O_8$ crystallizes within space group $I4_1cd$ (No. 110) at 0T and at T=0.5K orders as we previously reported with a k = (0,0,1) propagation vector, where the magnetic moments on the Co^{2+} ions point predominantly along the c-axis with a small canting away from the axis.[15]. The magnitude of the ordered magnetic moment obtained from the refinement of μ=2.342(2)μ_B at T=0.5K agrees well with the saturation magnetization of 2.75μ_B obtained from previous measurements and is near to the 3μ_B expected for Co^{2+} spin only, high spin state S=3/2. Fig 1a shows the projection on the **bc**-plane where it shows 4 screw chains whose moments are antiferromagnetically aligned along each chain and along the **b**-axis whereas they align ferromagnetically along the **a**-axis (fig1b). The plot F_{calc} vs F_{obs} is presented in Fig 1c showing a good fit of the magnetic structure model. A small canting is observable from the refinement but further analysis has to be done. This magnetic structure is very similar to that observed in $BaCo_2V_2O_8$ and $SrCo_2V_2O_8$. As the magnetic field increases, the magnetic structure seems to be the same but with an increase on the moment along c-axis, with μ_c=2.652(4)μ_B at 13 T, but more analysis needs to be done to confirm this behavior.



Previous results of the magnetic field-temperature phase diagram of $PbCo_2V_2O_8$ for the field direction μ_0H || [110] were obtained from the positions of the extrema in the field derivatives of M(H) and temperature derivative of χ(T), and from the position of the peaks in C_p(T) and C_p(H). The critical temperature decreases smoothly with increasing field up to 8T. Starting from 8T one can clearly see a phase boundary to a new ordered phase, whose critical field increases to 10.4T as temperature is reduced to 0.06K. From this neutron scattering experiment we were able to measure with more precision the borders of this phase, revealing a higher T_N in the region from 8T to 14T. Fig 2 shows the points obtained from magnetization (on green) and heat capacity (black), the pink open circles show the preliminary results obtained from neutron scattering but more analysis must be done.

References

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