OAK RIDGE National Laboratory

Hands-on polarized powder data from POWDER (HB-2A) $Ho_2Ti_2O_7$

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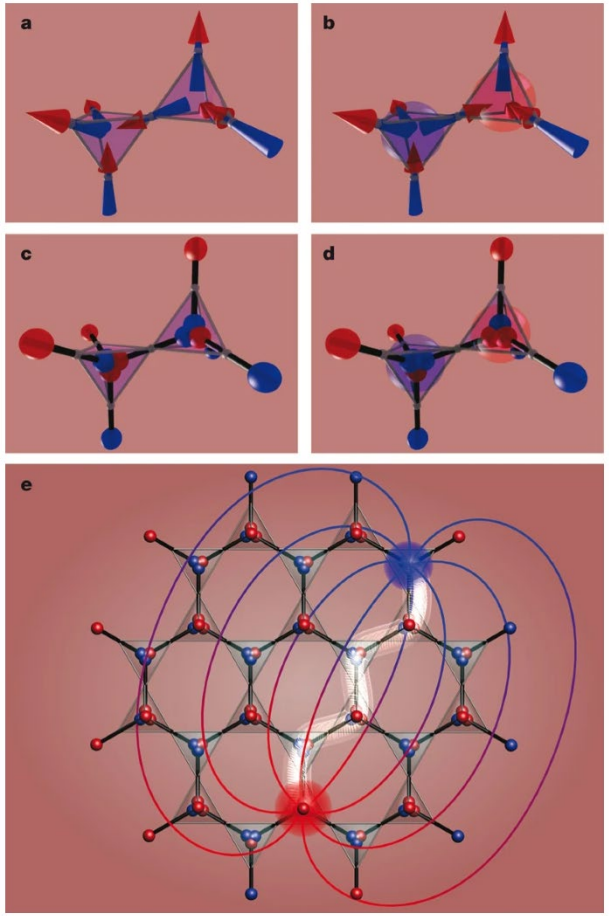
U.S. DEPARTMENT OF ENERGY

Background: $Ho_2Ti_2O_7$

- The pyrochlore $Ho_2Ti_2O_7$ is characterized by Ising spins along the <111> axis on a frustrated lattice.
- In zero field there is no long-range order, instead short range "spin-ice" order gives rise to exotic properties, including magnetic monopoles.
- The local anisotropy encodes and drives the underlying physics.
- **This example shows how half-polarized measurements on a powder can provide details on the local anisotropy.**
  - **In this case, the powder data yields the same information as a single crystal analysis. See DOI:10.1103/PhysRevResearch.1.033100 for more details.**
  - **The goal of this exercise is to find the site susceptibility tensor**

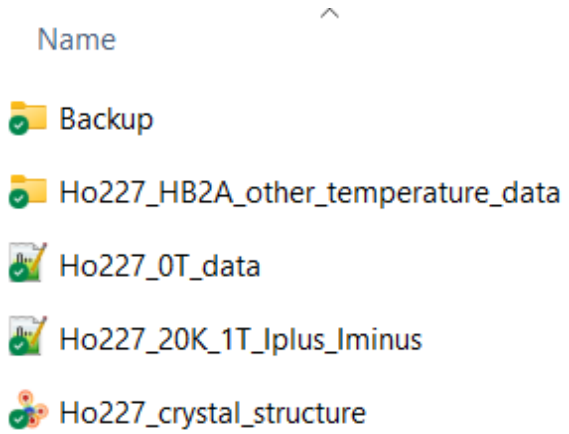


$$\chi = \frac{\boldsymbol{m}}{\boldsymbol{H}} = \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{12} & \chi_{22} & \chi_{23} \\ \chi_{13} & \chi_{23} & \chi_{33} \end{pmatrix}$$

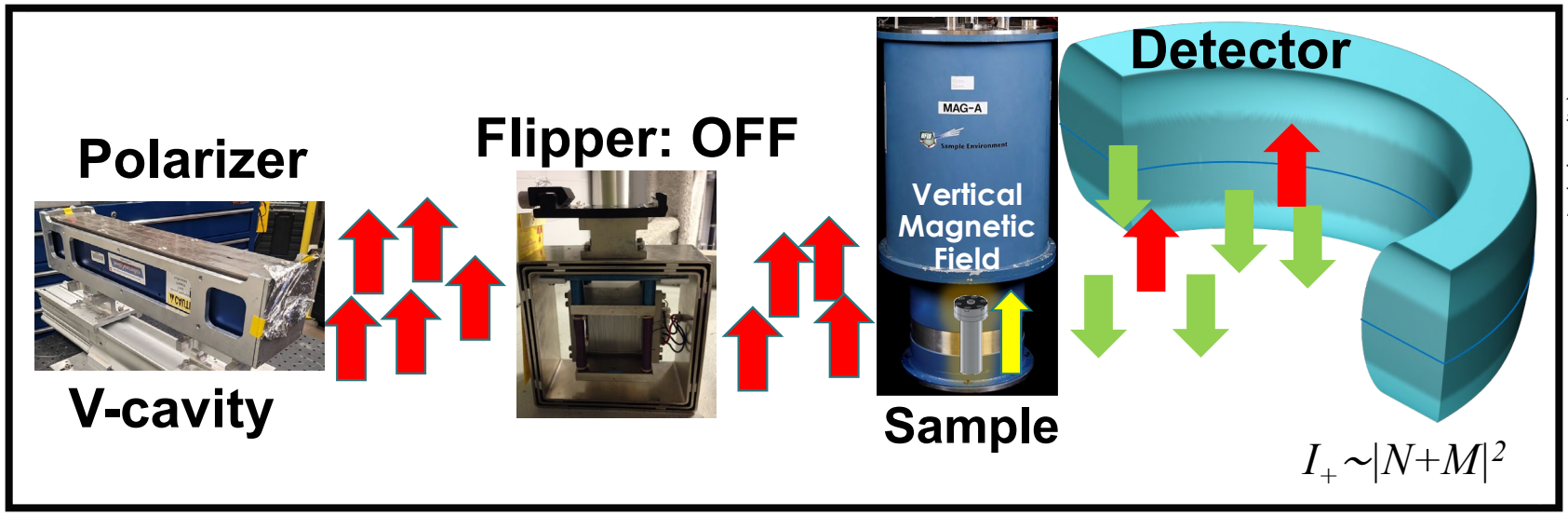
S. Calder, Hands-on Polarized Powder data from POWDER (HB-2A).

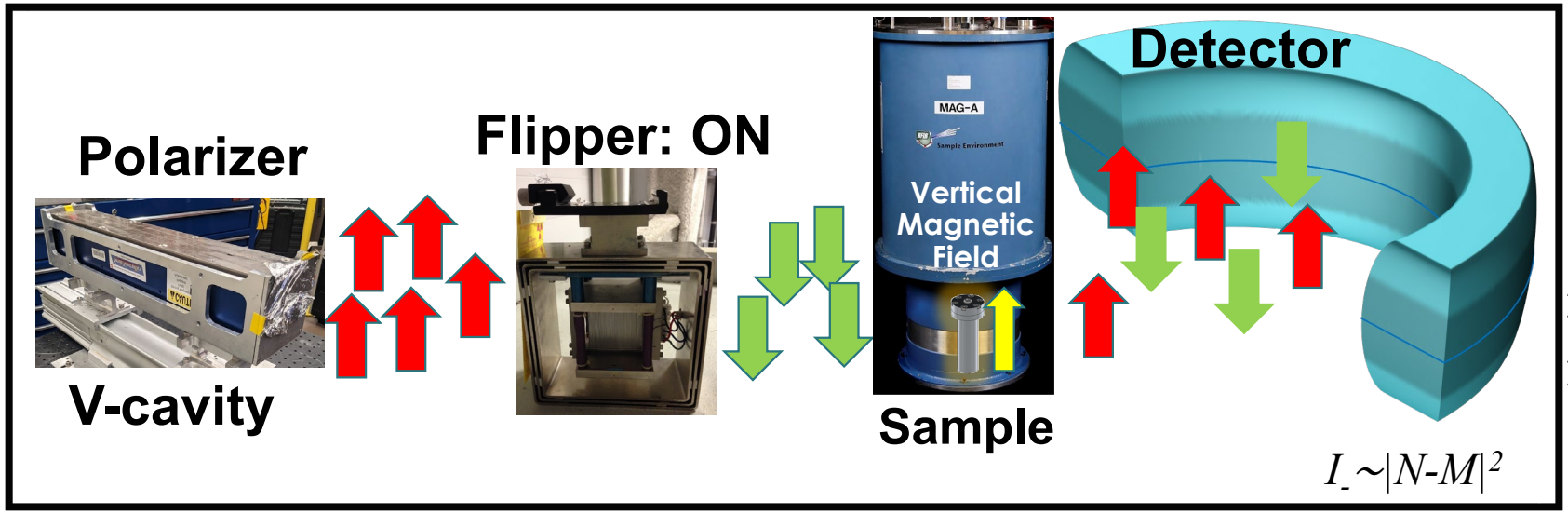
Details of this $Ho_2Ti_2O_7$ example

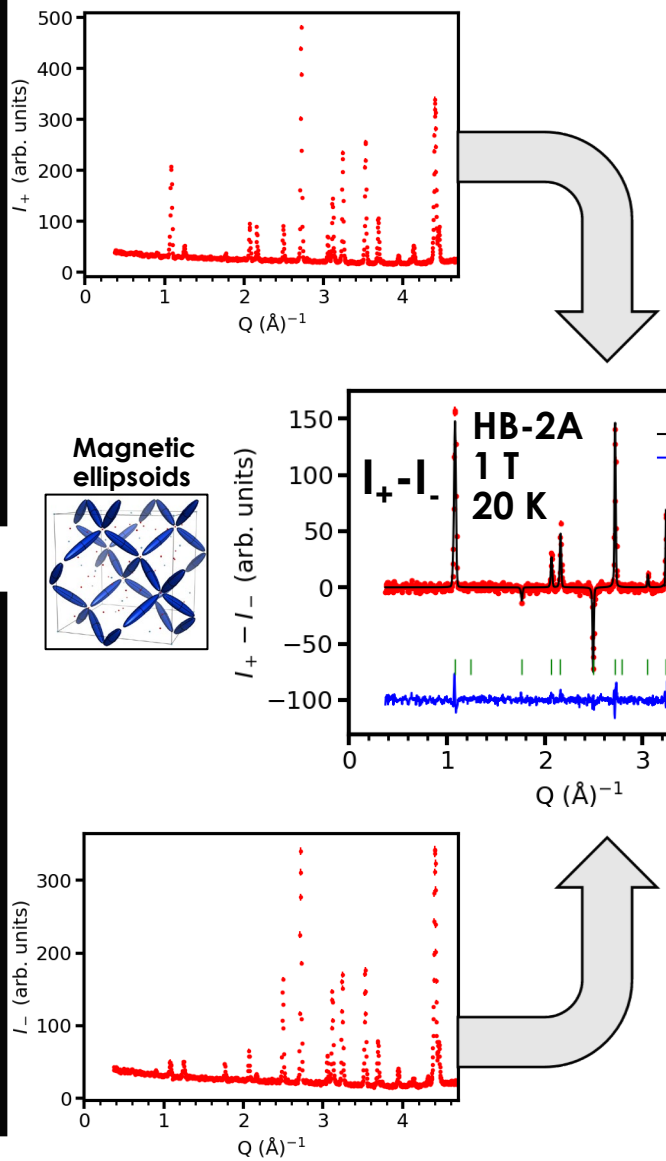
- Polarized neutron powder diffraction data collected on HB-2A at HFIR in a magnetic field of 1 T at several temperatures from 1.5 K to 100 K using using λ= 2.41 Å.
  - ~ 3 gram $Ho_2Ti_2O_7$ polycrystalline rod
  - Counting time per temperature/field was ~3 hours
  - This example will look at 20 K, 1T data with flipper on and flipper off
  - Other temperatures are included to follow the change in anisotropy
- Files included in example:
  - Datafiles:
    - **Ho227_20K_1T_Iplus_Iminus.dat**
    - **Ho227_0T.dat**
  - Crystallographic Information File: **Ho227_crystal_structure.cif**



Half-polarized Neutron Powder Diffraction: HB-2A







S. Calder, Hands-on Polarized Powder data from POWDER (HB-2A).

$Ho_2Ti_2O_7$ Powder Example

- This example will use crysPy to determine the local site susceptibility in the following steps:
  - Step 1: Installing and running CrysPy
  - Step 2: Refine unpolarized, non-magnetic $Ho_2Ti_2O_7$ crystal structure using CrysPy
  - Step 3: Refine the polarized $Ho_2Ti_2O_7$ data using CrysPy
  - Step 4: Interpreting the results

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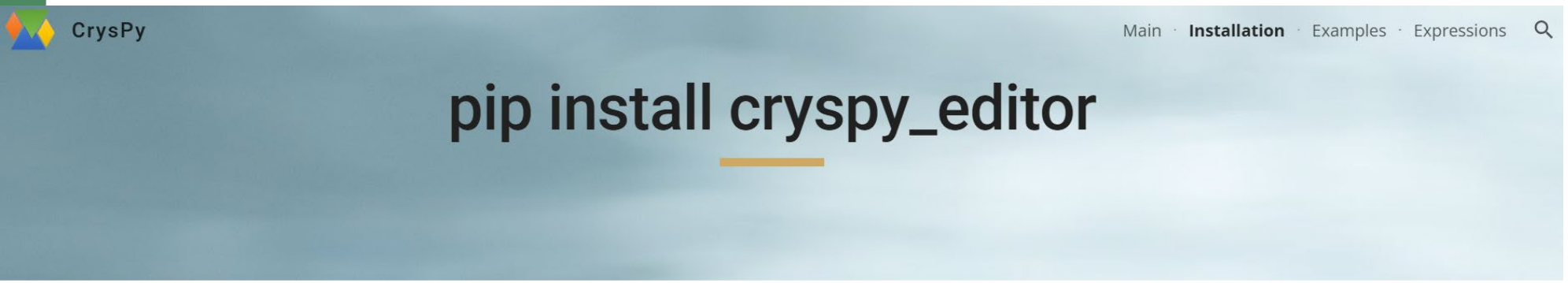
Step 1: Installing and running CrysPy https://sites.google.com/view/cryspy/



Stable version (PIP)

The stable version of CrysPy and "CrysPy Editor"can be downloaded through pip (python 3 has to be installed):

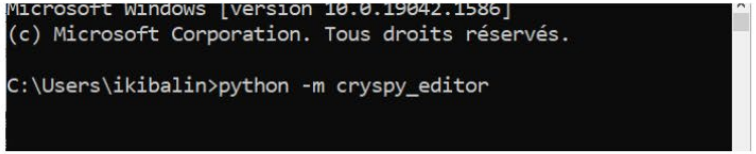
```
python -m pip install cryspy_editor
```

to upgrade the library type:

```
python -m pip install --upgrade cryspy cryspy_editor
```

To run "CrysPy editor" type in console:

```
python -m cryspy_editor
```



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CrysPy can refine polarized as well as unpolarized data on both powders and single crystals.

Neutron and x-ray data.

Python based with GUI

$Ho_2Ti_2O_7$ Powder Example

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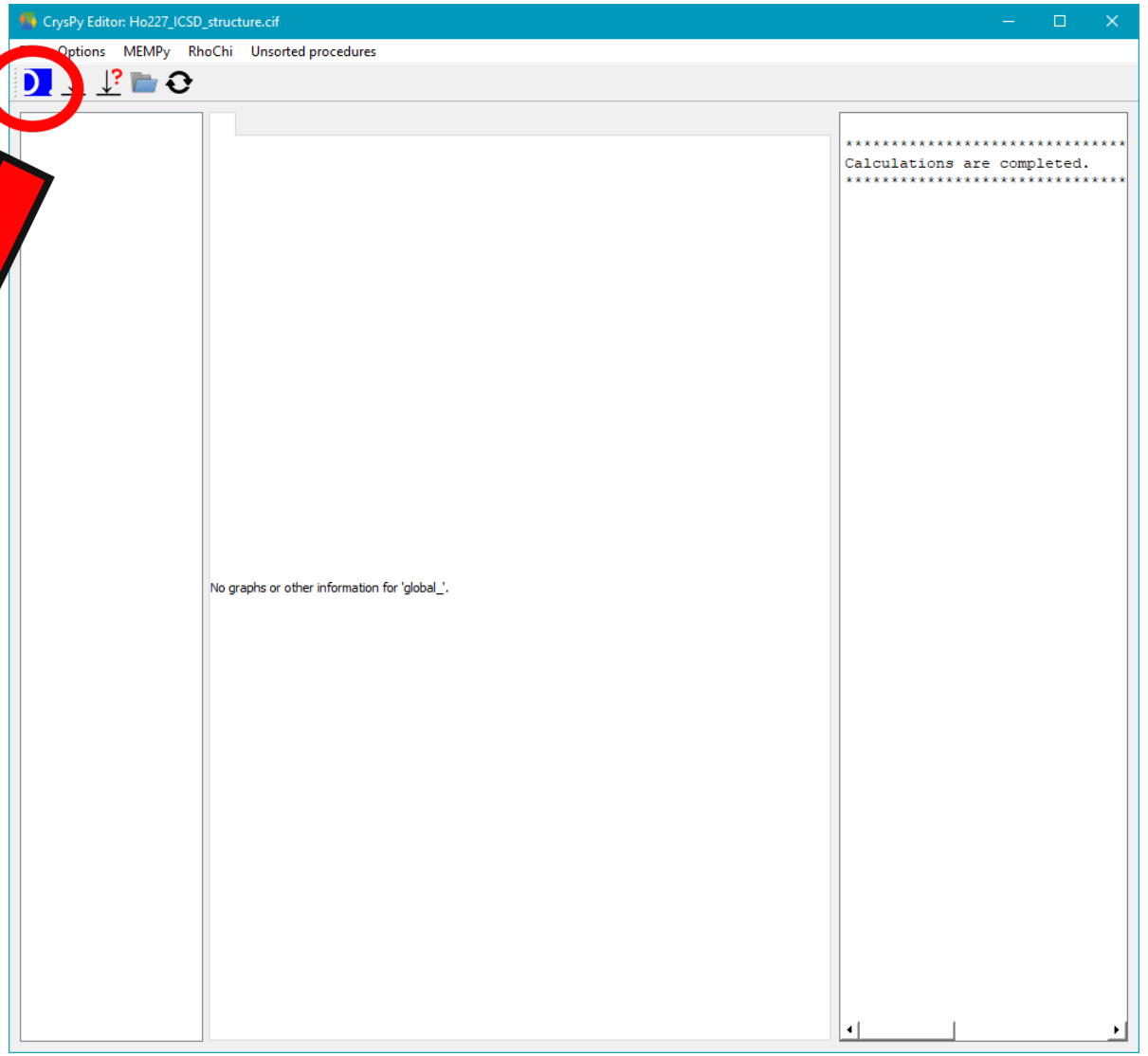
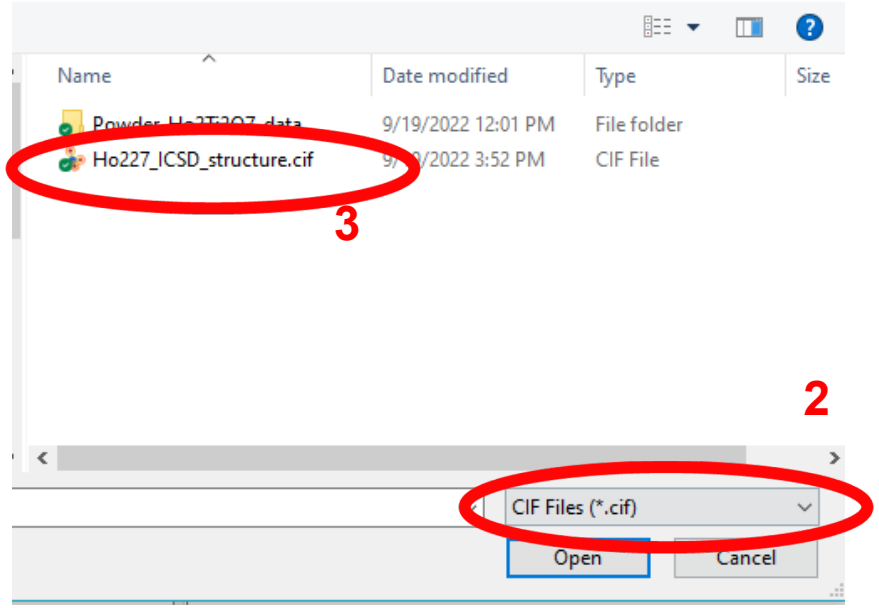
  - Step 4: Interpreting the results

