

## RMCPow PRACTICAL

### Modelling the magnetic structure of MnO

Files needed are found in the same zip file as this instruction. Copy or extract the files into a suitable directory and follow the instructions below. The different programs used can be accessed with the shell program called **WinNFLP**, available at [ftp://ftp.studsvik.uu.se/Pub/WinNFLP/Win\\_NT/](ftp://ftp.studsvik.uu.se/Pub/WinNFLP/Win_NT/). To run, click on the WinNFLP icon, a browser window will then appear. Select your working directory by clicking on a file in that directory (any will do). Now you can access the programs you want to run from the menus **Useful** or **RMC**. If you do not have the WinNFLP program it is still possible to run the RMCPow simulations since all necessary input files are provided here.

The general approach of the practical is as follows:

- The average magnetic structure of MnO in the long-range ordered antiferromagnetic phase at 15 K will be determined.
- Investigation of the short-range magnetic order at 130 K, i.e. just above the Neel temperature  $T_N=120$  K.

**The times given in this practical, for running RMCPow is real time and NOT CPU time. The times may have to be increased on a slow computer. The output you get, will probably not be identical to the output in this example. The plotting and programs used in this practical can also be done with in principle any standard plotting program, such as Origin or SigmaPlot, since most data is easily imported as ASCII files.**

**1.** The model is based on the neutron scattering data obtained on the Studsvik Liquid and Amorphous Diffractometer SLAD. The low temperature data is in the file `mno15.fq`. Plot it with the program `FqPlot`.

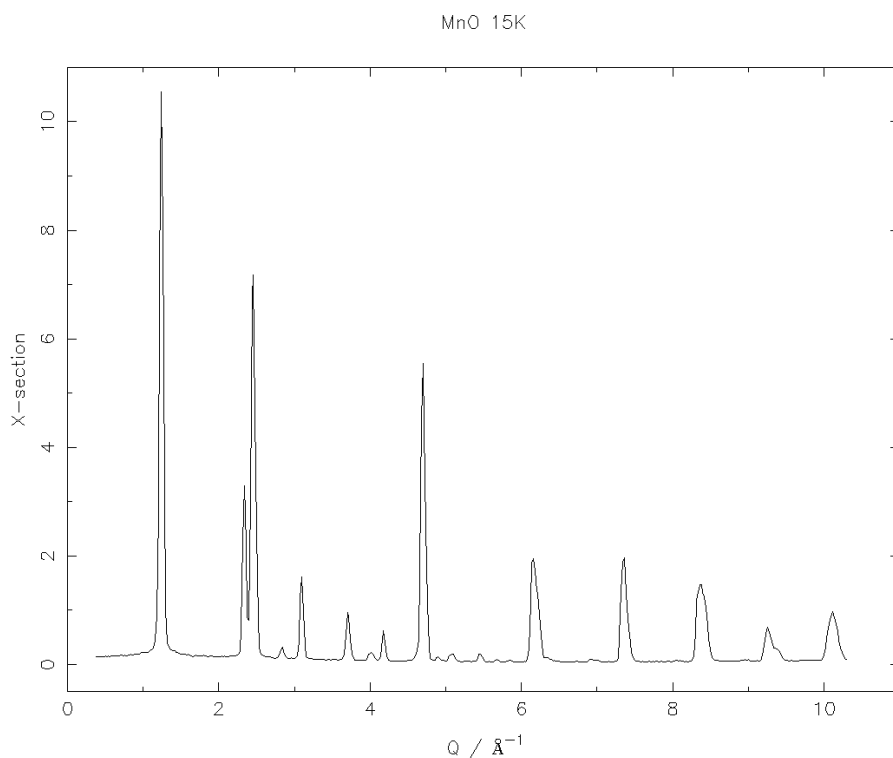
**Useful → User programs → FqPlot.exe**

When the `FqPlot` program starts type:

```
Filename [.fq] > mno15.fq
mno15.fq
Format? G(ENIE)/T(EXT)/D(ATA) > d
Graphics device/type (? to see list, default/W9):<RETURN>
```

Now you should have a plot on the screen. To plot with other limits select the `FqPlot` text input window and type

```
Change limits? [T/F] > t
Data limits are           : .020 10.300  .000 10.558
Current plotting limits are : .000 12.000 -.528 11.086
New limits > 0 10 0 11
```



To exit type

```
Change limits? [T/F] > f
Hardcopy output? [T/F] > f
Type <RETURN> to close graphics window # 1 <RETURN>
```

If you want a hardcopy (PostScript file) output type

Hardcopy output? [T/F] > **t**  
 Graphics device/type (? to see list, default/W9): **name/ps**  
 Output to name  
 where **name** will be the PostScript filename.

