Preparing a magCIF file of a magnetic Structure for MAGNDATA. Example 3: $PrFe_2Al_8$

Reference:
Nair et al., J. Phys.: Condens. Matter **29** (2017) 345801
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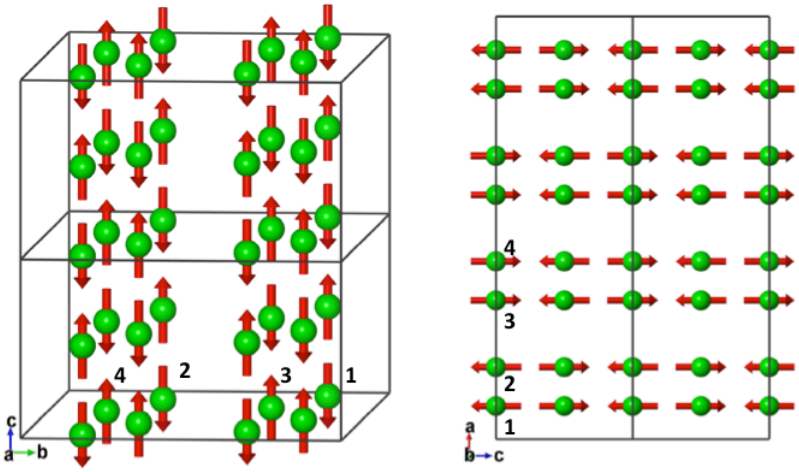
Parent Pbam structure at 300K in file: icsd_259673_parent_300K.cif
Although the authors have determined the positional structure at several lower temperatures, the article only gives full information on the one determined at 300K, which can be retrieved from the ICSD database. The corresponding cif file is attached.
Cell parameters at 150K and 25K are also reported. The magnetic diffraction data used is from a measurement at 2K.

The magnetic structure at 2K as reported in the reference:

The propagation vector is identified as (1/2,0,1/2) and a single irrep is assigned to the refined spin arrangement, using a rather arbitrary label Γ_1 coming from the software used (two alternative irreps were in principle possible). The structure is only described in Figure 4 within a 1x1x2 supercell with respect to the actual cell 2a,b,2c of the magnetic structure with respect to the parent cell:



In order to facilitate the identification of the proposed structure, we have ordered and labelled with numbers the Pr atoms within the parent unit cell. Notice that the moment values of these 4 atoms are sufficient to fully describe the magnetic ordering. The rest are determined by these four values through the value of the propagation vector (1/2,0,1/2), depending on the value +1 or -1 of $\exp(i2\pi\mathbf{k}.\mathbf{T})$, where **T** is the parent lattice vector relating the atom with one of the four numbered. The arrangement in the figure can be then described as - - + + along c for the four ordered Pr atoms.

Moment modulus: 2.5 μB at 2K and T_N= 4.5K

The information above, together with the provided CIF file is sufficient to produce a magCIF file that can be uploaded in MAGNDATA.

First of all, if you do not have them, download the instructions available in the webpage of MAGNDATA. There you will obtain a zip file that includes the pdf file: "instructions_to_prepare_mcif_file_v6.5.pdf". According to these instructions, our situation is "Case 3", as we do not have yet any magCIF file. For the simple magnetic structure of this example the most rapid approach is to follow the 1st Procedure for "Case 3", which is recommended in the instructions, and use the program MAXMAGN. This will be sufficient to produce an initial magCIF file and then follow the steps explained in the instructions for "Case 1", i.e. when one has already a magCIF file consistent with the MSG of the structure:

1) Use MAXMAGN: Introduce the k-vector and tick the option that "Structure data of the paramagnetic phase will be included". In the next step introduce the cif file with the reported position structure at 300K. The program will show that four maximal symmetries are compatible with the observed propagation vector, being described by four different magnetic space groups (MSGs). By clicking on the last column "Magnetic Structure" identify the only two MSGs, which allow a spin arrangement along c.

2) Putting values to the allowed moments and producing the corresponding mcif file, use the visualization tool of the program and identify that the two spin arrangements allowed by the first MSG in the list, which is compatible with moments along c (for the four atoms ordered 1,2,3,4 (with the additional constraint of equal moduli) are : + + - + and - + + + .
None of these arrangements corresponds to the one reported. However, this possible maximal symmetry listed by the program should not be fully discarded if there are other possible conjugate subgroups, yielding domain related arrangements, which will be different, but equivalent. This is the case here, because the point group of the structure decreases with respect to the paramagnetic phase. This can be checked by clicking on the button "alternatives (domain-related)". The program will show that there is an alternative conjugate subgroup, which can be chosen and corresponds to an alternative equivalent description of the magnetic structure. But before checking if the reported structure corresponds to one of these possible alternative descriptions for this maximal symmetry, it is convenient to check first all MSGs chosen by the program by default, so we go to the next step.

3) Do the same as in step 2, for the second MSG in the list that is compatible with moments along c. Identify that the two possible arrangements constrained to equal moments allowed by this MSG are: + + - - and - + - +. The first one is the one proposed in the paper. Thus we have identified the MSG of the reported structure. Save therefore the corresponding mcif file for further processing.

4) You have saved in the previous step a magCIF file of the model corresponding to the magnetic structure that is proposed in the paper. You have therefore the necessary file to go now to the process that is explained as "Case 1" in the instructions, and obtain the desired magCIF file ready for uploading in MAGNDATA.

5) The magCIF to be prepared has a small part about information on the irreps that are active. It is can be left untouched, without information, if wished. But it is recommended that you fill it using Get_mirreps. You should obtain with this program that only one 2-

dim irrep is compatible with the magnetic arrangement of the structure, as stated in the paper, but this irrep is restricted along a special direction. Therefore the four basis vectors corresponding to this irrep should be combined in a specific form to yield the observed MSG, reducing the number of free parameters to two. The constraint to a common moment modulus for all sites, is however not forced by the restriction to a single irrep or by the restriction to a MSG within this irrep. It is an additional restriction introduced in the refined model. This additional assumption of equal moduli reduces then the number of free parameters to one.

6) According to the .mcif file that has been prepared for uploading in MAGNDATA, the system has decreased its point group symmetry by a factor two (apart from breaking time reversal) with respect to the paramagnetic phase. There must be therefore an alternative equivalent description of the magnetic structure, which would equally fit the experimental data, and corresponds to a spin arrangement related with the reported one by the lost symmetry operations. This equivalent arrangement can be easily obtained using MVISUALIZE: upload in the program MVISUALIZE the mcif file that you have prepared, corresponding to the reported structure with arrangement - - +. Use the button "Domain related equivalent descriptions" to obtain at the bottom of the output a table with the two possible equivalent structures. The first one is the one of the input and the second one, as indicated in the table, can be obtained by applying the lost operation {2y| ½ ½ 0}. You can obtain a mcif file and visualize this equivalent structure by clicking on the button "Show". If you visualize this structure you will see that the arrangement is now: + + + +. This arrangement represents exactly the same magnetic structure! It is just represented differently with respect to the paramagnetic structure. *This example shows that one should be very careful when trying to derive from the relative signs of the moments in a magnetic structure a specific local FM or AFM coupling between the corresponding moments. As shown here, this relative sign is in many cases not absolute and depends on the description of the structure.*