Step 2: Refine the unpolarized, non-magnetic crystal structure using CrysPy

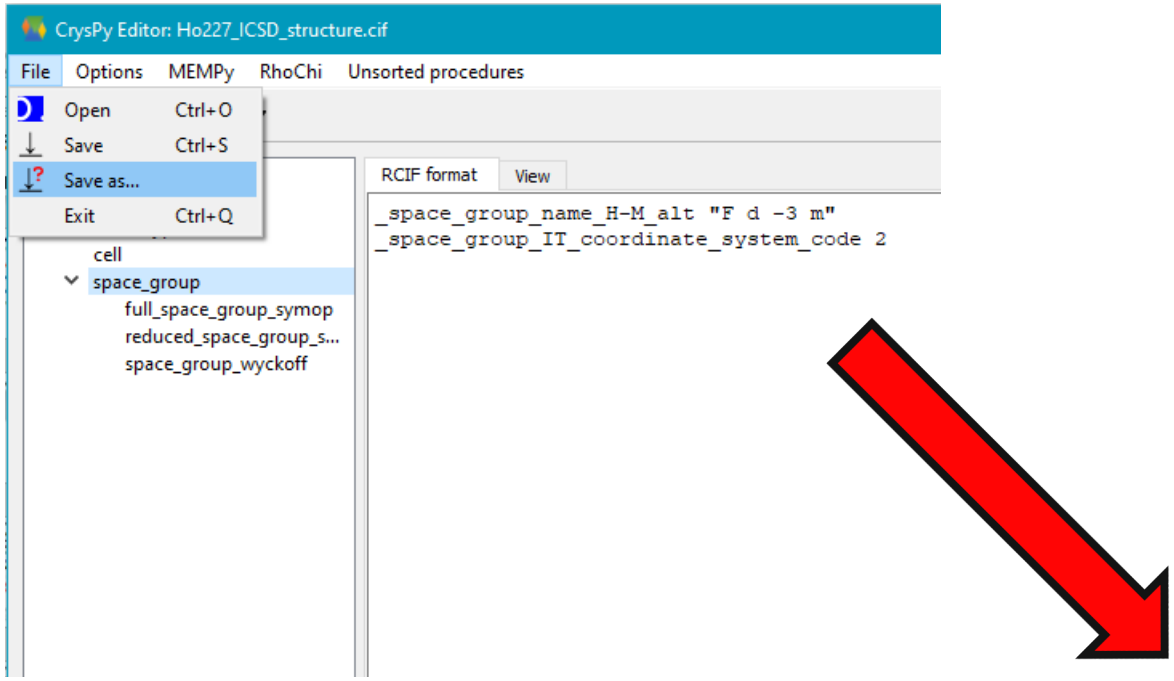
- In the CrysPy_Editor
  - **1.** Open a new file.
  - **2.** you can select a .rcif (CrysPy file) or a standard .cif file format.
  - **3.** We will start with a .cif for $Ho_2Ti_2O_7$ obtained from ICSD.

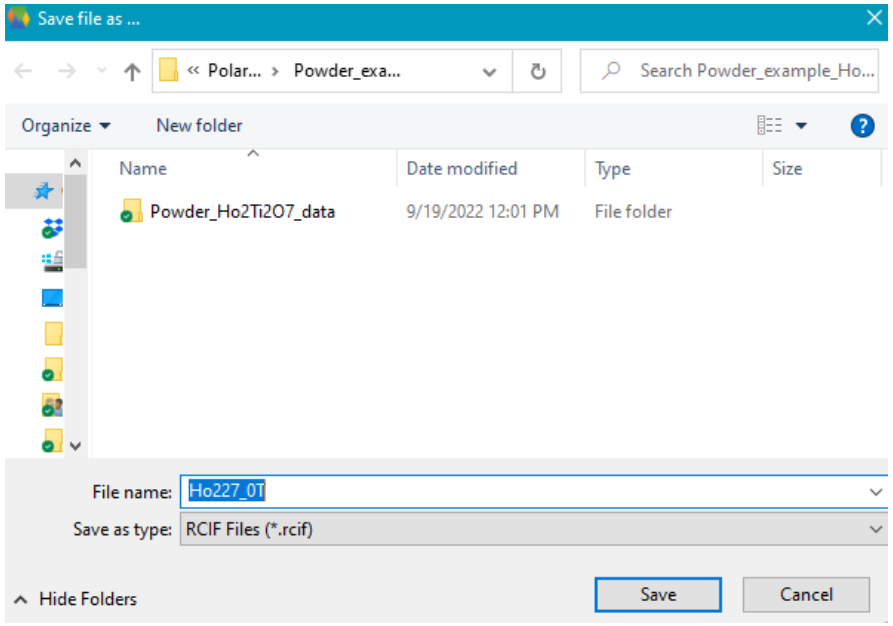




Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- Save this in .rcif format

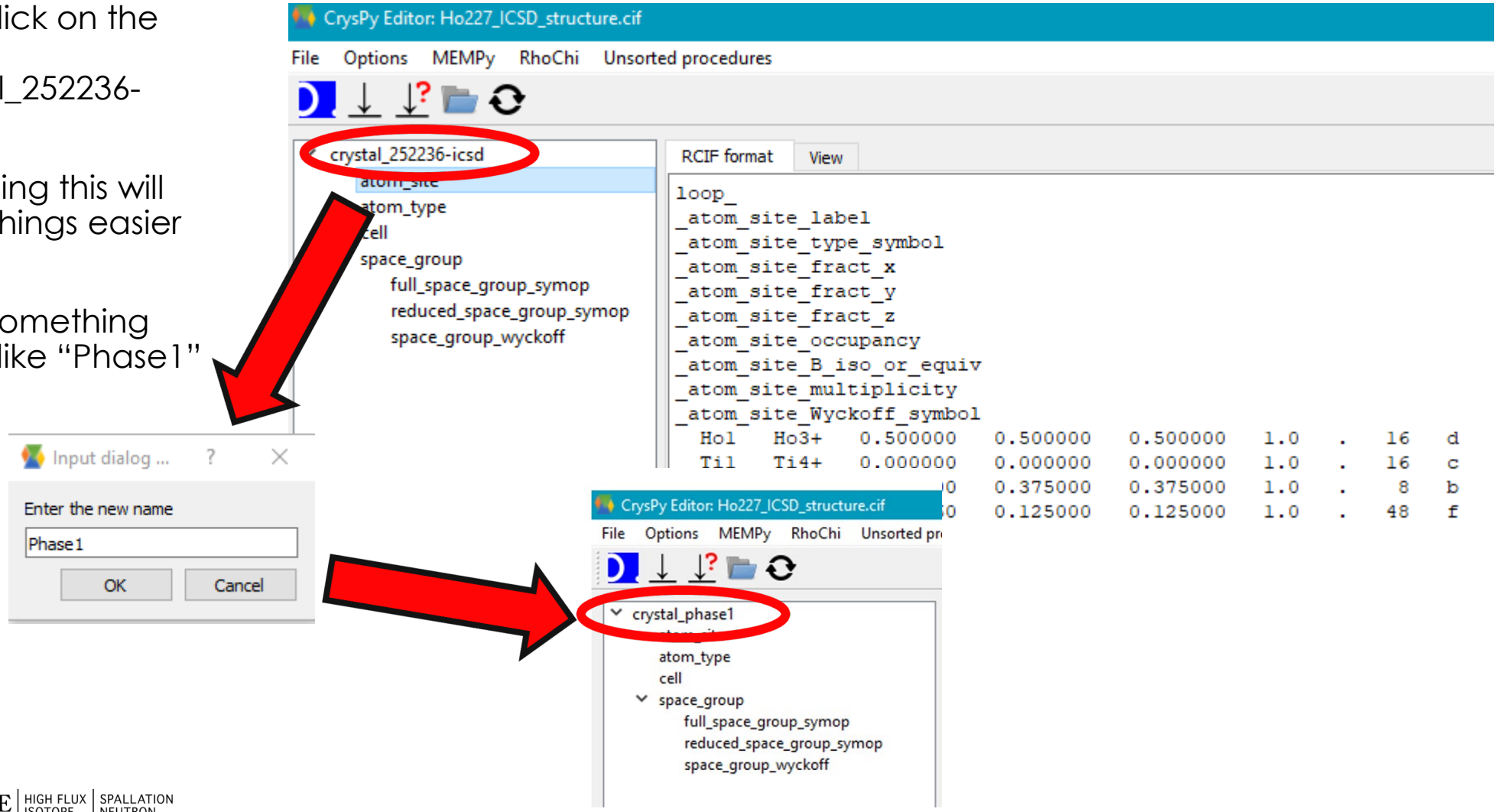




Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- This structure parameters are loaded into the crysPy_Editor from the cif

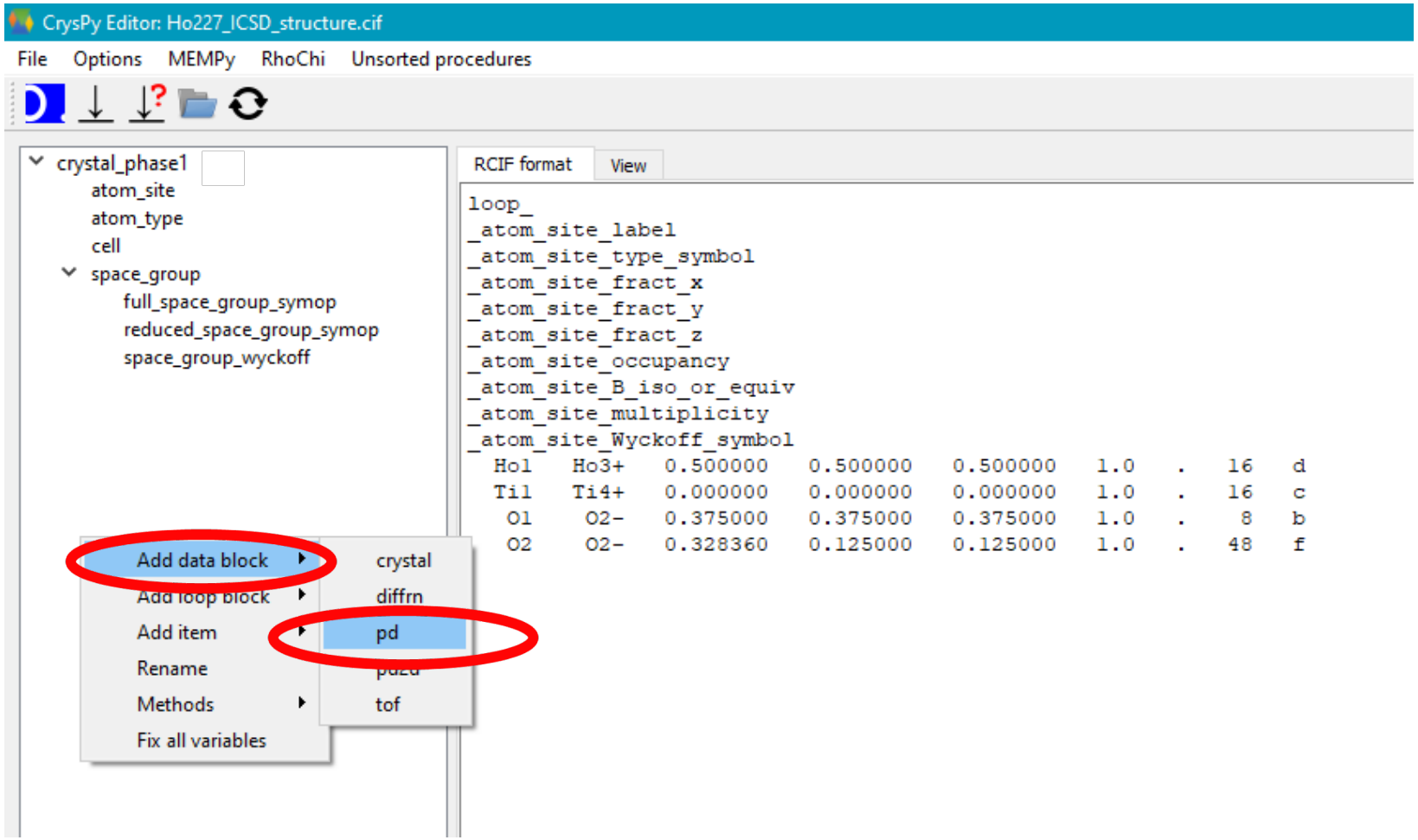
- Right click on the name "crystal_252236-icsd".
- Renaming this will make things easier later.
- Call it something simple like "Phase1"



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- We now want to add the data and instrument parameters to refine the data.

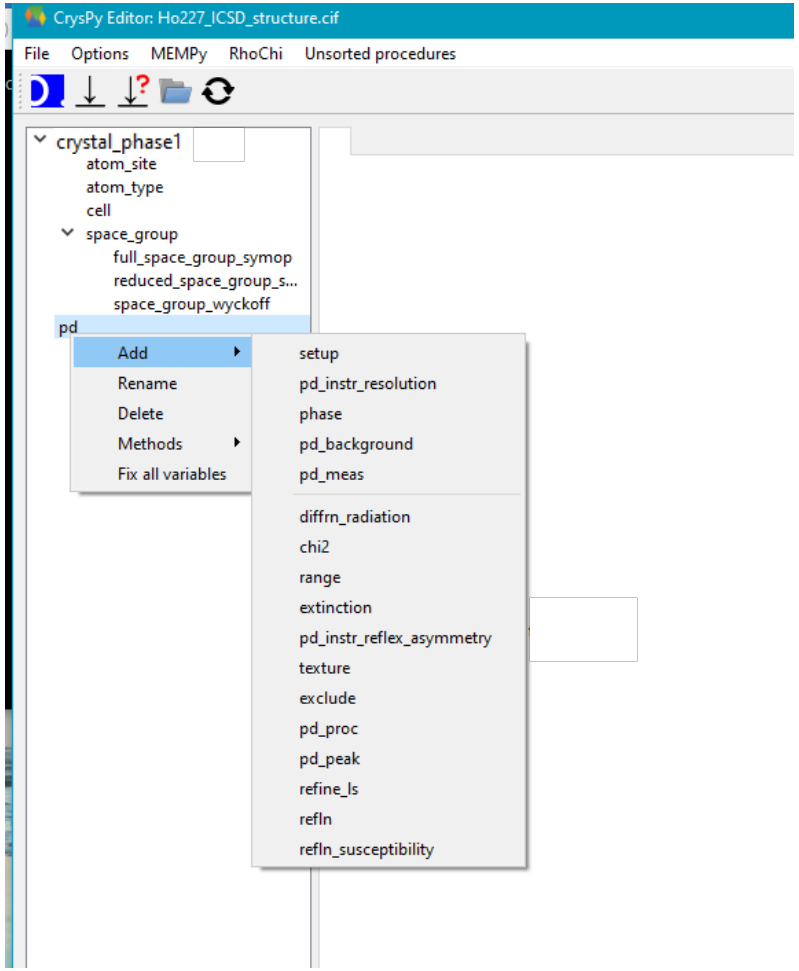
  - **1.** Right click on the left hand panel → Add data block → pd



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- We now want to add the data and instrument parameters to refine the data.

