A visual guide for crystal and commensurate magnetic structure refinement using Mag2Pol

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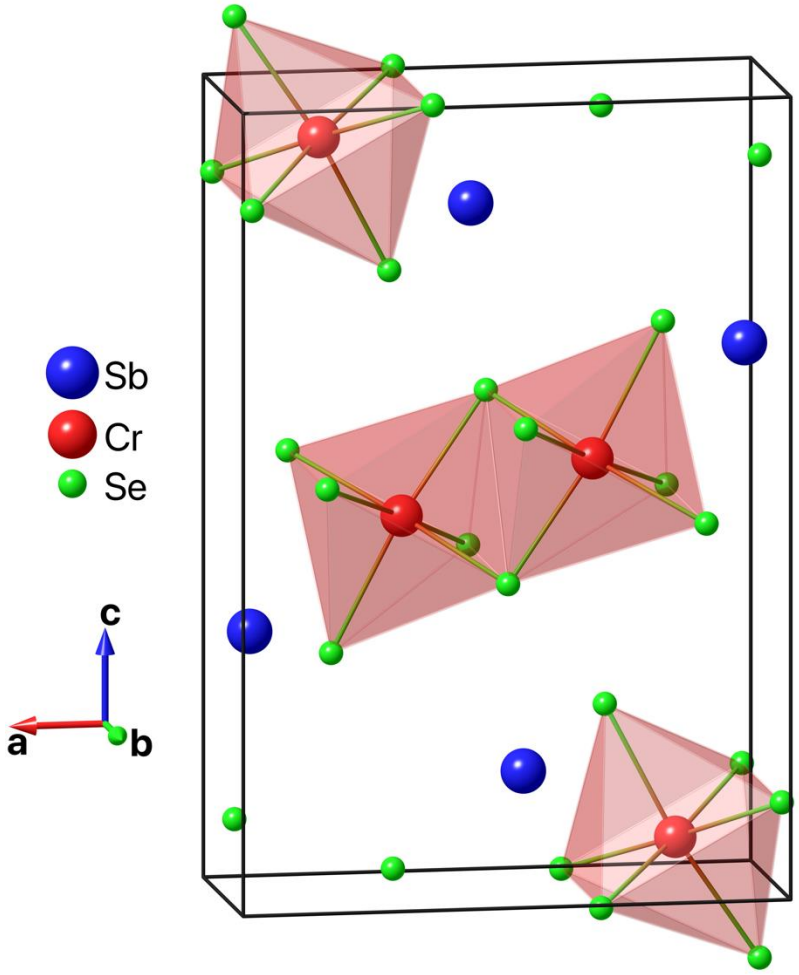
Details of the compound, data and software



Orthorhombic structure: *Pnma* (No. 62)

a = 9.143086 Å, b= 3.784552 Å, c=13.416915 Å

Time-of-flight diffraction data at POWGEN:

PG3_42702-2_300K.dat (Paramagnetic phase @ 300 K)

PG3_42704-2_10K.dat (Magnetically ordered phase @ 10 K)

Instrumental resolution file:

PG2018B_HighRes_60Hz_b2_Ddep.irf

Crystal Information File for crystal structure:

CrSbSe3.cif

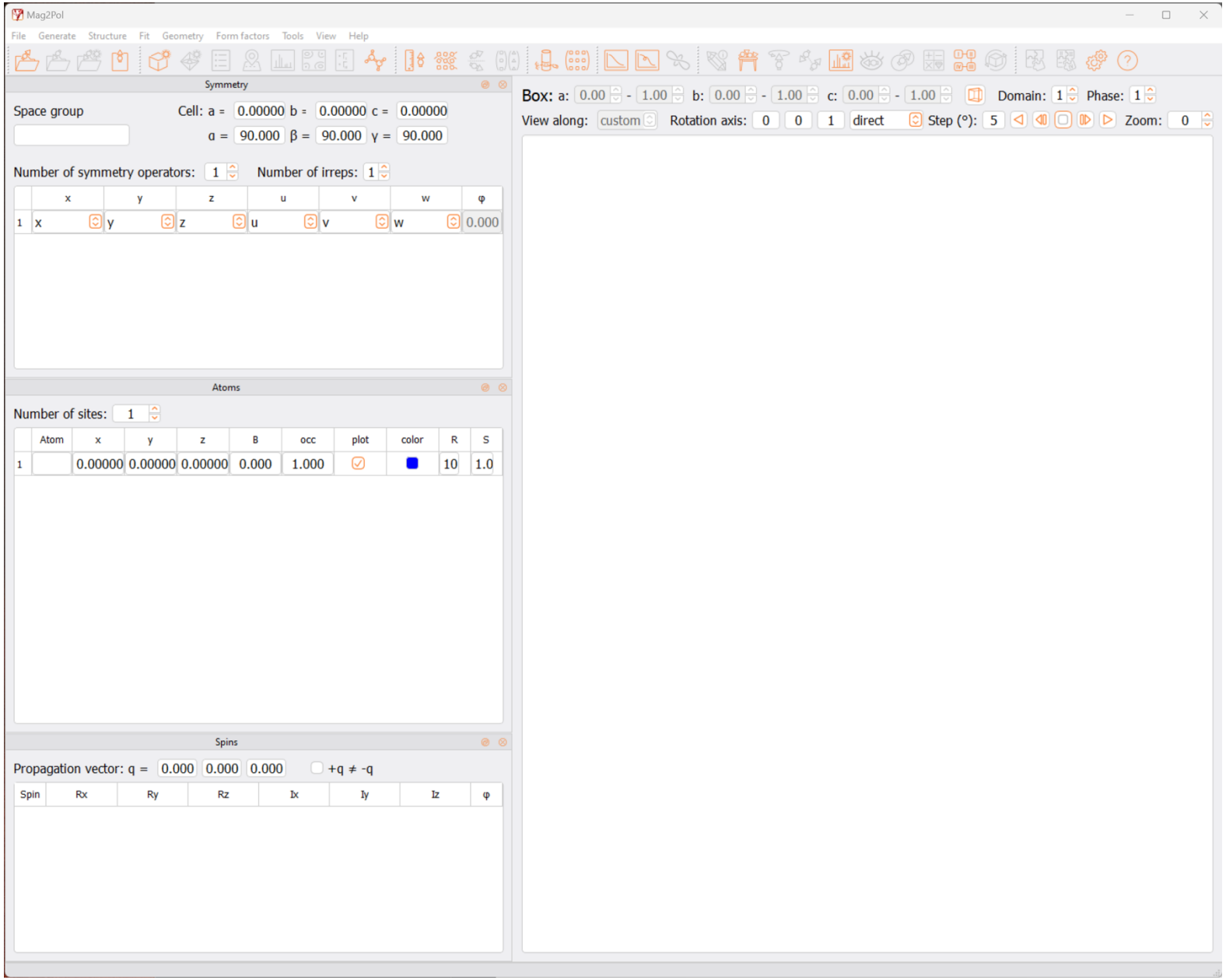
Mag2Pol version used: 7.0.4

The main interface of Mag2Pol

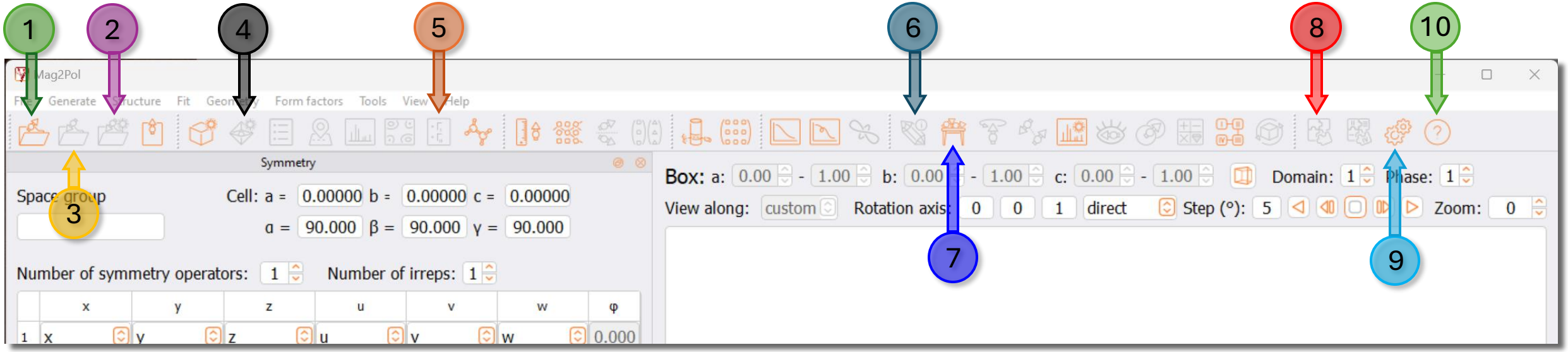
The main window of Mag2Pol - GUI

Running the Mag2Pol application, the main window ‘Mag2Pol’ will open. By default, the interface may look slightly different. You can modify this under the menu ‘View’

This window should remain open as long as any other Mag2Pol utility or window is running. The current version of Mag2Pol will not save the project automatically, so make sure to save any project before closing this window.



Menu items (icons) on the main window relevant for this tutorial



1 **Open**: Load a structure file (cif) or an existing Mag2Pol project (xml).

2 **Save as**: Save a new Mag2Pol project (as *.xml).

3 **Save**: Append the currently working project with the same name.

4 **Bonds**: Opens a new window that is used to modify the visuals of the structural model [*active after loading the structure file, if it is not active even after loading the cif file, press Ctrl+U (on Windows) or Cmd+U (on Mac)*].

5 **Irreducible representations:** Opens a new window that provides details of irreps and lets you select one or more irreps for the refinement of magnetic structure (active when at least one magnetic ion is defined)

6 **Sample info:** Opens a new window that provides details of, lattice and magnetic structures and the results of the last refinements.

7 **Spacegroup tables:** Opens a window on which you can select magnetic space group (Shubnikov group) instead of irreps, to refine the magnetic structure.

8 **Fit:** Opens the window where you can choose the data file, and instrument resolution file, and perform the refinements.

9 **Settings:** Opens a new window that lets you modify a few important settings about GUI, a graphical rendering of plots and figures, and also the refinement procedure.

10 **Manual:** Opens Mag2Pol manual in pdf format.

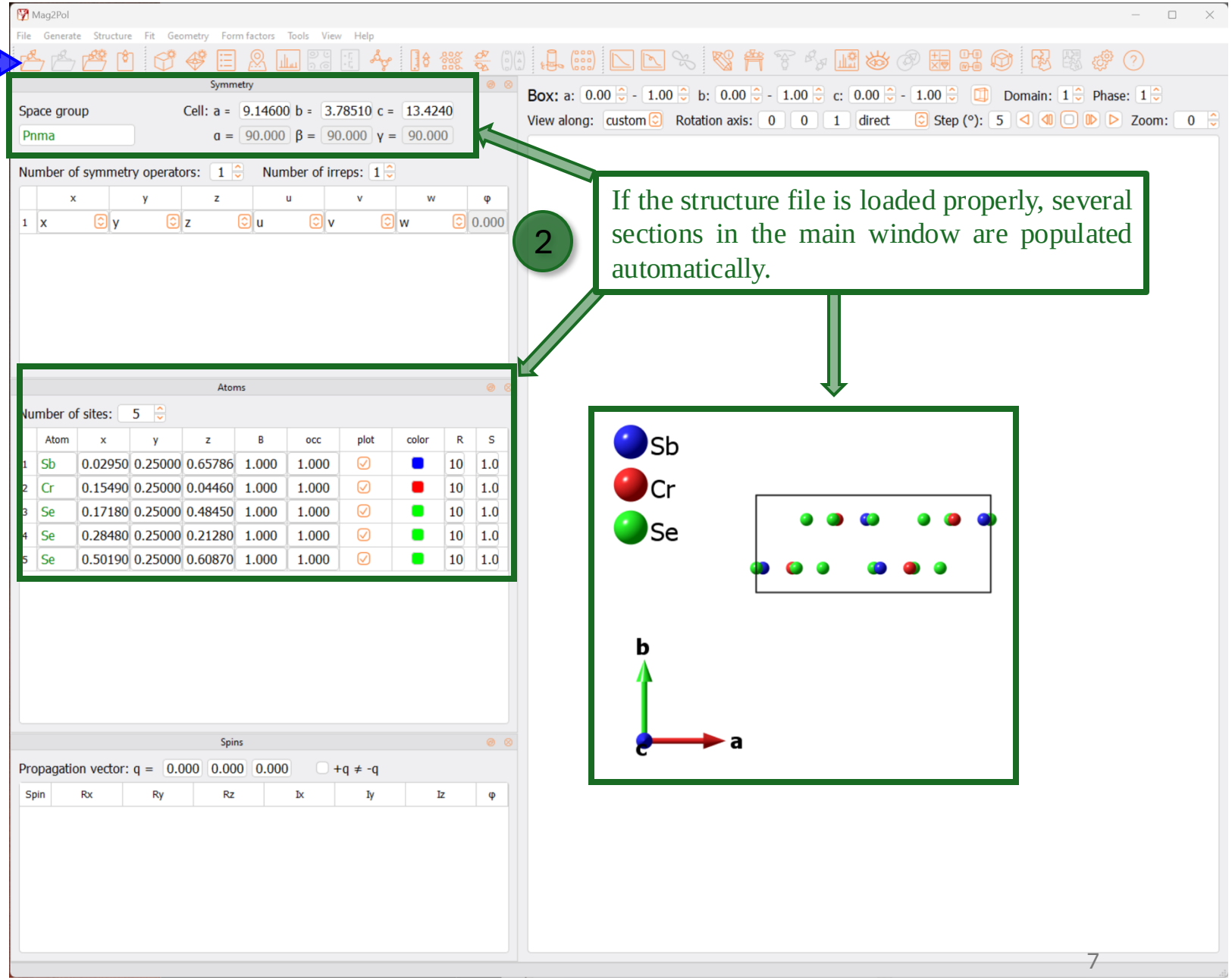
Refining high temperature NPD data, collected in paramagnetic phase

Step 1: Loading the structure or an existing project

The structure file **(*.cif, *.mcif) or existing Mag2Pol project file (*.xml)** can be loaded by clicking this icon or through the menu tree: File→Open

In this case: **CrSbSe3.cif**

1

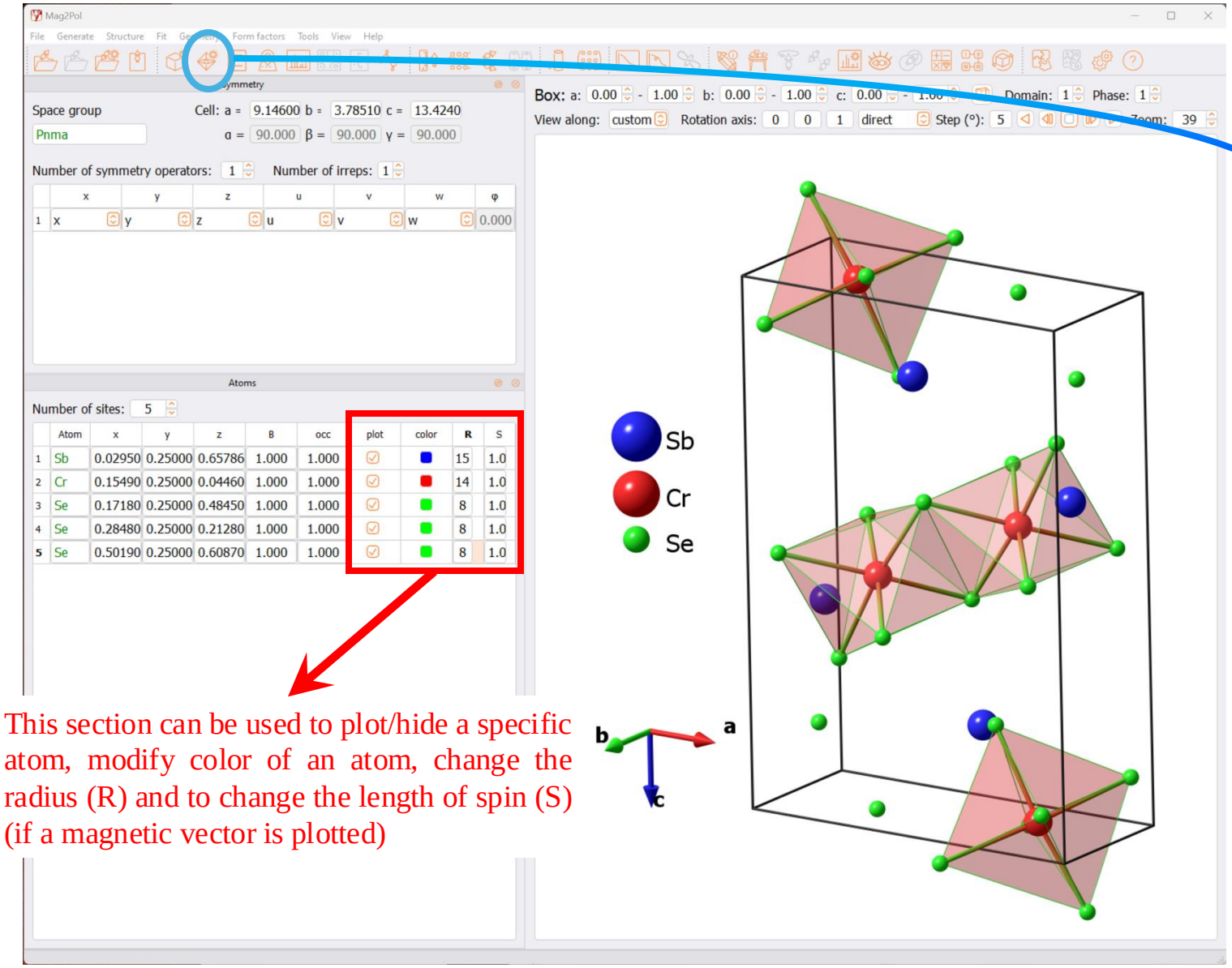


2

If the structure file is loaded properly, several sections in the main window are populated automatically.

Optional: Editing structure plot to produce publication-quality figure

Click on **Bonds** icon, a new window 'Bonds and polyhedra' will pop out. You can define and edit the bonds and polyhedral of the graphic here. If the Bonds icon is inactive even after loading the structure file it can be activated by pressing key board short-cut, Ctrl+U (on Windows) or Cmd+U (on Mac).



This section can be used to plot/hide a specific atom, modify color of an atom, change the radius (R) and to change the length of spin (S) (if a magnetic vector is plotted)

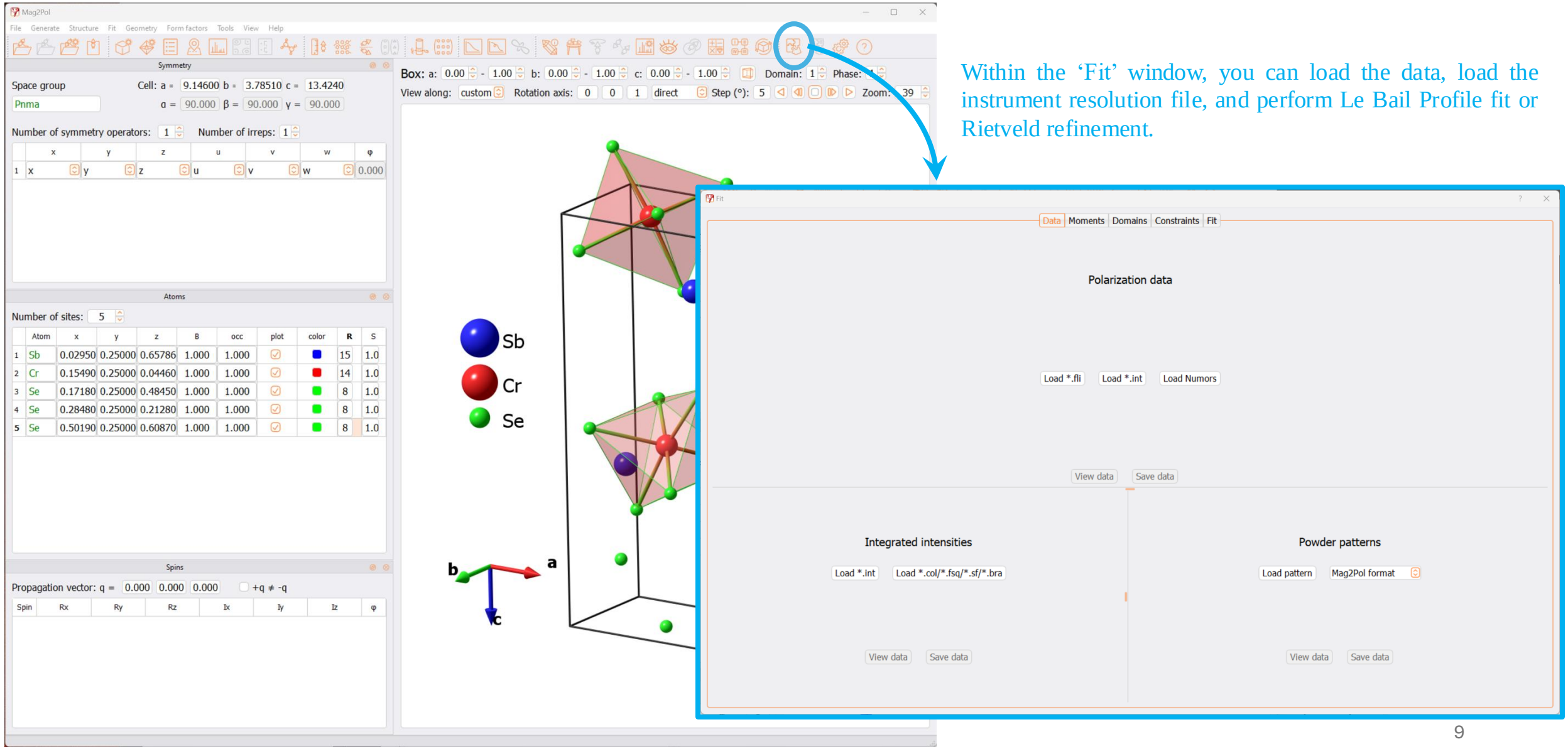
Once you are satisfied with the structure plot, you can export it as *.png following the menu:

File→Export→Graphic

It is also possible to export the graphic as *.cif and *.mcif (in the case of magnetic structure).

Step 2: Enter the refinement (Fit) window

To load the experimental data we need to open the ‘Fit’ window, by clicking on the icon as shown in the screenshot below (or the keyboard shortcut ‘Ctrl+F’).

