Experimental report

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Title: Disclosing the magnetic structure of PrBaMn2O6 and NdBaMn2O6 single crystals.

Research area: Materials

This proposal is a new proposal

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Samples: PrBaMn2O6
NdBaMn2O6

Instrument	Requested days	Allocated days	From	To
D9	9	8	08/06/2021	16/06/2021

Abstract:

We want to determine the magnetic structure of PrBaMn2O6 and NdBaMn2O6. Both samples are perovskites with layered ordering of Pr(Nd) and Ba atoms along the c-axis. They are isostructural at high temperatures adopting a tetragonal unit cell (space group P4/mmm). They undergo magnetic transitions around room temperature and significant differences in the magnetic ground state have been found using neutron powder diffraction. A mixture of CE- and A-type magnetic structures were reported for the Pr-based compound whereas only A-type for the Nd-one. However, recent x-ray diffraction studies on single crystals of Nd-based sample reported contradictory results about a new kind of orbital ordering in the low temperature phase and the occurrence of new superstructure peaks in the low temperature phase not compatible with an A-type magnetic ordering. We have grown single crystals of both specimens with the aim of giving a definitive answer about the magnetic order developed by these compounds and the existence, or not, of a new type of orbital ordering.

Disclosing the magnetic structure of $PrBaMn_2O_6$ and $NdBaMn_2O_6$ single crystals.

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Single crystals of $PrBaMn_2O_6$ (PBMO) and $NdBaMn_2O_6$ (NBMO) were grown using the floating zone method from polycrystalline precursors [1] and one piece of each boule was selected for the present experiment. Unfortunately, the NBMO crystal developed some cracks during the cooling process and ended up breaking. This prevented its measurement and the experiment was focused on the PBMO sample.

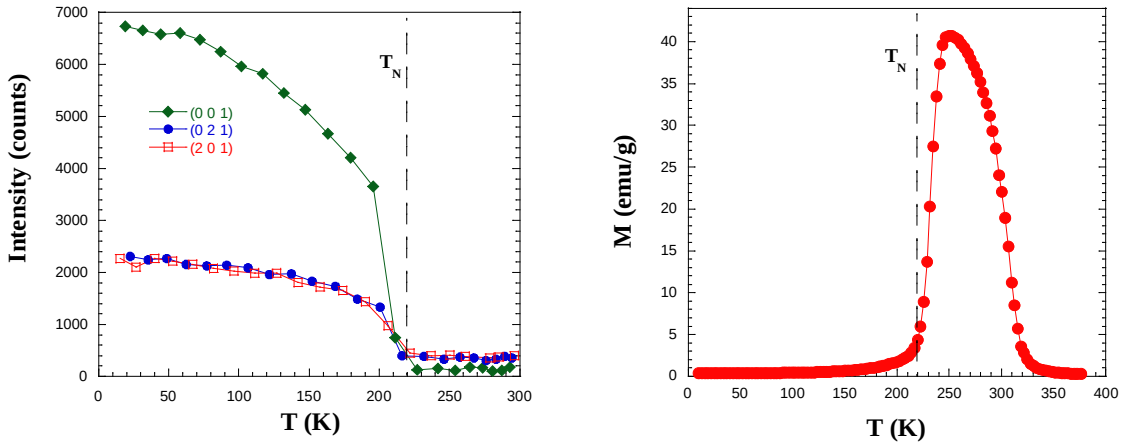

Fig. 1. Temperature dependence for the intensity of selected magnetic reflections (left). Magnetization as a function of the temperature for PBMO crystal (right).

Single-crystal neutron diffraction data were collected on the high resolution four-circle diffractometer D9 (ILL) using a wavelength of 0.832 Å. PBMO adopts an orthorhombic crystal structure at low temperature with space group $P2_1am$ (No. 26) and lattice parameters $\sqrt{2}\mathbf{a}_p \times \sqrt{2}\mathbf{a}_p \times \mathbf{c}_p$ ($\mathbf{a}_p$ =primitive cubic cell; $\mathbf{c}_p = 2\mathbf{a}_p$). The Q-scans performed on the PBMO crystal at 12 K found no magnetic contribution following the propagation vector $\mathbf{k}$= (½, ½, ½). No reflections were observed to suggest that the **c**-axis is doubled. This ruled out the existence of a CE-type magnetic contribution in this sample in opposite to a previous publication based on neutron powder diffraction measurements [2]. Therefore, the magnetic ground state in PBMO follows the propagation vector $\mathbf{k}$= (0, 0, 0). The main magnetic peak corresponds to the (0, 0, 1) reflection disclosing that the magnetic moments are ordered in the *ab*-plane. The magnetic peaks follow the reflection conditions $h+k$=even