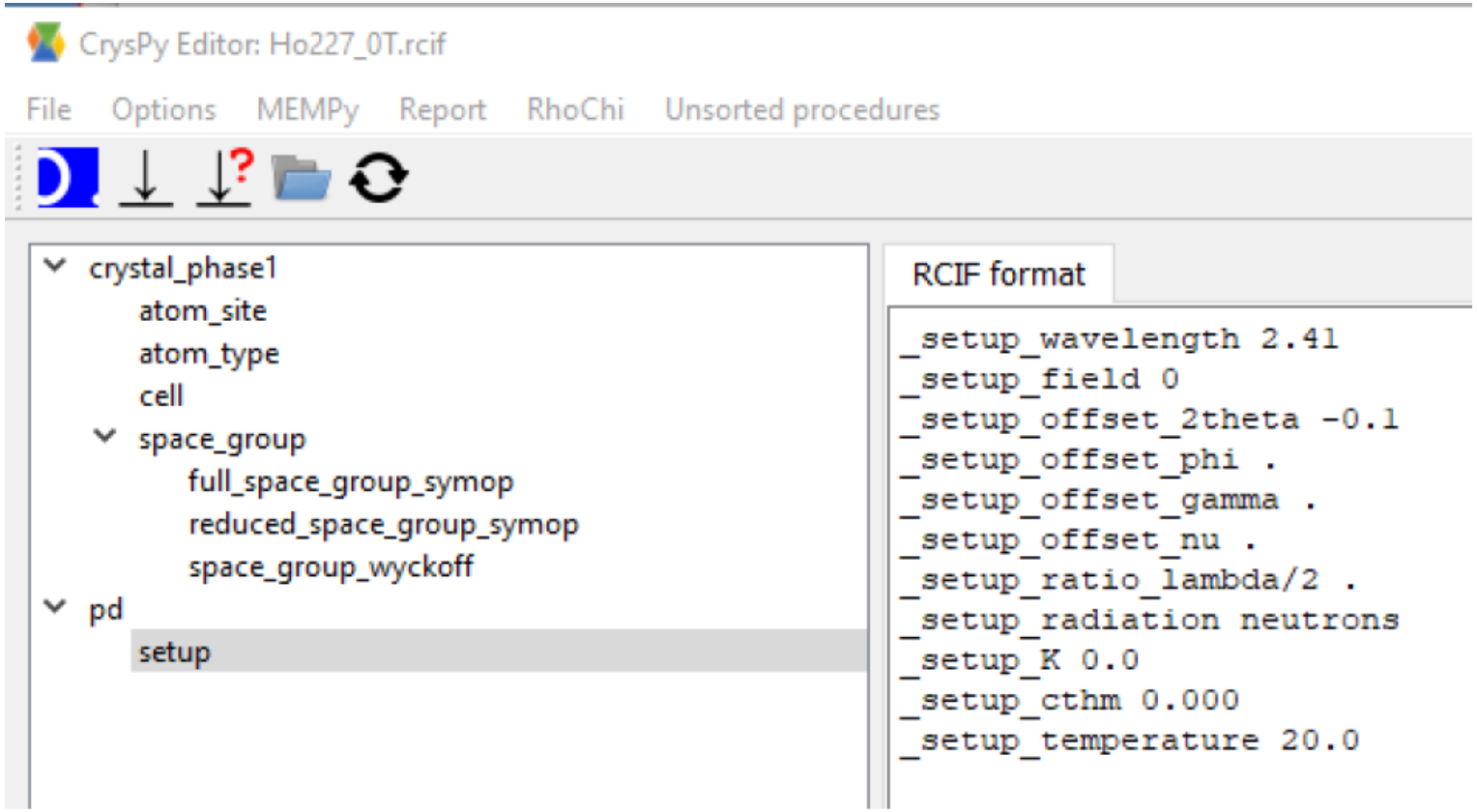
  - Right click on “pd” to select the subcategories.
  - We will add these one at a time.



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- **pd→ setup**

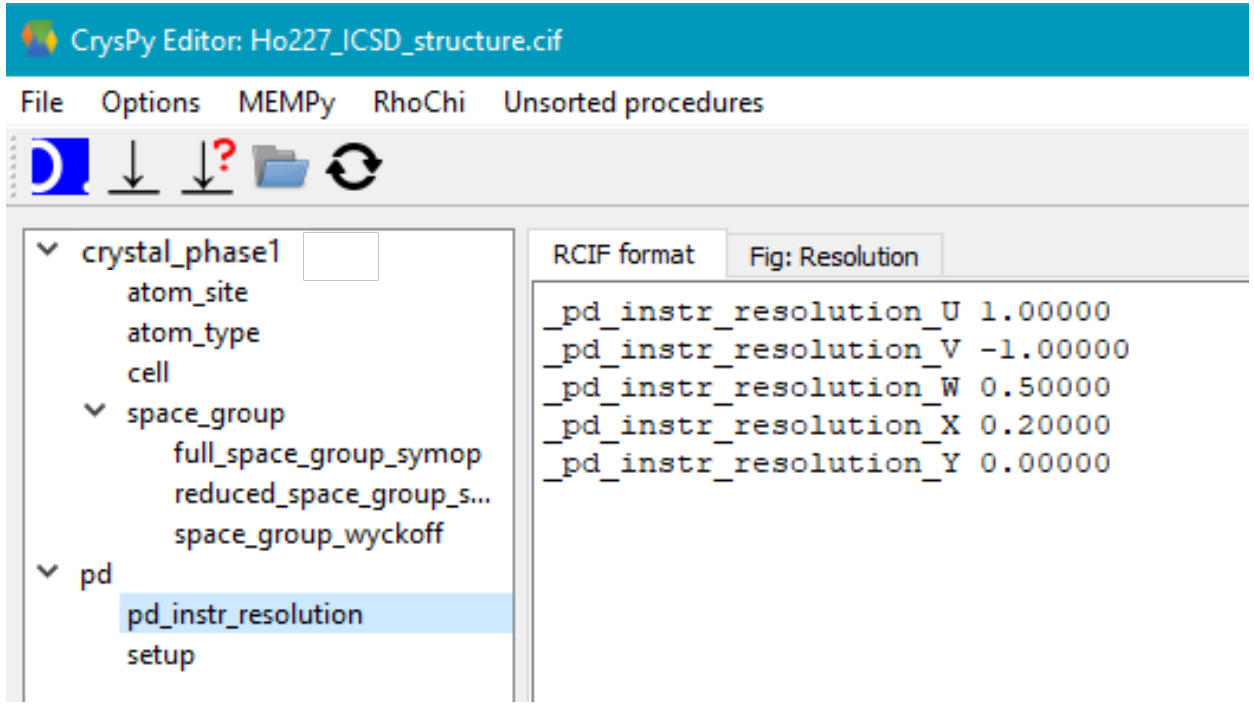
- We will first refine the zero-field data. So set field "0"
- The wavelength used on HB-2A was 2.41
- From calibration measurements the 2theta offset of the detector was -0.1
  - This should be discussed with instrument team



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- **pd→ pd_instr_resolution**

- Starting profile peak shape parameters.
- These will depend on instrument.
  - Ask instrument scientist for reasonable values or calibration data

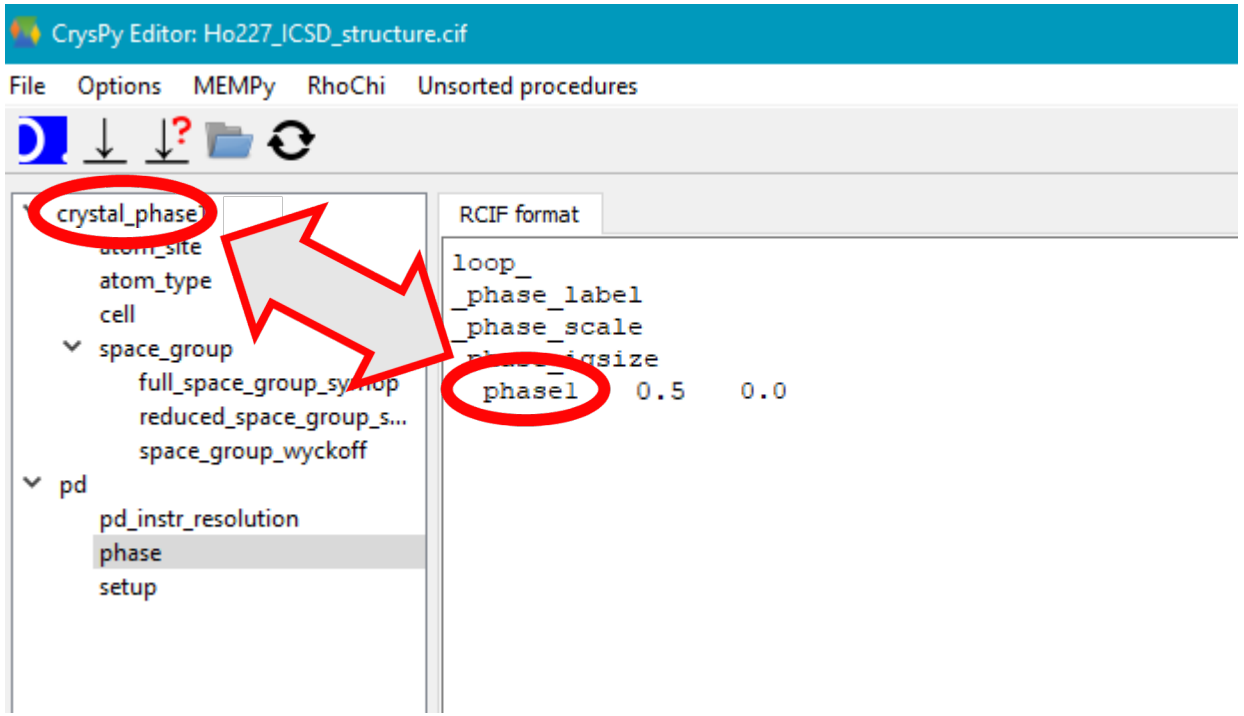


Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- **pd→ phase**

- Make sure the phase_label matches the crystal name!!!!
  - "phase1"

- The starting value for the scale is based on a best guess. This will be refined.

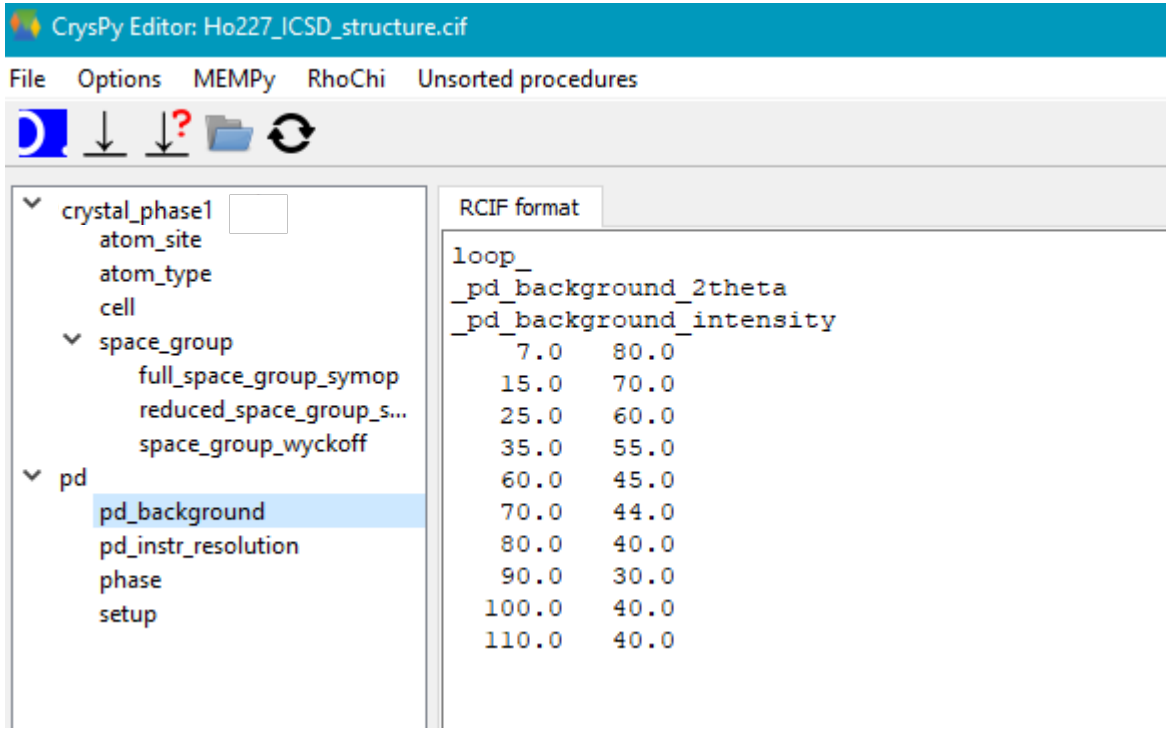


Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- **pd→ pd_background**

- Check the data file and put in some reasonable background starting values.

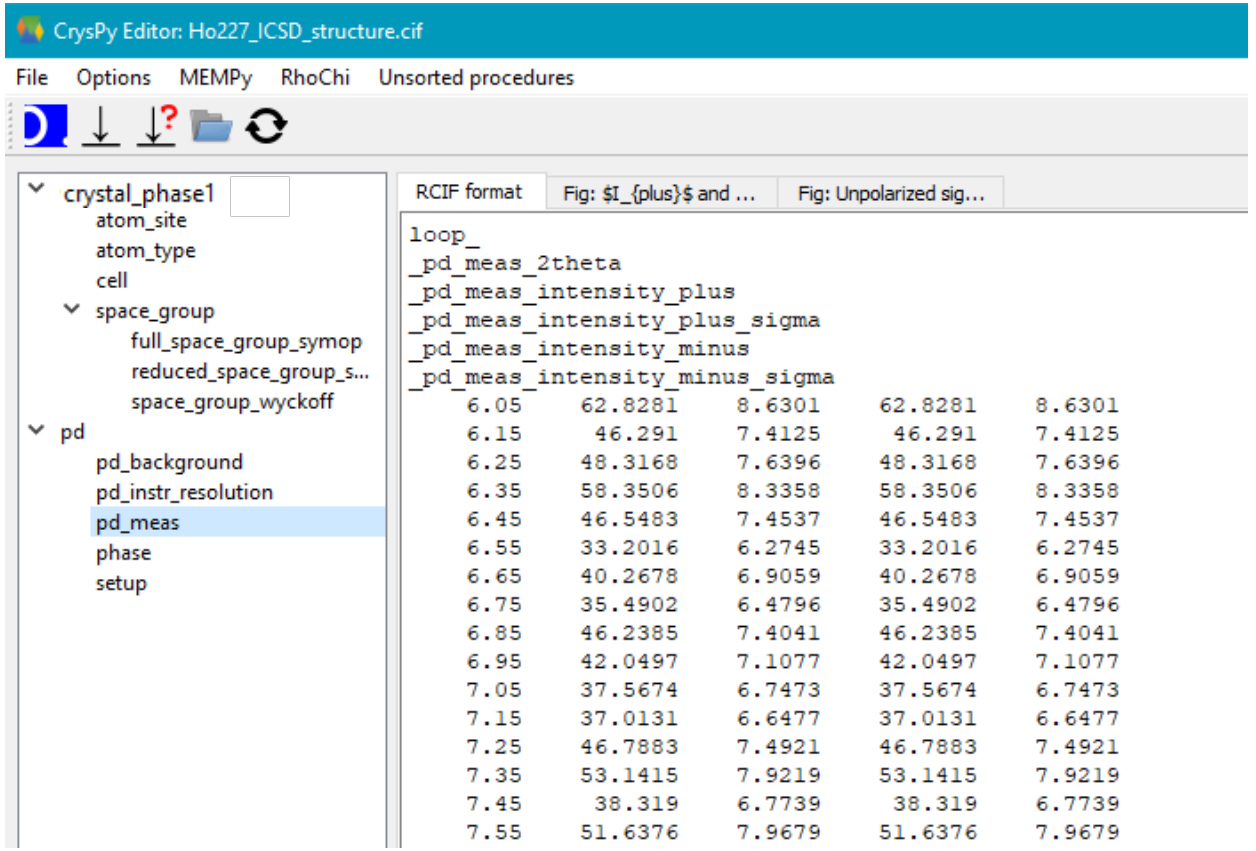
- *NOTE: Be careful not to refine a 2theta position that is excluded.*



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

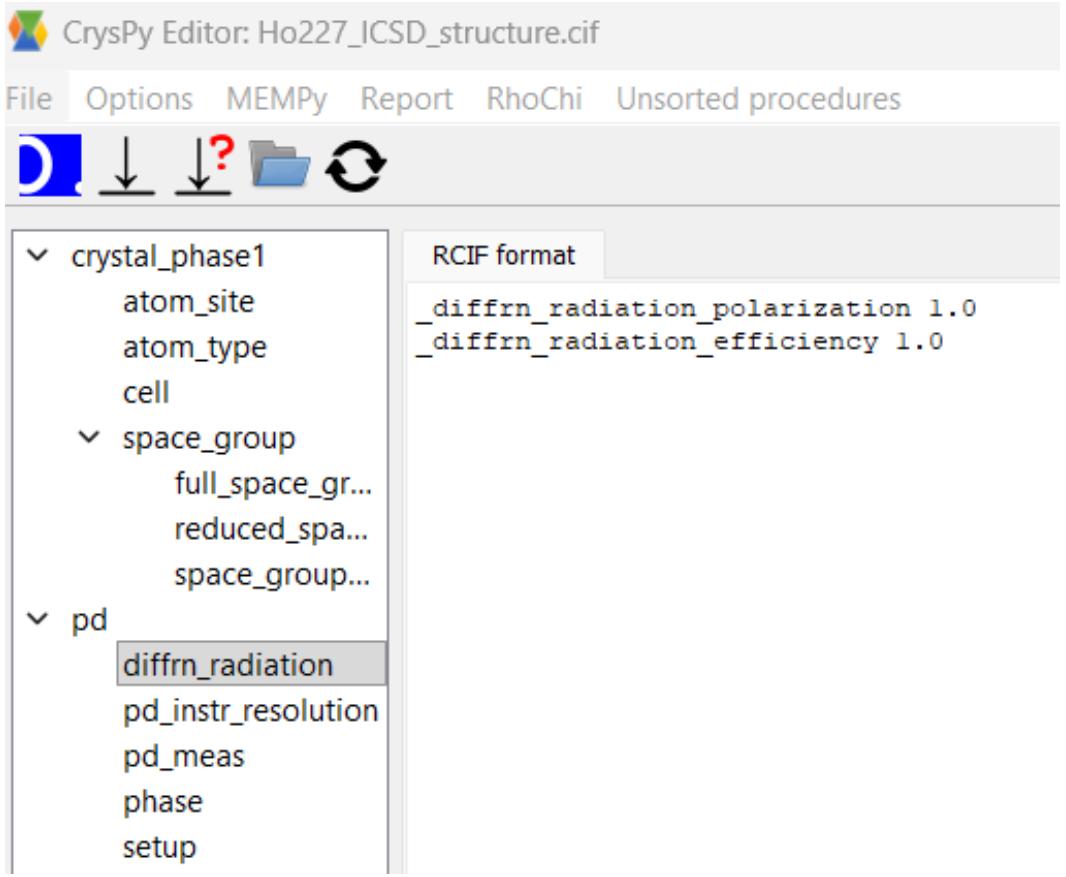
- **pd→ pd_meas**

- Add the data. The 0T data is saved in the example folder and named "Ho227_0T_data.dat".
- Copy and paste the data over everything there in the "pd_meas" tab
- NOTE: we add both plus and minus data, even though this is 0T. This makes it easier to use this 0T refined phase and background values for the actual polarized data in STEP 3.



