Experimental report

14/11/2022

Proposal: **5-42-566** **Council:** 4/2021

Title: Searching for magnetic defects in the two-dimensional honeycomb lattice compound BaCo2(AsO4)2

Research area: Physics

This proposal is a new proposal

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Samples: BaCo2(AsO4)

Instrument	Requested days	Allocated days	From	To
D33	3	3	01/10/2021	04/10/2021

Abstract:

We propose to re-explore the magnetic structure of BaCo2(AsO4) in the context of Kitaev physics, following recent theoretical proposal to realise bond-dependent interaction in cobalt-based honeycomb lattice systems. BaCo2(AsO4), in particular, exhibits a peculiar magnetic structure at zero field, consisting of pseudo chains along the b-axis alternating and forming an incommensurate ++-- pattern along a*. Under a magnetic field applied along b, this structure becomes metastable and highly 2D. Correlations lengths are highly anisotropic within the honeycomb layers, despite its perfectly regular structure. The presence of magnetic domain walls was proposed to explain these features. We would like to look for signatures of magnetic defects using SANS in zero-field and under a magnetic field (0-0.5 T) at several temperatures. We request 3 days on D33 with a wavelength of 6A to perform this study.

ILL 5-42-566 : Searching for magnetic defects in the two-dimensional honeycomb lattice compound $BaCo_2(AsO_4)_2$

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1/ Motivations

The $BaM_2(XO_4)_2$ compounds -where M = (Co, Ni) and X = (As, P)- are a very nice playground where to explore the properties of 2D-XY magnetism[1]. This series is currently receiving a renewed interest thanks to its crystal structure, made of stacked honeycomb layers formed by the M ions. In particular, Co-based systems such as $BaCo_2(AsO_4)_2$ (BCAO) are believed to be good candidates for practical realizations of the so-called Kitaev model, which requires bond-dependent anisotropic interactions favored by the entanglement between spin and orbital degrees of freedom[2].

The ground state (GS) magnetic structure of BCAO was first assumed to be helimagnetic (HM) with spins lying in the honeycomb plane, in view of its incommensurate ordering wavevector **k** = (0.27, 0, -1.33) and the very strong planar anisotropy of its magnetic susceptibility. This "simple" planar HM ordering was however disproved by a thorough 3D neutron polarimetry study, summarized in the recent work of Regnault *et al.*[3]. It was shown that the GS of BCAO is likely formed by a sequence of pseudo spin chains, stacked in a "++--" sequence w.r.t. the **a*** axis in zero field, which evolves towards a ferrimagnetic (FIM) state under a magnetic field applied in the honeycomb plane. This transitions should be accompanied with the proliferation of domain walls (DW) between the six possible FIM sequences ("++-", "+--", *etc.*). This picture is in fact more consistent with the observation of (gapped) spin waves with energy minima at the zone center (as opposed to a Goldstone mode at **Q** = **k**) and can be checked by inspecting the evolution of the mesostructure formed by the elongated DW.

In this context, we have proposed to study BCAO using SANS, which is the most relevant technique to study extended magnetic textures. We expected to observe streaks of magnetic scattering, corresponding to the formation of 1d ferromagnetic (FM) DW, thereby explaining the continuous evolution of the z-component of **k** from the GS to the FIM state and confirming the puzzling pseudo-chain structure.

2/ Experiment

$\vec{H} \parallel \vec{b} \perp \vec{k_i}$ **–** We started the experiment with temperature-scans in zero applied field to check for the evolution of magnetic SANS between the ordered and paramagnetic (PM) regimes. The scattering curves are isotropic and can be well described by power laws Q^{-n} with an exponent n ≈ 3.7, indicating that the signal is dominated by nuclear scattering. As shown in **Fig. 1a**, an inspection of the temperature-dependence of the integrated SANS intensity reveals no obvious change in the small angle regime (0.004 < Q < 0.03 $Å^{-1}$), while weak anomalies are observed when considering the large Q regime (0.03 < Q < 0.1 $Å^{-1}$) only. This indicates that we are likely probing the evolution of the tail of the zeroth order magnetic satellite (located at Q ≈ 0.53 $Å^{-1}$, *i.e.* out of range accessible on D33).