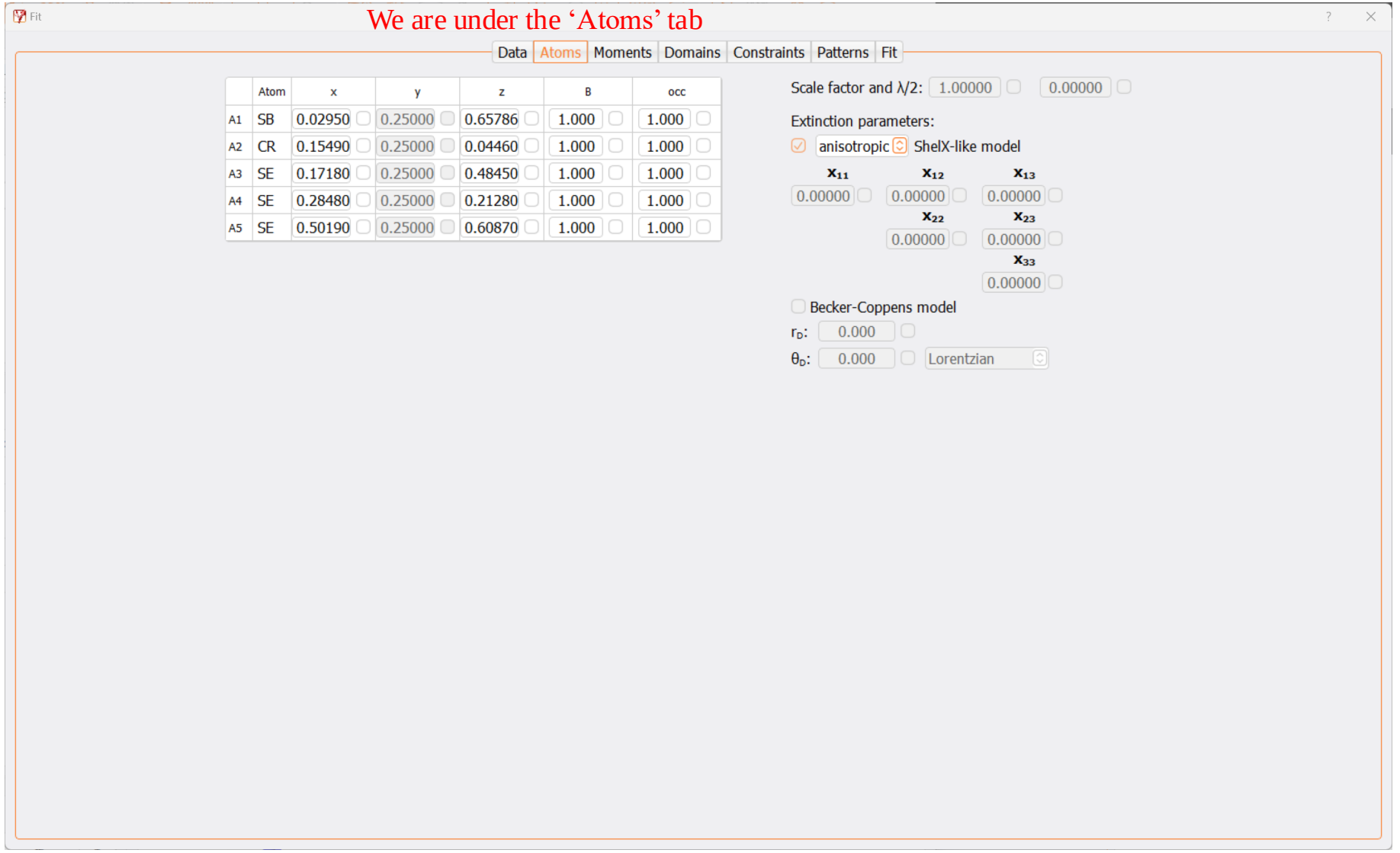


Step 2a: Load the experimental data

The data can be loaded under the ‘Data’ tab of the ‘Fit’ window. Mag2Pol can accept three kinds of data sets. In this example, we will start with ‘Powder diffraction data’ of $CrSbSe_3$ collected at 300 K (Paramagnetic phase). **Follow the order indicated below.**

Step 2b: Confirm all the atoms are loaded properly

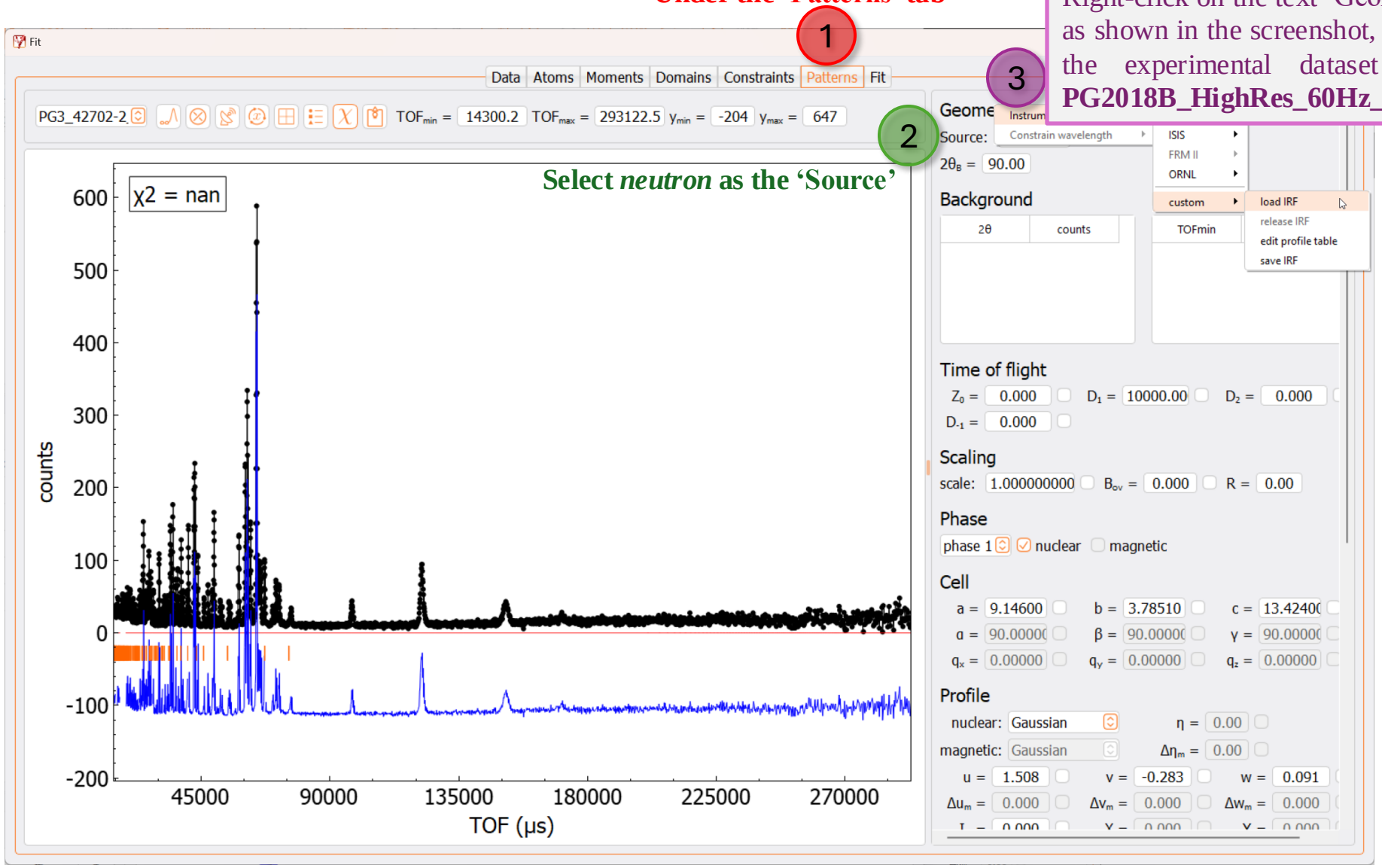
Under the ‘Atoms’ tab, check if all the atoms are loaded properly and the occupancies nominally follow the chemical formula. Do not select any of these parameters for refinement, at this stage.



Step 2c: Load the instrument resolution (irf) file

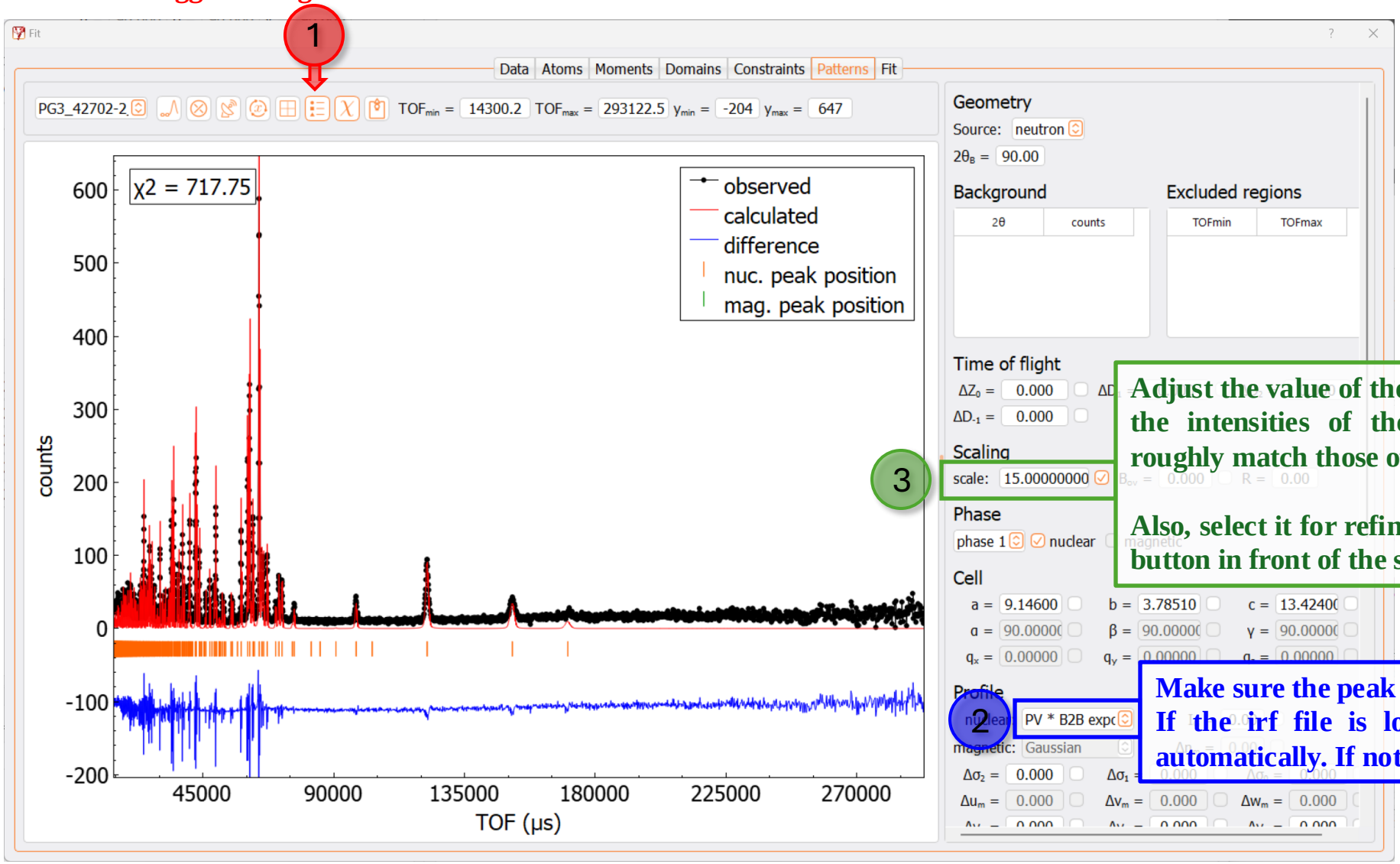
Under the 'Patterns' tab

Right-click on the text 'Geometry' and follow the menu items as shown in the screenshot, to load the appropriate irf file for the experimental dataset (data bank). In this case: **PG2018B_HighRes_60Hz_b2_Ddep.irf**



Step 2d: Check the peak function and set an appropriate scale factor

Toggle the legend ON/OFF



Adjust the value of the 'scale' factor so that the intensities of the calculated pattern roughly match those of observed data.

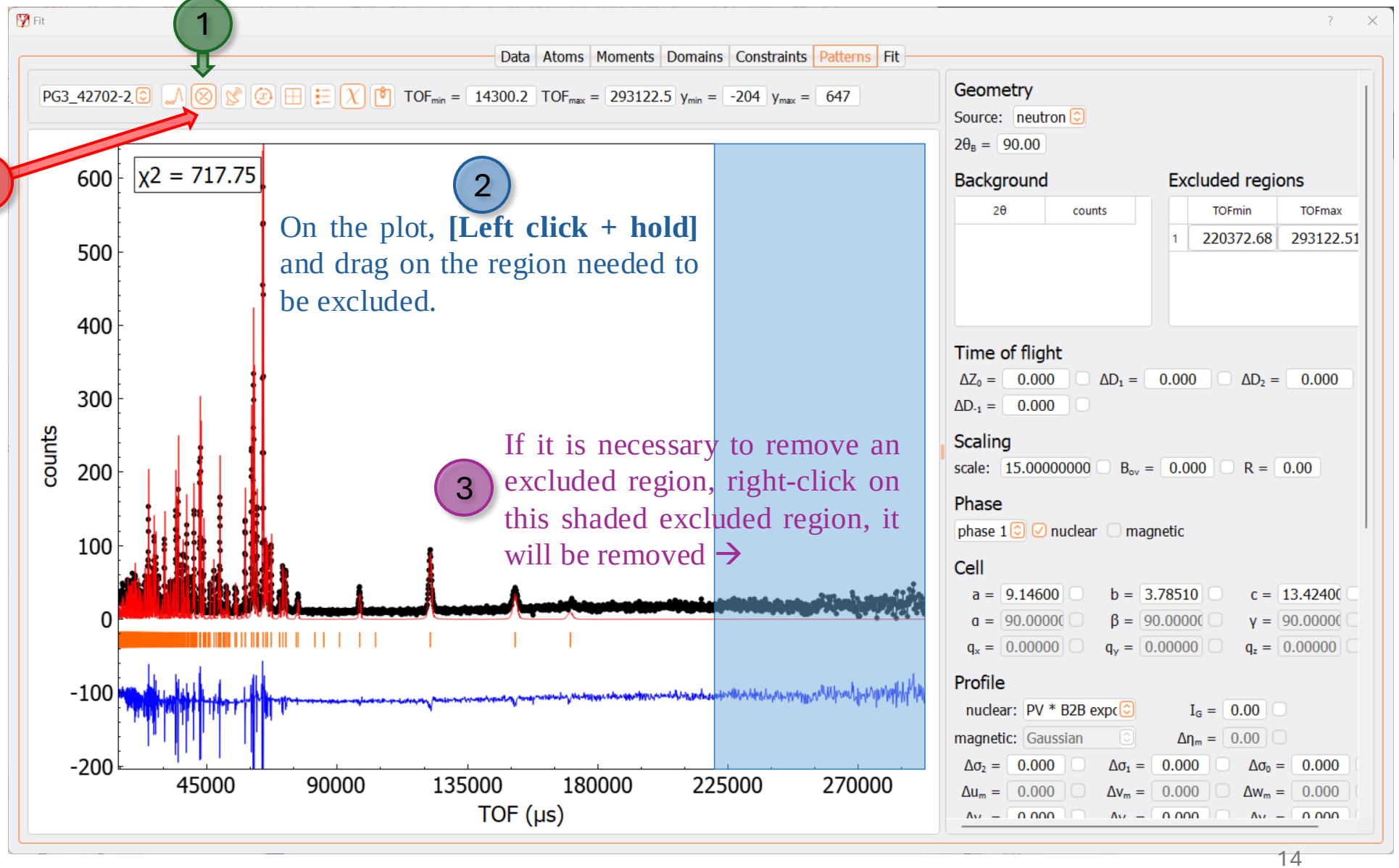
Also, select it for refinement, by ticking the button in front of the scale factor value.

Make sure the peak function is: *PV * B2B exponential (Tof1)*
If the irf file is loaded properly this should be updated automatically. If not choose it manually.

Step 2e: Select the region to exclude from the fit 'Excluded region'

Toggle the 'excluded regions' ON

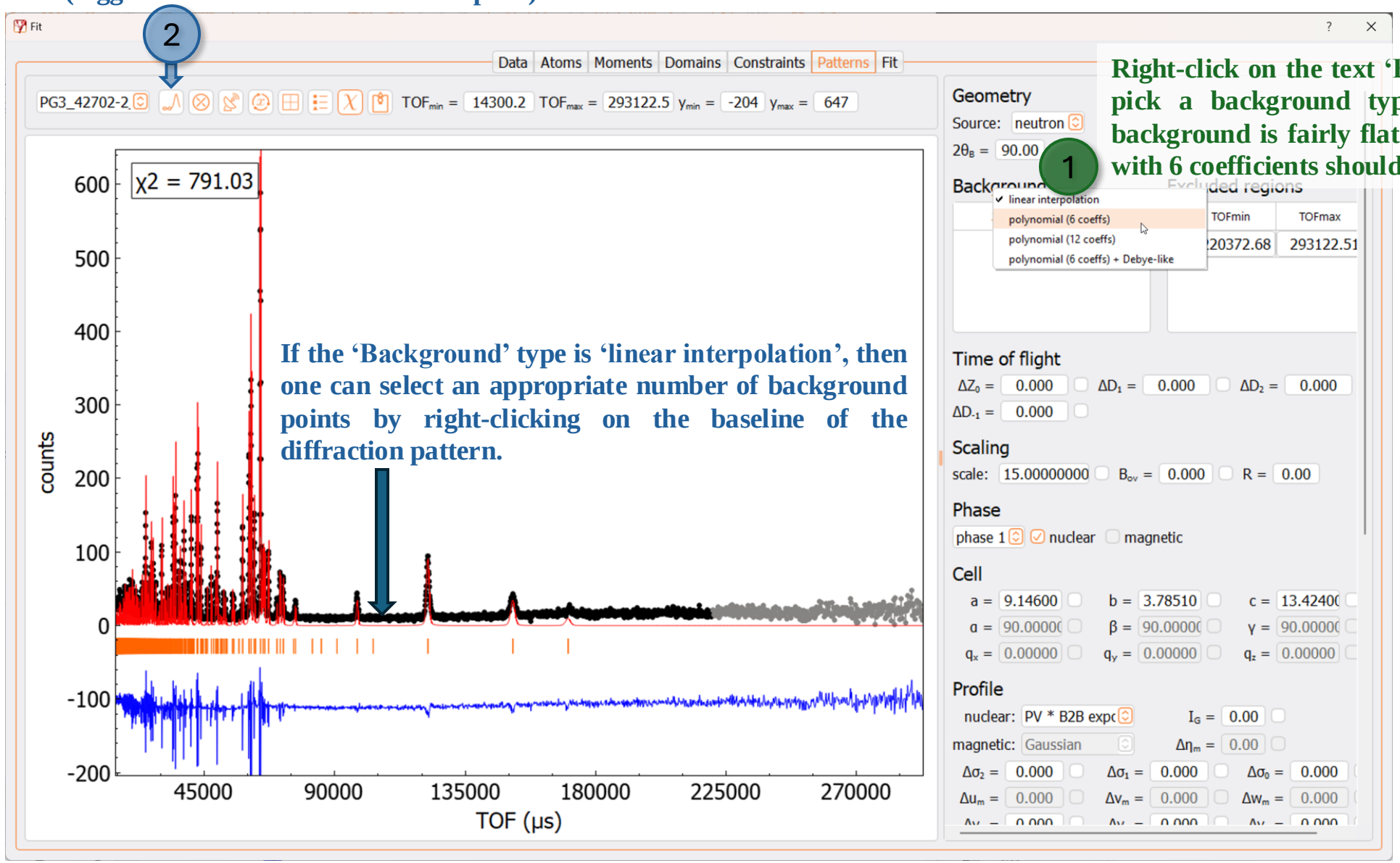
Toggle the 'excluded regions' OFF



Step 2f: Select the background

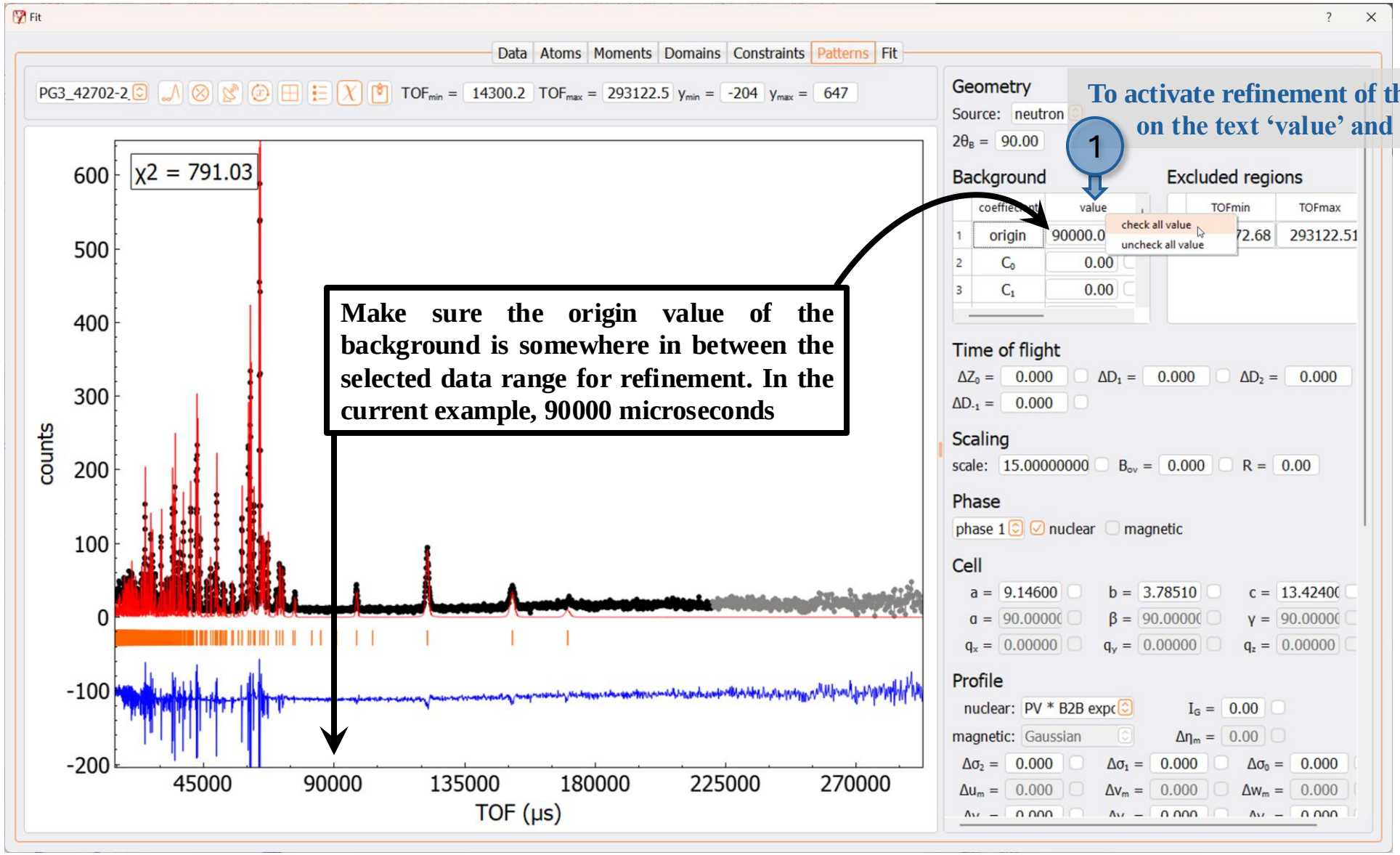
(Only for BG type 'linear interpolation')

For manual background selection toggle this ON (toggle it OFF after selection is complete)



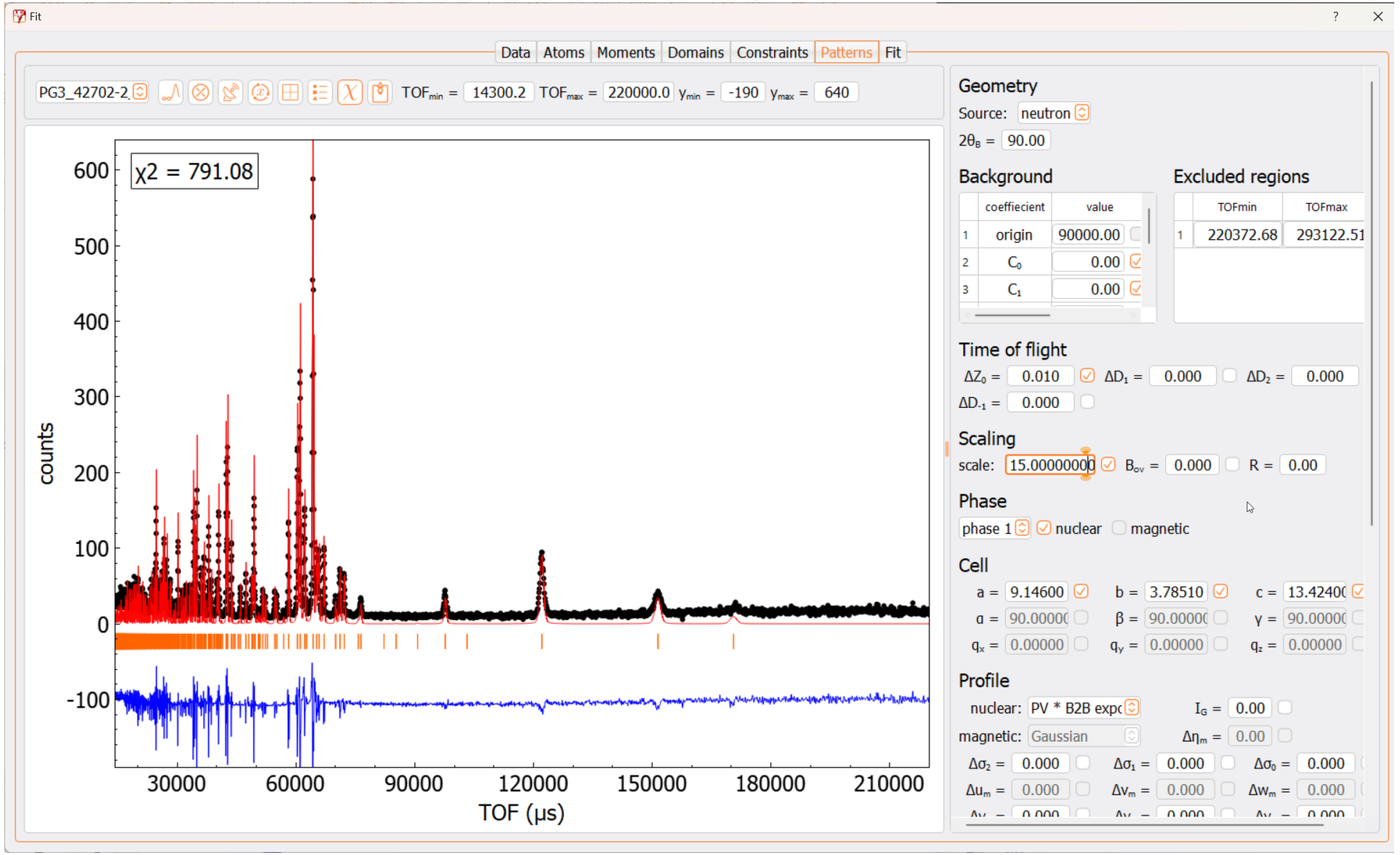
Right-click on the text 'Background' and pick a background type. In this case background is fairly flat so a polynomial with 6 coefficients should be sufficient.