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- **pd→ diffrn_radiation**

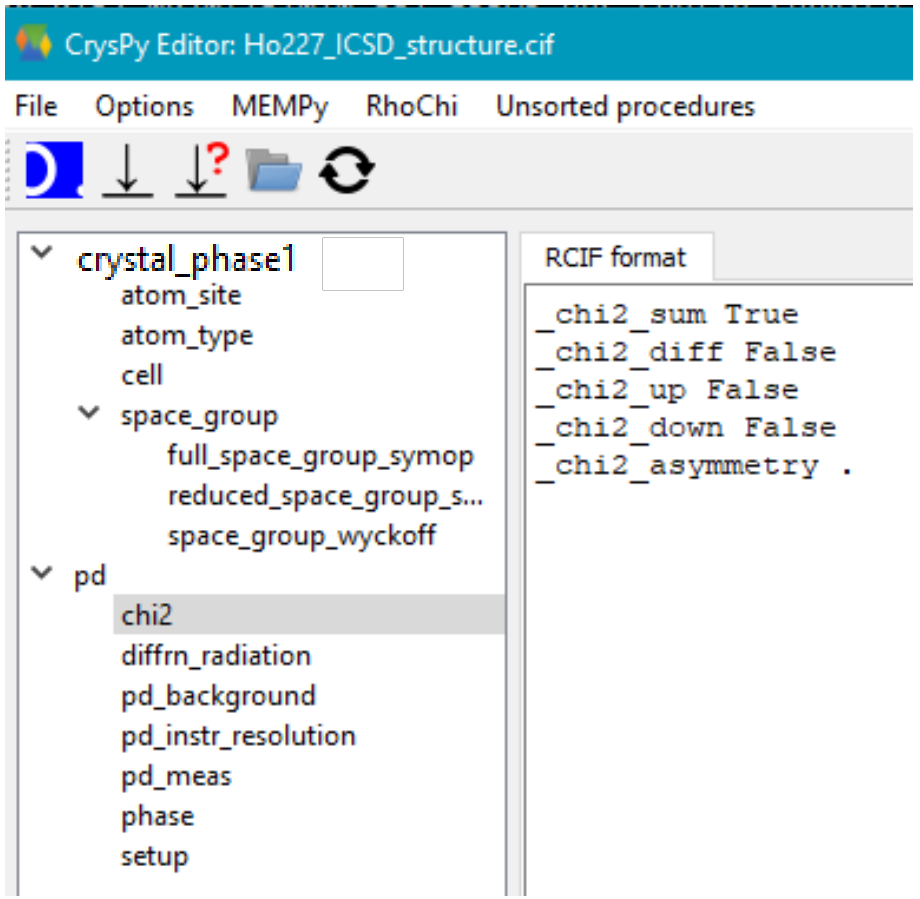


- This is for the beam polarization.
- The incident beam polarization should be known for the specific instrument.
- *NOTE: for cases where the sample depolarizes the beam this can be refined or even better experimentally determined.*
- The data in step 2 data is 0 T and polarization is not being considered. So set to standard values shown.
  - We will discuss this further when looking at the polarized data in Step 3

Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

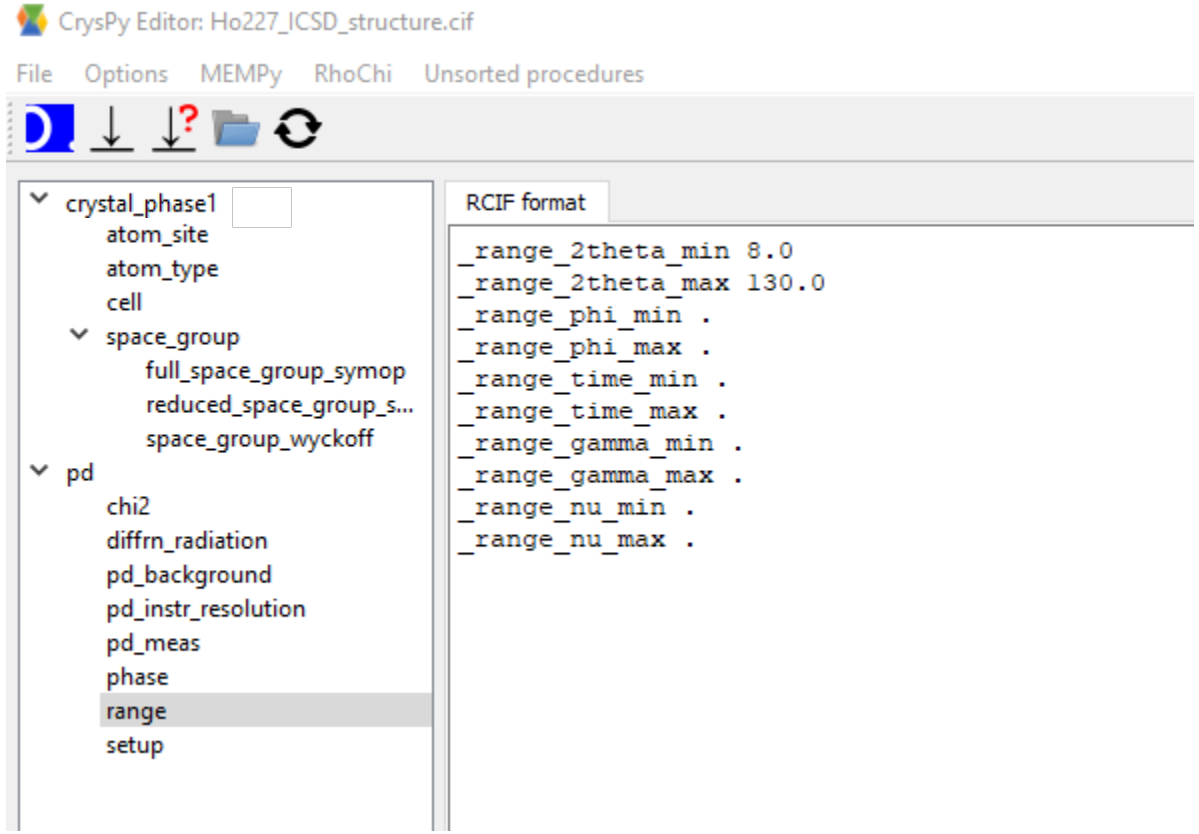
- **pd→ chi2**

- This allows you to flag which data is refined.
  - chi2_sum is minus + plus (so recovers the unpolarized data)
  - chi2_diff is the difference. This will be used in STEP 3 when refining the polarized data.



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- **pd→ range**
- Range of data to consider

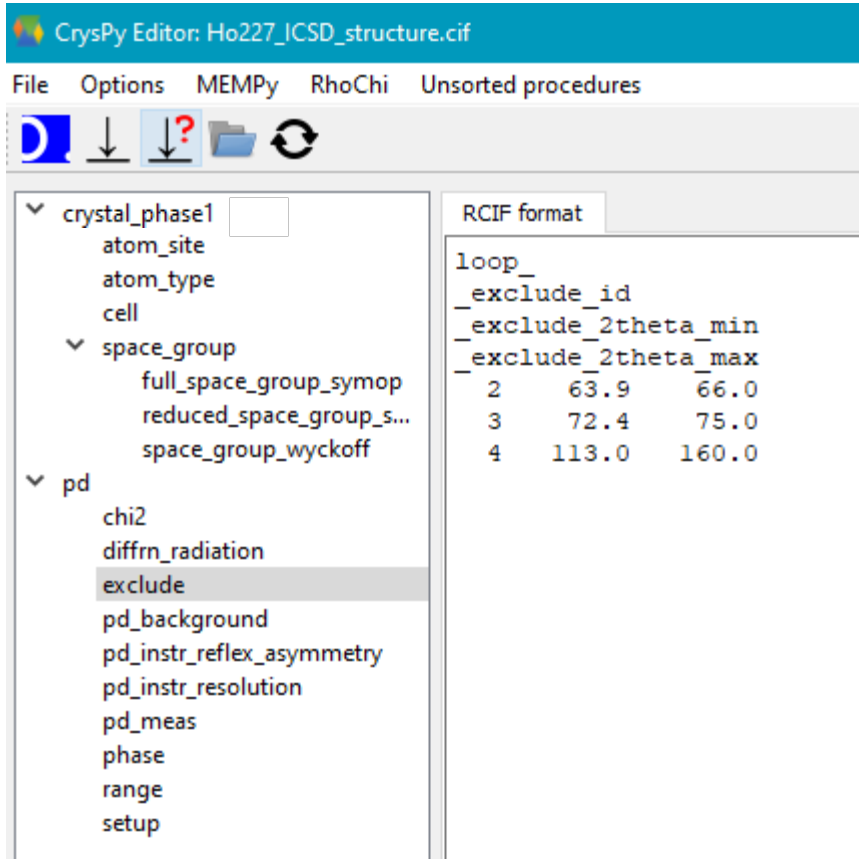


Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- **pd→ exclude**

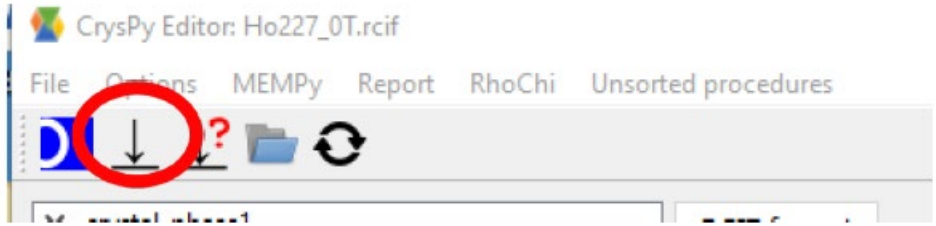
- If there are regions of the data that you want to exclude, this is added here.

- *NOTE: it is best to refine all the data. But in this case we will remove the peaks from the Al sample can*



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

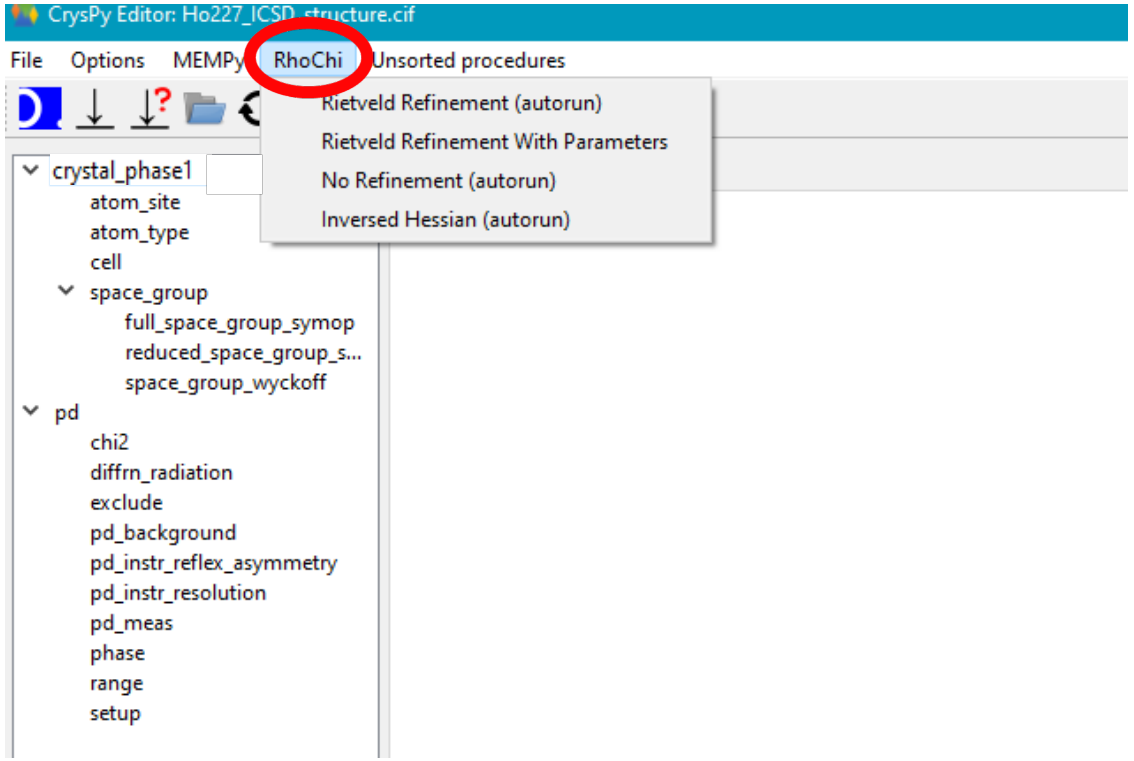
- This is the end of the subcategories that you need to add.
- Hit the save button



- When you run crysPy more subcategories will appear.
- These contain information on the model (reflection list, goodness of fit, etc).

Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

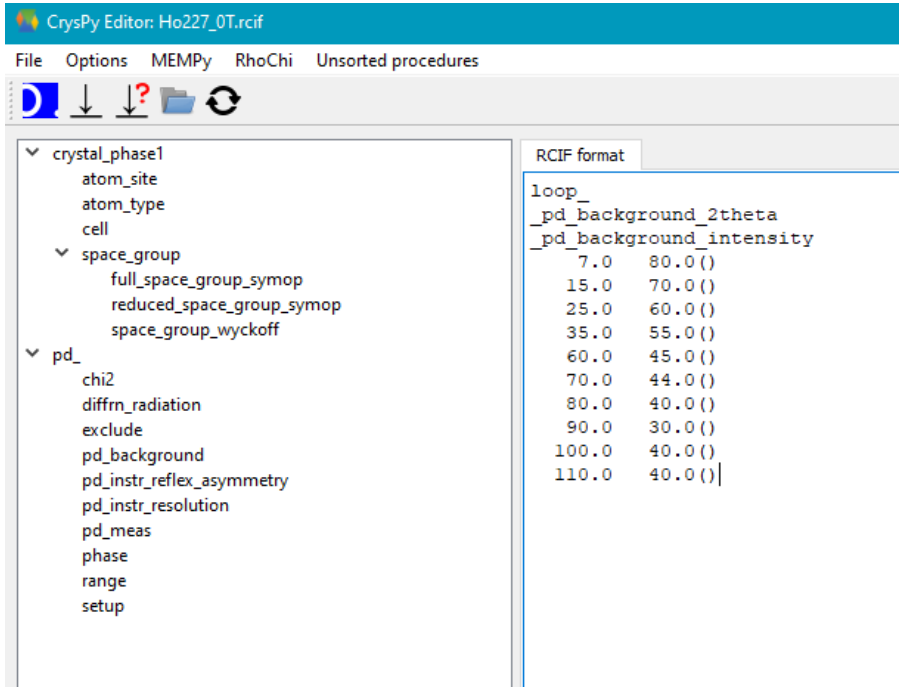
- The refinement is run using the "RhoChi" drop-down menu
- First run "No Refinement (autorun)" to check if the initial parameters are close.



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- We're now ready to refine the parameters.
- To set a variable to refine put a () after it.
- For categories with lots of values you can right click and select "refine all variables"
  - Do this for the background

NOTE: There might be an error when you run the refinement. But there is a quick fix!



