Experimental report

16/09/2019

Proposal: 5-31-2571 **Council:** 4/2017

Title: magnetic structure determintion of(Ln1/3Mo2/3)2AlC(Ln = Nd, Dy, Tb) by unpolarised neutron powder diffraction

Research area: Physics

This proposal is a new proposal

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Samples: (Nd1/3Mo2/3)2AlC
(Tb1/3Mo2/3)2AlC
(Dy1/3Mo2/3)2AlC

Instrument	Requested days	Allocated days	From	To
D20	3	4	07/06/2018	11/06/2018
D2B	2	0		
D1B	0	0		

Abstract:

Magnetic MAX phases are promising new materials for fundamental studies as well as spintronic applications. Very recently, we have synthesized the first MAX phases containing rare earth elements, (Ln1/3Mo2/3)2AlC. We have done thorough structural and magnetic characterizations. This series crystalize in a monoclinic C2/c structure, in which Ln form quasi-one dimensional chains. We have found a rich variety of magnetic behavior in this series. (Ln1/3Mo2/3)2AlC, Ln=Nd, Tb and Dy are especially intriguing. Preliminary neutron diffraction study on the (Nd1/3Mo2/3)2AlC sample suggests a chiral order. Here we propose to do high-resolution measurements on D2B to parameterize the magnetic structure of the three compounds, and High-flux measurements to determine the magnetic structures as a function of temperature at D20.

Magnetic structure of $(Mo_{2/3}Ln_{1/3})_2AlC$ (Ln = Nd, Dy, Tb) by unpolarised neutron powder diffraction

MAX phases are a group of atomically laminated materials based on a transition metal (M) which can be partially substituted with a rare earth, an A-group element (A), and carbon or nitrogen (X), combining the characteristics of metal and ceramic [1]. We have synthesized rare earth elements containing MAX phases, $(Mo_{2/3}Ln_{1/3})_2AlC$, and systematically studied their magnetic behaviors [2]. We were interested in the magnetic structure of Ln= Nd, Dy, Tb.

Based on magnetization and heat capacity measurements, Ln = Nd orders below T_N=7.6 K; Ln = Dy shows two magnetic transitions at T_{N2} =16 K and T_{N1} =13 K, respectively; Ln = Tb shows two magnetic transitions at 28 K and 20 K, respectively. We aim to determine the magnetic structures of these materials by neutron powder diffraction.

We performed neutron powder diffraction at D20 on three samples, $(Mo_{2/3}Ln_{1/3})_2AlC$ (Ln = Nd, Dy, Tb). 5g of Ln = Nd and Tb are used, while about 1g Ln = Dy are used. In this experiment, we systematically characterized the crystal structure and magnetic structure of all three samples. First, we used 1.54 Å wavelength with 90° takeoff angles to collect data to refine the crystal structure at each temperature. Then we used 2.41 Å wavelength with 42° takeoff angles down to 1.5 K for magnetic structure refinement.

We determined the magnetic structures of $(Mo_{2/3}Tb_{1/3})_2AlC$ and $(Mo_{2/3}Dy_{1/3})_2AlC$. Their magnetic structures are similar. The low temperature structure below T_{N1} is antiferromagnetic with $\boldsymbol{k}$ =(0, ½, 0). The structure consists of alternating magnetic and nonmagnetic Tb in the *a*-*c* plane along *b* axis. Between T_{N1} and T_{N2} , it orders in a spin density wave structure with $\boldsymbol{k}$=(0, 0.617, 0) for $(Mo_{2/3}Dy_{1/3})_2AlC$ and $\boldsymbol{k}$=(0, 0.605, 0) for $(Mo_{2/3}Tb_{1/3})_2AlC$. Schematics for the two structures are shown in Fig. 1.