Step 2g: Activate refinement of background

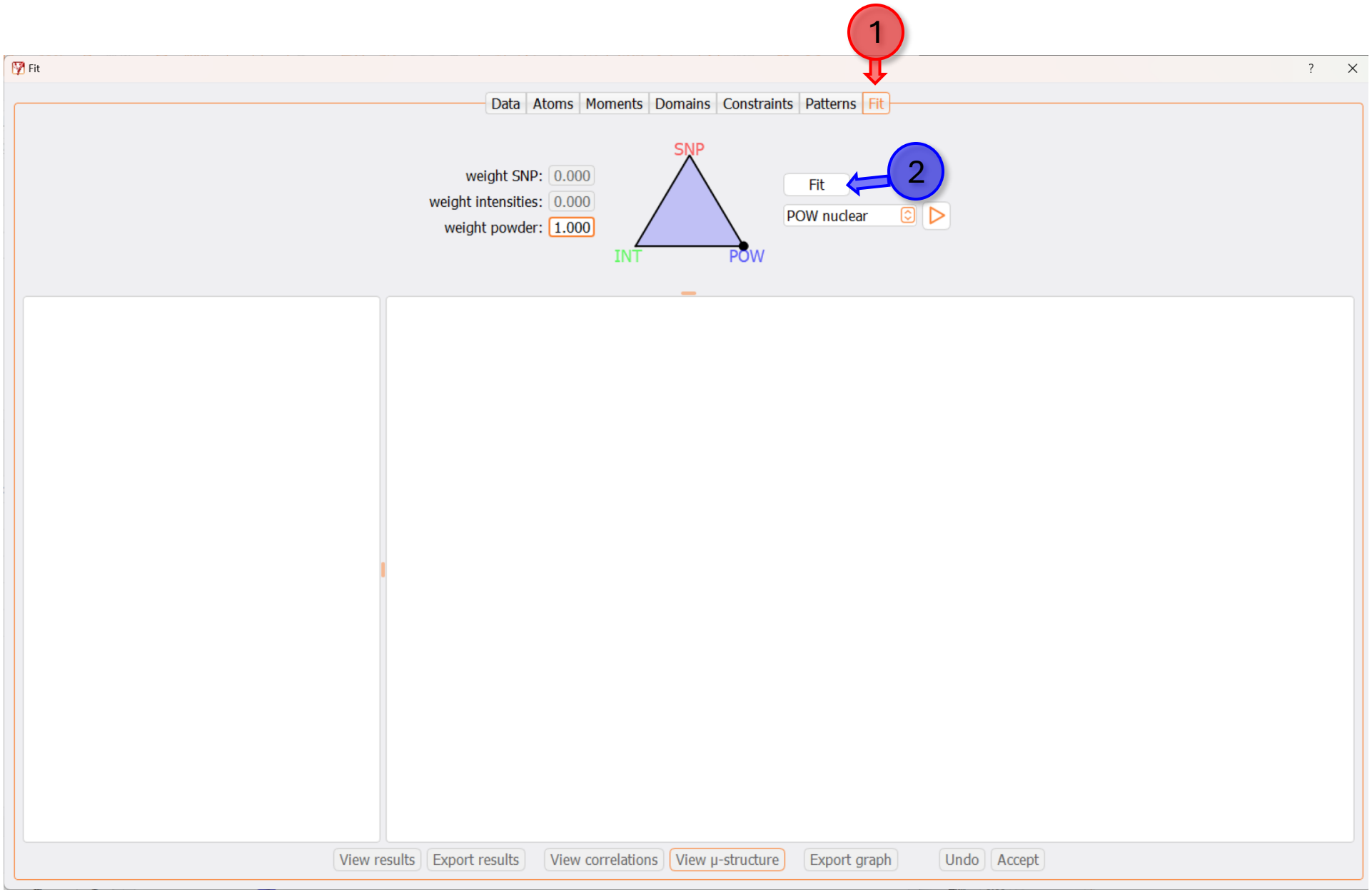


To activate refinement of the background, Right-click on the text 'value' and select 'check all value'

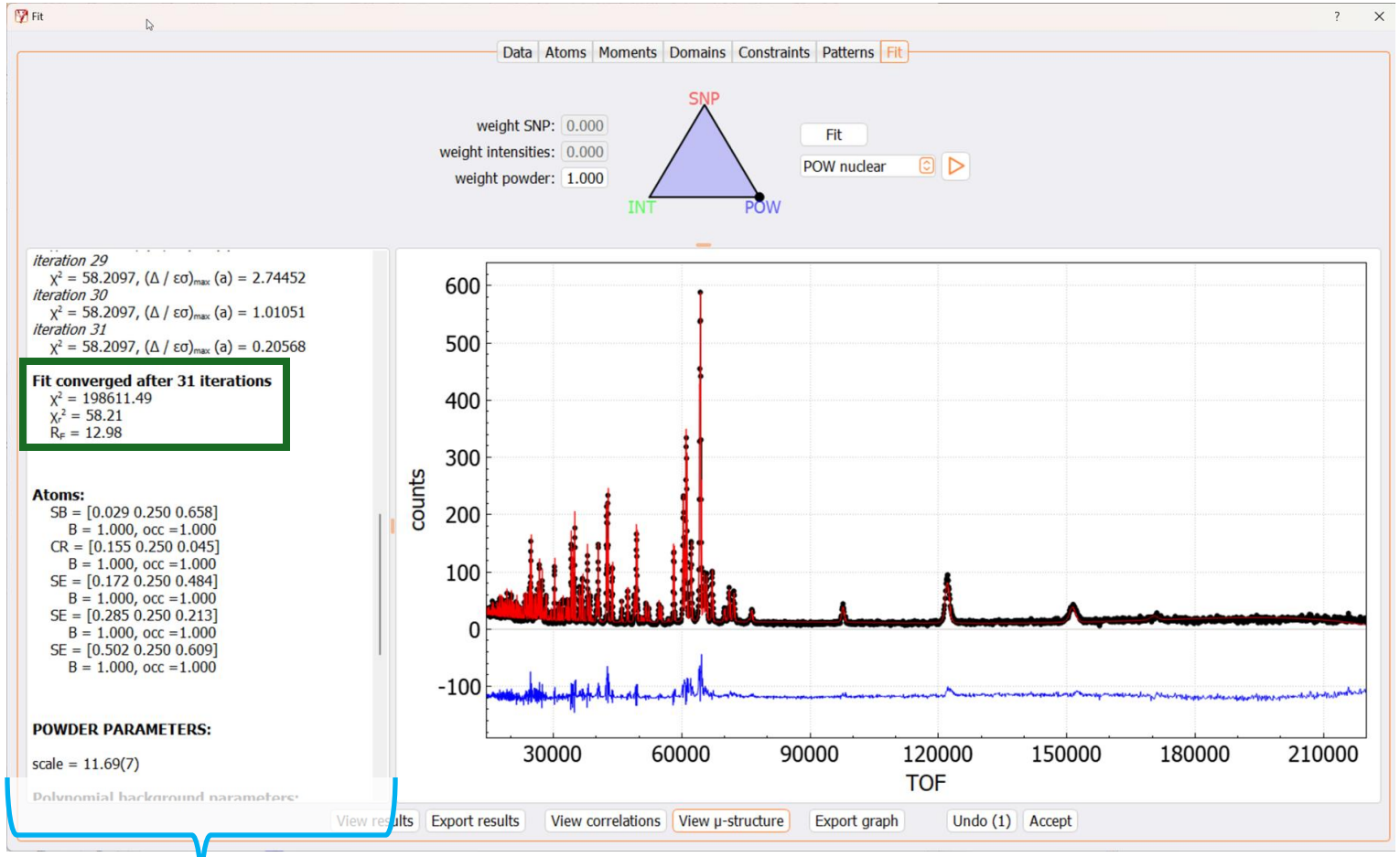
Step 2h: Select Cell parameters and ΔZ_0 for refinement

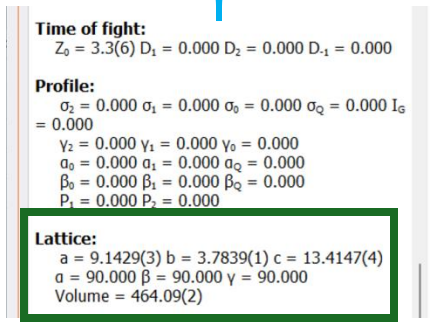


Step 3a: Run the first refinement run



Step 3b: Results after the first run of the refinement

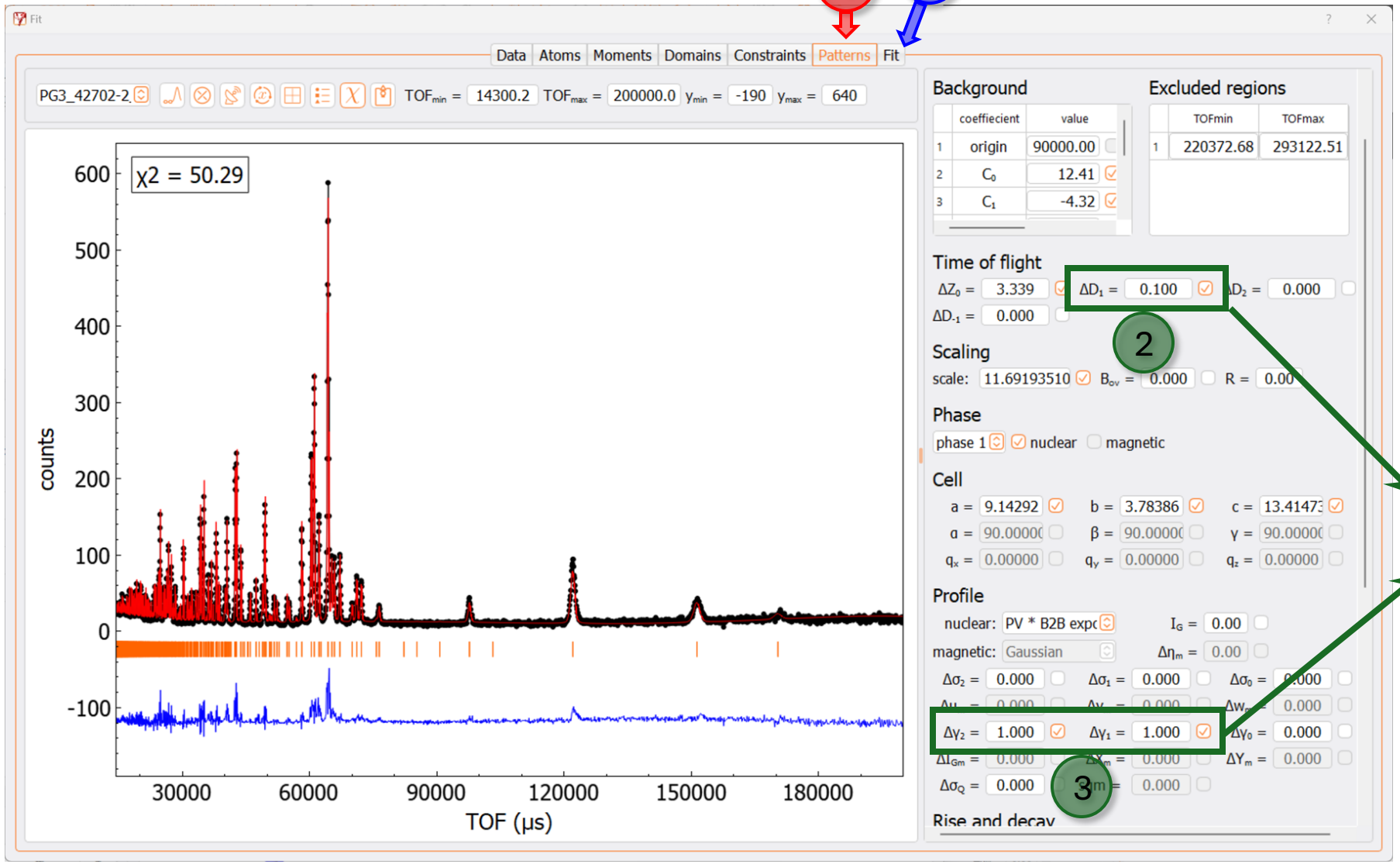




Convergence reached after 31 cycles! Discrepancy parameters and all the refined parameters can be found in the left window.

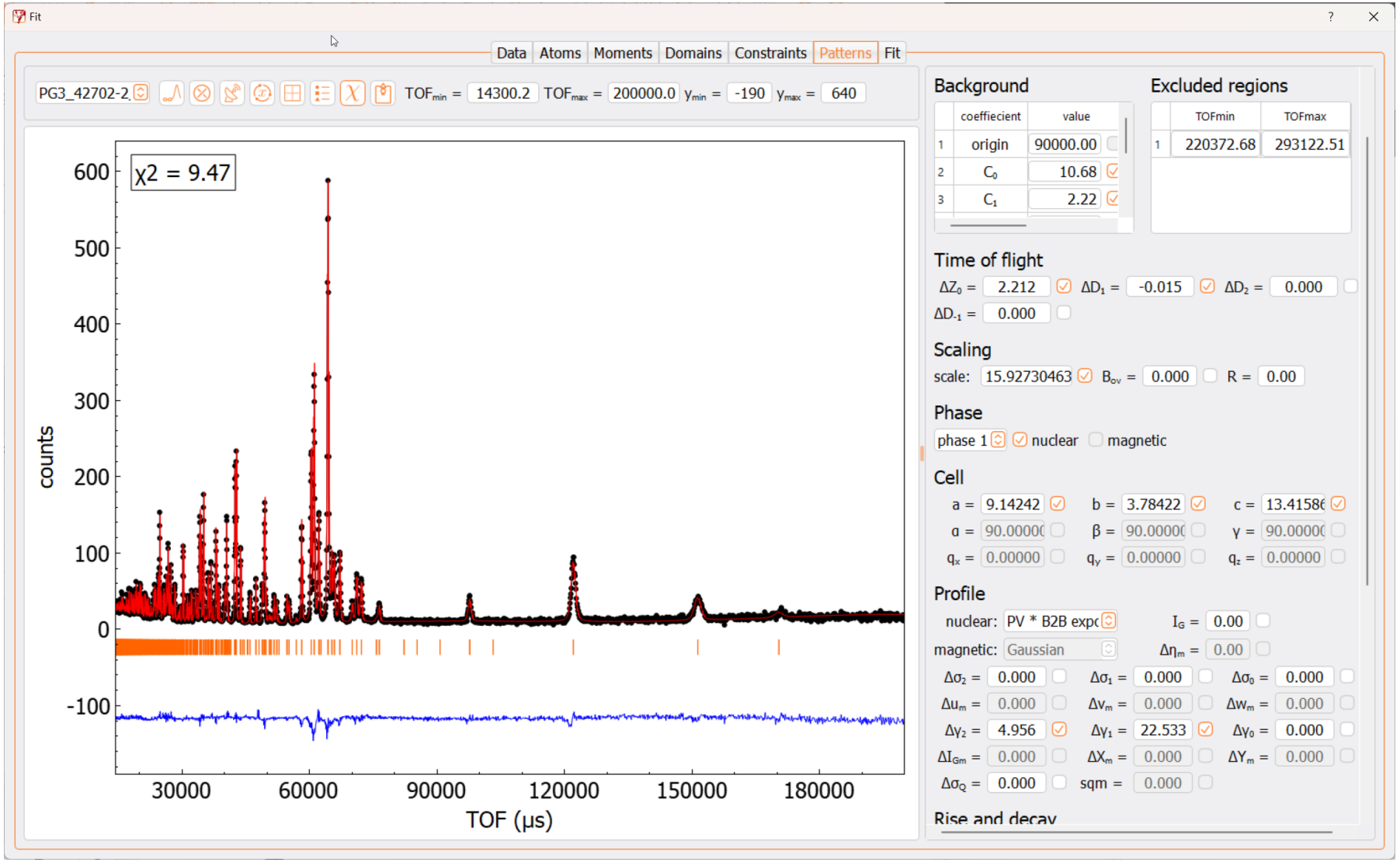
Step 3c: Refine instrument parameters

4 Back to the fitting tab and run fitting again till the fit converges.

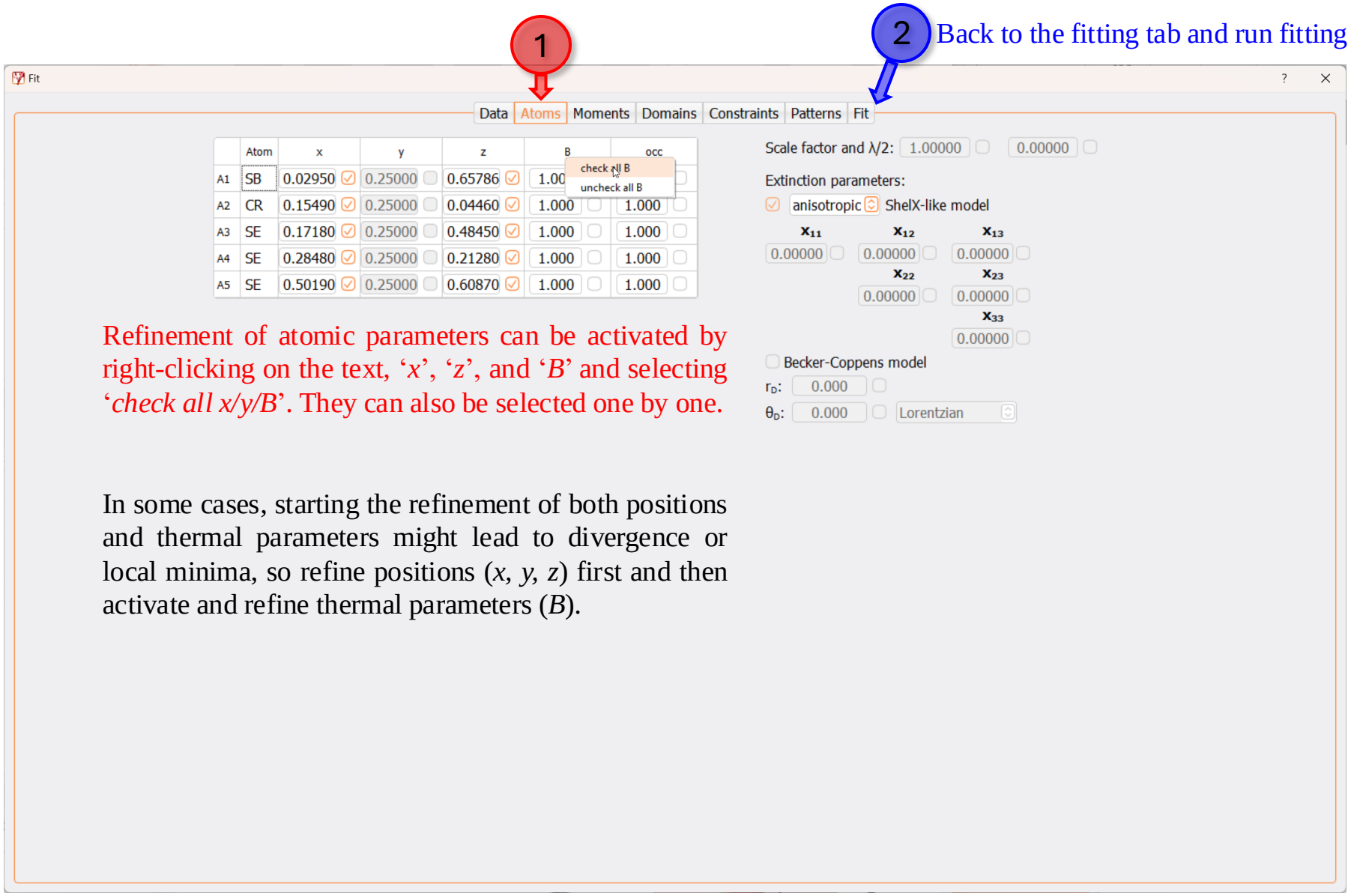


In the current version, it is necessary to choose a small value when starting the refinement of a new parameter. Leaving them to be zero won't work.

Step 3d: Significant improvement in the fit



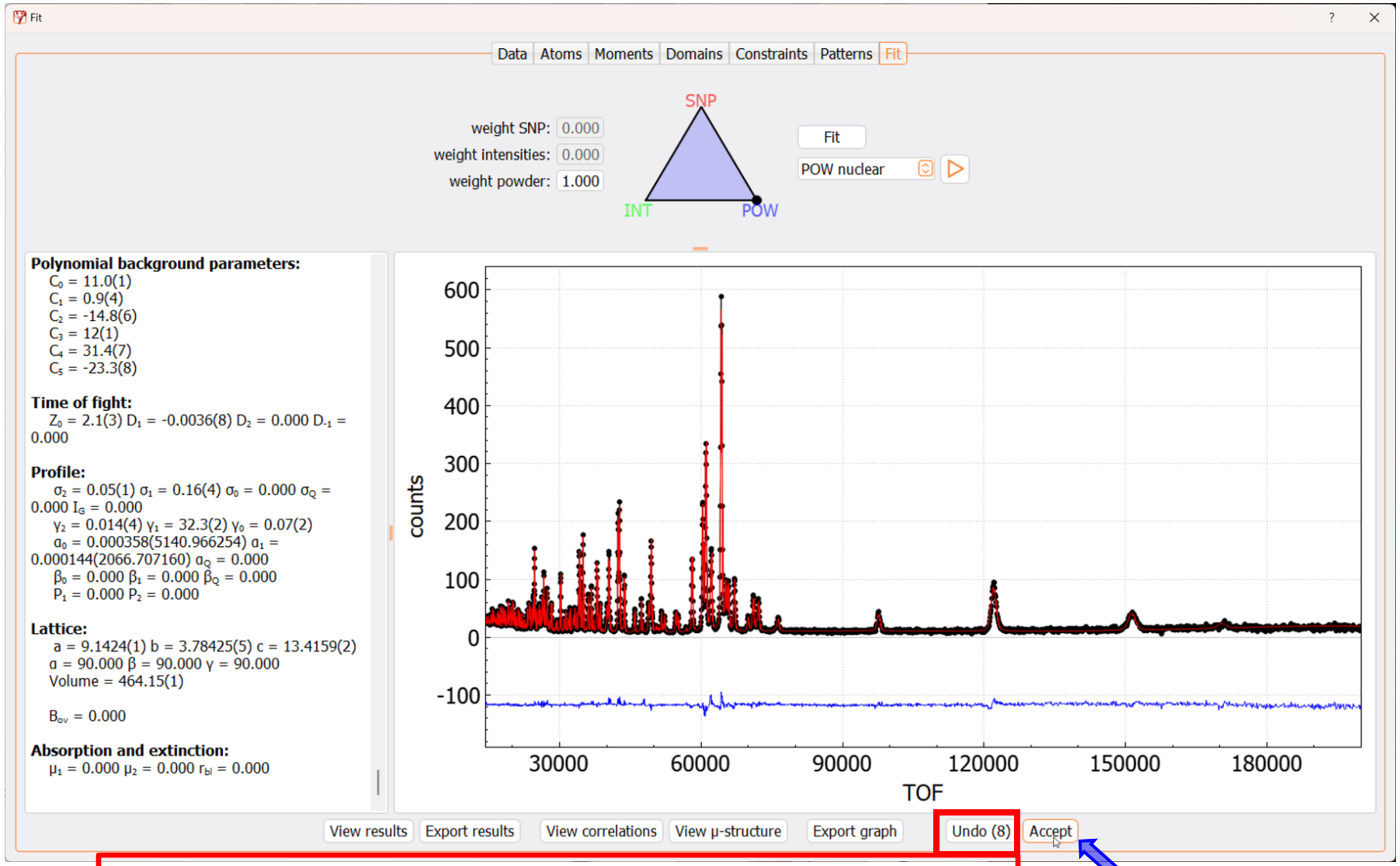
Step 3e: Refine atomic parameters



Refinement of atomic parameters can be activated by right-clicking on the text, '*x*', '*z*', and '*B*' and selecting '*check all x/y/B*'. They can also be selected one by one.

In some cases, starting the refinement of both positions and thermal parameters might lead to divergence or local minima, so refine positions (*x*, *y*, *z*) first and then activate and refine thermal parameters (*B*).

Step 3f: The fit looks good; accept this as final refinement result.