Potential bug in this version of CrysPy (March 2026)

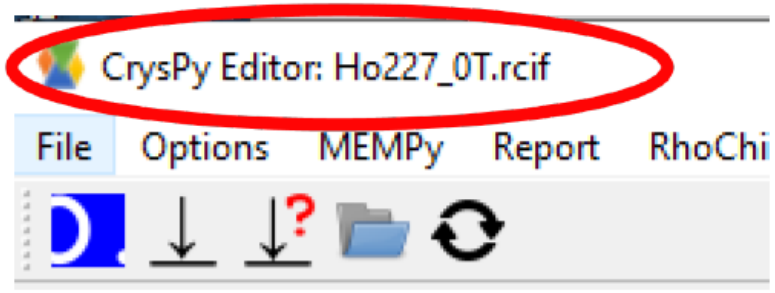
- If you see this error, then reopen the ".rcif" file **[SEE NEXT SLIDE]**

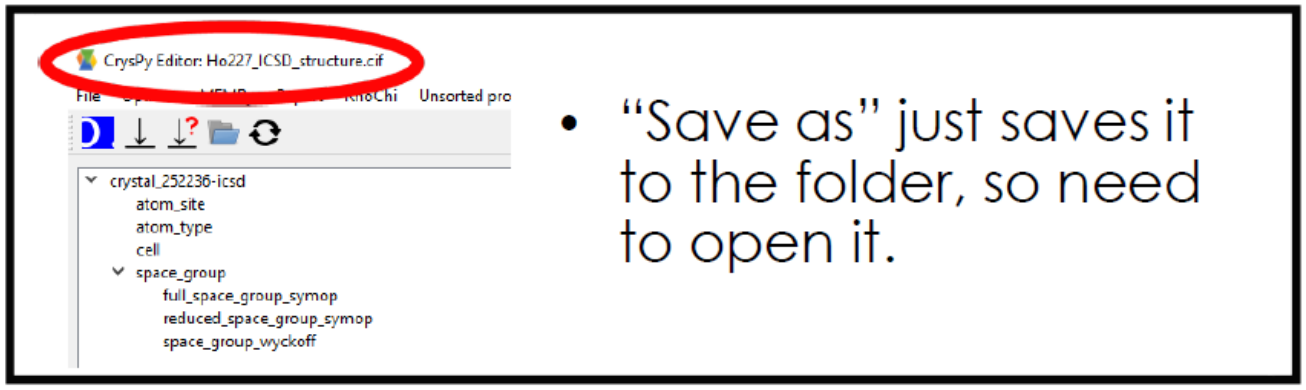
```
************************************************
Rietveld refinement by CrysPy (module RhoChi)
************************************************
Derivatives are calculated numerically.
Preliminary calculations...
************************************************
Calculations are completed.
************************************************
Result of function is
ERROR DURING PROGRAM EXECUTION

Traceback (most recent call last):
  File "C:\Users\cr9\AppData\Local\anaconda3\lib\site-
packages\cryspy_editor\cl_thread.py", line 127, in run
    out = func(*arg)
  File "C:\Users\cr9\AppData\Local\anaconda3\lib\site-
packages\cryspy\procedure_rhochi\rhochi.py", line 73, in
rhochi_rietveld_refinement
    dict_out = rhochi_rietveld_refinement_with_parameters(
  File "C:\Users\cr9\AppData\Local\anaconda3\lib\site-
packages\cryspy\procedure_rhochi\rhochi.py", line 95, in
rhochi_rietveld_refinement_with_parameters
    chi_sq, parameter_name, dict_in_out, res =
rhochi_rietveld_refinement_by_dictionary(
  File "C:\Users\cr9\AppData\Local\anaconda3\lib\site-
packages\cryspy\procedure_rhochi\rhochi_by_dictionary.py", line 126, in
rhochi_rietveld_refinement_by_dictionary
    print(f"Started chi_sq per number of points is {chi_sq/n_point:.2f}.
")
ZeroDivisionError: division by zero
```

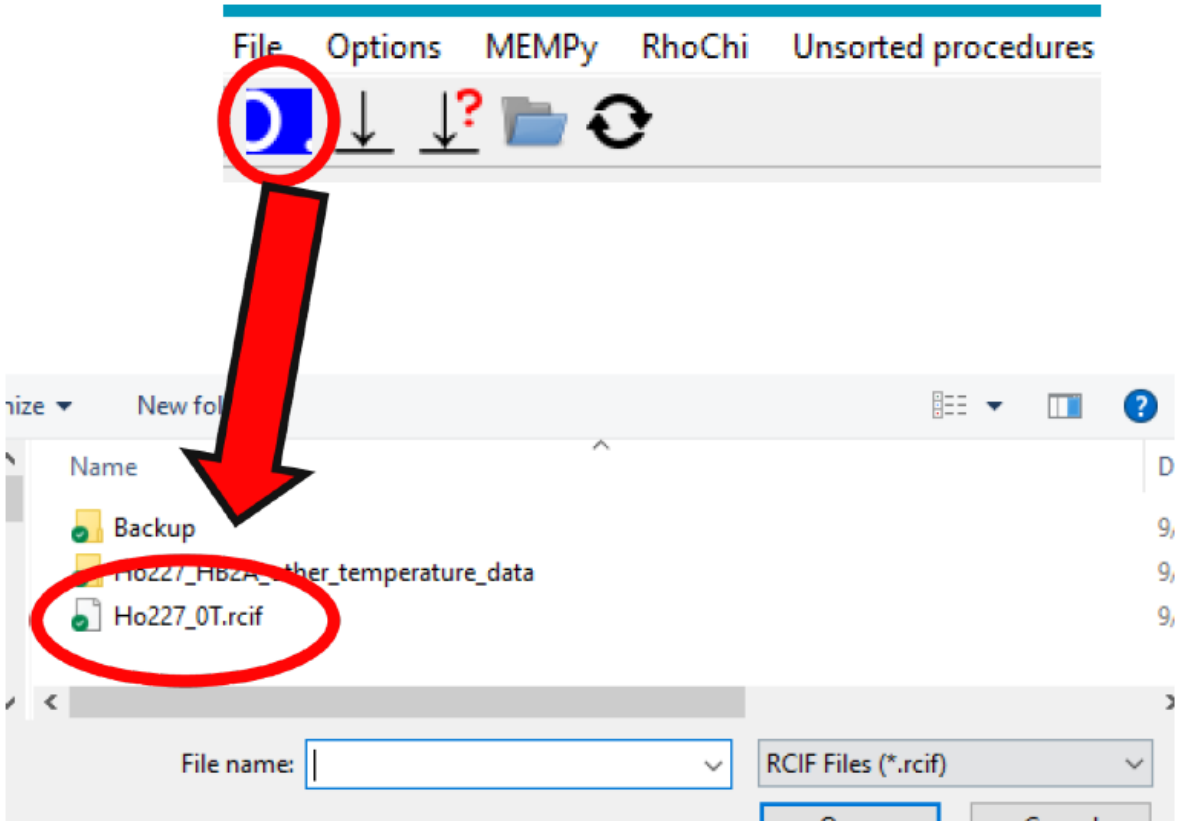
Potential bug in this version of CrysPy (March 2026)

- If the displayed filename ends in ".cif" then you need to open the saved .rcif file
- Now the top left of the program should say:





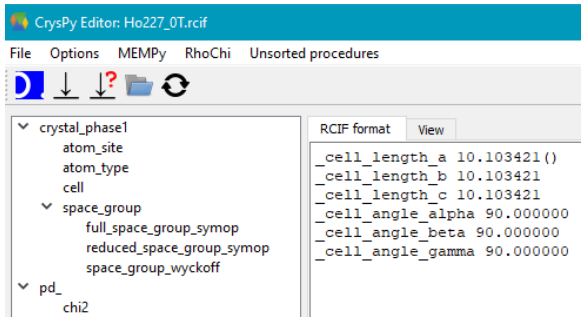
- "Save as" just saves it to the folder, so need to open it.

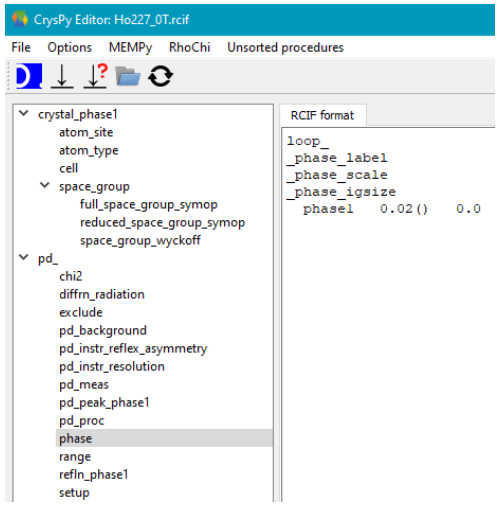


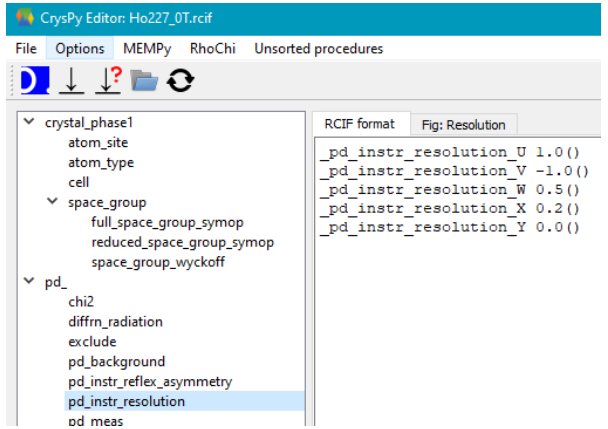
- *NOTE: It is often easier to edit a known working file when starting a new project, rather than setting everything up from the beginning.*

Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- Now refine more parameters to improve the model:
  - Cell (lattice constants)
  - Phase (scale),
    only refine the first number.
    Keep second 0.
  - pd_instr_resolution (peak shape)
  - Background

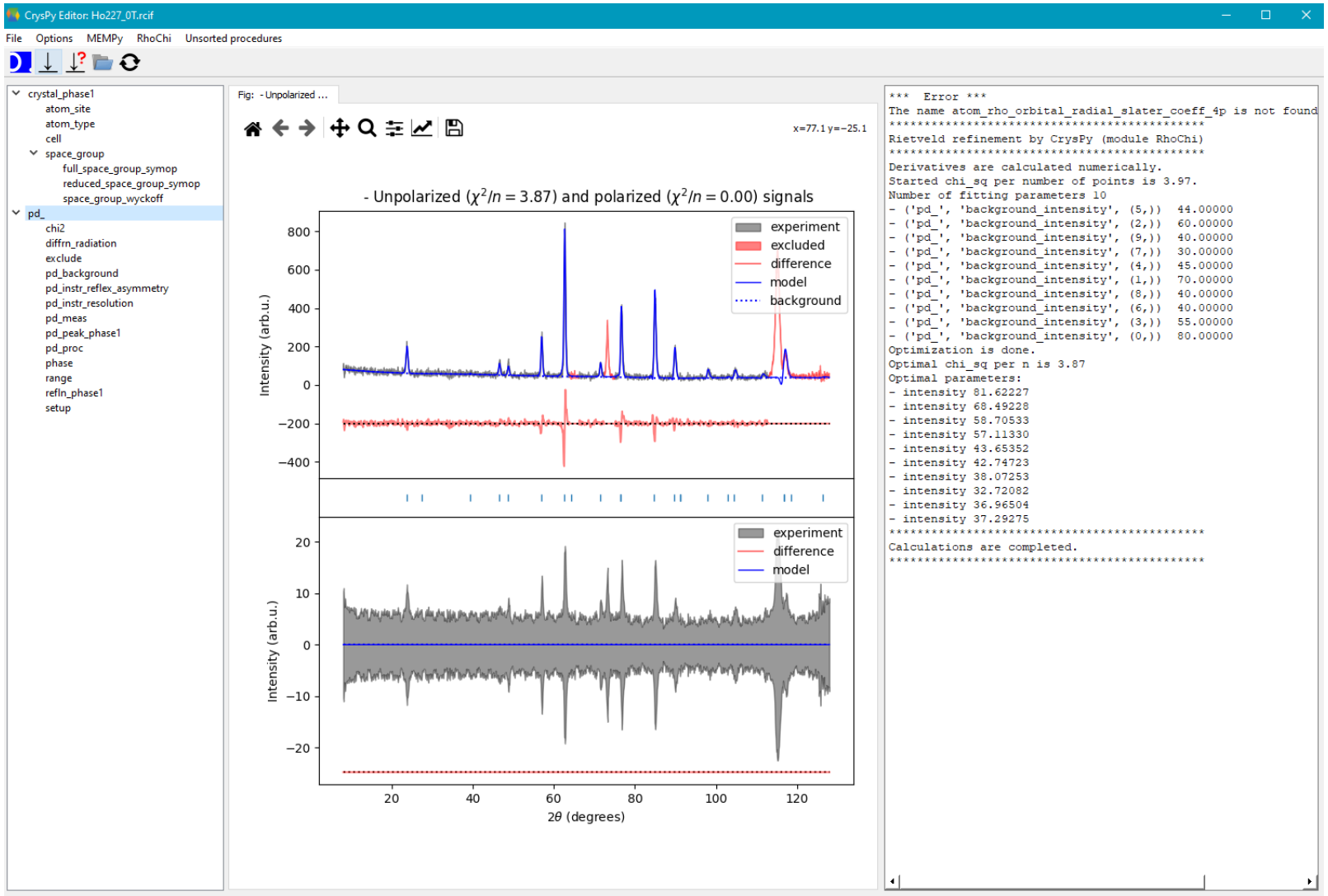






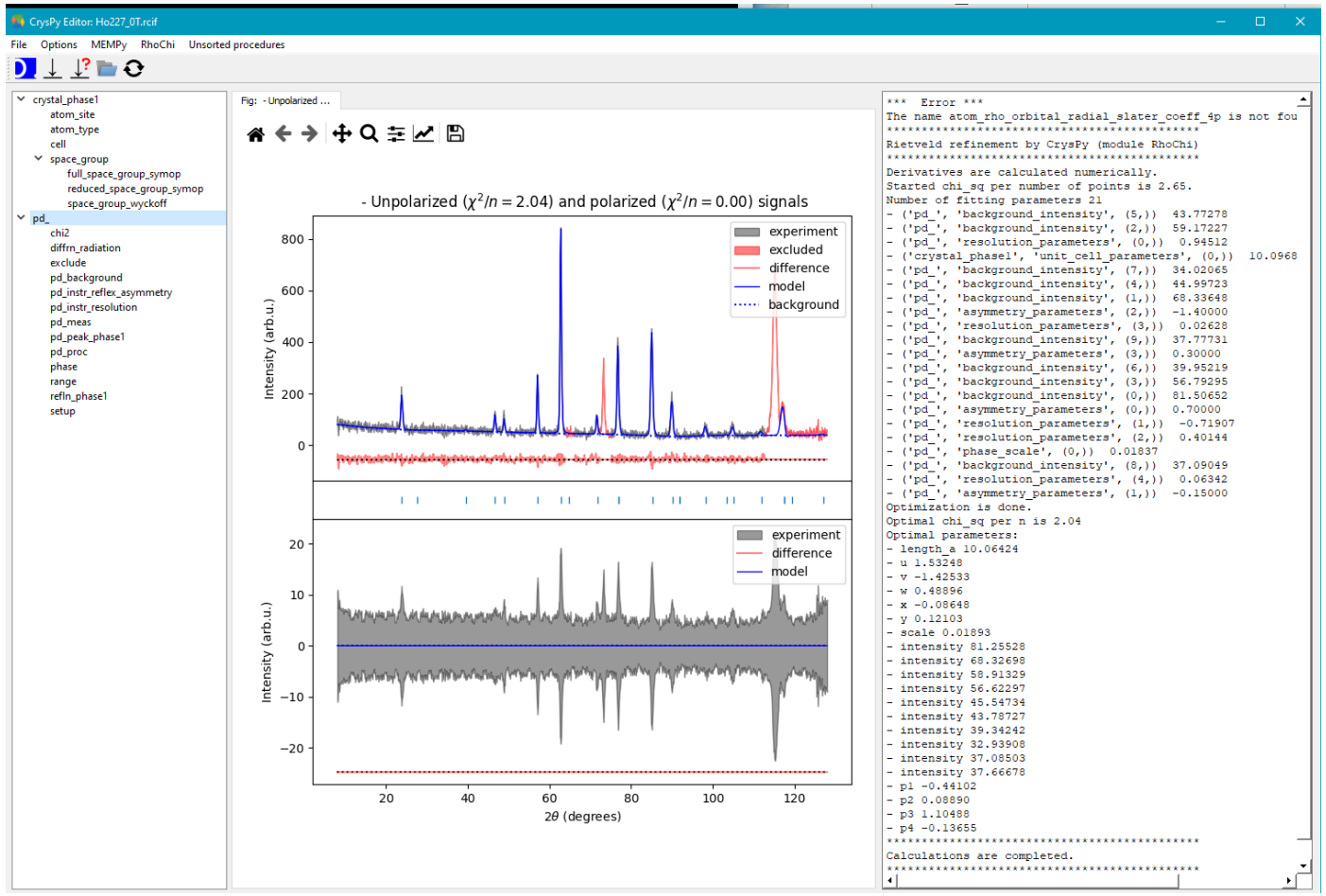
Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- Run the refinement
  - "RhoChi" → Rietveld Refinement (autorun)
  - The right-hand side should go yellow and show the variables refining.
- Once complete, check the data and model pattern by clicking on the "pd_" category.
- The model and data are very close
- NOTE: The bottom panel (difference) is zero. This is unpolarized data and so is correct!



Step 2: Refine the unpolarized, non-magnetic crystal structure using crysPy

- The fit is very good (by eye and for Chi^2).
- Now we can fix (i.e. no longer refine) the instrument parameters (phase, U,V,W,X, etc) when refining the polarized data.
- If this STEP 2 of refining unpolarized zero field data is not done then it can be hard to interpret the polarized difference data.
- This becomes more important the weaker the difference signal or the more change to scattering with an applied magnetic field.



$Ho_2Ti_2O_7$ Powder Example

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  - Step 1: Installing and running CrysPy
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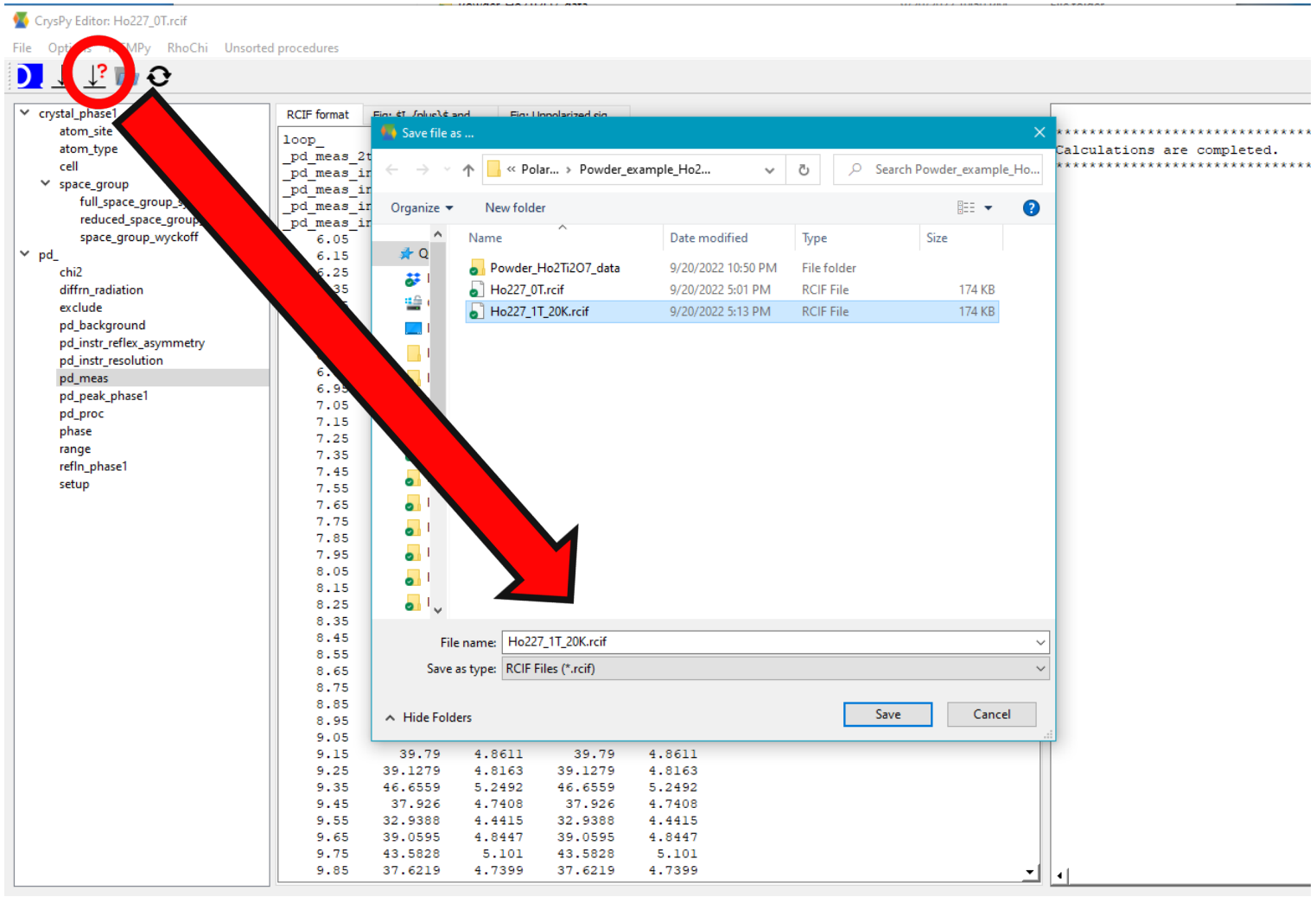
  - **Step 3: Refine the polarized $Ho_2Ti_2O_7$ data using CrysPy**
  - Step 4: Interpreting the results