**2.** In the classic experiment by Shull et al [1] the possibility of using neutron diffraction to determine magnetic structures was first demonstrated. It was observed that below the Neel ordering temperature extra Bragg peaks (w.r.t. to the nuclear peaks seen at ambient temperature) occurred in the diffractogram. We are now going to confirm their results using the reverse Monte Carlo algorithm as implemented in the RMCPOW program. We start with a lattice of multiple unit cells with atoms placed on equilibrium positions and initially random oriented spins. To create the atomic configuration we use the Crystal program. As input we give the name of an input file, **mno15a.atom**, containing relevant parameters. These include the space group number (we use the cubic symmetry group Fm-3m to create initial positions although the actual cell has a slight rhombohedral distortion as given by the cell angles g1,g2,g3), lattice parameters, the number of unit cell copies along each edge etc. Use an editor (like Notepad) to look at the input file.

```
.false.          ! do not apply to an existing cfg
225 1            ! fm_3m spg, setting 1
4.432 4.432 4.432 90.62 90.62 90.62 ! a1,a2,a3,g1,g2,g3
 1 2 2 2         ! isym,n1,n2,n3
n                ! no displacements
 2              ! No. of atom types
 1              ! No. of Mn sites
4a 0.0 0.0 0.0 ! Wyckoff site,x,y,z
 1              ! No. of O sites
4b 0.5 0.5 0.5 ! Wyckoff site,x,y,z
.true.          ! RMC coordinates
```

Run the program: **Useful → CFG programs → Crystal.exe**. Give **mno15A.atom** as input file and **mno15A.cfg** as output. Have a look at the new file mno15A.cfg if desired. Note that there are 64 (32 Mn and 32 O) atoms in the configuration.

**3.** Next we are going to create the random spin configuration. This is done with the RandomSpins program and all we need to do is **Useful → CFG programs → RandomSpins.exe** and type

```
Number of magnetic types > 1
Number of particles of type 1 > 32

Average magnetisation is 6.353857E-02
in direction 4.808538E-01 3.022465E-01 -8.230593E-01

Output file [.scfg] > mno15A.scfg
```

The final file needed to start the simulation is the control input file, mno15A.dat:

MnO 15K

```

0 0 ! ncoll,ncycles
100 0 60 60 ! iprint,iplot,timelim,timesav
8.864 8.864 8.864 90.62 90.62 90.62 ! UC parameters
0 0 0 0 0 0 ! UC parameter flags
1 1 1 1 ! isym,na,nb,nc
2 1 ! ntypes,ntypesm
1 ! attypes
0.0 0.0 ! delta
40.0 ! deltam
0 0.0 ! nswap,swapfrac
0 0.0 ! nref,reffrac
2.39 1.69
      2.29 ! closest approaches
0 ! ncoord
0 ! nasc
0 ! nval
0 ! naval
0 ! navm
0 ! nsc
0 ! nasc
0 ! nmpot
1 0 0 ! 1 neutron, 1 xray constraint
5.0 0.2 ! rdwt,swdt
2 0 ! ijob,iexists
0.0008 0.0033 ! msd,Tabs
      0 0
4.54 ! muz
0 ! flag
mno15.fq
0.5 7.0 ! Qmin,Qmax
0 ! nexcl
3 0.005 ! isig,sigma
1 0 ! nbpol,nbfix
0.0 1 ! alpha0,flag
1.0 1 ! beta,flag
1.1126 1.0 ! wav,fwmax
2.1265 -1.4302 0.4953 0.0 0.0 ! ur,vr,wr,ig,eta
      0 0 0 0 0 ! ur,vr,wr,ig,eta flags
0 ! wextn
1 ! Mn noccn
1.0 -3.73 0.40 ! Mn coccn,bbar,siginc
1 ! O noccn
1.0 5.803 0.0 ! O coccn,bbar,siginc
7.0 ! q2m
1 ! Mn noccn
1.0 5.65 ! Mn2+ (3d5) cocc,mueff
0.4020 17.7870 0.6156 6.0950 0.0000 -8.9310 0.0 0.0
-0.0170 ! form factor parameters (a1,b1,...,c)

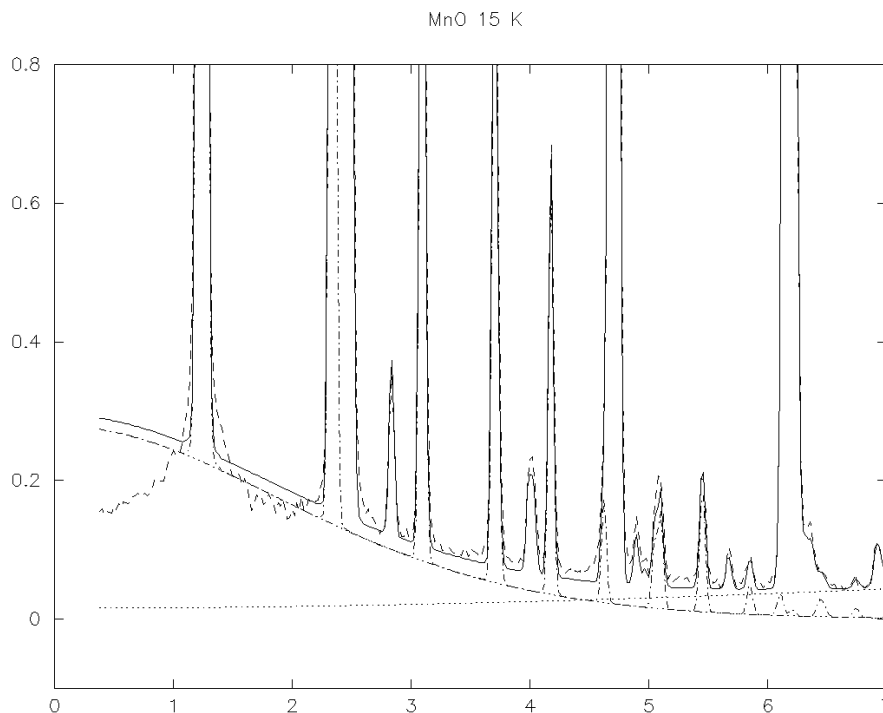
```

A full description of the control file is given in the RMCPOW manual. Here we just make a few comments on some parameters:

- Lattice parameters and number of unit cells are given for a double nuclear unit cell. This is because from the occurrence of the extra magnetic peaks we expect the magnetic cell to be doubled w.r.t. to the nuclear cell and so we must allow for the corresponding points in the reciprocal lattice to be considered as Bragg peaks.
- For `ijob=2` the effect of atomic displacements are considered using an analytical approach, hence atomic positions are not changed.
- Likewise the effect of magnetic disorder is given by the `muz` parameter which is the ordered component of the magnetic moment. This parameter is expected to be  $gS$  at low temperature, for a spin  $S$ . For a pure  $Mn^{2+}$  ion  $S=5/2$  but covalency decreases this value somewhat. `mueff = 5.65`, the total moment, is then given by  $g\sqrt{S(S+1)}$

Now we can start the simulation: **RMC → RMC programs → RMCPOW.exe** and give **mno15A.dat** as input file. This will take 5 minutes or about 3000 generated moves to converge. On the screen you see how  $\chi^2$  decreases as the fitted  $F(Q)$  approaches the experimental  $F(Q)$ . You should obtain a  $\chi^2 \sim 170$ . Exit the program using **File → Exit** and save the results.

Now we can look at the fit with the RMCPlot program: **Useful → Plot programs → RMCPlot.exe** and give **mno15A.out** as the file to plot. In the plot you will find the experimental and RMCPOW fit curves as well as various contributions to the RMCPOW fit.



Make a hardcopy or exit RMCPlot as in the FqPlot program.