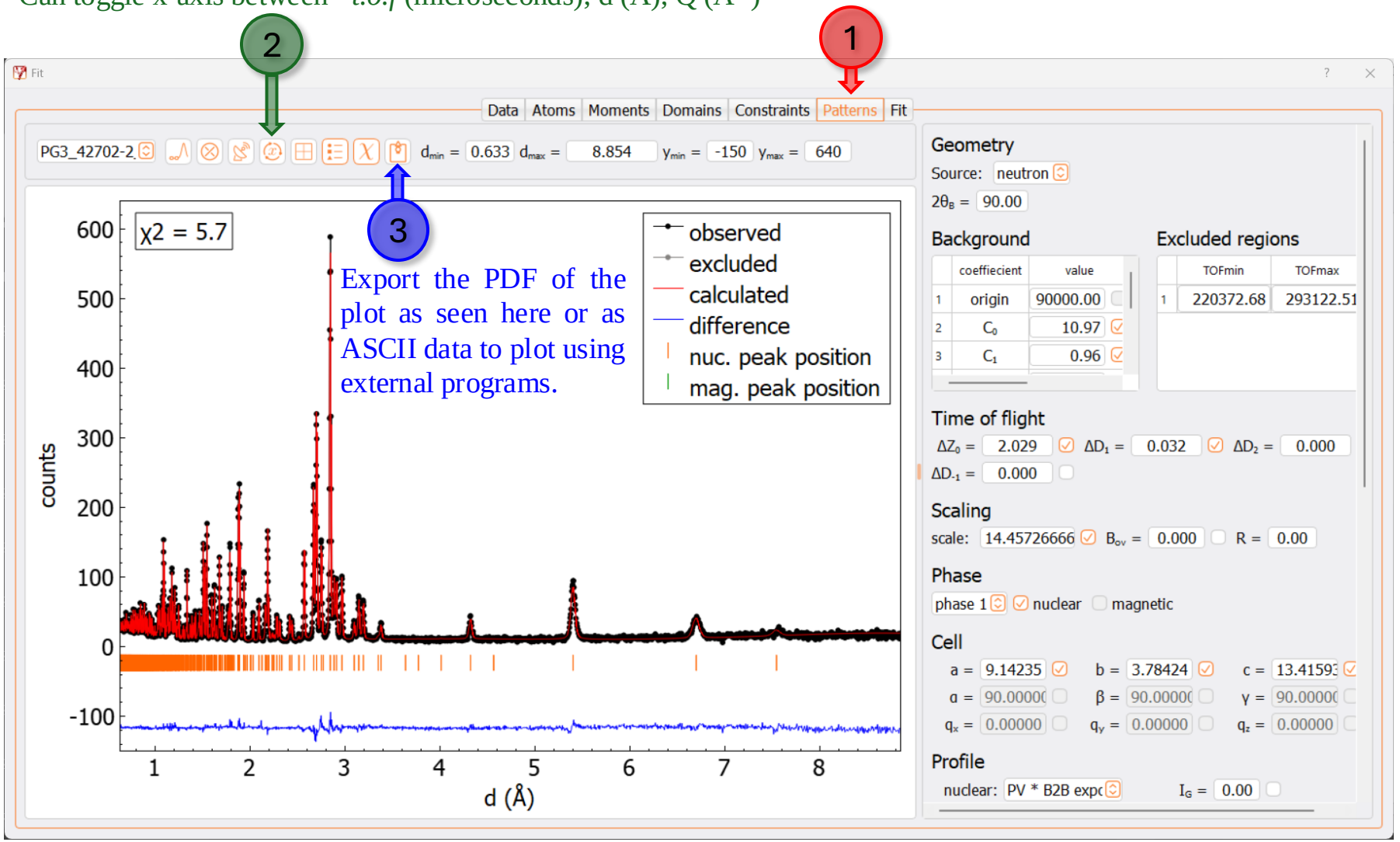


At any stage, if the fit diverges, you can recover previous states by pressing 'Undo' button. It can go back as many steps as you want, all the way back to the initial state of the refinements of the current session.

1 Accept the fit results by clicking on this button.

Step 3g: Export the refinement profile as a PDF or ASCII file.

Can toggle x-axis between - *t.o.f* (microseconds); d (Å); Q (Å^{-1})

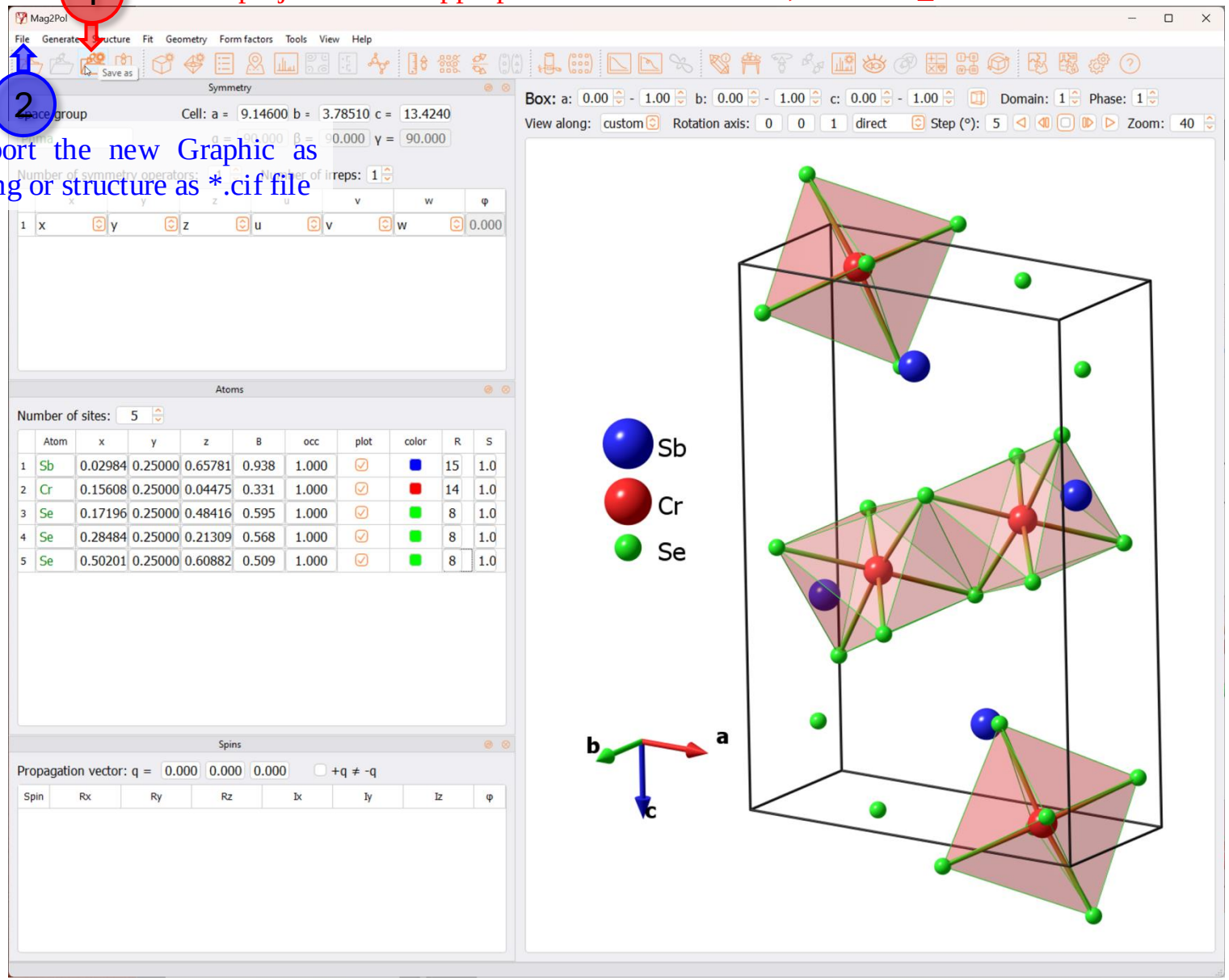


4 Close the 'Fit' window, and go back to the main Mag2Pol window.

Step 3h: Save the Mag2Pol project (*.xml). It's a very crucial step!

1 Save the project with an appropriate name. In this case, **CrSbSe3_300K.xml**

2 Export the new Graphic as *.png or structure as *.cif file

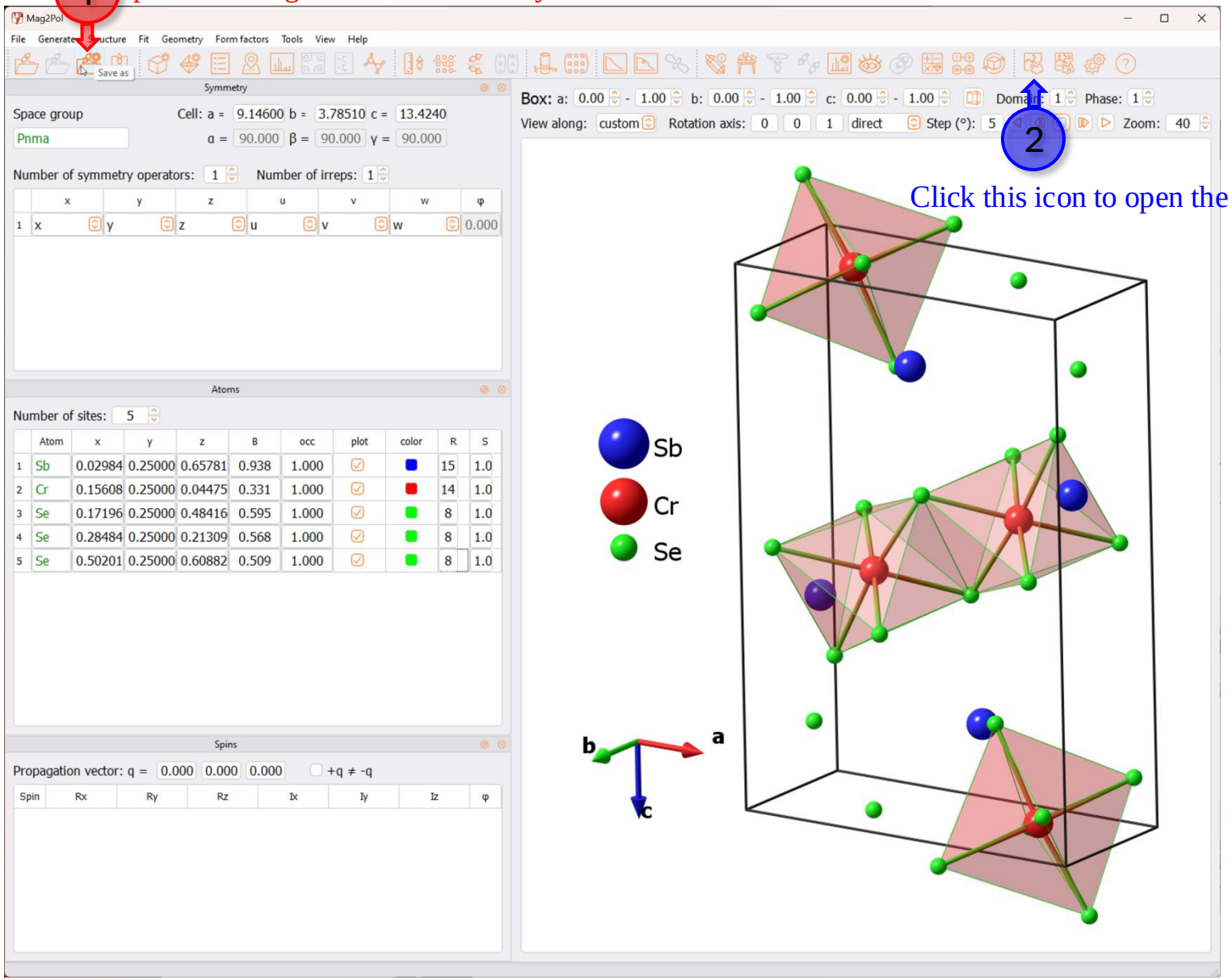


In the current version of Mag2Pol, the project (*.xml) file will have all the refined parameters and absolute paths for data and irf files. The project file can be reloaded without any issue later. But if it is opened in a different computer with a different folder structure, it will complain about missing data and irf files file and prompts to choose the file path. Choose the right file format and indicate the file path; it will reload the project in its original.

Refining low temperature NPD data, collected in the magnetically ordered phase

Step 4a: Start with the project created for the high-temperature phase in Step 7f.

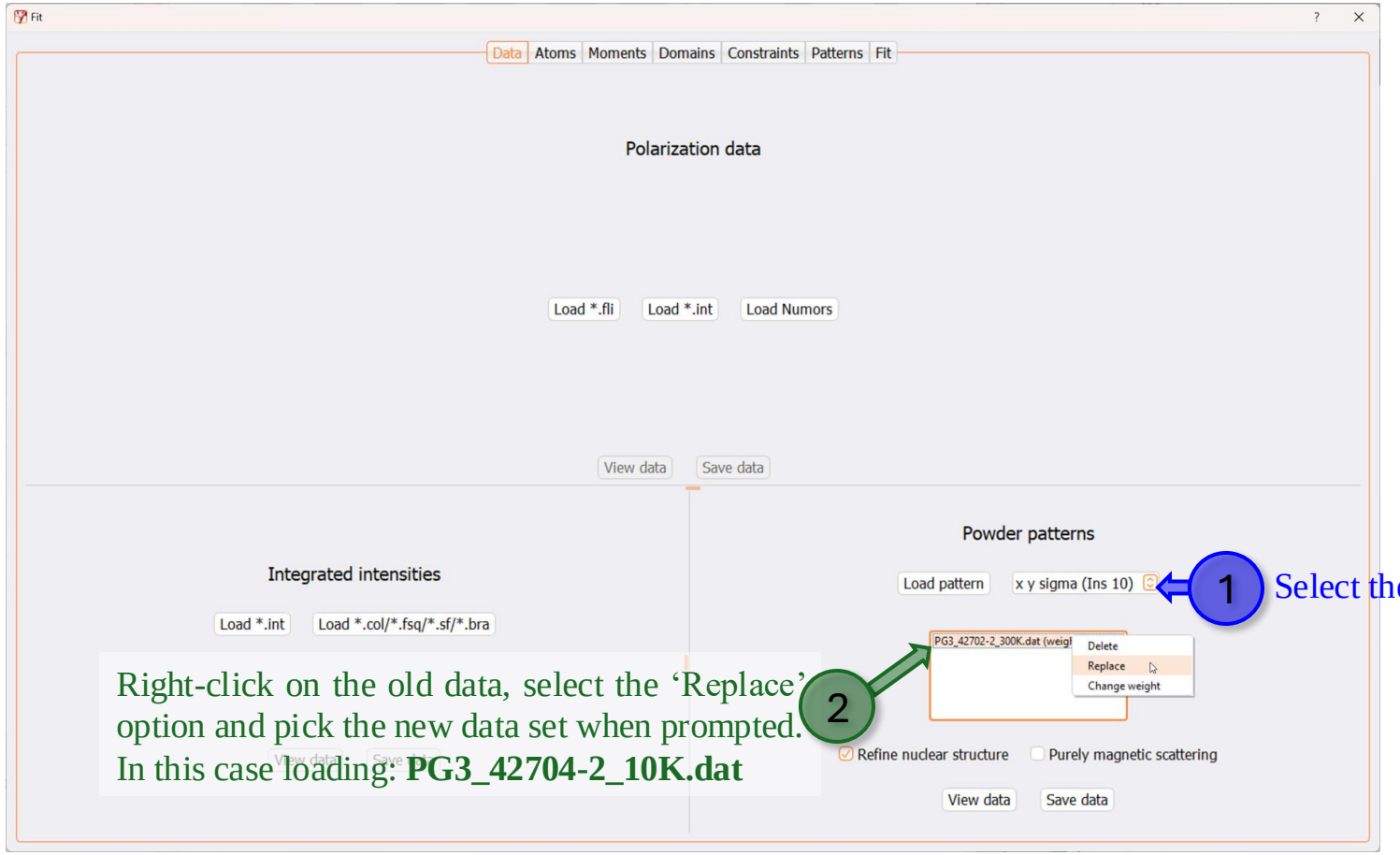
Save as a new project, with appropriate name for the low-temperature phase. To begin with, we will only consider lattice contribution.



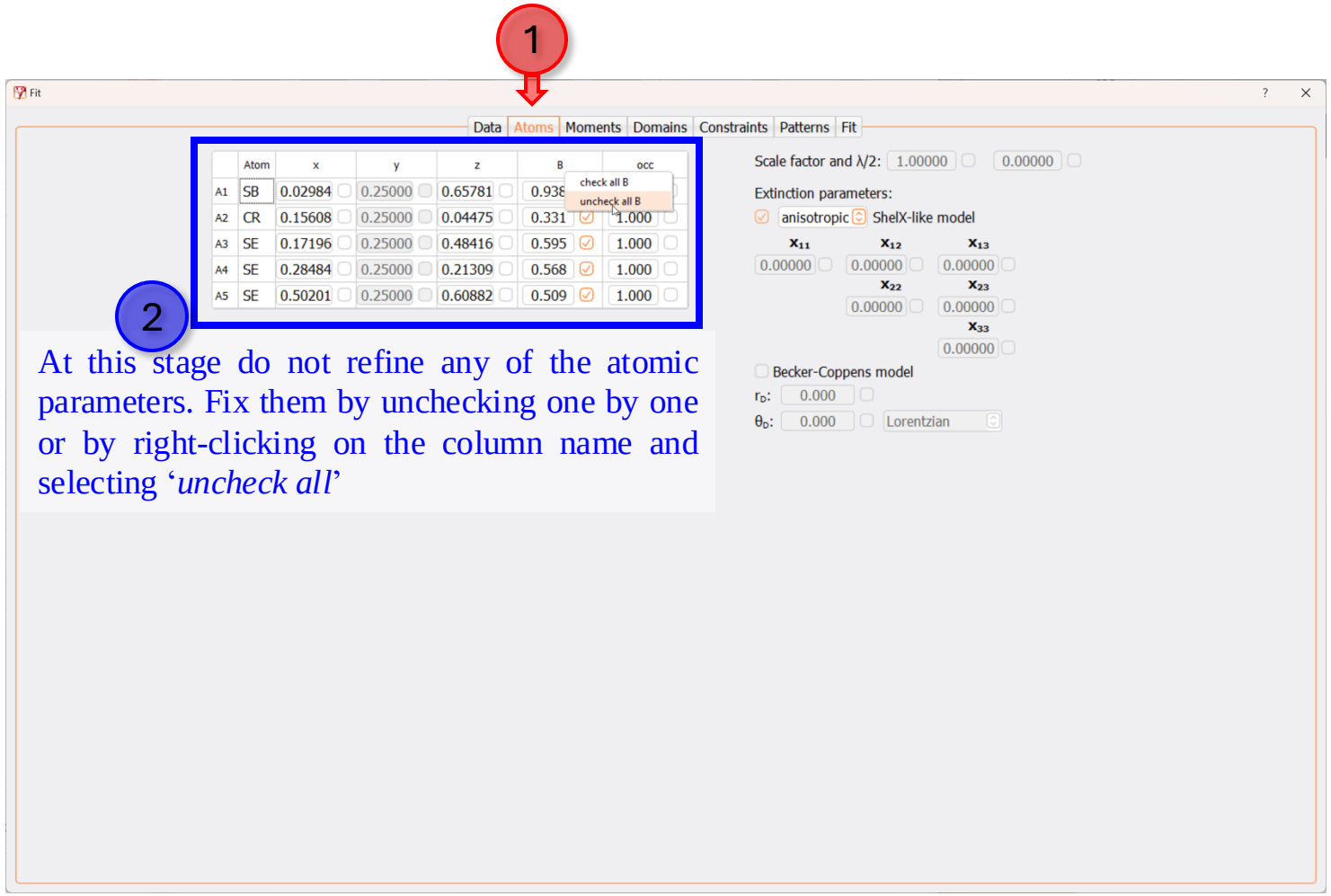
Click this icon to open the 'Fit' window.

We will be using the refinement results of high-temperature data as a starting point for the refinement of low-temperature data.

Step 4b: Replace the old data with the new data.

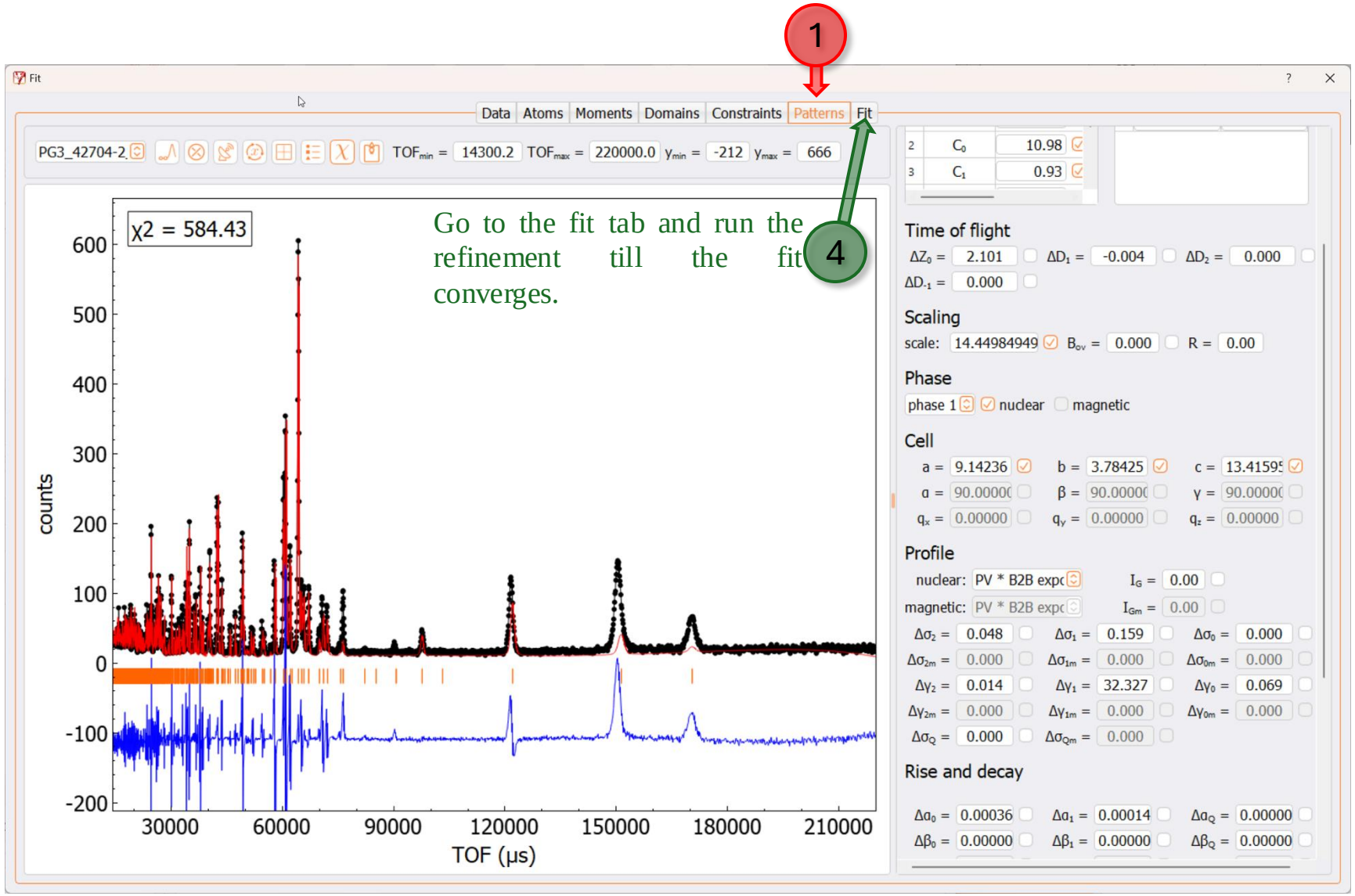


Step 4c: Fix all the atomic parameters



At this stage do not refine any of the atomic parameters. Fix them by unchecking one by one or by right-clicking on the column name and selecting '*uncheck all*'

Step 4d: Refine the scale and Cell parameters



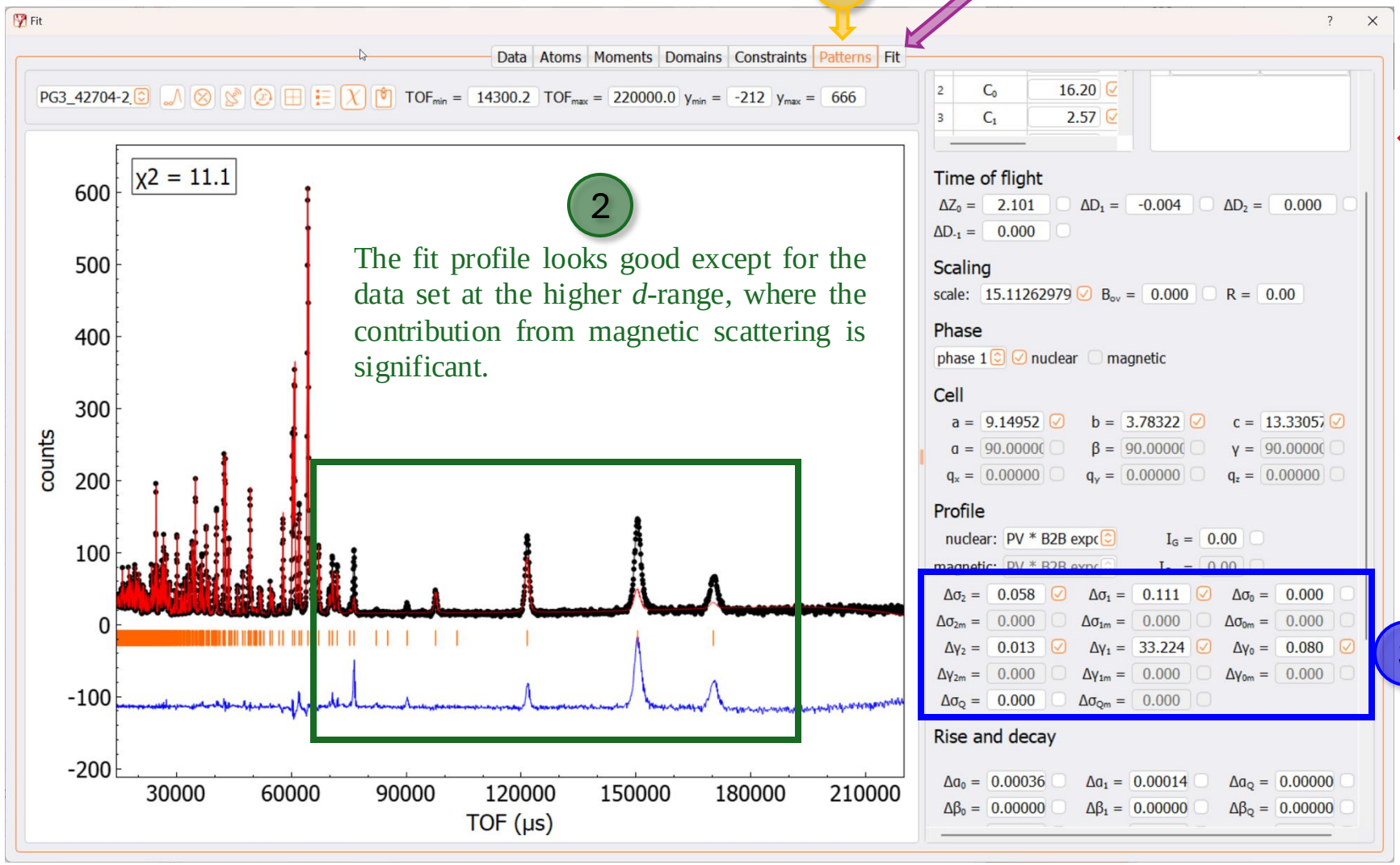
2

NOTE: If the new data set that was loaded in this section is measured with a different center wavelength (data bank) than the previous data set, then make sure to load the corresponding irf file. And need to refine the profile, Time of flight, and Rise and decay parameters as necessary, before proceeding to the next point (3).