and *l*=odd, suggesting an A-type magnetic ordering of the Mn moments. The temperature dependence of the most important magnetic peaks reveals that they appear at T_N as can be seen in Figure 1.

A symmetry analysis unveiled two possible magnetic structures compatible with our results. Using the Bertaut's notation [3], they correspond to $G_xA_yC_z$ and $A_xG_yF_z$ structures. The occurrence of forbidden nuclear reflections revealed that our crystal was twinned with an even mixture of two domains, namely *abc* and *ba-c*. In this way, the forbidden reflection (odd, 0, *l*) is allowed in the second domain as (0, odd, -*l*). This fact prevents to discern between the two magnetic models. Figure 2 shows the best fit obtained with the second magnetic structure, $A_xG_yF_z$, which exhibits somewhat better reliability factors (R_F^2= 2.6, χ^2=6). The magnetic structure is also displayed in Fig. 2. The refined Mn moment was 3.11(2) μ_B/Mn, close to the one expected for a mixed valence Mn (around 3.5).

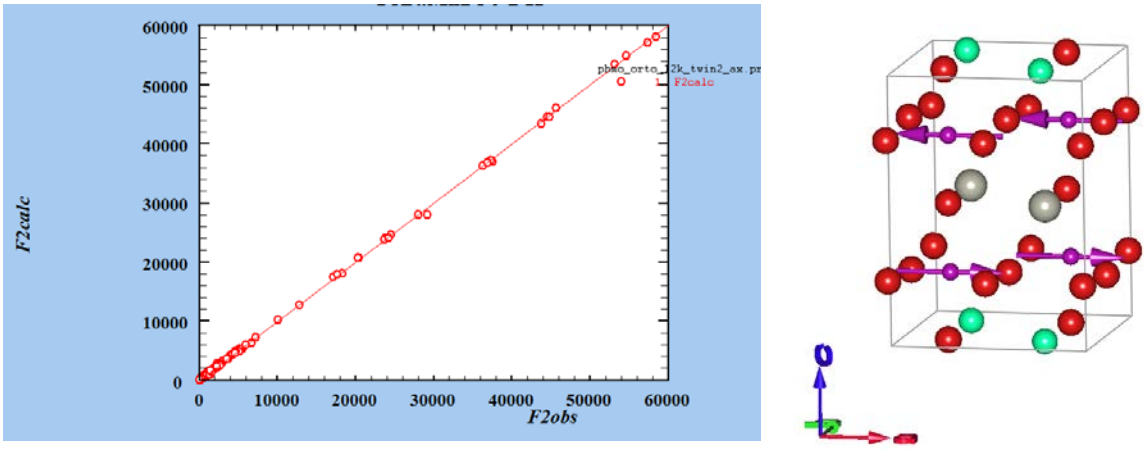

Fig. 2. Left: Calculated versus observed values of the squared intensities for refined single crystal neutron diffraction data collected at 12 K for PBMO following the model $A_xG_yF_z$. Right: Corresponding magnetic structure.

Finally, in order to take advantage of the resulting beam time, a third crystal, $SmBaMn_2O_5$, was measured as a reference. This layered perovskite with oxygen vacancies showed a G-type magnetic ordering of the Mn moments at low temperature. Details of the structural and magnetic study can be found in a recent publication [4].

References.

[1] J. Blasco *et al.* Phys. Rev. B **103**, 064105 (2021).
[2] T. Nakajima *et al.*, J. Phys. Soc. Jpn. **72**, 3237 (2003).
[3] E.F. Bertaut, Acta Crystallogr. Sect. A **24** 2177 (1968).
[4] J. Blasco et al. Mat. Res. Bull. **150**, 111780 (2022).