We can also view the spin configuration using the ConfPlot program: **Useful → Plot programs → ConfPlot.exe** and at the ConfPlot prompt type

```

> open mno15A.cfg
Reading file: mno15a.cfg
Configuration contains      64 atoms of  2 types.
> spin mno15A.scfg
Reading file: mno15a.scfg
Configuration contains      32 spins of  1 types.
> dir -2 1 1
> dot -2 1 1
> spl 1 1
Warning: Atom type  2 contains different number of atoms
and spins
Configuration: mno15a.cfg
Spin config. : mno15a.scfg
PLEASE WAIT...
Type: 1

```

The dot command tells ConfPlot to colour code the spins from red to blue depending on their projection on the chosen direction. As can be seen spins are now ordered in alternating sheets along (111) with the spins pointing in the [111] plane (try also to plot an initial random configuration!). The actual direction can vary but usually the simulation ends up with the spins along one of the  $\{-2,1,1\}$  directions, i.e.  $\perp$  [111]. A closer investigation reveals that each spin has 6 parallel and 6 anti-parallel nearest neighbours whereas next nearest neighbours are all anti-parallel. Try repeating this simulation a few times and/or running it for a longer time to check the repeatability.

4. While this is running you can start preparing for modelling the diffuse (magnetic and nuclear) scattering at  $T > T_N$ . We will now use another approach where we are explicitly going to introduce atomic displacements into the model. First we will try this on the low temperature data and then go on to the higher temperature. Because atomic displacements even at 130 K are small and mostly of a non-correlated character we will introduce harmonic modes of suitable amplitude into the initial configurations and keep the atom positions fixed since this will save computing time considerably. Such an atomic configuration (with  $6 \times 6 \times 6$  nuclear unit cells) can be made using the Crystal program and the **mno15B.atom** input file. Note that there is now a line with parameters for creating harmonic displacements. These are chosen to give a good fit with the mostly nuclear scattering at high Q. Run Crystal and give **mno15B.cfg** as output.

To save further time, we will also start from an ordered spin configuration, using the SpinSym program. From our results in section 2. we can deduce that the magnetic structure is described by a propagation vector  $\mathbf{q} = \frac{1}{2}(\mathbf{a}^* + \mathbf{b}^* + \mathbf{c}^*)$ . The magnetic moments of all spins are then related to the four in the nuclear unit cell by a factor  $\exp(i\mathbf{q}\mathbf{R})$  which in this case takes values  $\pm 1$ . As input to SpinSym we use the file **mno15B.spin**:

```

6 6 6           ! No. of unit cells for each direction
1              ! No. of propagation vectors
0.5 0.5 0.5     ! propagation vector, in recip. UC units
.false.        ! no sine components
1              ! No. of spin types
.true.         ! align

```

```

4           ! No. of Mn spins in UC
  1.0  1.0 -2.0 ! spin direction at (000)
-1.0 -1.0  2.0 ! spin direction at (½½0)
-1.0 -1.0  2.0 ! spin direction at (½0½)
-1.0 -1.0  2.0 ! spin direction at (0½½)

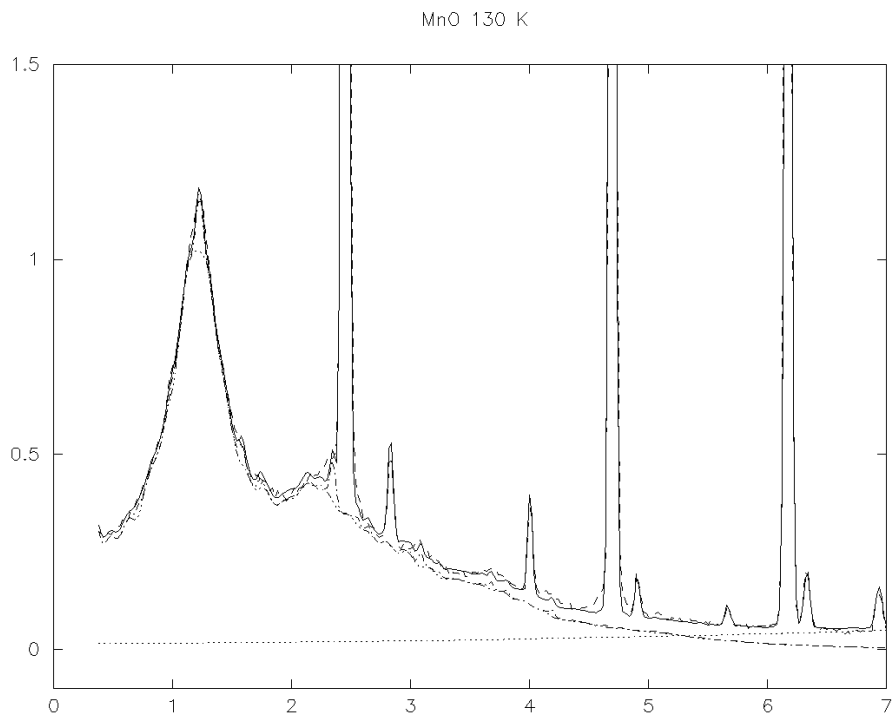
```

align=.true. means that all spins are aligned with the corresponding reference spin in the first copy of the nuclear unit cells. Look at the new mno15B.scfg to confirm that this is really true.