In the current case, both 300 K and 10K data sets were collected with the same center wavelength neutrons, so no need to change the irf file.

3 Fix all parameters except the '*scale*' and '*Cell*' parameters.

Step 4e: Refine profile parameters as necessary



1

2

The fit profile looks good except for the data set at the higher *d*-range, where the contribution from magnetic scattering is significant.

3

Select profile parameters for the refinement

4

Go to the fit tab and run the refinement till the fit converges.

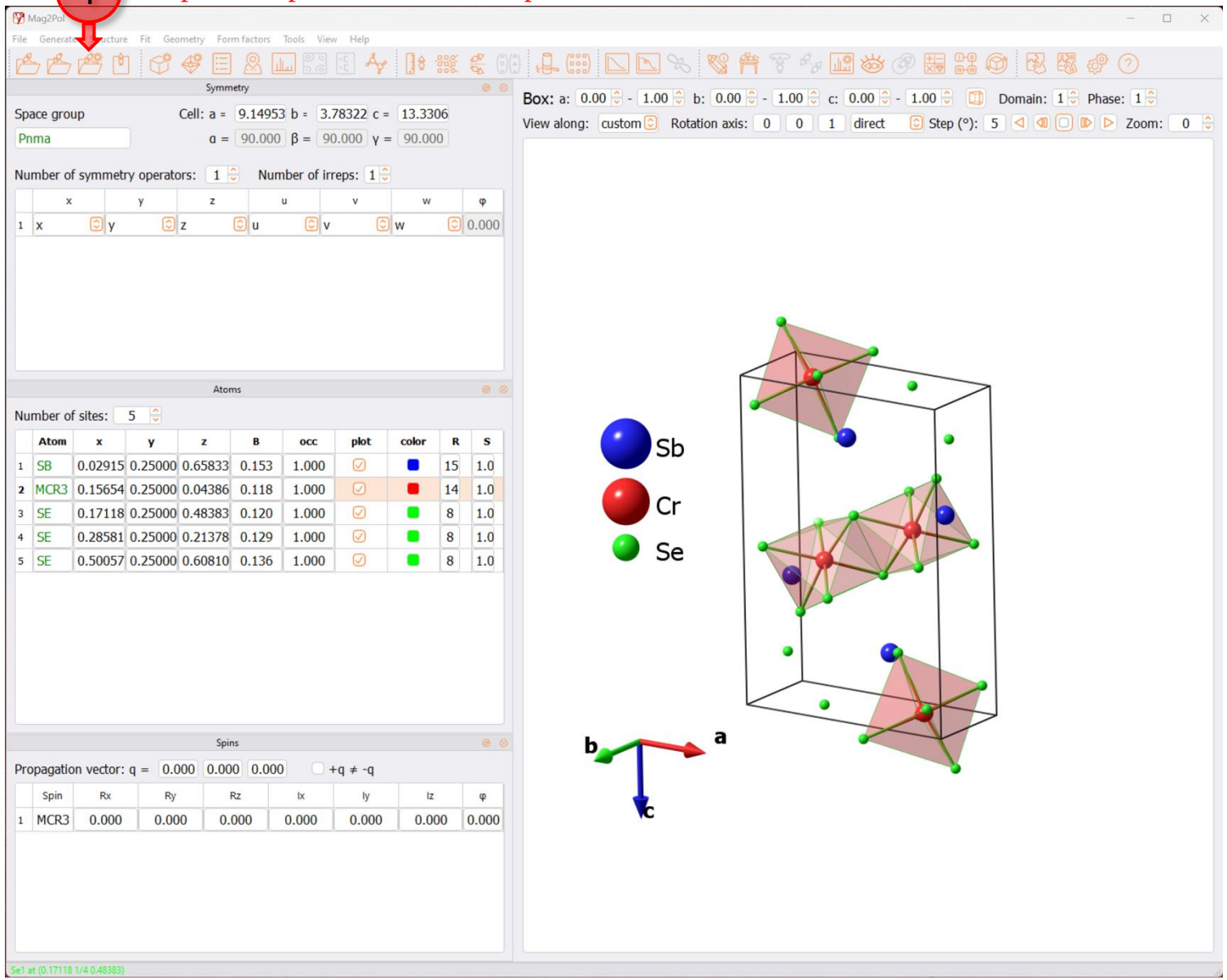
5

After convergence is reached click on the 'Accept' button *(under Fit tab) and* close this 'Fit' window.

Step 4f: Back to the main window of Mag2Pol

1

Save as a new project, with an appropriate name for the low-temperature phase. In this example, CrSbSe3_10K_Lattice.xml



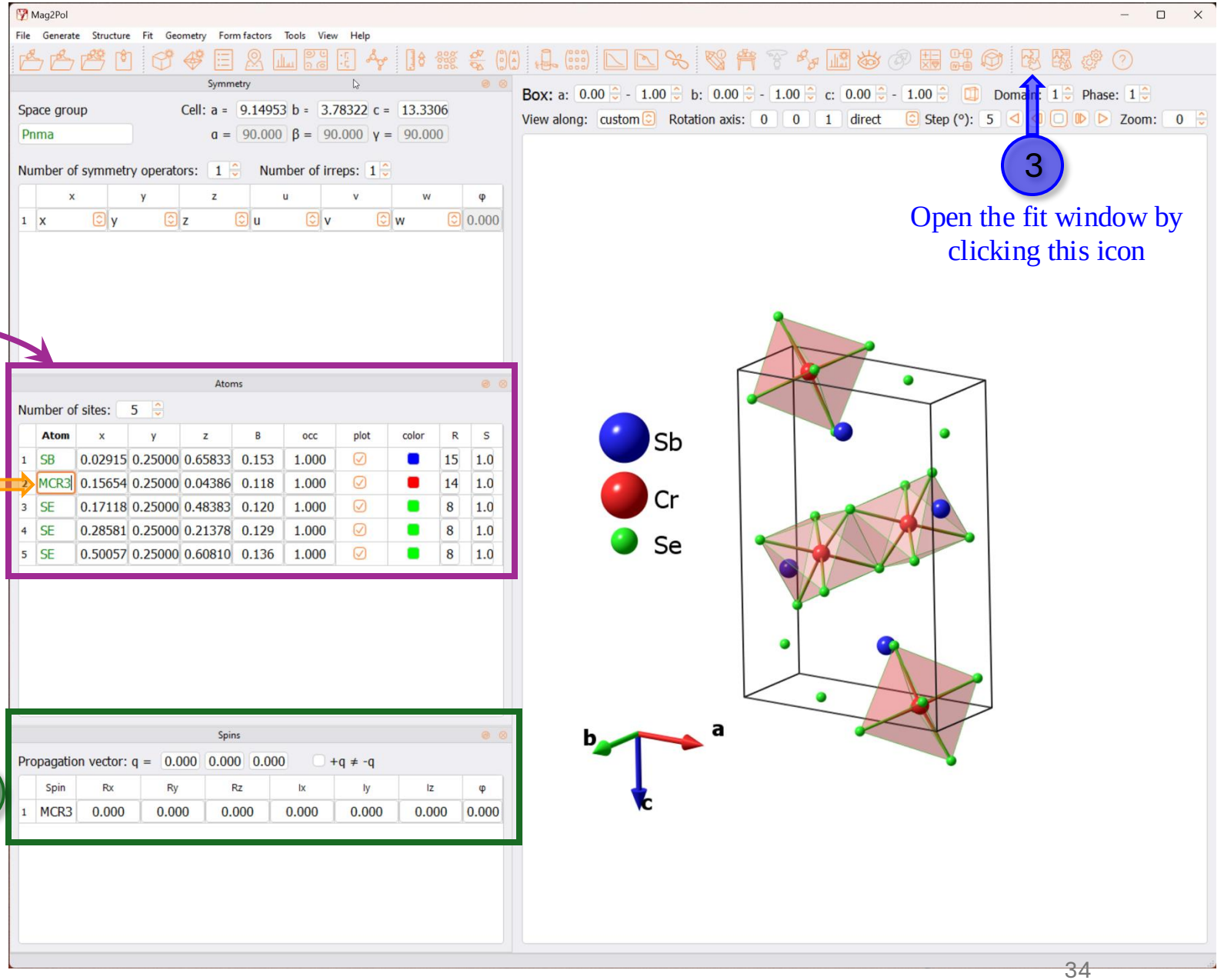
We have refined only the lattice contribution so far, and now we can define the magnetic ion and proceed with the refinement of the magnetic structure.

Refinement of magnetic structure

Step 9: Define the magnetic ion

Under the ‘Atoms’ section:
Depending on the type of magnetic contribution, simply add the prefix M or J to the magnetic ion followed by the ionic state. In this case only Cr^{3+} is magnetic.
After this, update the structure by the key board shortcut [Ctrl+U] (on Windows) or [Cmd+U] (on Mac)

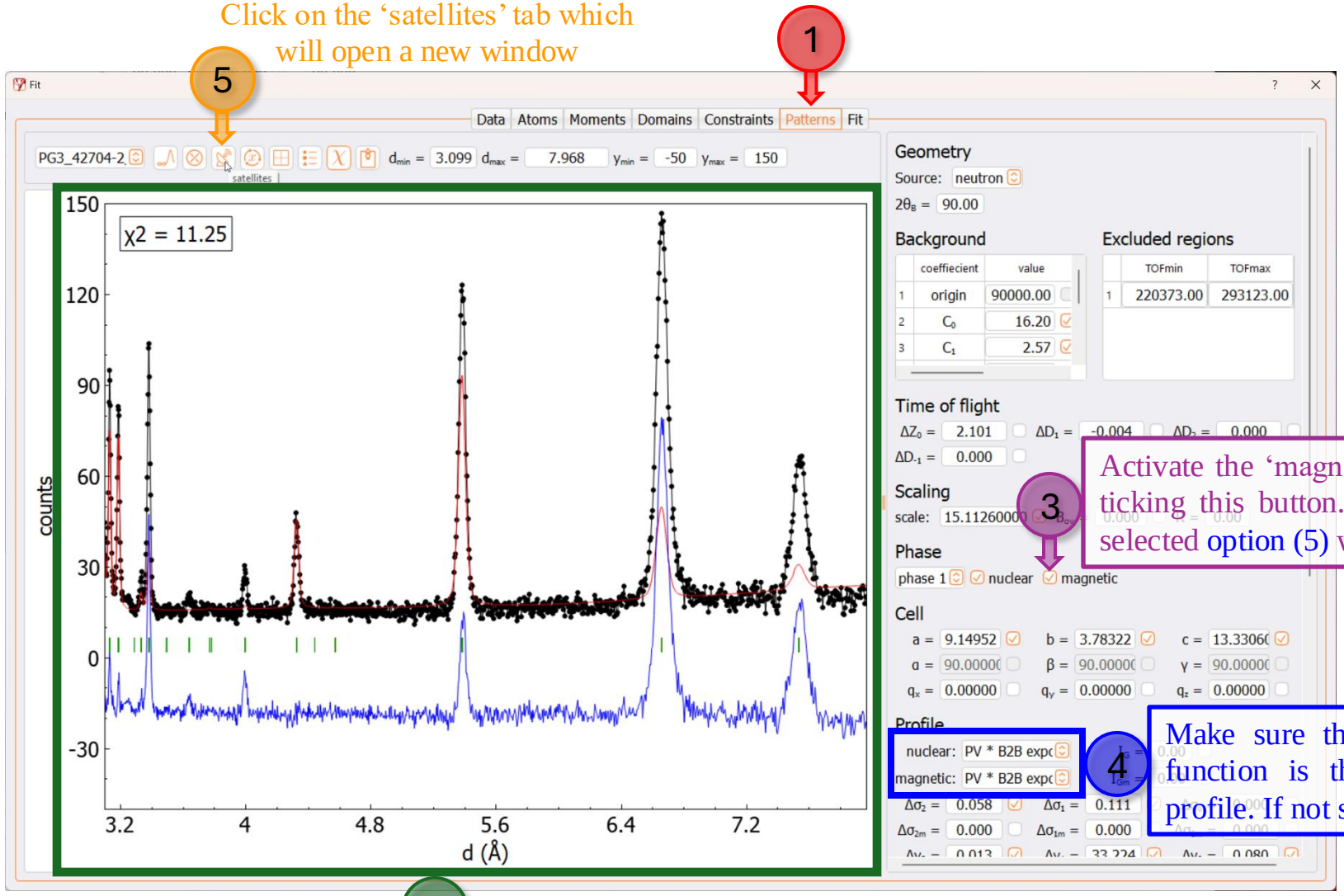
As soon as a Magnetic atom is defined under the ‘Atoms’ section, the ‘Spins’ section will be populated automatically with the Magnetic ions. By default, the Propagation vector is set to (0, 0, 0). Leave everything default.



Searching for propagation vector (q-vector) from the satellite peaks

Step 5a: Zoom into the diffraction pattern and activate ‘Satellites’ window

Click on the ‘satellites’ tab which will open a new window

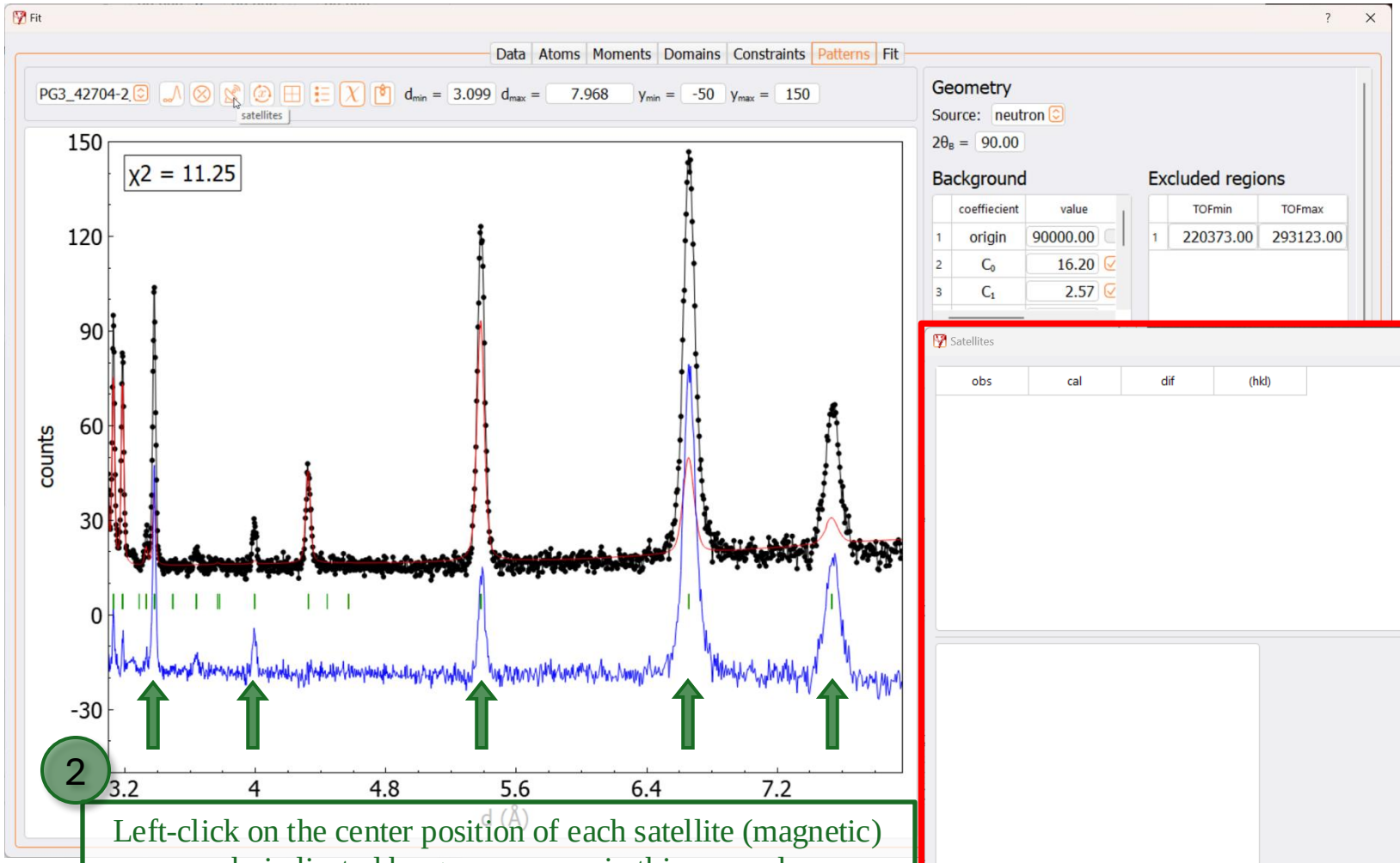


Activate the ‘magnetic’ option by ticking this button. If this is not selected option (5) will not work.

Make sure the magnetic peak profile function is the same as the nuclear profile. If not select it manually.

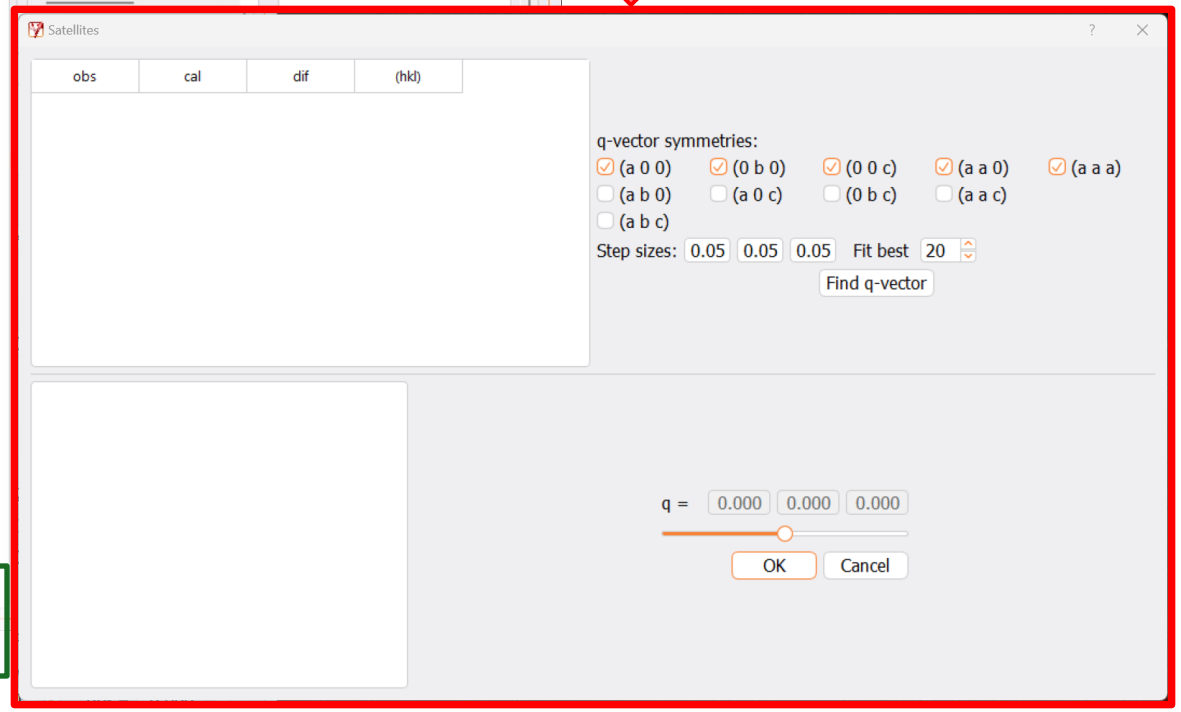
Zoom into the section of the diffraction pattern where you see magnetic peaks (at higher *d*-range)

Step 5b: Select satellite (magnetic) peaks

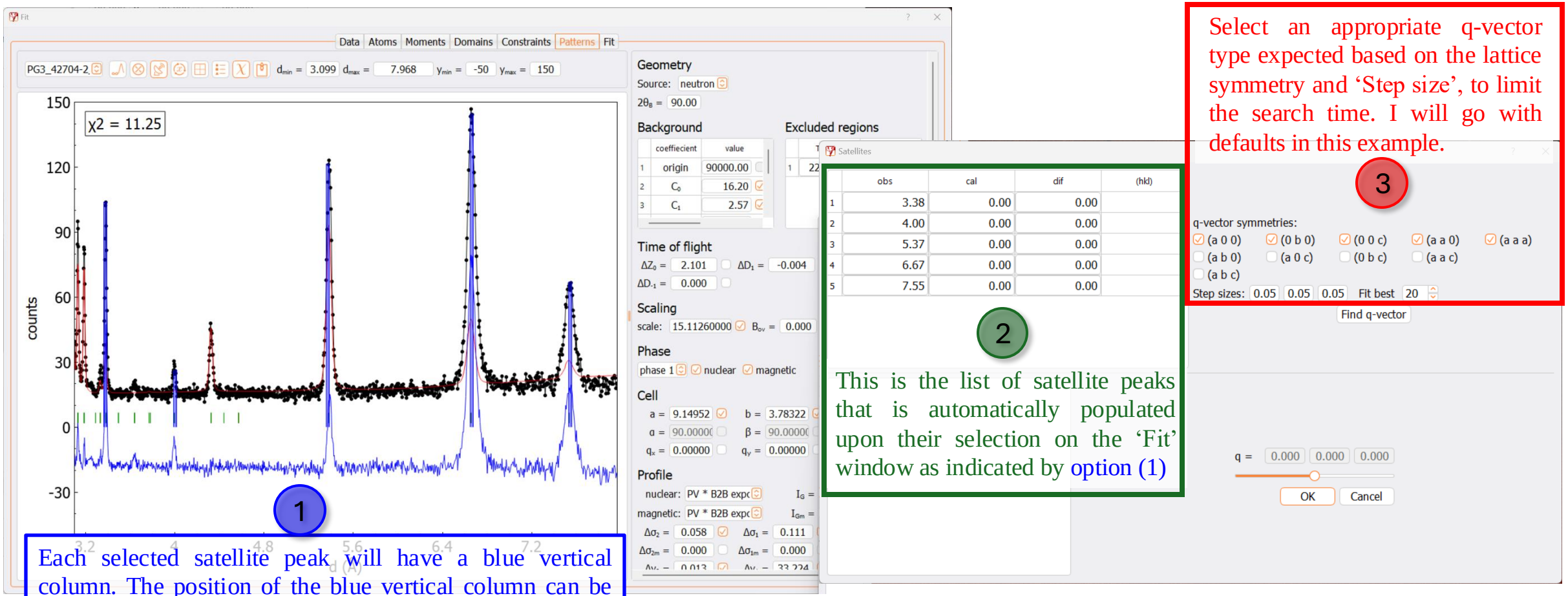


2 Left-click on the center position of each satellite (magnetic) peak, indicated by green arrows in this example.

1 Move this 'Satellites' window away so that the diffraction profile on the 'Fit' window is accessible.



Step 5b: Select satellite (magnetic) peaks

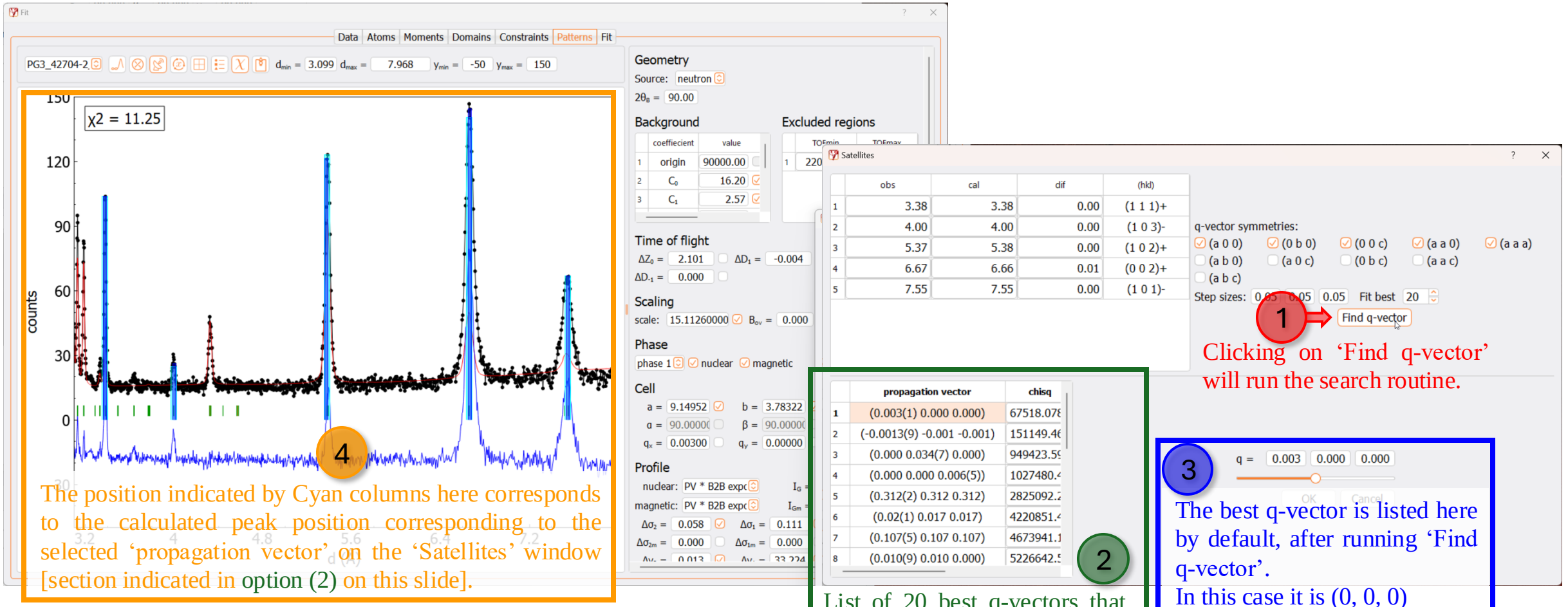


Each selected satellite peak will have a blue vertical column. The position of the blue vertical column can be altered using a mouse (left-click + hold + move) and can be removed by right-clicking on it

This is the list of satellite peaks that is automatically populated upon their selection on the 'Fit' window as indicated by option (1)

Select an appropriate q-vector type expected based on the lattice symmetry and 'Step size', to limit the search time. I will go with defaults in this example.

