Experimental report

Proposal: 5-31-2941	**Council:** 10/2022

Title: Crystallographic and magnetic structures of the new honeycomb material BaCo2-xMgx(AsO4)2 (x=0, 0.1, 0.3)

Research area: Physics

This proposal is a new proposal

Main proposer: **Manila SONGVILAY**

Experimental team: Armand DEVILLEZ
Virginie SIMONET
Manila SONGVILAY

Local contacts: Claire COLIN

Samples: BaCo2(AsO4)2
BaCo1.7Mg0.3(AsO4)2
BaCo1.9Mg0.1(AsO4)2
BaCo2(AsO4)2, BaCo2-xZnx(1sO4)2 (x= 0.1, 0.3)

Instrument	Requested days	Allocated days	From	To
D1B	5	4	26/05/2023	30/05/2023

Abstract:

BaCo2(AsO4)2 is a compound comprising a honeycomb lattice of magnetic Co2+ ions. It exhibits exotic magnetic properties, with an incommensurate magnetic order below 5 K, which is still not fully understood. In particular, it displays anisotropic in-plane magnetic correlations lengths, which remain unexplained so far as no structural distortion of the honeycomb planes has been reported. Moreover, recent neutron inelastic measurements reveal magnetic excitations incompatible with an incommensurate magnetic order, thus highlighting discrepancies between the static and dynamic properties. In order to further understand these discrepancies, we propose to disturb the magnetic ground state and study the effect of non-magnetic substitution in the newly synthesized compounds BaCo2-xMgx(AsO4)2 (x = 0, 0.1 and 0.3) and perform a detailed temperature dependence study of the structure and magnetic propagation

Crystallographic and magnetic structures of the new honeycomb material BaCo$_{2-x}$Mg$_x$(AsO$_4$)$_2$ (x = 0, 0.1 0.3)

Note: We decided to measure Zn-substituted samples instead as our preliminary X-Ray and magnetisation measurements indicated that the Zn compounds were less likely to contain magnetic impurities. The evolution of the ordering temperature seems to follow the same trend (see fig. 1) as for the Mg-substituted compounds.

Context and scientific goal

BaCo$_2$(AsO$_4$)$_2$ is a quasi-two-dimensional magnet, comprising honeycomb planes formed by magnetic Co^{2+} ions. It exhibits an unusual magnetic order below T$_N$ = 5 K, which consists in a colinear spin arrangement, yet described by an incommensurate propagation vector **k** = (0.26 0 -4/3). In particular, it displays anisotropic in-plane magnetic correlation lengths, which remain unexplained so far, as no structural distortion of the honeycomb planes has been reported. Moreover, recent neutron inelastic measurements reveal magnetic excitations incompatible with an incommensurate magnetic order, thus highlighting discrepancies between the static and dynamic properties. In order to further understand these discrepancies, this experiment aimed at studying the effect of non-magnetic substitution in the newly-synthesized compounds BaCo$_{2-x}$Zn$_x$(AsO$_4$)$_2$ (x = 0, 0.1 and 0.3). The goal of the experiment was two-fold:

- From preliminary magnetisation measurements, we could observe a decrease of T$_N$ with Zn content, showing that non-magnetic substitution on the Co^{2+} sites may disrupt the magnetic ordering. Therefore, we performed detailed temperature dependence measurements using λ = 2.52 Å in order to follow the evolution of the magnetic propagation vector as a function of Zn content, and check for any sign of diffuse scattering indicating the presence of short-range correlations.
- As no detailed structural study has been carried out so far, we also performed higher resolution measurements using λ = 1.28 Å in order to study the effect of Zn doping on the Co-Co distances and Co-O-Co bonds. Indeed, observing any change in the honeycomb structure may help to rationalise the changes in the magnetic order.
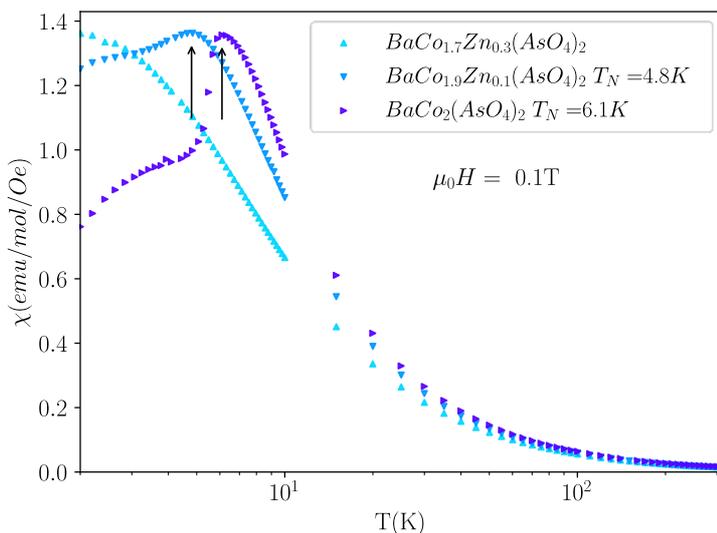

Fig. 1. (a) Susceptibility curves as a function of temperature for the three compositions. The inset shows the decrease in the Néel temperature (black arrow) as a function of Zn content.