The RMCPOW control file mno15B.out is now slightly modified. We have increased the number of unit cells, and because of the magnetic unit cell we have doubled the lattice parameters. Parameters for average displacements, msd and muz parameters, are no longer needed and therefore removed and ijob is consequently set to zero. Start RMCPOW with this input file and run for 20 minutes or about 7000 generated moves. This should be sufficient to reach convergence at  $\chi^2 \sim 12$ .

Before you start to analyze the result also create a configuration with harmonic displacements for the simulation of the 130 K data, use **mno130B.atom** as input to Crystal. The experimental data is now in the file mno130.fq. Inspection of this file show that there is now essentially no magnetic Bragg scattering. We therefore use a random spin initial configuration, now with 864 Mn spins, save this as **mno130B.scfg**. Start a RMCPOW simulation for the 130 K data with **mno130B.dat** as input. The convergence is now somewhat slower but you should reach  $\chi^2 \sim 110$  after 30 minutes or about 8000 moves.

After convergence plot the fits with RMCPLOT. Note the oscillations at low Q in the magnetic scattering. This is due to the limited size of the configuration.



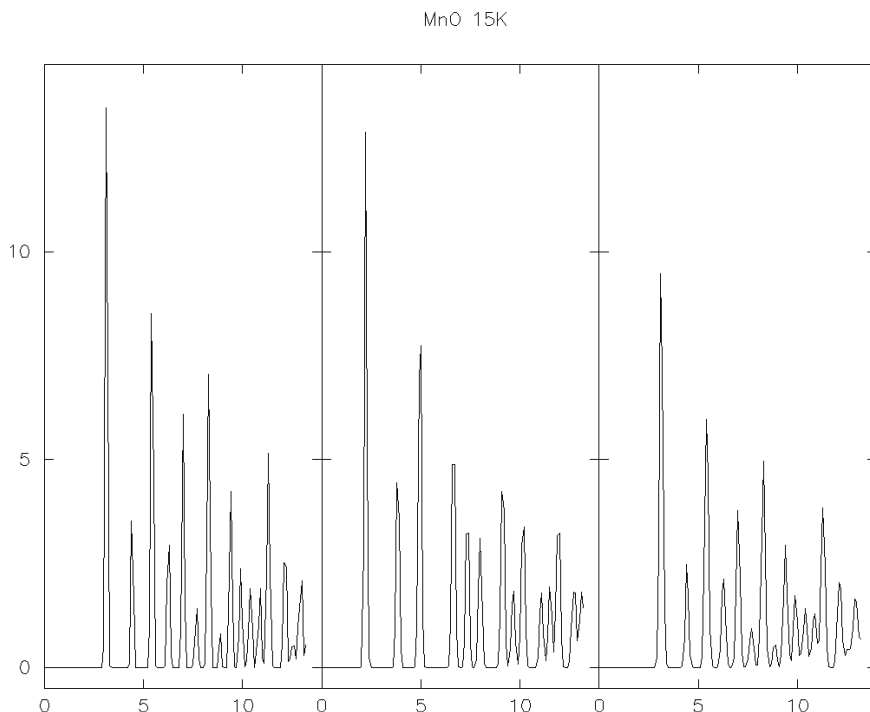
**5.** We are now going to use some of the configuration analysis programs to look at the local order. First we will calculate the partial pair correlation functions for the fitted configuration. Start the program **Useful → Analyze Cfg programs → GrSq.exe** and type

```
Parameter file [.par] > <RETURN>
r spacing, rmax      > .1 100
Q min, Q spacing, Qmax > 0 0 0
No. of configs       > 1
Totals               [T,F] > f

Configuration file [.cfg] > mno15b
Output file        [.grs] > mno15b
```

Use RMCPlot to display the result, selecting mno15b.grs for input. The three panels show the Mn-Mn, Mn-O and O-O partials respectively. Compare the plots with the corresponding for mno130b. Try enlarging the plots and check if there are any signs of correlated displacements, i.e. narrower nearest neighbour peaks.