Step 5c: Run the q-vector search, and note down the best q-vectors



The position indicated by Cyan columns here corresponds to the calculated peak position corresponding to the selected 'propagation vector' on the 'Satellites' window [section indicated in option (2) on this slide].

Clicking on 'Find q-vector' will run the search routine.

List of 20 best q-vectors that fit the selected satellites best, Populated automatically after running 'Find q-vector'.

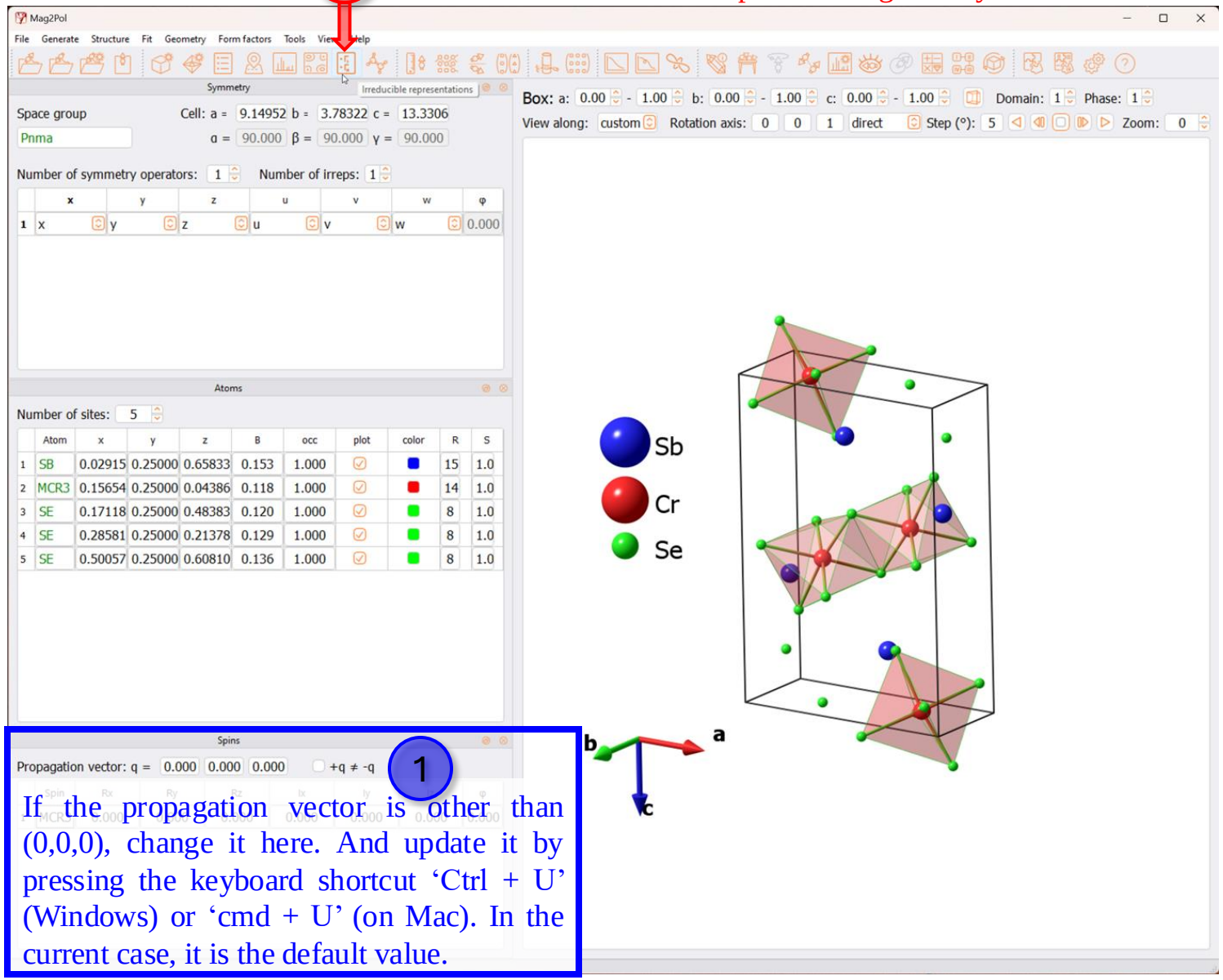
The best q-vector is listed here by default, after running 'Find q-vector'.
In this case it is (0, 0, 0)

5 Note down all the propagation vectors that you wish to try out. Close both the 'Satellites' and 'Fit' windows. And go back to the main window of Mag2Pol.

Refinement of Magnetic structure

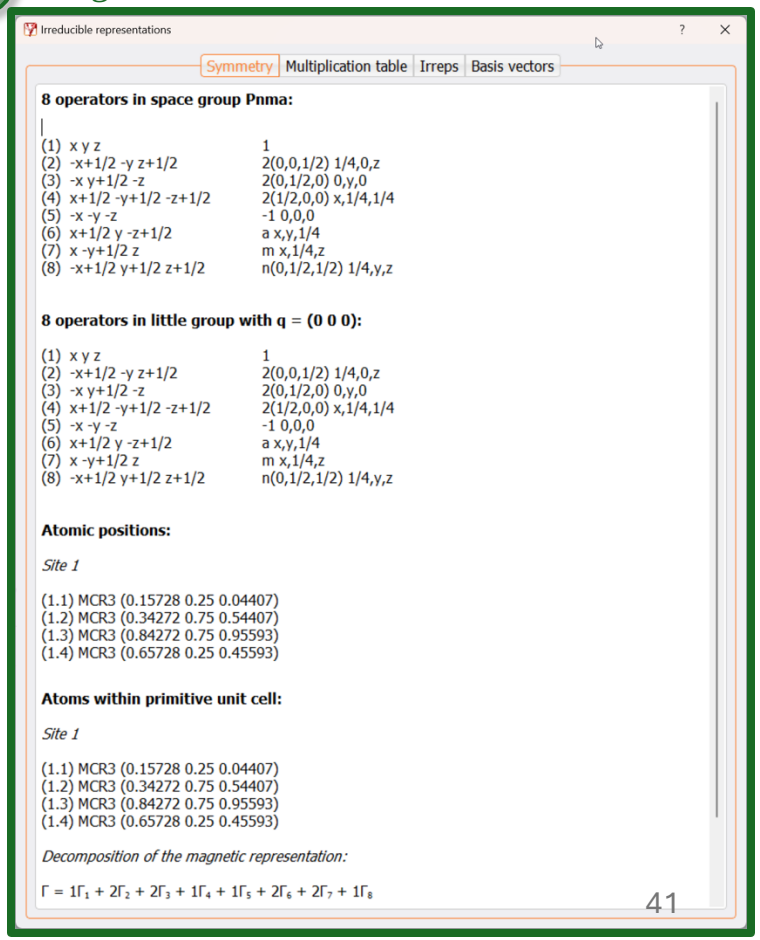
Step 6a: Update propagation vector and open the ‘Irreducible representations’ window

2 Clicking on this icon will open a new window, ‘Irreducible representations’. This may not be active if the structure is not updated using the keyboard shortcut after updating the *q*-vector.



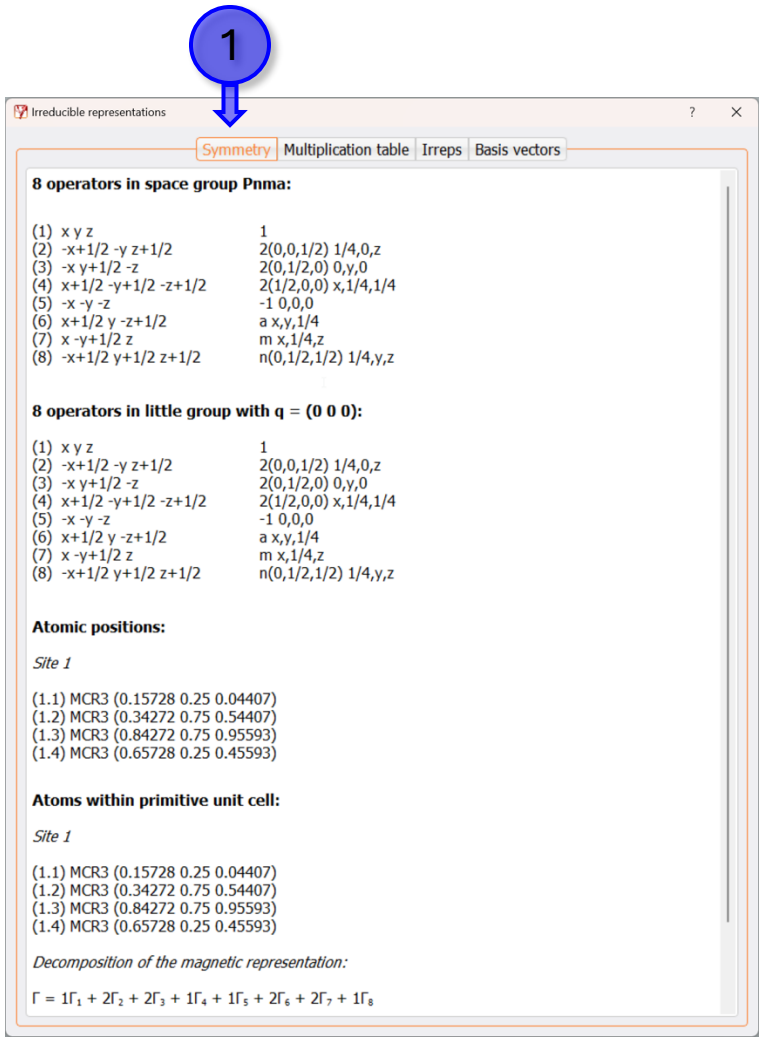
1 If the propagation vector is other than (0,0,0), change it here. And update it by pressing the keyboard shortcut ‘Ctrl + U’ (Windows) or ‘cmd + U’ (on Mac). In the current case, it is the default value.

3 The newly opened ‘Irreducible representations’. Contains details of magnetic representations for the given wave vector.



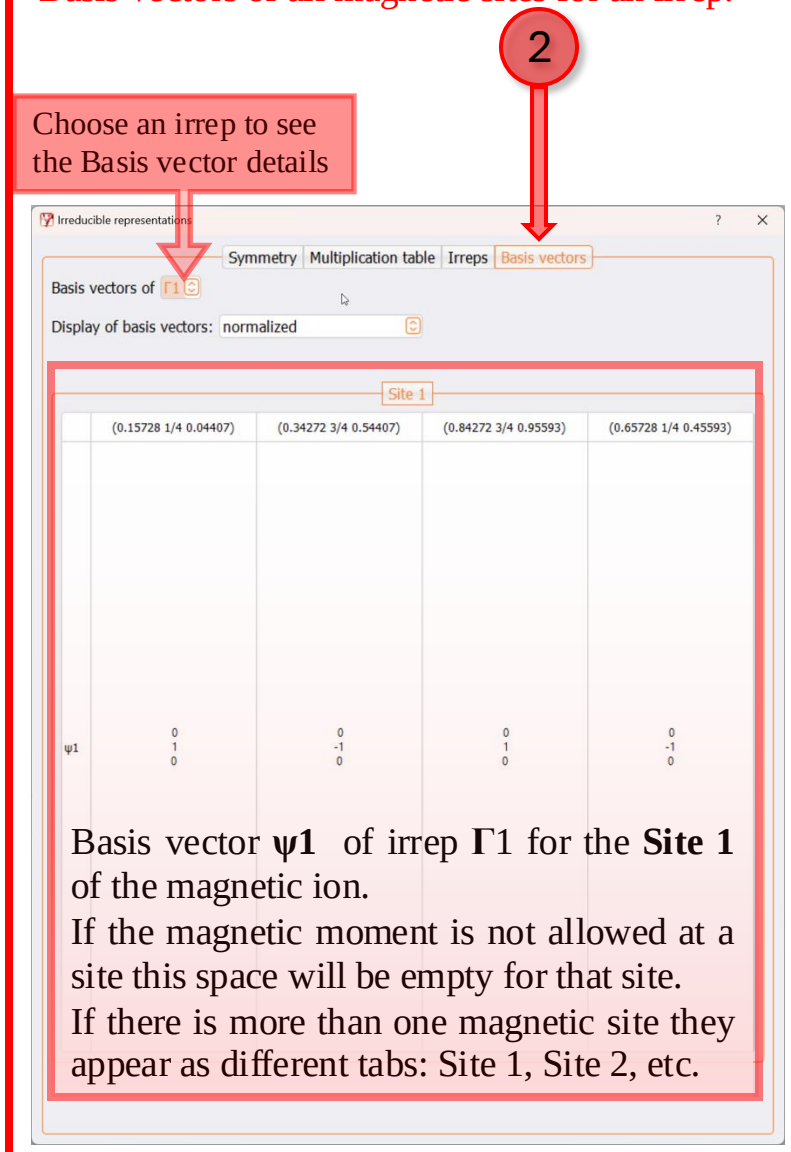
Step 6b: On the 'Irreducible representation' window select an irrep (Γn) for the refinement

The **'Symmetry'** tab provides some details of symmetry analysis and magnetic representation



The **'Basis vectors'** tab provides some details of Basis vectors of all magnetic sites for an irrep.

Choose an irrep to see the Basis vector details



Basis vector **ψ1** of irrep **Γ1** for the **Site 1** of the magnetic ion.

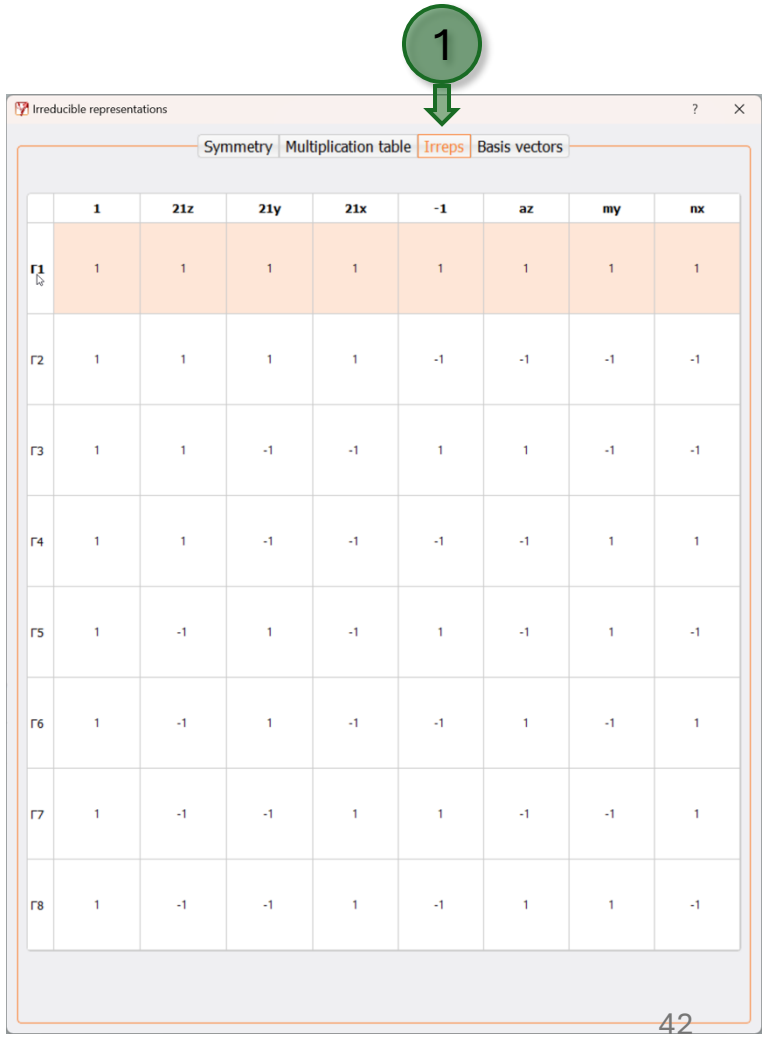
If the magnetic moment is not allowed at a site this space will be empty for that site.

If there is more than one magnetic site they appear as different tabs: Site 1, Site 2, etc.

The **'Irreps'** tab lists all irreps.

Right-click on one of the irreps symbols (Γ1 in this example), consequently click on 'Use Γ1'

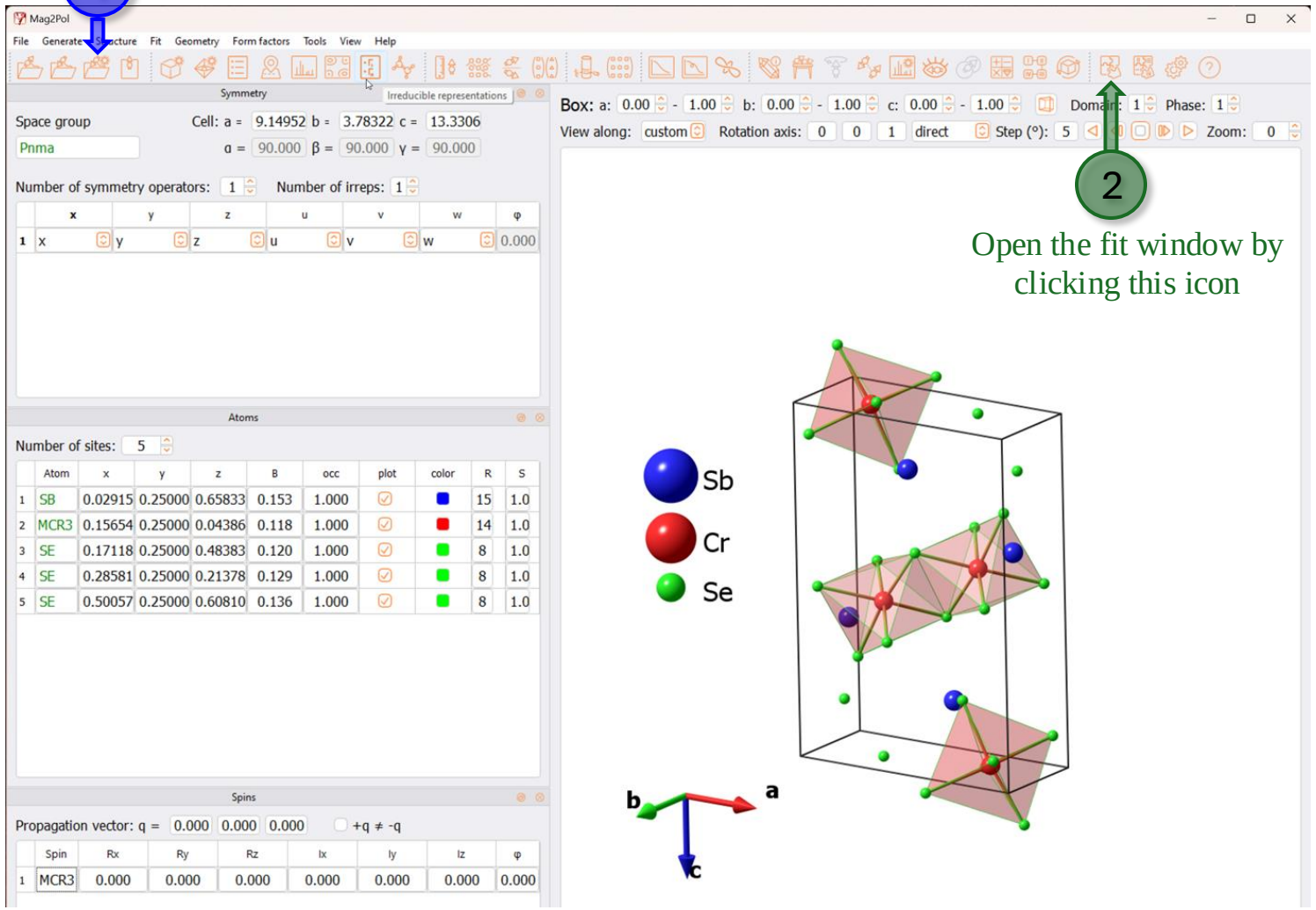
Go back to the main window of Mag2Pol



Step 6c: Back to the main window, save the project

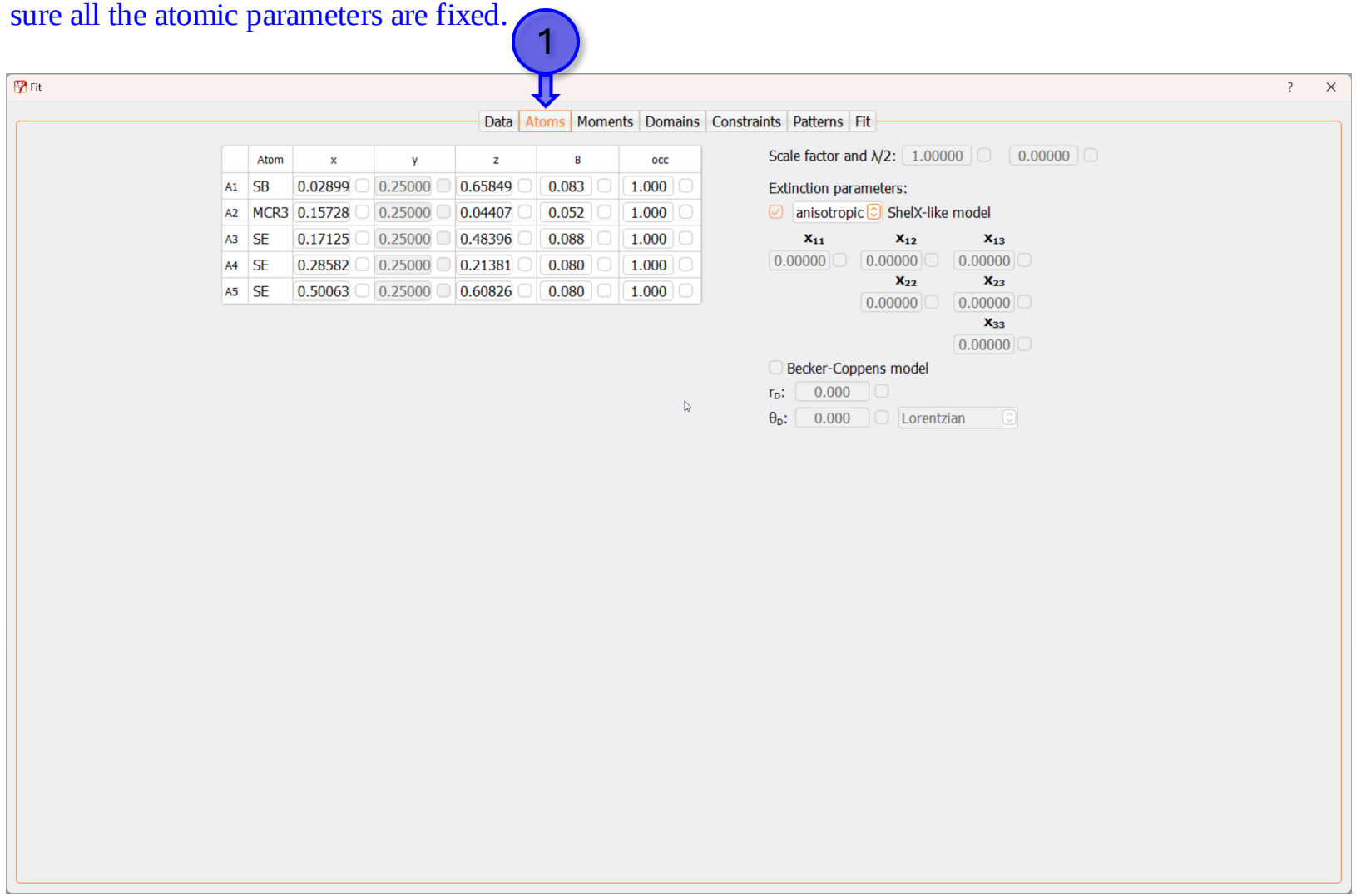
Save as a new project indicating irrep in the file name.