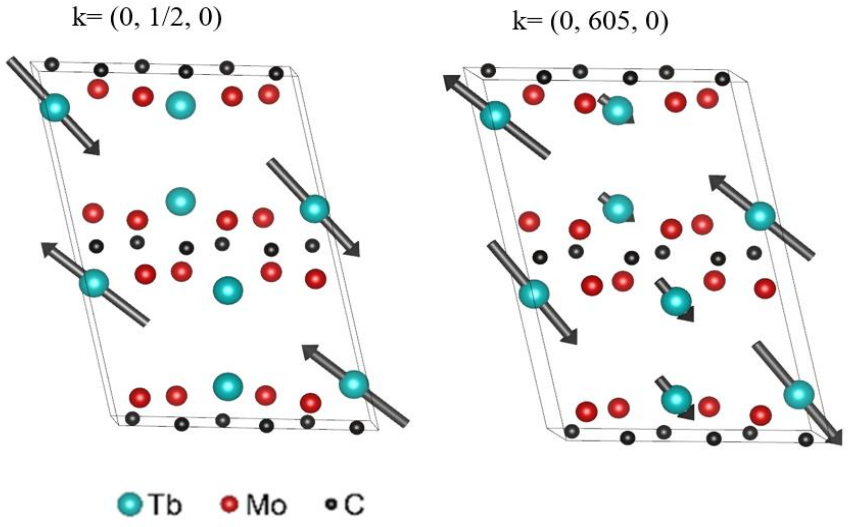

Fig. 1 schematic structures of $(Mo_{2/3}Tb_{1/3})_2AlC$. Below T_{N1}=20 K, it orders in an antiferromagnetic structure with alternating magnetic and nonmagnetic Tb in the *a*-*c* plane. Between 20 K and 28 K, it orders in a spin density wave structure.

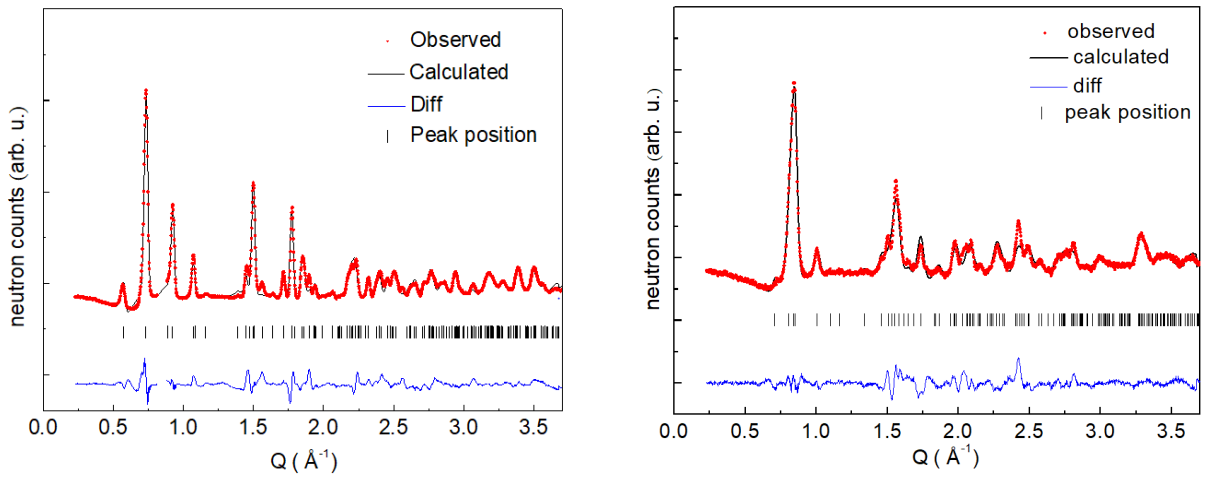

Fig. 2 neutron powder diffraction of $(Mo_{2/3}Dy_{1/3})_2AlC$ **a**. magnetic scattering at 1.5 K. **b**. magnetic scattering at 14 K. The nuclear scattering was subtracted by the data measured at 18 K.

Figure 2a. shows the magnetic scattering of $(Mo_{2/3}Dy_{1/3})_2AlC$ at 1.5 K. The peaks can be indexed with $\boldsymbol{k}$=(0, 0.5, 0). Symmetry analysis for this type of $\boldsymbol{k}$ with *C*2/*c* space group can be found in Ref.[2]. By considering all possible magnetic model compatible with the symmetry, the best solution is found by considering φ_1, φ_3, and φ_6 of Γ_2 with magnetic R-factor 12.7%. Figure 1b shows the magnetic scattering at 14 K. The peaks can be indexed with $\boldsymbol{k}$=(0, 0.617, 0). The $\boldsymbol{k}$ is close to the one found for $(Mo_{2/3}Tb_{1/3})_2AlC$, $\boldsymbol{k}$= (0, 0.605, 0). After considering possible models, the best solution is φ_1, φ_3, and φ_6 of Γ_2 with magnetic R-factor 10.7%. Refinement on $(Mo_{2/3}Tb_{1/3})_2AlC$ gives similar R-factor for each structure.

Results from this experiment will be reported together with results obtained from another experiment at ORNL in which we did neutron powder diffraction in magnetic field on these three materials. Manuscript is in final preparation. Results on $(Mo_{2/3}Nd_{1/3})_2AlC$ are found to be more complicated. We couldn't determine its propagation vector, and we plan to do single crystal neutron diffraction to determine its propagation vector and then refine the powder diffraction data.

REF:

[1]. Q. Tao etc. Nature communications 8, 14949, 2017

[2]. Q. Tao etc. Chemistry of Materials 31 (7), 2476-2485, 2019