Experimental report

19/10/2022

Proposal: **7-01-563** **Council:** 4/2021

Title: Understanding the giant barocaloric effects in Mn3AN using inelastic neutron scattering

Research area: Materials

This proposal is a new proposal

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Experimental team:

Local contacts: Bjorn FAK

Michael Marek KOZA

Samples: Mn3GaN

Instrument	Requested days	Allocated days	From	To
PANTHER	2	2	05/10/2021	08/10/2021

Abstract:

Materials with large caloric effects offer the opportunity to replace current cooling technologies that require the use of hazardous gases. Barocalorics are particularly appealing, compared to the more well established magneto- and electrocalorics, due to the relative ease with which hydrostatic pressure can be applied. Giant barocaloric effects (BCE) have recently been observed in the geometrically frustrated antiperovskite Mn3GaN at the 1st-order antiferromagnetic transition (TN). Our results on Mn3NiN have revealed yet larger entropy changes, which we interpret as due to differences in the nature of the electronic hybridisation with Mn between the Ni and Ga compounds. We recently performed inelastic neutron scattering experiments on Mn3NiN and Mn3SnN (the latter has zero BCE) at MERLIN to further understand the BCE and found an anomalous phonon temperature dependence close to TN. To further understand these results we plan to measure these features in Mn3GaN to allow a direct comparison with our previous data. This information will provide valuable insight into the origins of the large BCE in these materials and therefore aid our search for yet larger effects in other Mn3AN.

Understanding the giant barocaloric effects in Mn_3AN using inelastic neutron scattering

Summary:

Materials with large caloric effects offer the opportunity to replace current cooling technologies that require the compression of hazardous gases. Whilst magnetocaloric materials driven using magnetic fields are currently the most developed for applications, there is growing interest in barocaloric materials driven using hydrostatic pressure. The latter offers many advantages, but perhaps most important is the relative ease and cost of creating mechanical pressure compared to magnetic field generated by Nd-based permanent magnets. Giant barocaloric effects (BCE) have recently been observed in the geometrically frustrated antiperovskite Mn_3GaN at the 1^{st}-order antiferromagnetic transition (T_N), enhanced by suppressed spin fluctuations in the ordered phase [1]. Our recent results on the closely related Mn_3NiN have revealed yet larger entropy changes, which we interpret as due to differences in the nature of the electronic hybridisation with Mn between the Ni and Ga compounds [2]. We recently performed inelastic neutron scattering experiments on Mn_3NiN and Mn_3SnN (the latter has zero BCE) at MERLIN to further understand the BCE, the first such experiments on Mn-based antiperovskites, and found an anomalous phonon temperature dependence close to T_N. To further understand these results we plan to measure these features in Mn_3GaN to allow a direct comparison with our previous data. This information will provide valuable insight into the origins of the large BCE in these materials and therefore aid our search for yet larger effects in other Mn_3AN compositions.

The experiment:

The aim of the experiment was to observe the temperature dependence of the phonons in Mn3GaN, particularly close to the transition. A ~4g sample, previously measured on MERLIN, ISIS, was loaded into a standard aluminium can. Data were collected using E_i = 110 and 40meV to capture both the higher and lower phonons, respectively, at temperatures between 1.5 and 330K (see Figure 1). Data collected with E_i = 30 and 19meV did not provide any further information, so after these runs empty cans were measured for background subtraction.

The data show an interesting temperature dependence of the low energy phonons close to the transition temperature (T ~ 280K). We are currently analysing the data, particularly by extracting the temperature dependence of the phonon lifetime and energy and comparing with our other data collected on Mn_3NiN and Mn_3SnN at MERLIN, ISIS (the flux at Ei = 40meV here is significantly better than MERLIN). We are planning one further inelastic neutron scattering experiment on a doped $Mn_3(A,B)N$ sample, so as to compare the phonon spectra with undoped (Mn_3AN) materials. We plan to publish the PANTHER data once this further experiment has been completed.

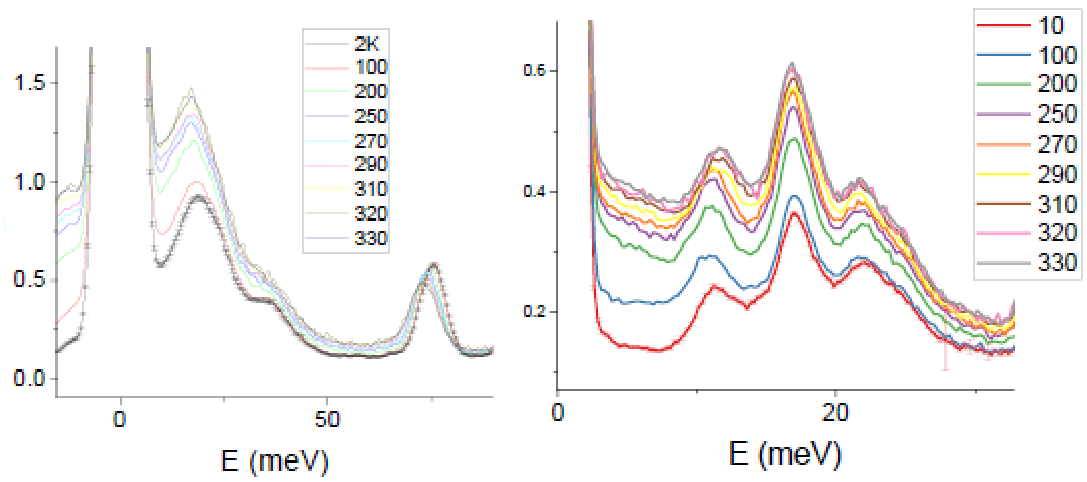

Figure 1: (left) data collected with E_i = 110 meV. (right) data collected with E_i = 40meV.

References:

[1] D. Matsunami et al., Nat. Mater. **14**, 73 (2014).
[2] D. Boldrin et al., Phys. Rev. X **8**, 041035 (2018).