For example, for this example, the project can be saved as **CrSbSe3_10K_Mag_G1.xml**

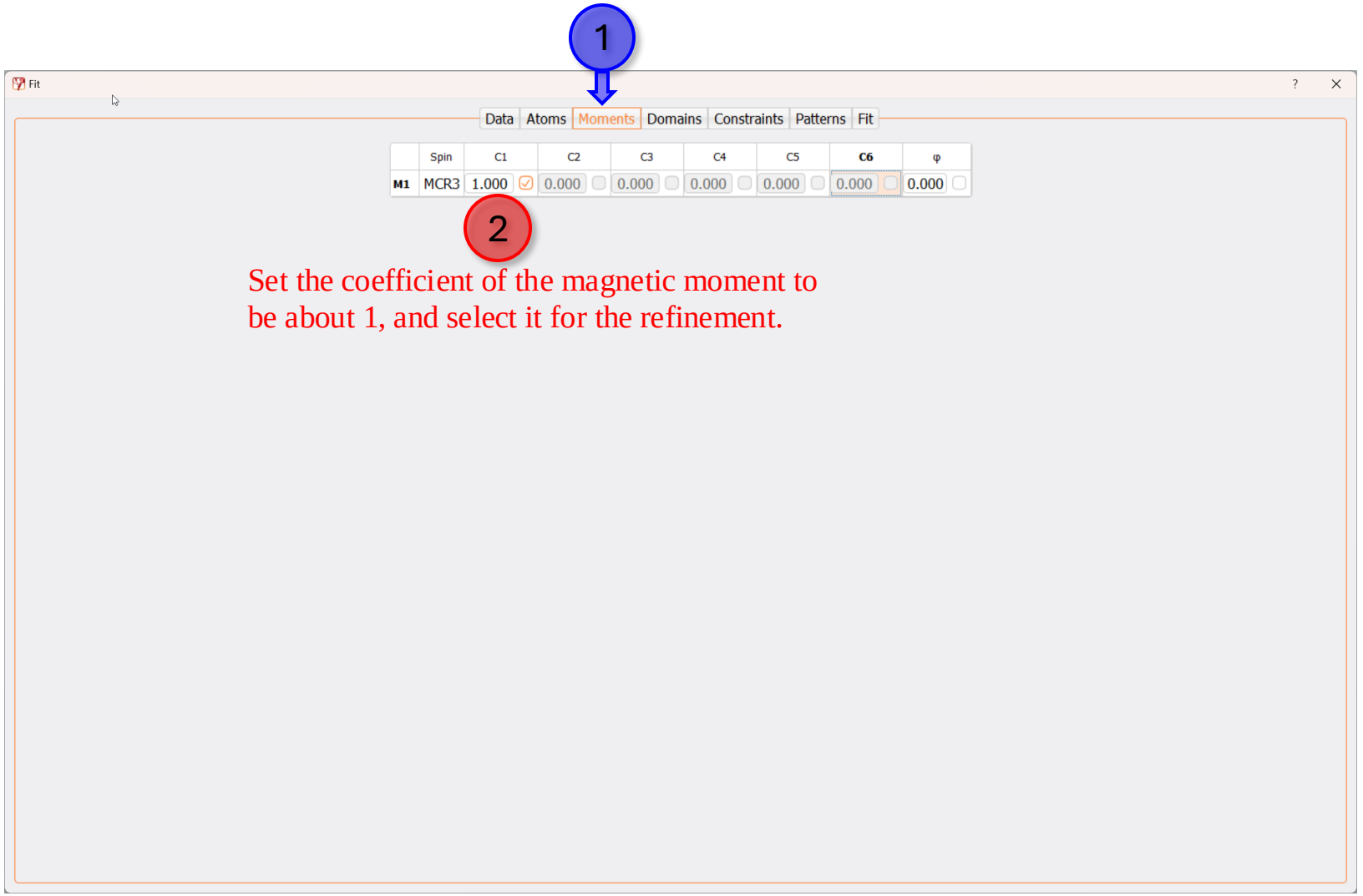


Step 6d: Fix all atomic parameters

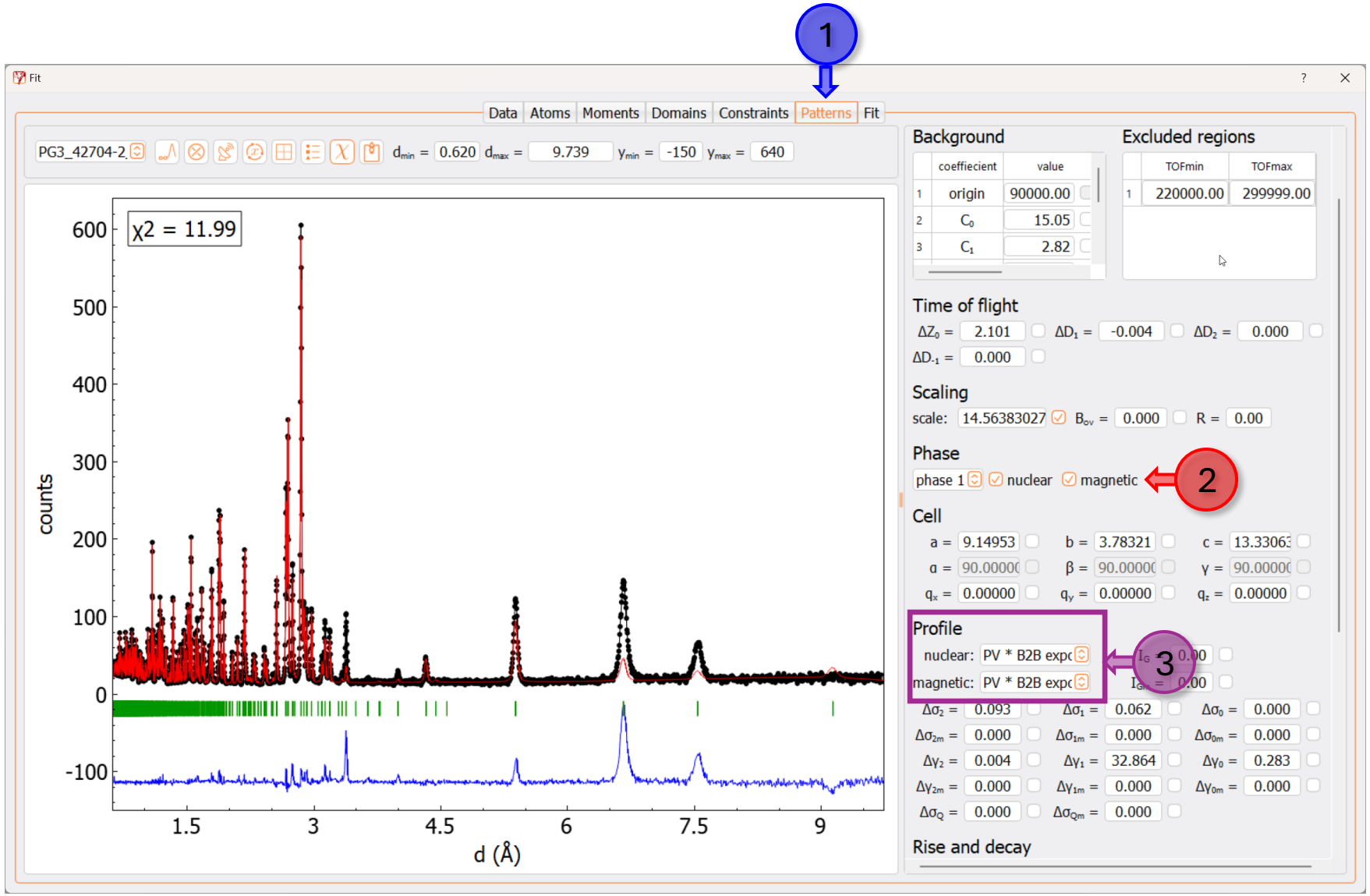
On the ‘Fit’ window, under the ‘Atoms’ tab, make sure all the atomic parameters are fixed.



Step 6e: Fix all atomic parameters



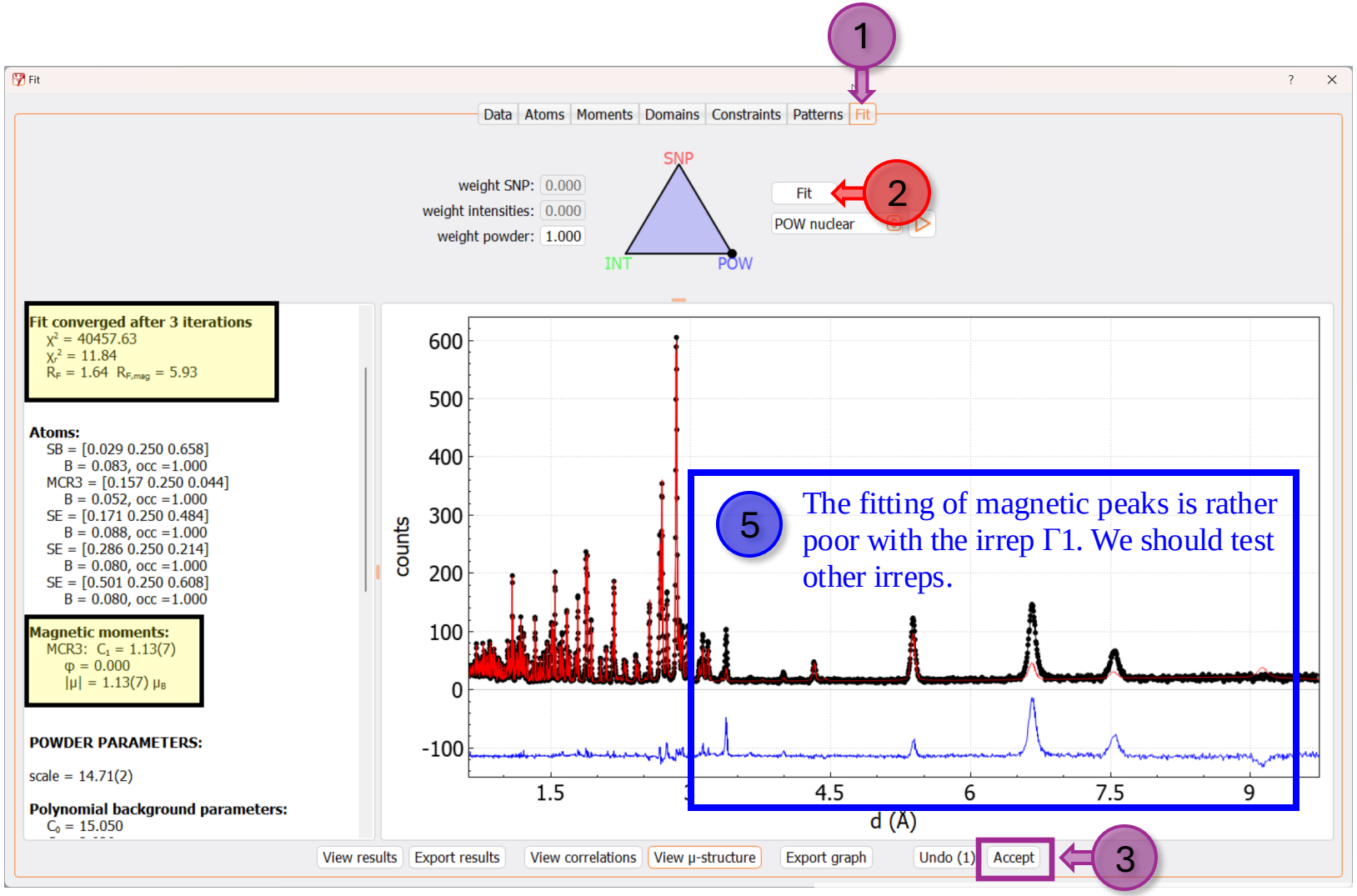
Step 6f: Fix all profile parameters



Fix all parameters except the ‘scale’ parameter.

Also, make sure both ‘nuclear’ and ‘magnetic’ phases are active and have the same Peak profiles, ***‘PV * B2B exponential (Tof1)’***

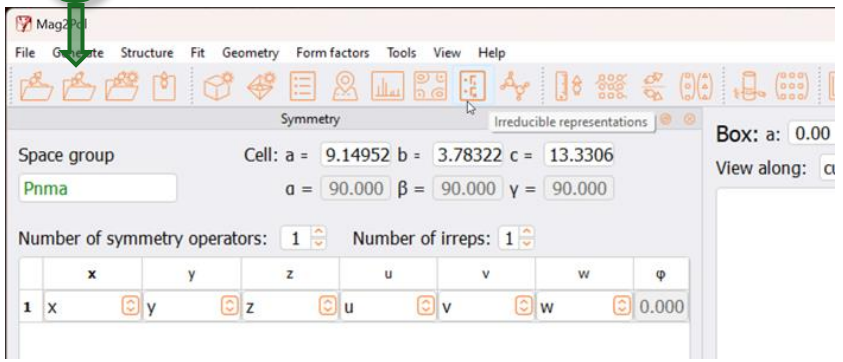
Step 6g: Perform fit



Once the convergence is reached, accept the fit results by clicking this button.

After accepting the refinement results on the 'Fit' window (keep this window open), go to the main window and click on the save button, which appends the previously saved project, **CrSbSe3_10K_Mag_G1.xml**

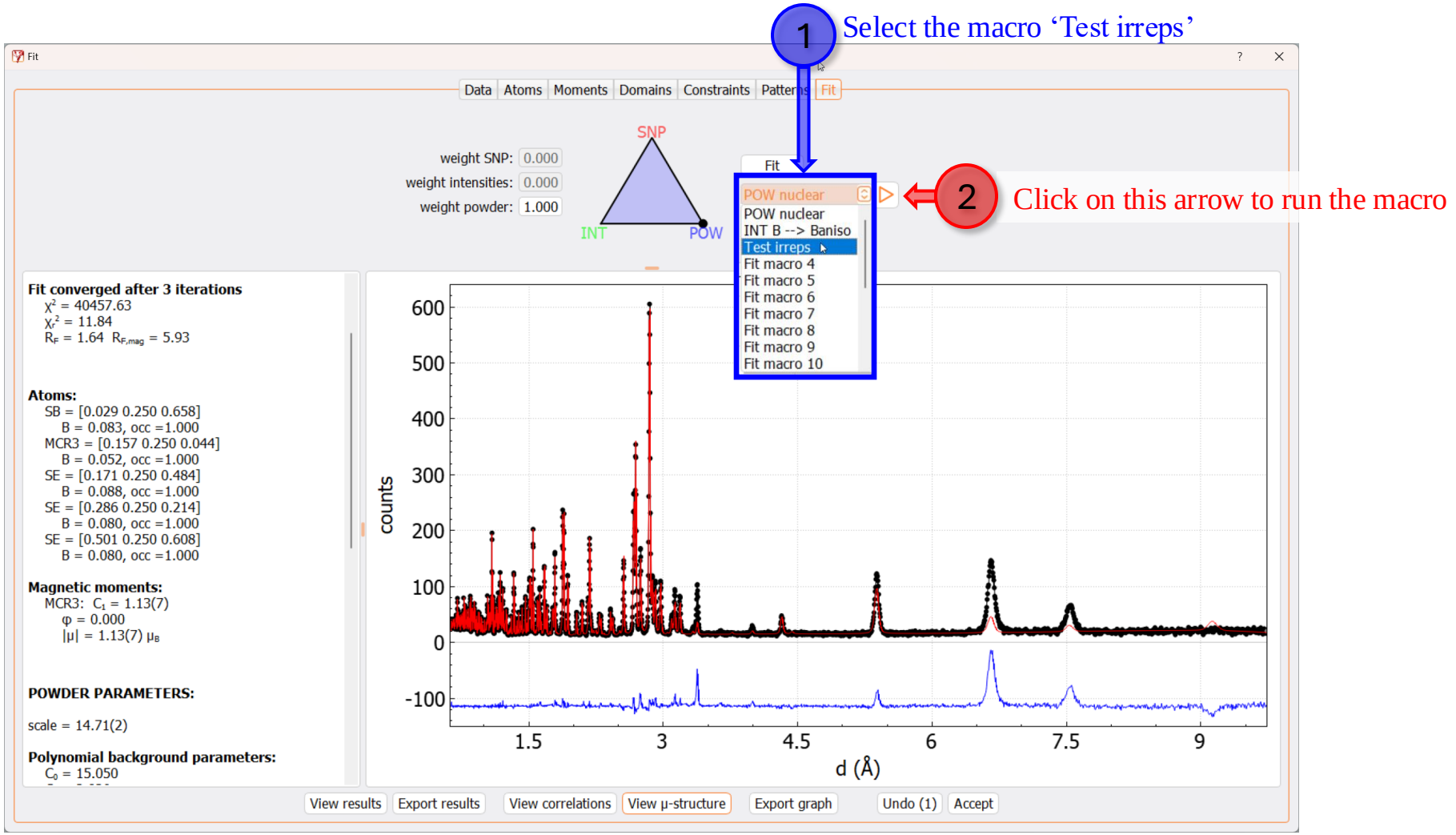
4



Test other irreps

Step 7a: Test all irreps by running built-in *Fit macros*

We can test each irrep one by one by selecting them in the main window of the Mag2Pol. But this can also be done much faster using the built-in fit macro ‘Test irreps’.



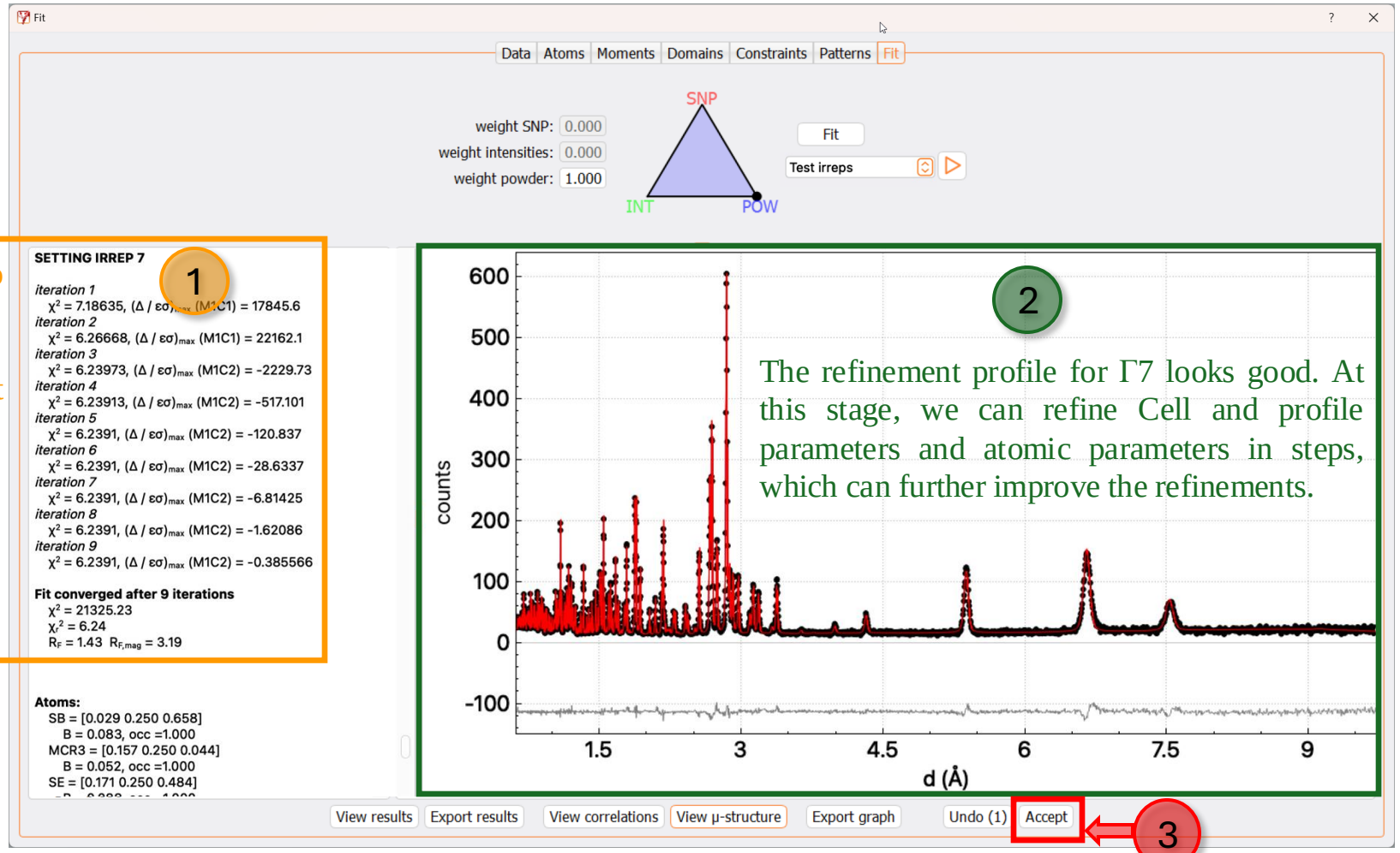
Step 7b: The macro ‘Test irrep’ ends with the best irrep model

The macro ‘Test irreps’ goes through all irreps and, at the end, reverts to the refinement of the best fitting irreps.

For the current example, the macro tested all 8 irreps.

In the end, it went back to the best model irrep, Γ7

For this model at this stage,
χ^2 = 6.24, $R_{F,\,mag}$ = 3.19



Important note:

The gamma notation between different programs may not match, so it is always a good idea to report the unambiguous, ‘basis vectors’.

For example, Γ3 in the SARAh program is equivalent to Γ7 on Mag2Pol

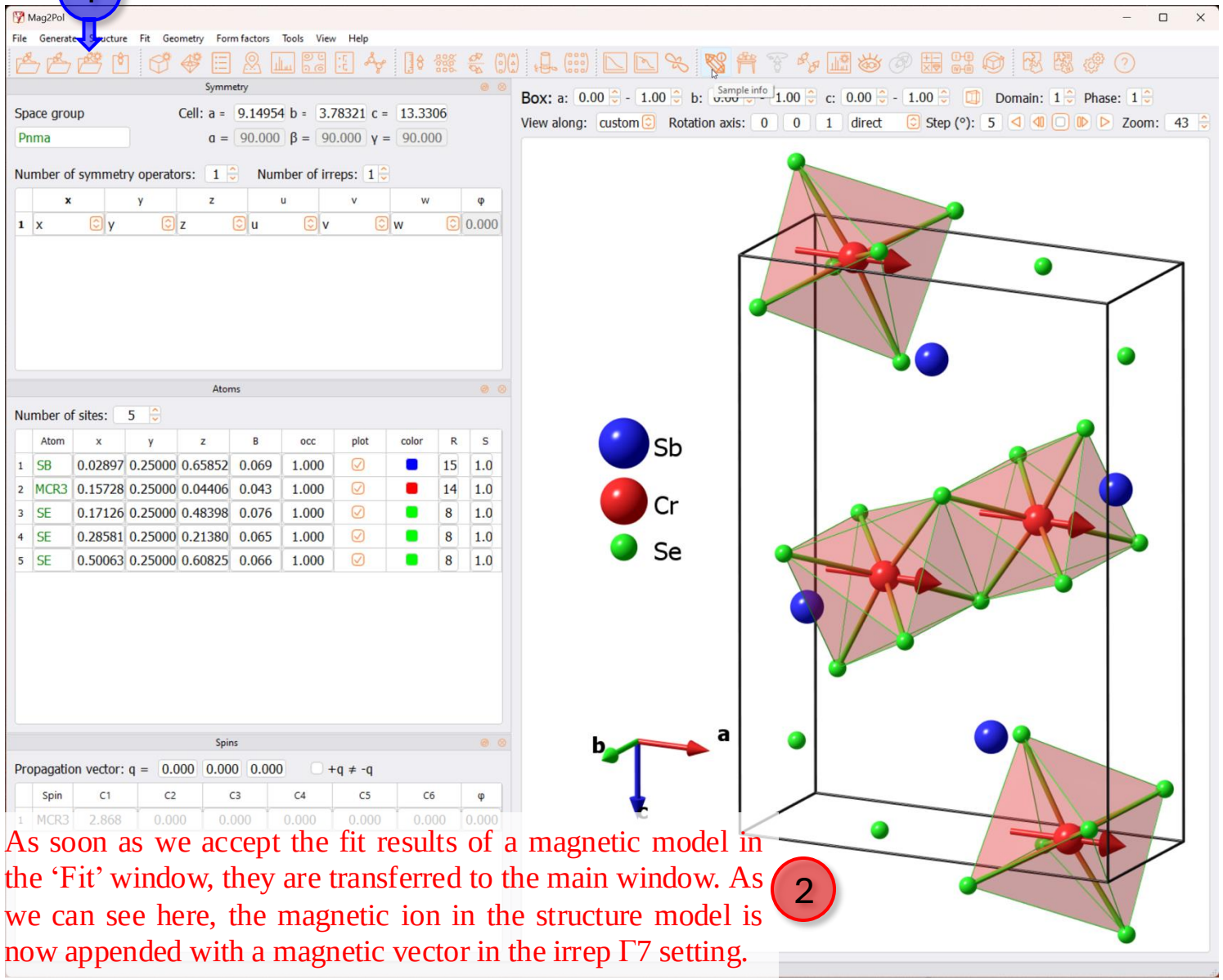
Once the final refinement of the irrep model is satisfactory, accept and transfer the fit results to the main window by clicking on the ‘Accept’ button.