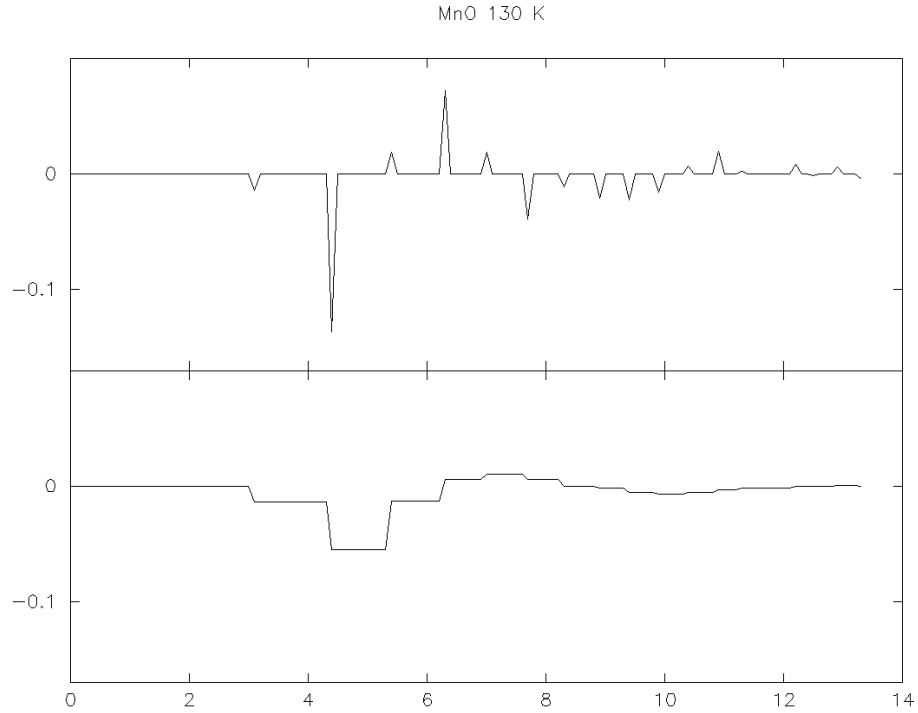




The local magnetic order is most directly seen with the MuMuR program which calculates the spin-spin correlation function  $\mu\mu(r)$ , i.e. the average cosine of spin pairs separated a distance  $r$ . Run **Useful → Analyze Scfg programs → MuMuR.exe** and type

```
No. of configurations > 1
Configuration file(s)      [.cfg*] > mno15b
Spin configuration files(s) [.scfg*] > mno15b
Maximum radius of a sphere within the config. is 8.88
Maximum distance to a corner of the config. is 15.386
dr and rmax > .1 100
No. of magnetic types > 1
Atom and magnetic type > 1 1
Minimum no of pairs in bin > 1
Tube, layer or spherical [T/L/S) > s
Output file [.mm,.gg] > mno15b
```

Plot the resulting  $\mu\mu(r)$  with the RMCPlot program. Select **mno15b.mm** as input. You will now see a plot with two panels. The top contains the (differential)  $\mu\mu(r)$  and in the bottom panel an integrated version is displayed. Compare with the corresponding for 130 K. Although now there is considerable overlap of various shells and, due to the loss of long-range order at 130 K, there is no unique direction for correlations it can be seen that first neighbour shells are clearly more correlated than far neighbours. In particular we observe that nearest neighbours now have on the average an anti-parallel alignment, which was not the case at low temperature when ordered correlations cancelled.



**6.** It is also instructive to look at the spin-spin correlations in various directions. This is effected by choosing the tube option in MuMuR, for which correlations are only calculated for spin pairs along some specified direction e.g. [100]. We can also take the (pseudo- in case of 15 K data) cubic symmetry into account by applying appropriate symmetry operations so that all e.g. {100} directions are averaged (give space group and setting 225 and 1 resp.). If we want to monitor the spin-spin correlations averaged over each neighbour shell it can be useful to make reference atomic configurations without atomic displacements so that each shell gives a point-like contribution to  $\mu\mu(r)$ . The two \*ref.atom files are supplied for this reason. Finally, we can do spin configuration plots of these larger configurations with ConfPlot.