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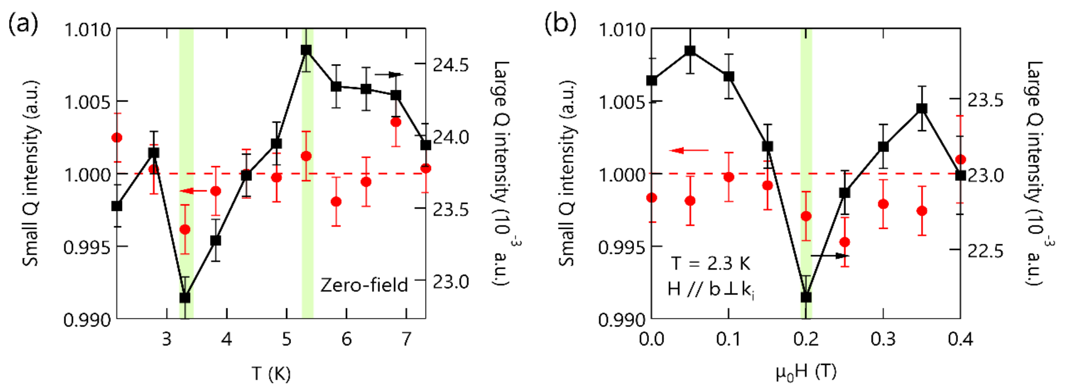
[1] L.-P. Regnault and J. Rossat-Mignod in *Magnetic Properties of Layered Transition Metal Compounds* (L.J. de Jongh, Ed.), p. 271 (1990)
[2] M. Songvilay *et al.*, Phys. Rev. B **102**, 224429 (2020)
[3] L.-P. Regnault *et al.*, Heliyon **4** (2018) e00507



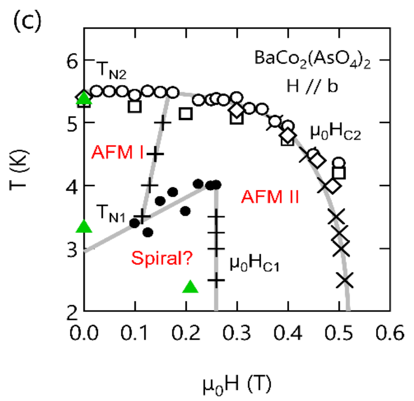


Fig. 1 – (a) Temperature-dependence of the SANS intensity in zero applied field. **(b)** Field-dependence of the SANS intensity at T = 2.3 K. **(c)** Phase diagram from Zhong *et al.*[4]. Green triangles represent the positions of the weak anomalies observed by SANS.

The same kind of observation can be made when considering the field-dependence of the SANS intensity at low temperature, where a weak minimum appears close to the critical field μ_0H_{C1} above which the FIM phase is stabilized (**Fig. 1b**). We report in **Fig. 1c** the (T,H) positions of these anomalies on the phase diagram deduced from susceptibility and specific heat measurements given by Zhong *et al.*[4]. We find that they more or less coincide with *(i)* "HM" → AFM I transition temperature T_{N1}, *(ii)* the AFM I → PM transition temperature T_{N2} and *(iii)* to the lower critical field μ_0H_{C1}.

On the other hand, the field scans did not reveal any anisotropy in the SANS intensity at low Q, which would have been expected due to the establishment of field-induced correlations. It is therefore likely that the magnetic contrast was too small or that the FM-like correlations quickly develop over length scales which are too large to be probed using SANS (or a combination of these two effects).

$\vec{H} \parallel \vec{c} \perp \vec{k_i}$ **–** We then proceeded with the study of the effect of an out-of-plane magnetic field. In this configuration, the signal is unfortunately dominated by horizontal nuclear streaks due to the reflectivity of the honeycomb planes (**Fig. 2a**). Neglecting this sector of the scattering map, we still obtained nuclear-like power laws, with no clear sign for extra magnetic scattering around Q = 0 (**Fig. 2b**).

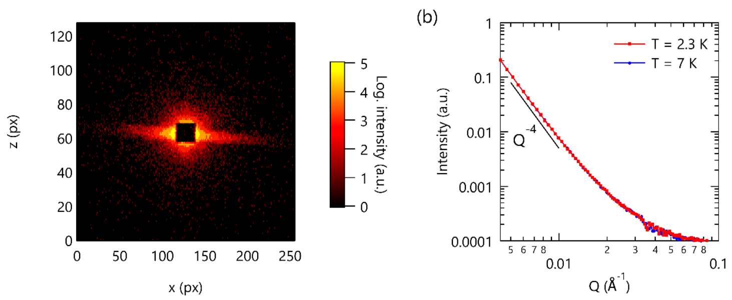


Fig. 2 – (a) Nuclear streaks observed in the configuration $\vec{H} \parallel \vec{c} \perp \vec{k_i}$. **(b)** Radial averaging of the SANS intensity excluding nuclear streaks, showing a power-law behavior with an exponent close to the Porod value.