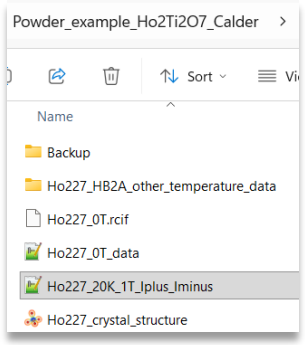
Step 3: Refine the polarized flipper on and flipper off data using using crysPy

- We will start with the .rcif file from STEP 2. Make sure all the refinement label "()" are removed so nothing is refining.
  - This can be done quickly by right clicking on "crystal_phase1" and select "Fix all variables". Do the same for "pd_", right click, select "Fix all variables"
- Rename the file to something else. We will call is "Ho227_1T_20K.rcif"
- **NOTE: If the displayed name doesn't change, then open the file (see slide 27 for similar bug)**

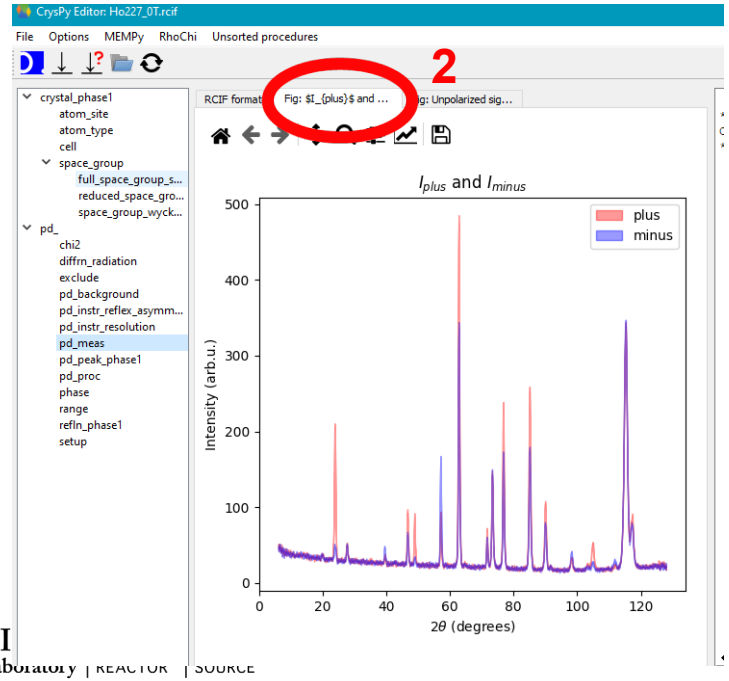


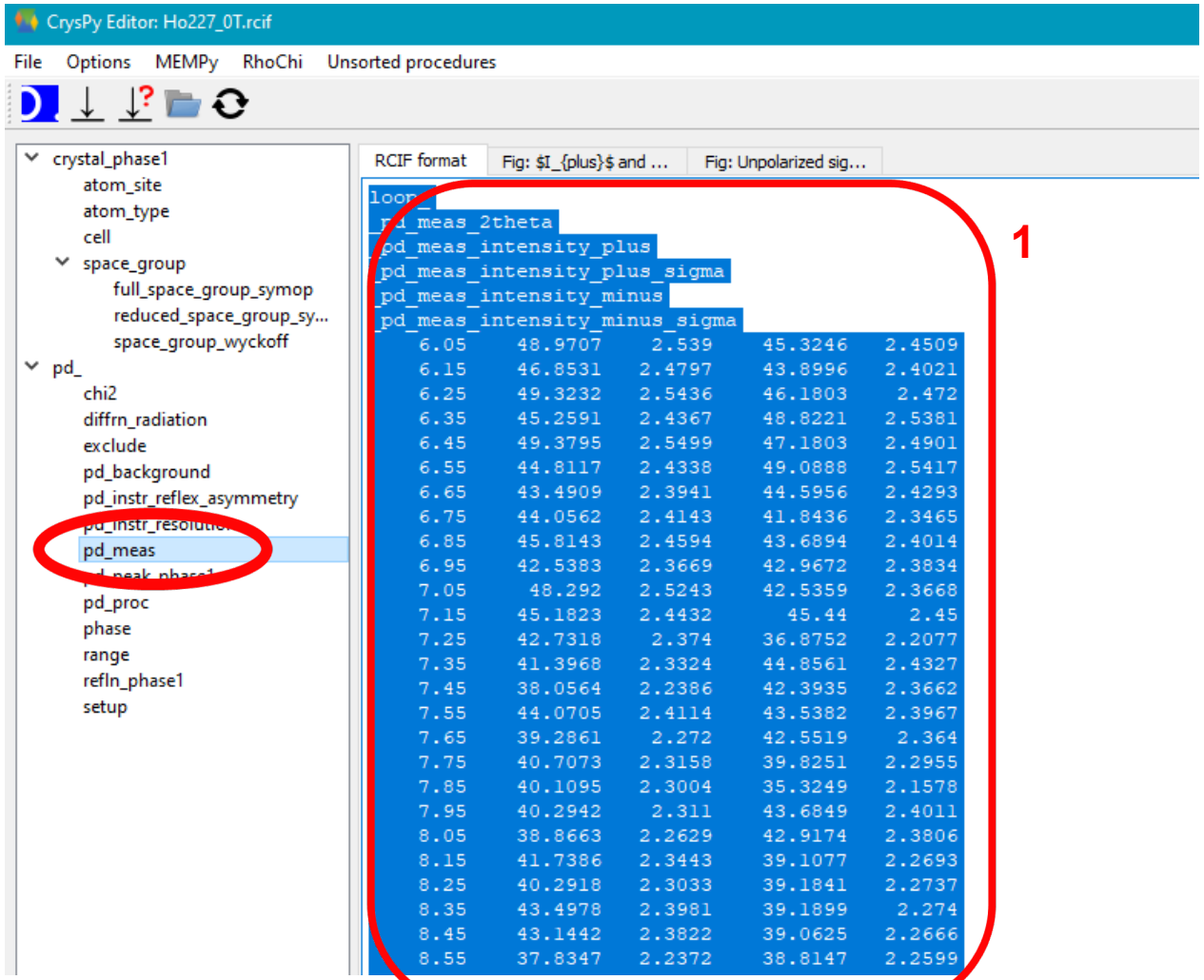
Step 3: Refine the polarized flipper on and flipper off data using using crysPy

- **Add the polarized data**



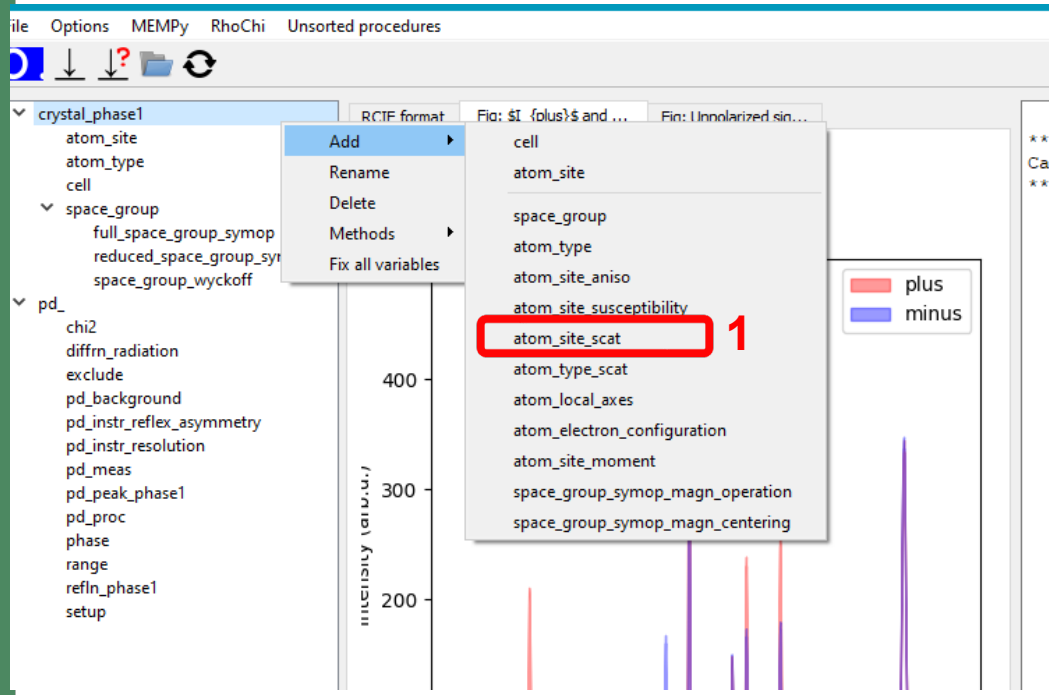
- **1.** Paste the data in the file "Ho227_20K_1T_Iplus_Iminus.dat" into the **pd_meas** category.
- **2.** You can look at the data in the "Fig:" tabs



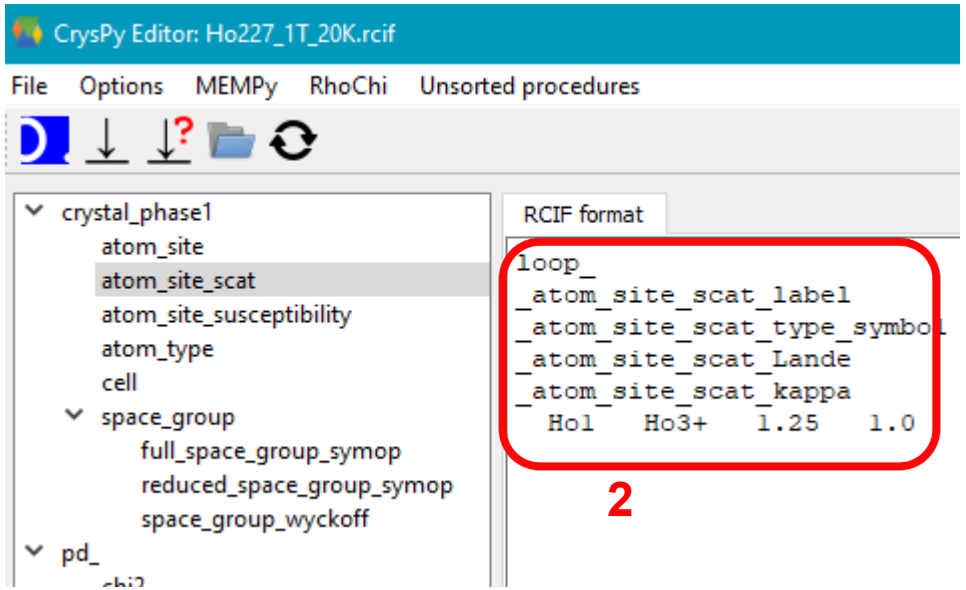


Step 3: Refine the polarized flipper on and flipper off data using using crysPy

- **Add the magnetic ion**



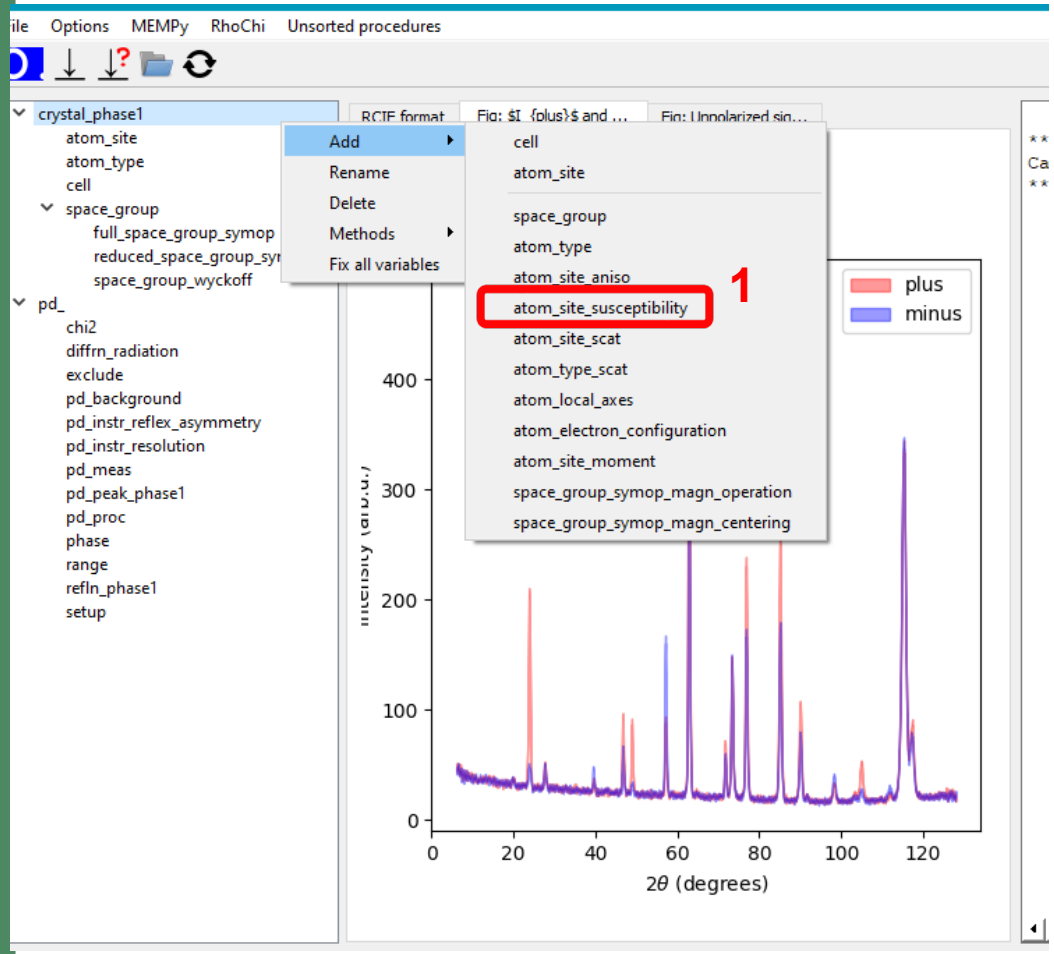
- **1.** Right-click on "crystal_phase1" → Add→ atom_site_scat
- **2.** Put in the Ho ion.
  - The lande-g needs to be looked up. For Ho^{3+} it is 5/4



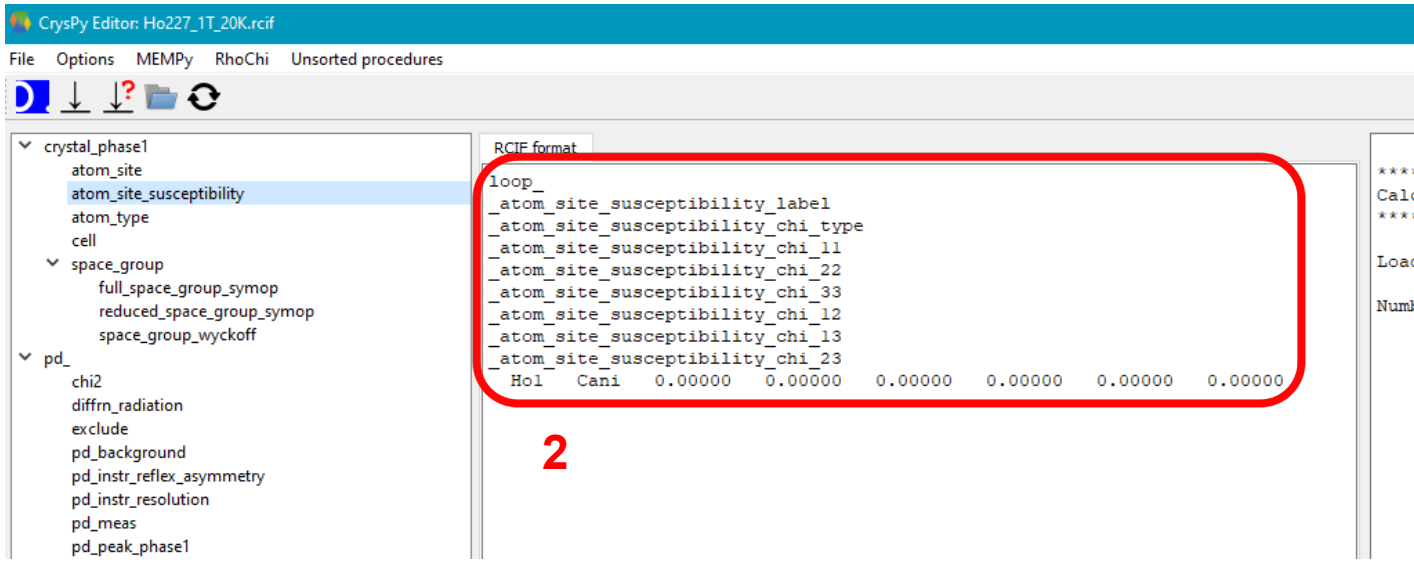
Ion	$4f^n$	L	L	J	Ground state	g_J	p^{th}	p^{exp}
La^{3+}	0	0	0	0	$^1S_0$	0	0	0
Ce^{3+}	1	1/2	3	5/2	$^2F_{5/2}$	6/7	2.535	2.4–2.7
Pr^{3+}	2	1	5	4	$^3H_4$	4/5	3.578	3.4–3.6
Nd^{3+}	3	3/2	6	9/2	$^4I_{9/2}$	8/11	3.618	3.4–3.7
Pm^{3+}	4	2	6	4	$^5I_4$	3/5	2.683	–
Sm^{3+}	5	5/2	5	5/2	$^6H_{5/2}$	2/7	0.845	1.3–1.6
Eu^{3+}	6	3	3	0	$^7F_0$	0	0	3.2–3.4
Gd^{3+}	7	7/2	0	7/2	$^8S_{7/2}$	2	7.937	7.9–8.0
Tb^{3+}	8	3	3	6	$^7F_6$	3/2	9.721	9.4–9.8
Dy^{3+}	9	5/2	5	15/2	$^6H_{15/2}$	4/3	10.646	10.5–10.7
Ho^{3+}	10	2	6	8	$^5I_8$	5/4	10.607	10.3–10.6
Er^{3+}	11	3/2	6	15/2	$^4I_{15/2}$	6/5	9.581	9.4–9.6
Tm^{3+}	12	1	5	6	$^3H_6$	7/6	7.561	7.3–7.6
Yb^{3+}	13	1/2	3	7/2	$^2F_{7/2}$	8/7	4.536	4.4–4.6
Lu^{3+}	14	0	0	0	$^1S_0$	0	0	0