Results

Fig. 2 shows the evolution of the diffraction data as function of Zn content (x = 0 in blue, x = 0.1 in red, x = 0.3 in green), measured at T= 1.6 K. As seen in the low-angle data, some magnetic Bragg peaks disappear with increasing *x*, while a broad diffuse scattering seems to grow at low two-theta.
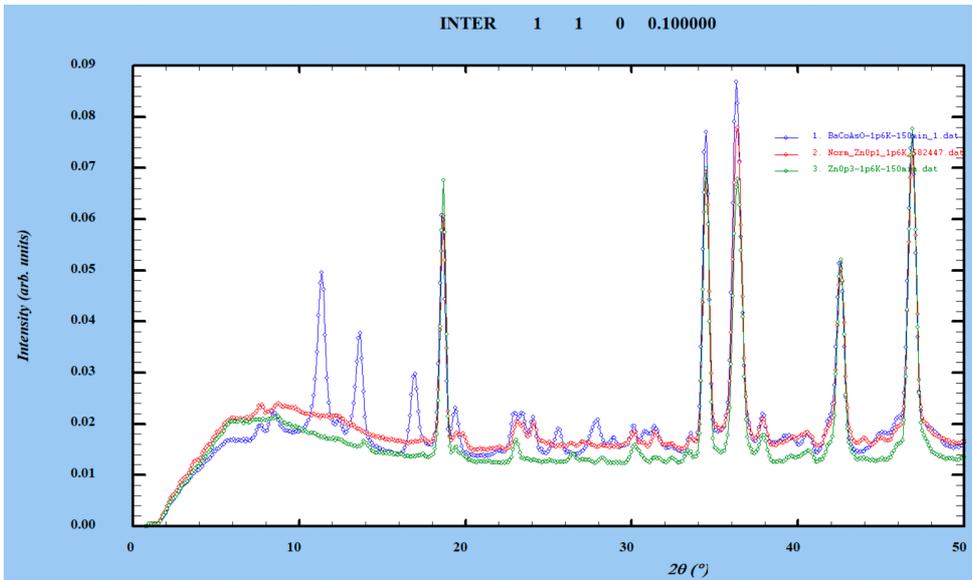
Fig. 2. Diffraction data at low angle for x=0 (blue), x = 0.1 (red) and x = 0.3 (green). The data was collected at 1.6 K with λ = 2.52 Å

Temperature dependence:

x = 0:

Fig 3. Displays the diffraction data of the pure compound BaCo2(AsO4)2 at several key temperatures. We can clearly observe several magnetic peaks which disappear at different temperatures. By following the temperature evolution of each peak, we could identify at least three different phases. As indicated by the arrows, four of these peaks belong to $BaCo_2(AsO_4)_2$. The others, however, seem to belong to other magnetic impurities.

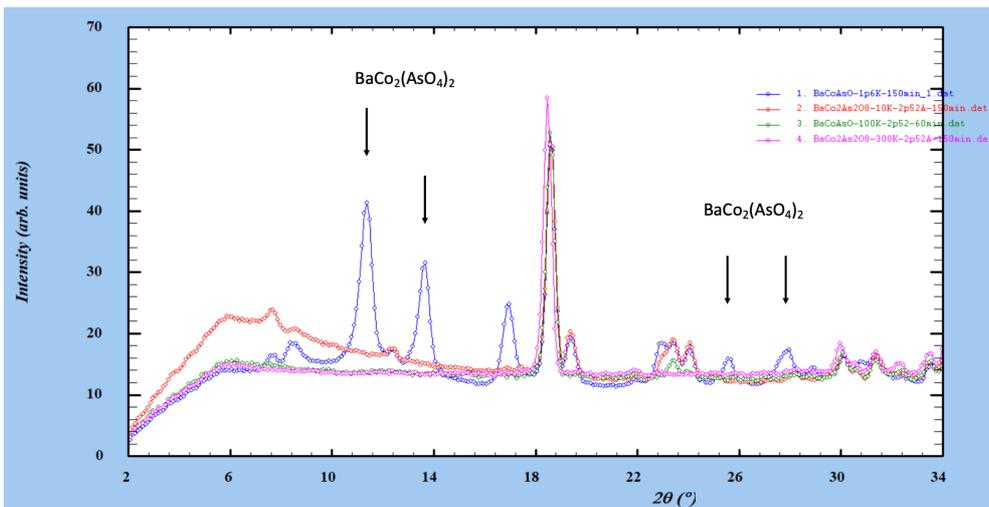

Fig. 3. Diffraction data at low angle for x=0, measured at T = 1.6 K (blue), 10 K (red), 100 K (green) and 300 K (magenta) with λ = 2.52 Å

x = 0.1 and 0.3:

As shown on figures 4 and 5, some of the magnetic peaks around 2θ = 22° which were identified as magnetic impurities in the pure compound are still there. Moreover, a broad diffuse signal has replaced the well-defined magnetic Bragg peaks associated to the magnetic order in $BaCo_2(AsO_4)_2$. This diffuse scattering decreases with increasing temperature so it may indicate that some short-range magnetic correlations have replaced the long-range magnetic ordering.
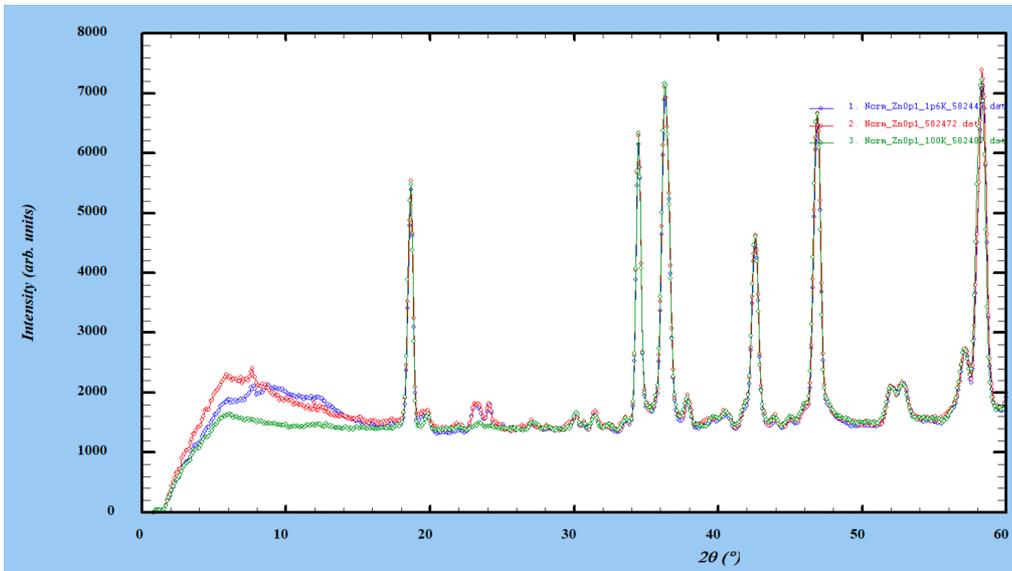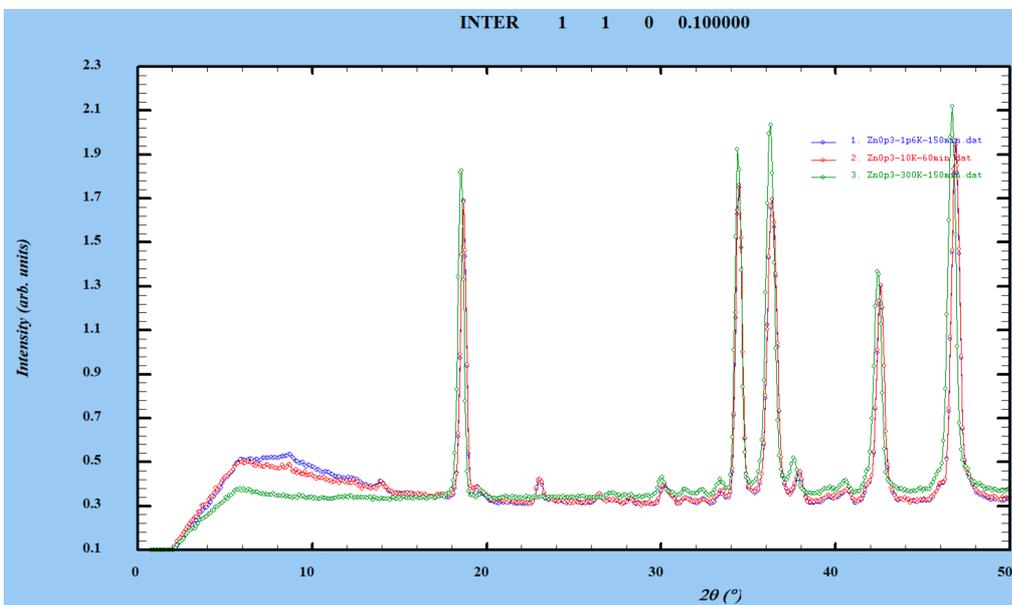
Fig. 4 and 5. Diffraction data at low angle for x=0.1 and 0.3, respectively, measured at T = 1.6 K (blue), 10 K (red) and 100 K (green) with λ = 2.52 Å

Conclusion

The presence of these magnetic impurities was not detected in our X-ray diffraction data and magnetisation measurements. Yet, the associated signal makes the neutron data analysis very difficult. The analysis of the data collected with λ = 1.28 Å is still in progress in order to identify the impurity phases but we will consider other types of substitution in order to improve the synthesis and obtain high quality samples.