[4] R. Zhong *et al.*, Sci. Adv. **6** (2020) eaay6953

$\vec{\boldsymbol{H}} \parallel \vec{c} \parallel \vec{\boldsymbol{k_i}}$ **–** In order to get rid of the nuclear streaks, we rotated the magnet in order to orient the honeycomb planes *perpendicular* to the incoming beam, while keeping the field *parallel* to the crystallographic **c** axis. In this configuration, the upper critical field is much larger than in the case of in-plane fields ($\mu_0 H_{C2} \approx 4$ T w.r.t. 0.5 T). We explored several instrument configurations, notably changing the neutron wavelength from 6 to 18 Å, to check whether *transverse* defects would form, having failed to find evidence for *longitudinal* DW.

Doing so, we noticed that the sample had moved down by ≈ 1 cm, due to the improper fixture of the sample stick. Having fixed it, we performed a field scan in the 0 ↔ 6 T range at λ = 18 Å and noticed the appearance of a new streak for fields larger than 1 T on the side detectors. This became more evident at λ = 6 Å, as displayed in **Fig. 3**. The strange feature of this streak was that its intensity remained constant for all fields above 1 T and only disappeared below 0.5 T.

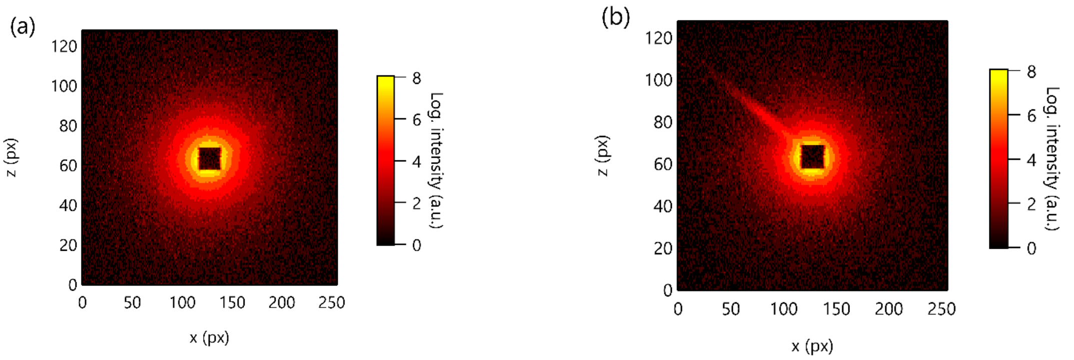


Fig. 3 – Scattering map obtained for a magnetic field applied perpendicular to the honeycomb plane for **(a)** $\mu_0 H = 0$ and **(b)** $\mu_0 H = 1$ T.

Since this particular field value did not correspond to any feature known from macroscopic measurements, we assumed that something had happened to the sample. We removed it from the cryomagnet and indeed found that it was broken, meaning that this streak could be due to the reflectivity on the surface of the fractured piece, which would move as a function of the applied field. The reason for this problem is unknown, but the glue might well have been damaged by the fall of the stick and the subsequent torque due to the applied field.

3/ Conclusions and outlook

We did not find any obvious evidence in the support of elongated defects forming in the magnetic structure of BCAO neither upon its transition from the "HM" to the FIM state, nor from the "HM" to the PM phase. Rather, weak anomalies in the "large Q" integrated intensity, corresponding to characteristic temperatures and fields deduced from macroscopic measurements indicate that we could only probe the low Q tail of the zeroth order magnetic satellite. *Unfortunately, the experiment had to be stopped due to the breaking of the sample*.

We however believe that there remain several points to explore in a future study, namely :

- Working with a shorter wavelength (not accessible on D33) to increase the Q range and study the shape of the diffuse scattering close to the lowest angle magnetic satellite,
- Using polarized neutrons to resolve the (weak) magnetic SANS anisotropy,
- Studying the SANS response under the above conditions over field cyclings at several temperatures to carefully explore the complex in-plane phase diagram (**Fig. 1c**) and the magnetic irreversibilities reported by Regnault *et al.*[1].