Step 3: Refine the polarized flipper on and flipper off data using using crysPy

- **Set-up the site susceptibility refinement for the magnetic ions**



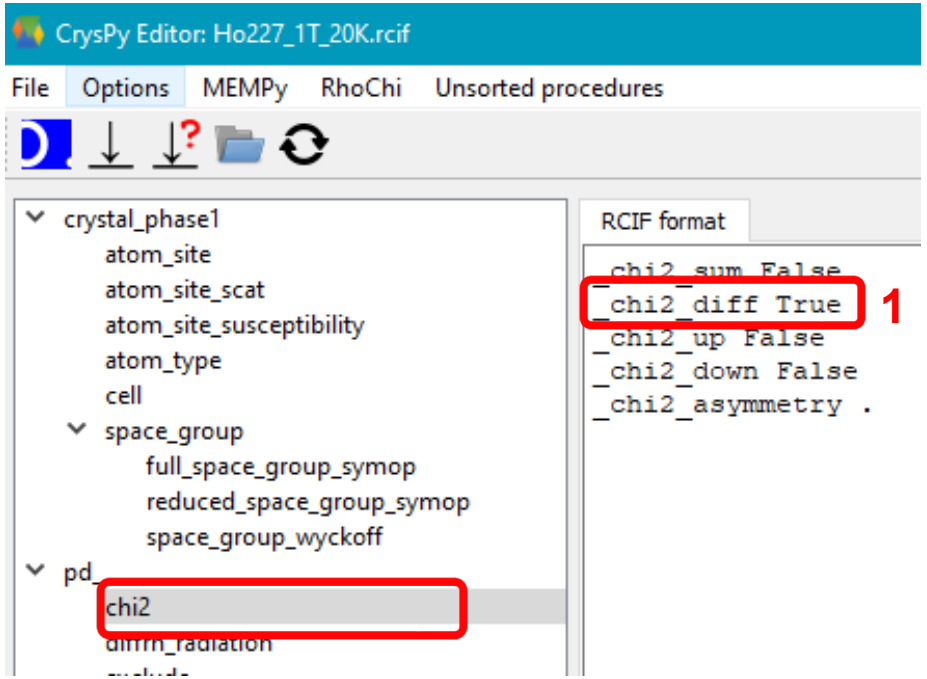
- **1.** Right-click on "crystal_phase1" → Add→ atom_site_susceptibility
- **2.** Put in the Ho ion.
  - C_{ani} will allow for an anisotropic susceptibility tensor. Cryspy will only refine symmetry allowed values. C_{iso} is isotropic (only first 3 terms)



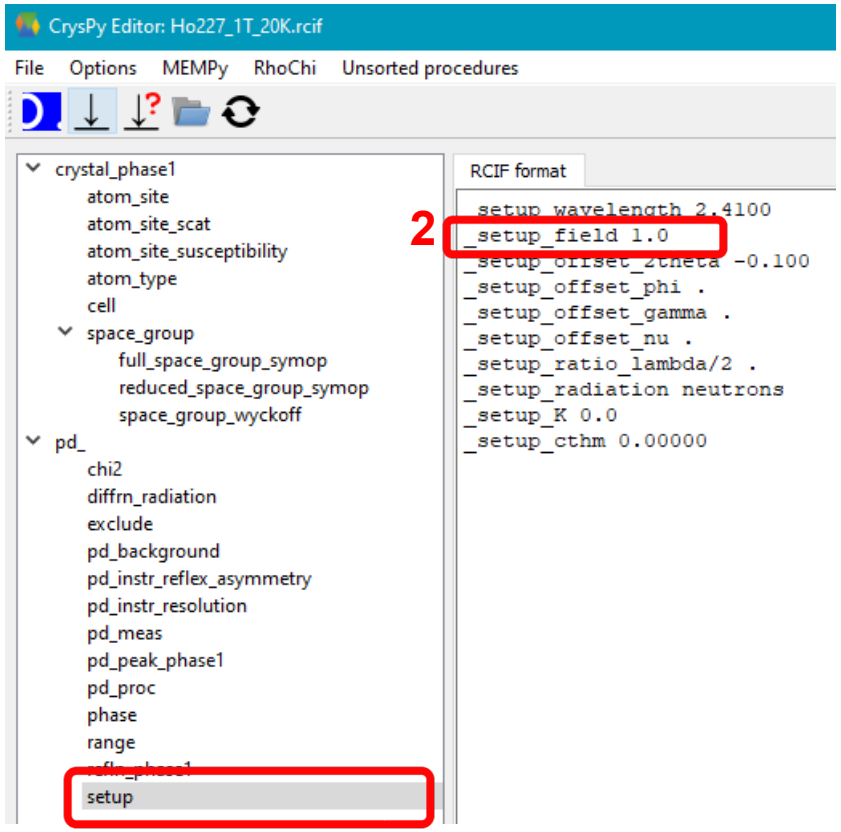
$$\chi = \frac{\boldsymbol{m}}{\boldsymbol{H}} = \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{12} & \chi_{22} & \chi_{23} \\ \chi_{13} & \chi_{23} & \chi_{33} \end{pmatrix}$$

Step 3: Refine the polarized flipper on and flipper off data using using crysPy

- **1** In chi2 set "_chi2_diff_" to True, since we want to refine the difference of spin up and spin down.
  - Later we can refine the sum as well.



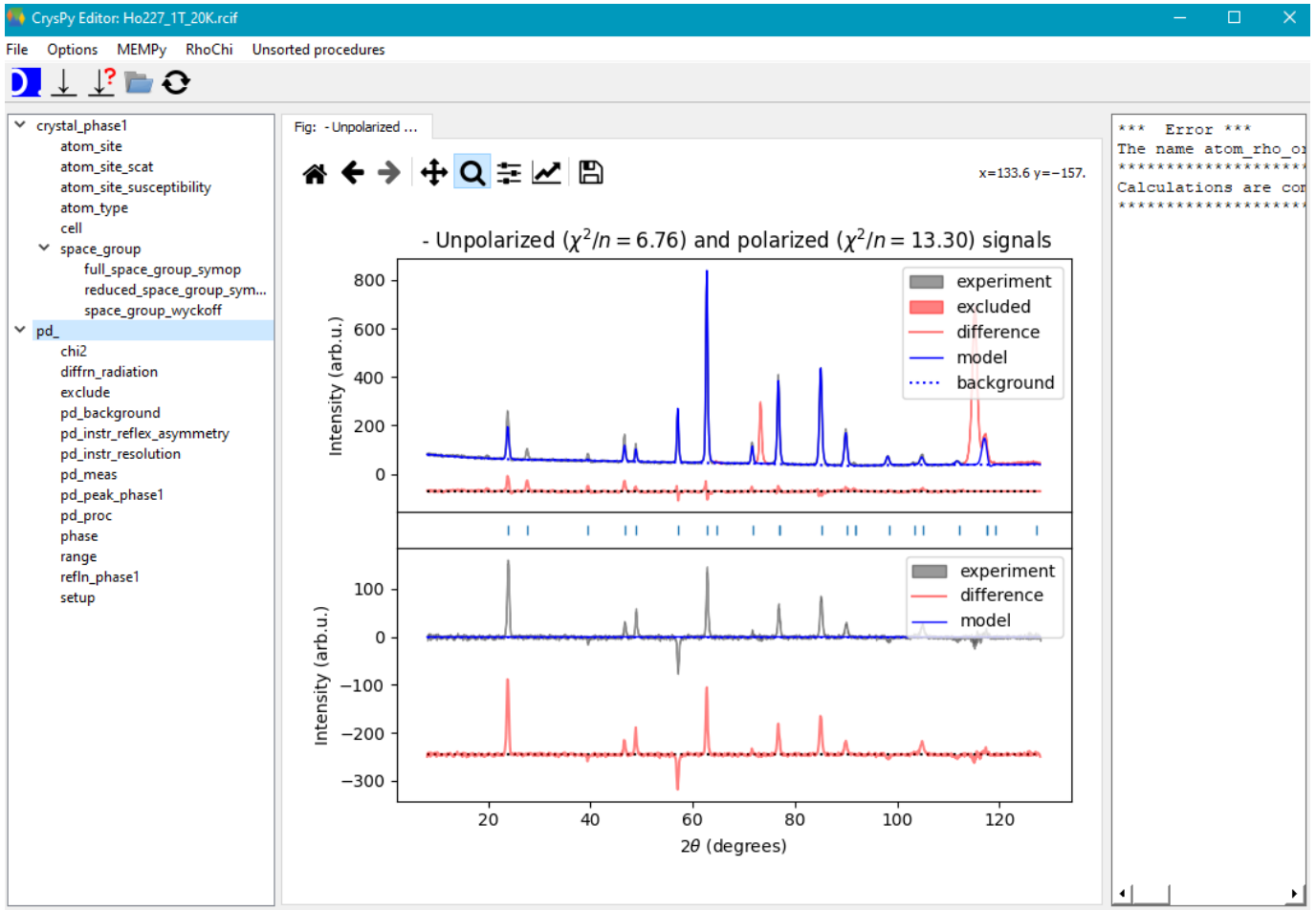
- **2.** Put in the field value used for the measurement in pd_ → setup
- A value of 1 Tesla was used



Step 3: Refine the polarized flipper on and flipper off data using using crysPy

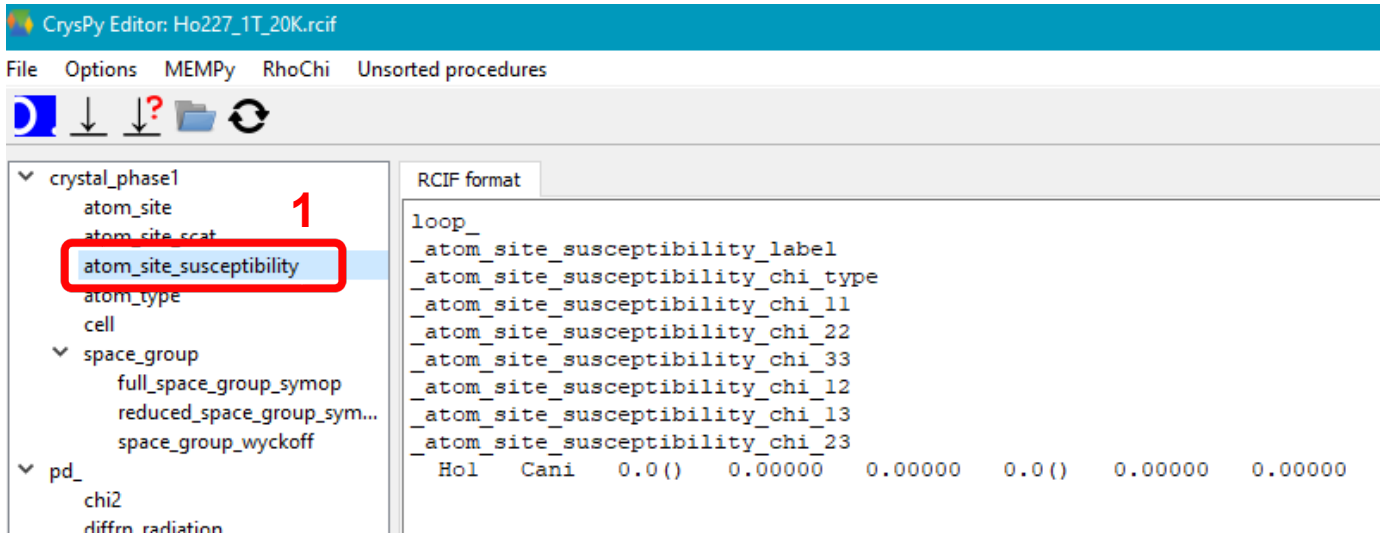
- Check everything is set-up correctly by running: RhoChi → No refinement (autorun) and look at the data against model
- The site susceptibility is zero, so the model for the difference is still a flat line.

- We're now ready to refine the local site susceptibility



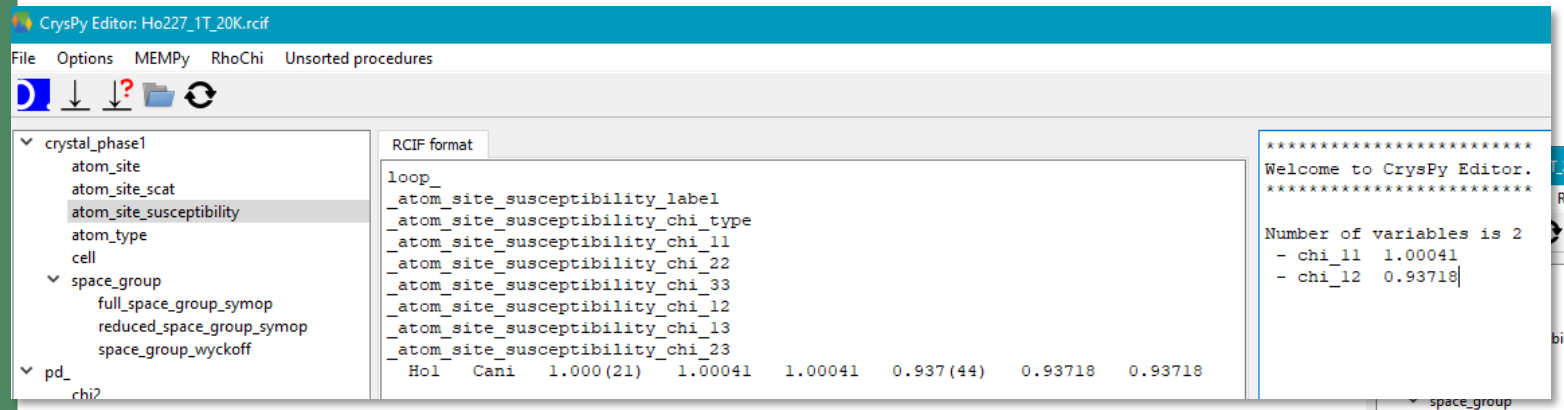
Step 3: Refine the polarized flipper on and flipper off data using using crysPy

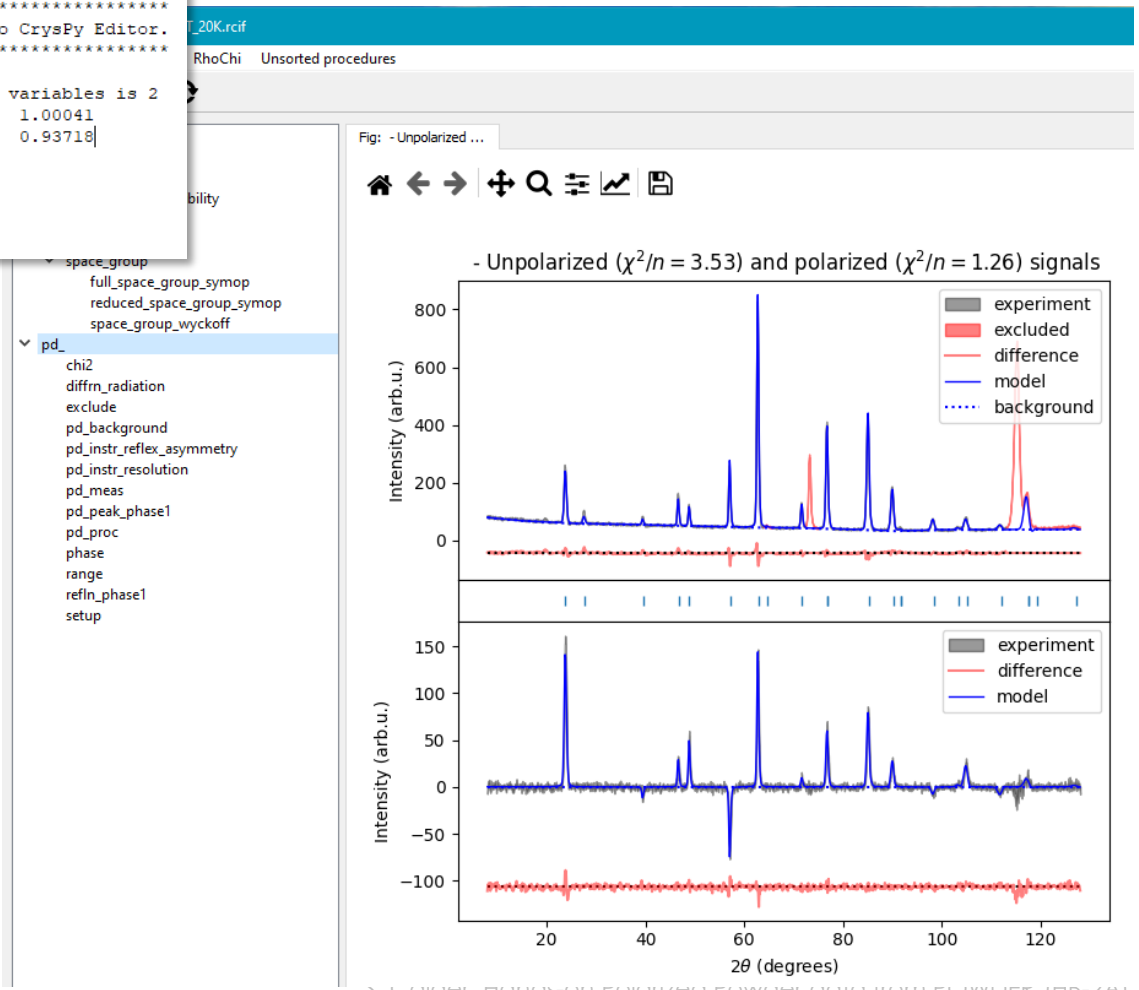
- **Refine the local site susceptibility tensor for $Ho_2Ti_2O_7$**
  - **1.** Right-click on "atom_site_susceptibility" → Refine all variables
  - The symmetry constraints in crysPy determine which matrix elements to refine.
    - Diagonal elements are constrained together
    - Off-diagonal elements are constrained together



Step 3: Refine the polarized flipper on and flipper off data using using crysPy

- **Refine the local site susceptibility tensor for $Ho_2Ti_2O_7$**



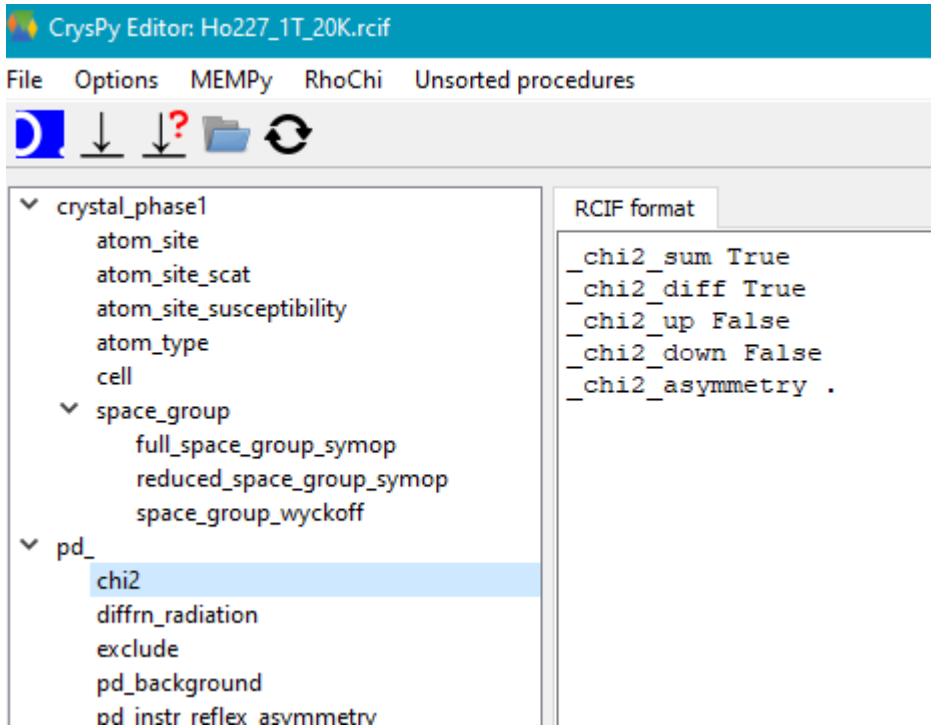


- The fit to the data looks very good!
- In the top panel the red peaks are the excluded Al sample holder peaks.

3. Calder, Hands-on Polarized Powder data from POWDER (HB-2A).

Step 3: Refine the polarized flipper on and flipper off data using using crysPy

- **Refine the local site susceptibility tensor for $Ho_2Ti_2O_7$**



- Now refine both the difference and sum by turning "_chi2_sum" to True

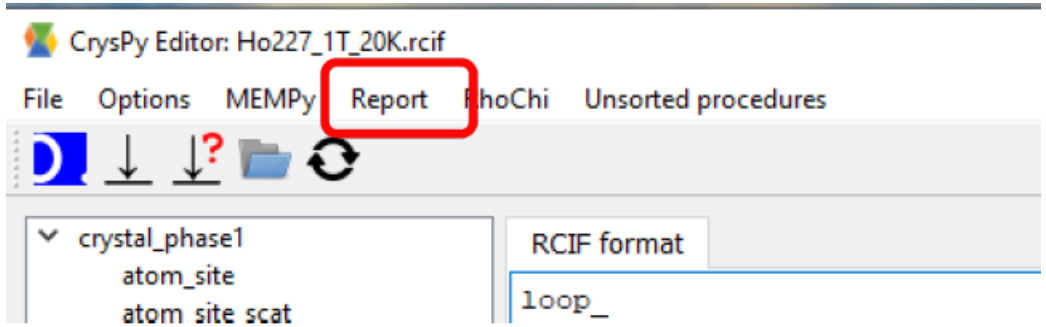
- Try refining further parameters to see how it influences the model.
  - Lattice constants, background,
- Also try "_diffrn_radiation_polarization"
  - This can be sensitive to depolarization or if the sample is no longer in the linear M/H regime (check the 1.5 K data as an exercise!)

$Ho_2Ti_2O_7$ Powder Example

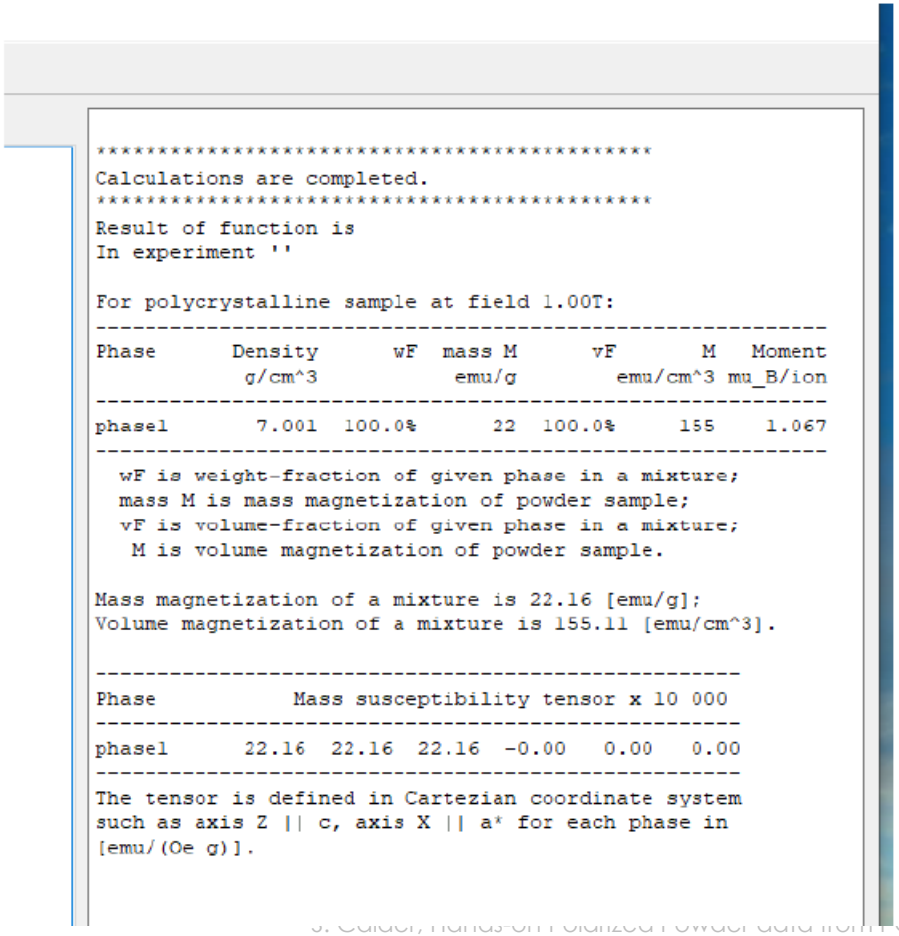
- This example will use crsyPy to determine the local site susceptibility in the following steps:
  - Step 1: Installing and running crysPy
  - Step 2: Refine unpolarized, non-magnetic $Ho_2Ti_2O_7$ crystal structure using crysPy
  - Step 3: Refine the polarized $Ho_2Ti_2O_7$ data using crysPy
  - **Step 4: Interpreting the results**

Step 4: Interpreting the results

- In the "Report" drop down menu run
  - Powder experiment (autorun)



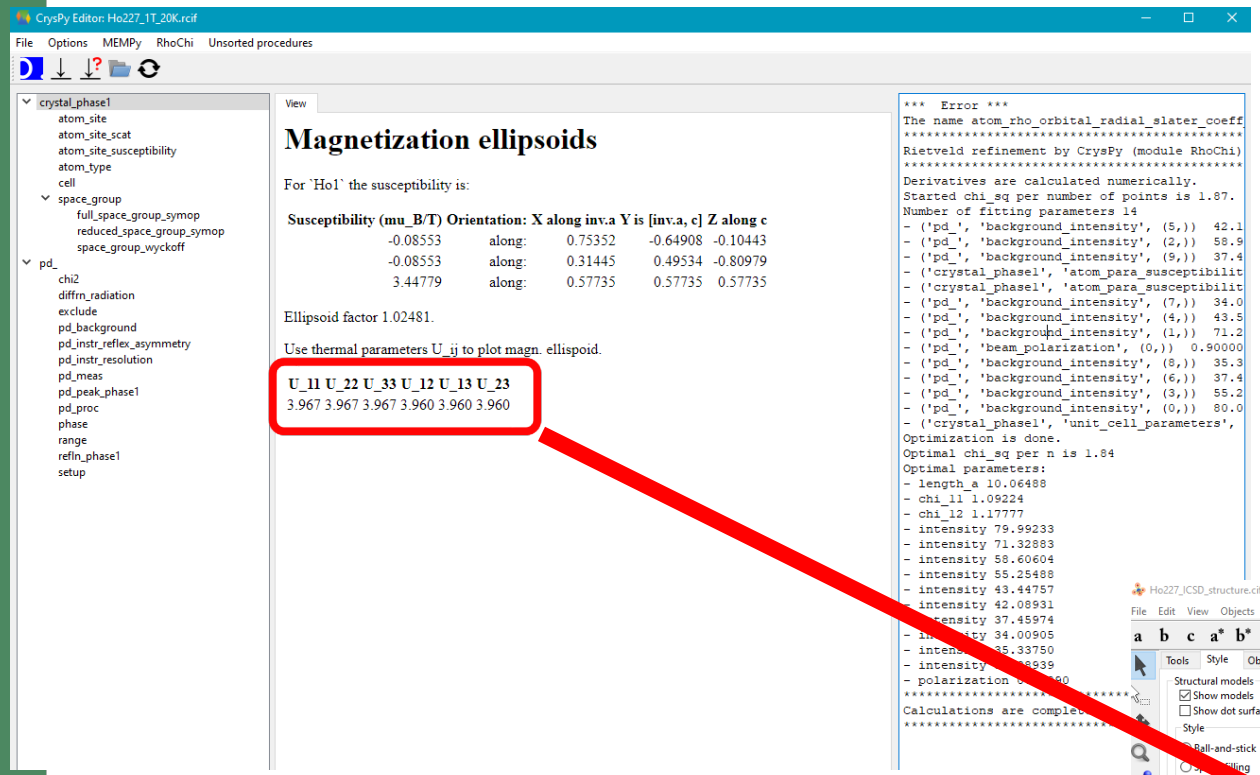
- This gives the moment. This value should agree with bulk M/H data for the sample at the same field and temperature.



```
**********************************************
Calculations are completed.
**********************************************
Result of function is
In experiment ''

For polycrystalline sample at field 1.00T:
------------------------------------------------------------
Phase      Density      wF  mass M      vF       M   Moment
           g/cm^3           emu/g        emu/cm^3 mu_B/ion
------------------------------------------------------------
phase1       7.001  100.0%      22  100.0%     155     1.067
------------------------------------------------------------
  wF is weight-fraction of given phase in a mixture;
  mass M is mass magnetization of powder sample;
  vF is volume-fraction of given phase in a mixture;
   M is volume magnetization of powder sample.

Mass magnetization of a mixture is 22.16 [emu/g];
Volume magnetization of a mixture is 155.11 [emu/cm^3].

-----------------------------------------------------
Phase           Mass susceptibility tensor x 10 000
-----------------------------------------------------
phase1      22.16  22.16  22.16  -0.00   0.00   0.00
-----------------------------------------------------
The tensor is defined in Cartezian coordinate system
such as axis Z || c, axis X || a* for each phase in
[emu/(Oe g)].
```

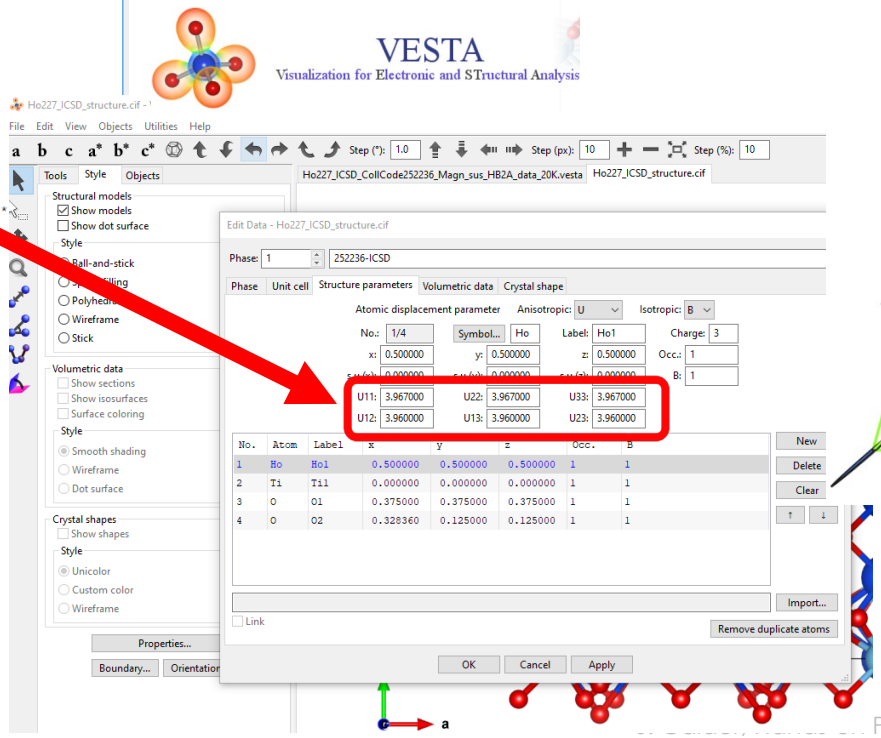
- Note: Care should be taken if there is more than one magnetic ion to make sure you are comparing the same numbers!

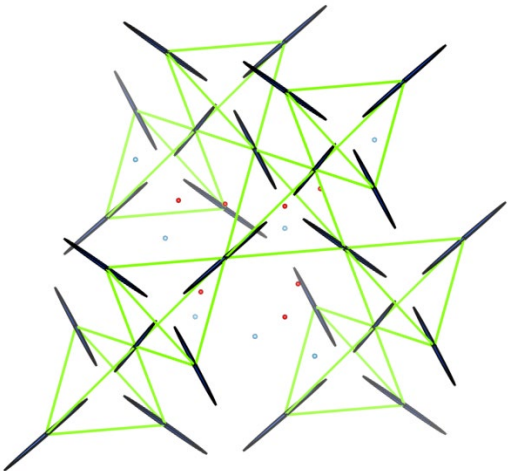
Step 4: Interpreting the results



$$\chi = \frac{\boldsymbol{m}}{\boldsymbol{H}} = \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{12} & \chi_{22} & \chi_{23} \\ \chi_{13} & \chi_{23} & \chi_{33} \end{pmatrix}$$

- Click on "crystal_phase1" to see the Magnetization ellipsoid details.
- These can be visualized using VESTA by populating the thermal parameters.
- The magnetization ellipsoids (black) are anisotropic and drive the all-in, all-out spin ice behavior of $Ho_2Ti_2O_7$





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Polarized Powder data from POWDER (HB-2A).

Step 4: Interpreting the results

DOI: 10.1103/PhysRevResearch.1.033100

PHYSICAL REVIEW RESEARCH **1**, 033100 (2019)

Local magnetic anisotropy by polarized neutron powder diffraction: Application of magnetically induced preferred crystallite orientation

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For the space group $Fd\bar{3}m$, the symmetry constraints imply that the local susceptibility tensor has only two independent matrix elements χ_{11} and χ_{12} and the principal axes of Ho magnetization ellipsoids are oriented along the four local ⟨111⟩ axes. Their lengths, given by $\chi_{\parallel} = \chi_{11} + 2\chi_{12}$ and $\chi_{\perp} = \chi_{11} - \chi_{12}$, were determined at each temperature. The thermal evolution of $\chi_{\parallel}$ and $\chi_{\perp}$ obtained by 2D Rietveld refinement on a polycrystalline sample is shown in Fig. 6 by solid symbols. Open symbols in the figure show the results of a previous study performed using polarized neutron diffraction on a single crystal [6]. One can see that the results of Rietveld refinement are in good agreement with the single-crystal ones and offer the same precision of the susceptibility parameters.

- The HB-2A results can be compared to other single crystal and powder measurements on $Ho_2Ti_2O_7$.
- Use CrysPy to fit the 1.5K, 50K and 100K data and follow the symmetry arguments in the paper to reproduce the temperature dependence of the susceptibility.

DOI: 10.1103/PhysRevResearch.1.033100

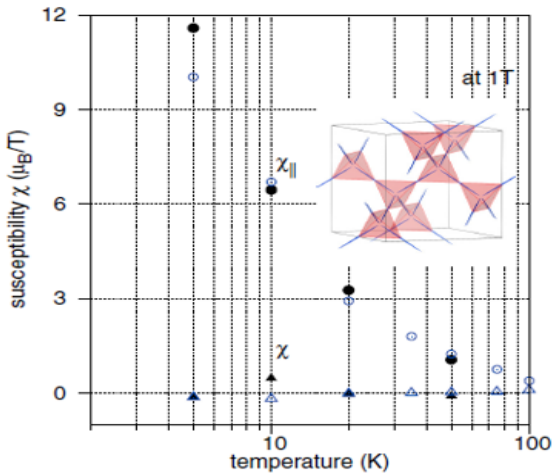

FIG. 6. Temperature dependence of the susceptibility components $\chi_{\parallel}$ (circles) and $\chi_{\perp}$ (triangles) for single (open symbols, taken from Ref. [6]) and powder (solid symbols) $Ho_2Ti_2O_7$ at 1 T. The inset shows Ho magnetization ellipsoids at 50 K.

HB-2A polarized analysis