For the irrep Γ7, at the final stage we get; χ^2 = 5.04, $R_{F,\,mag}$ = 2.86

Step 7c: Inspect the structure model and save the project

The last accepted fit results were for the irrep Γ7, so this project will be saved as, **CrSbSe3_10K_Mag_G7.xml**

1



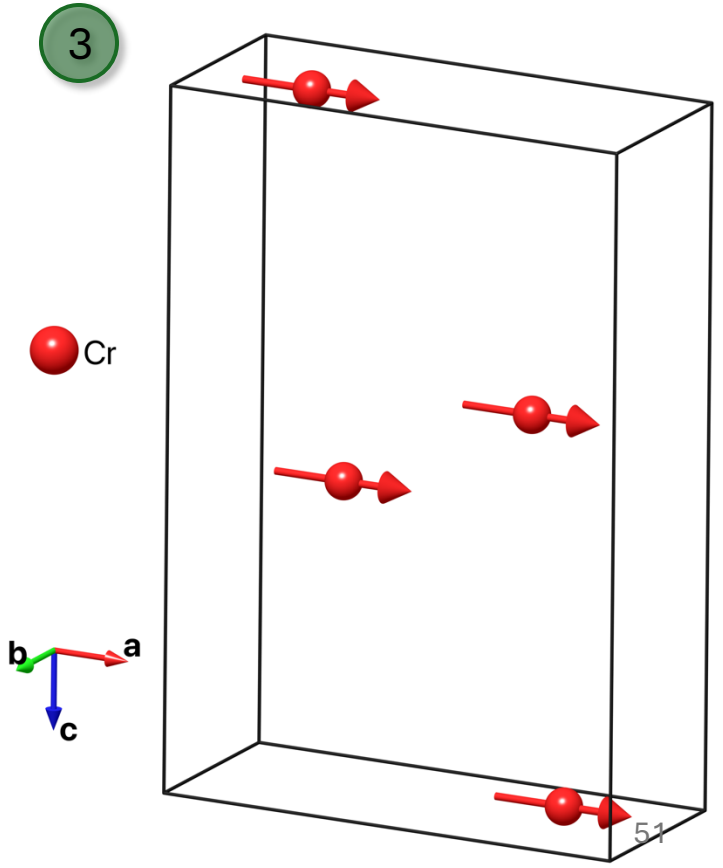
2

As soon as we accept the fit results of a magnetic model in the 'Fit' window, they are transferred to the main window. As we can see here, the magnetic ion in the structure model is now appended with a magnetic vector in the irrep Γ7 setting.

It is possible to produce high-quality graphics (*.png) from within the Mag2Pol. Checkout the Mag2Pol manual for more details.

Here is another representation of the magnetic structure model in Γ7 setting, produced using Mag2Pol.

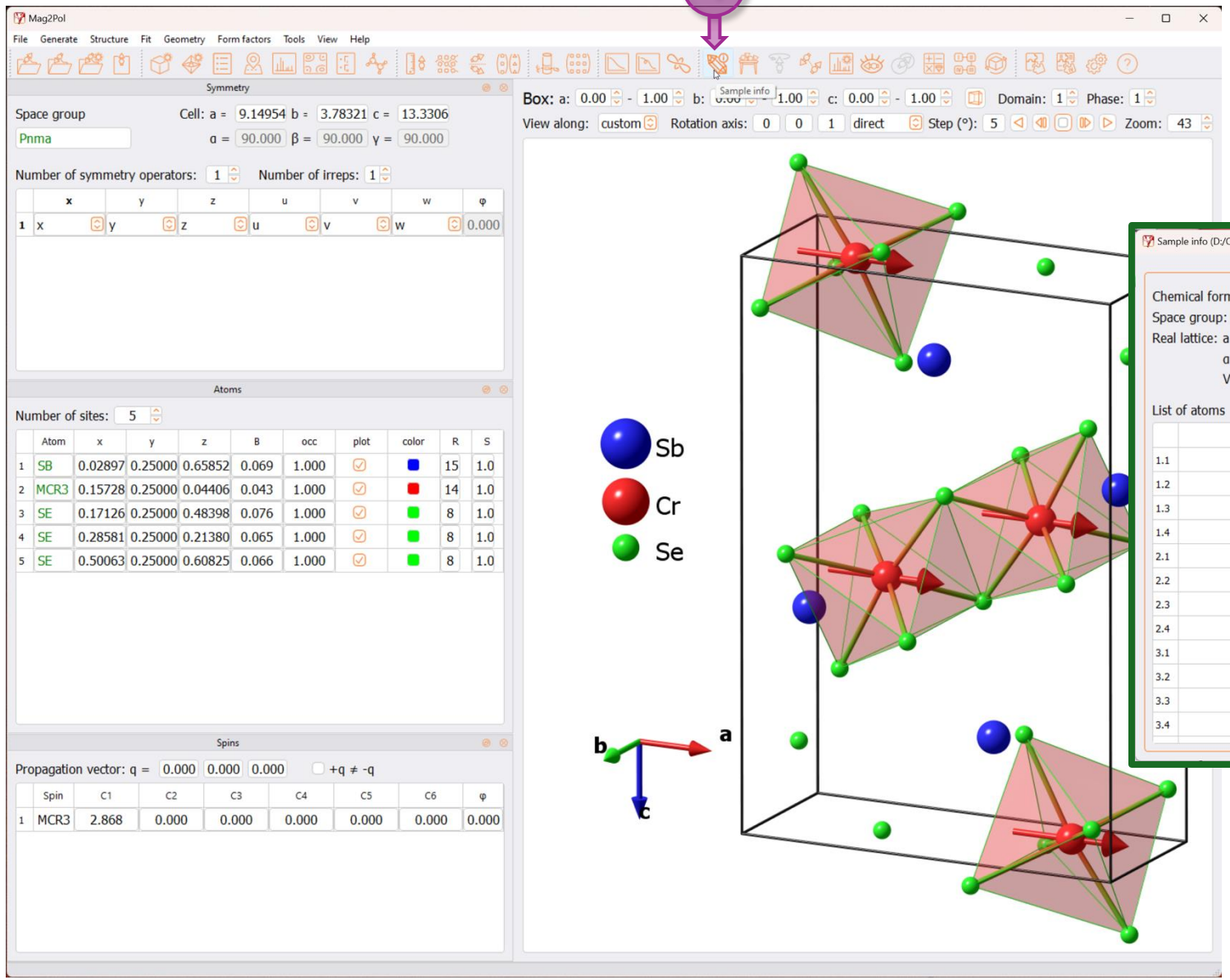
3



Step 7d: Inspect full refinement results

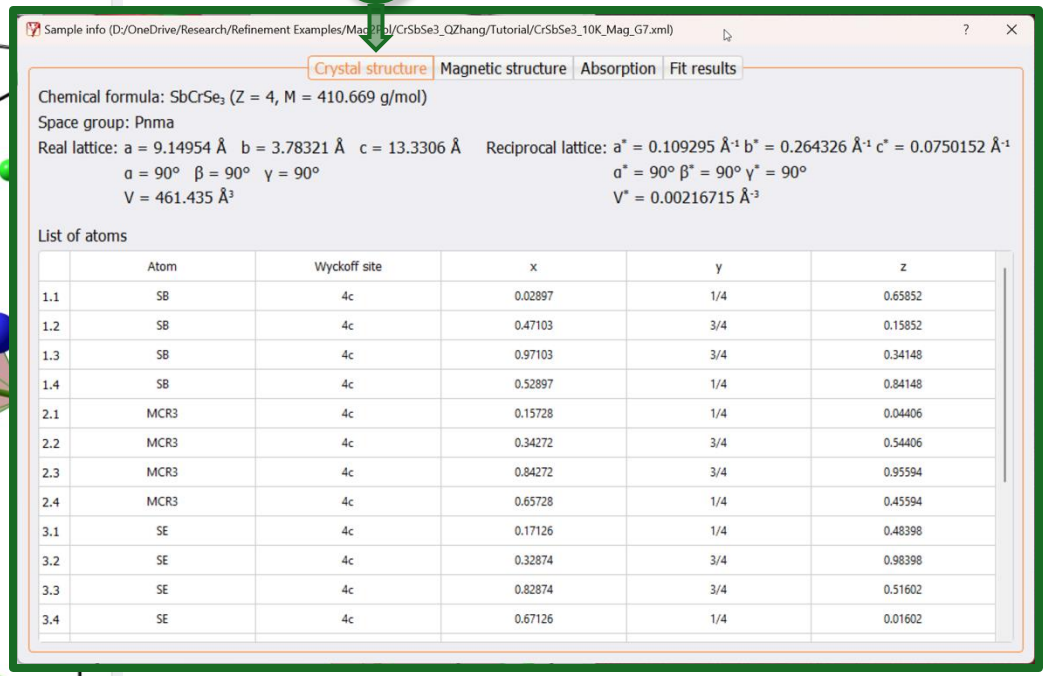
Click this icon to open ‘Sample info’ window, which contains detailed information of refined Crystal structure, Magnetic structure, and Fit results. Also indicates the path and name of the current project file.

1

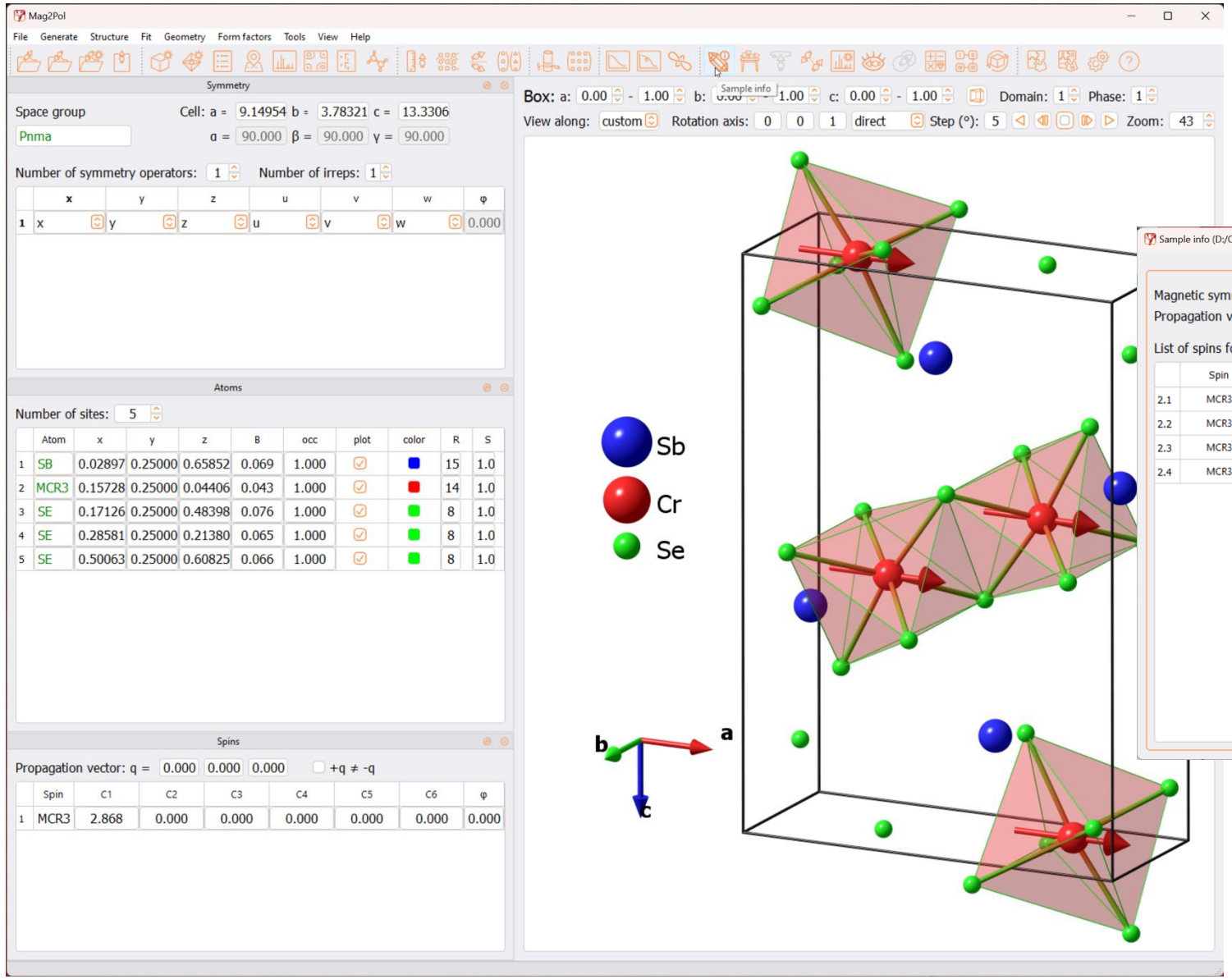


‘Sample info’ window, with the ‘Crystal Structure’ tab active.

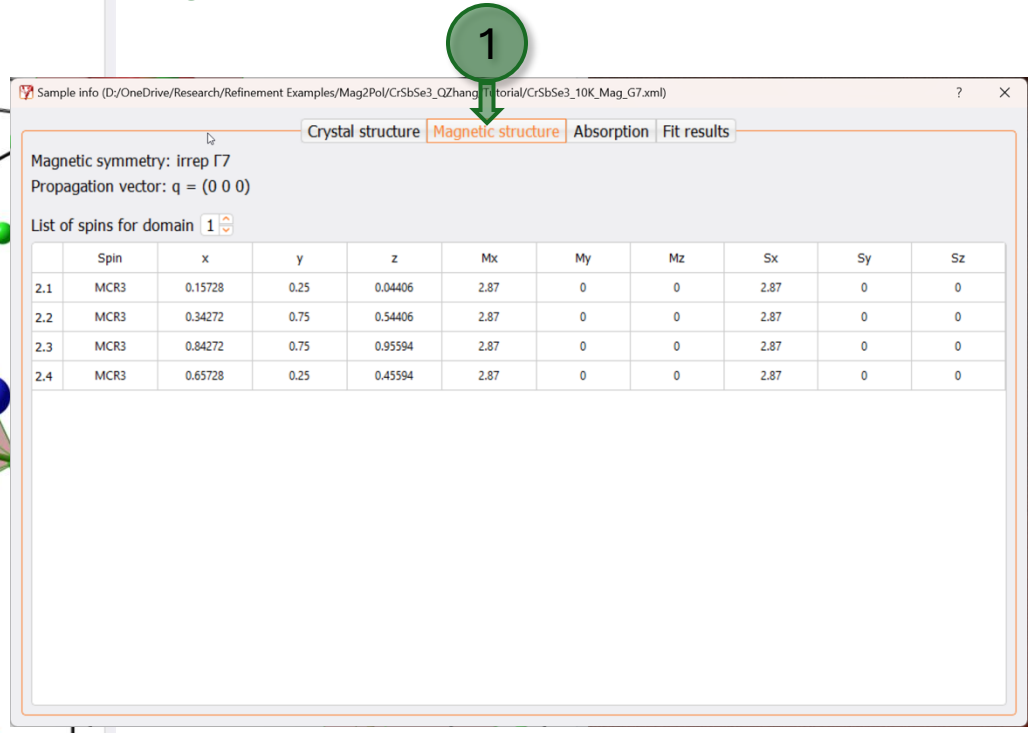
2



Step 7d: Inspect full refinement results

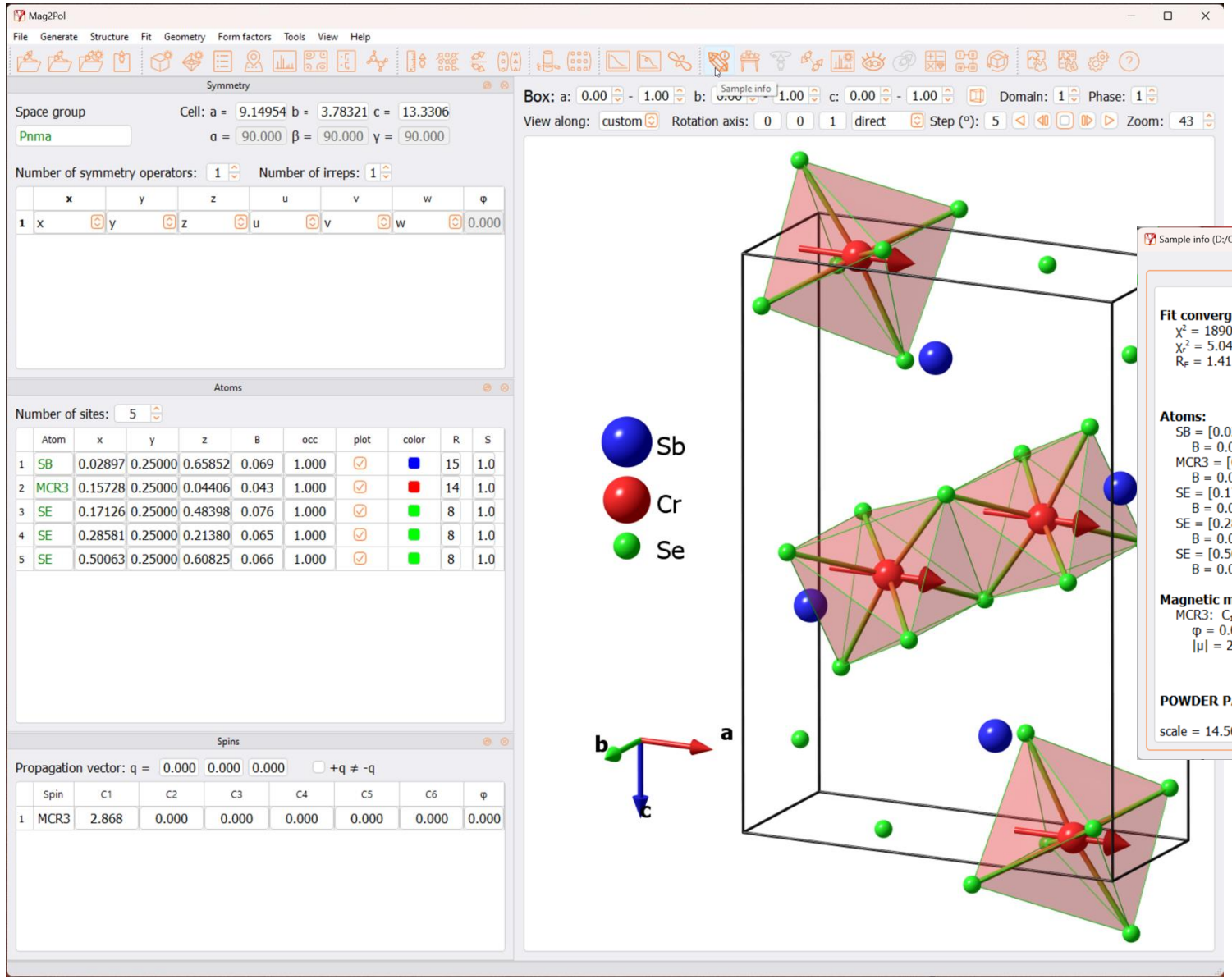


‘Magnetic structure’ tab information of magnetic model used in the last refinements.



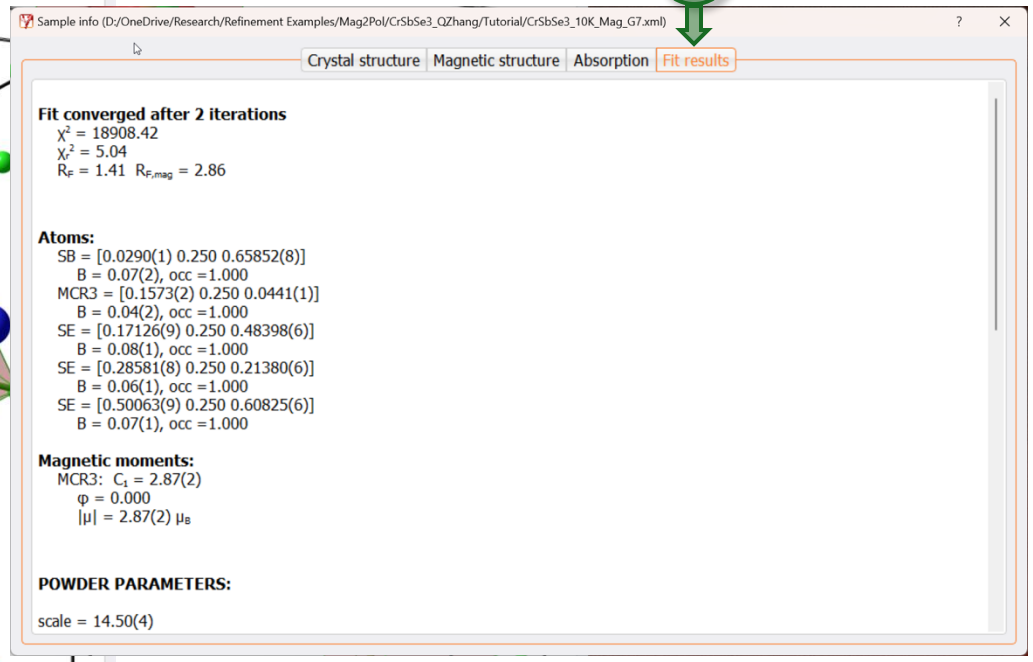
	Spin	x	y	z	Mx	My	Mz	Sx	Sy	Sz
2.1	MCR3	0.15728	0.25	0.04406	2.87	0	0	2.87	0	0
2.2	MCR3	0.34272	0.75	0.54406	2.87	0	0	2.87	0	0
2.3	MCR3	0.84272	0.75	0.95594	2.87	0	0	2.87	0	0
2.4	MCR3	0.65728	0.25	0.45594	2.87	0	0	2.87	0	0

Step 7d: Inspect full refinement results

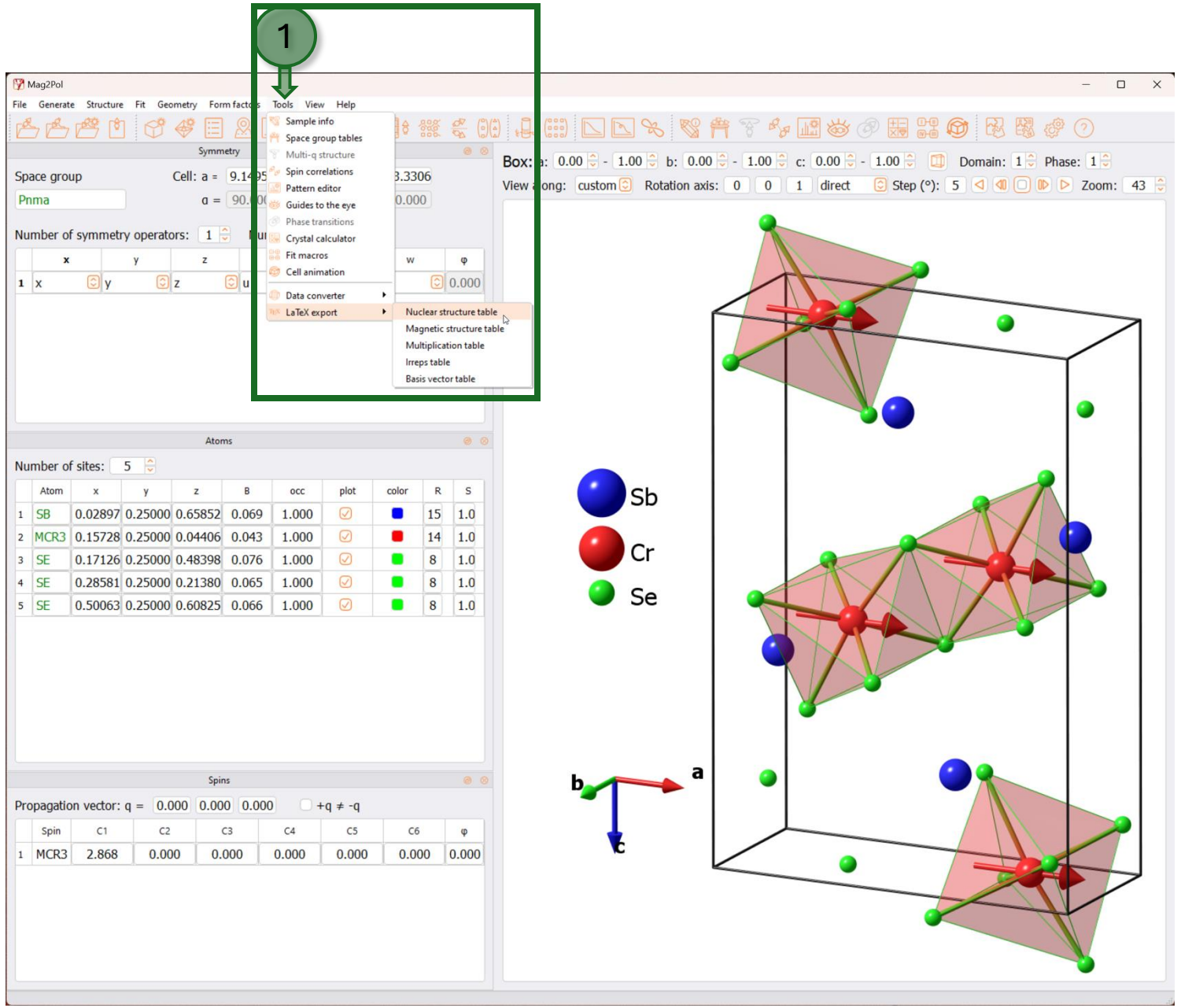


The 'Fit results' tab contains detailed refinement results of last refinements.

1



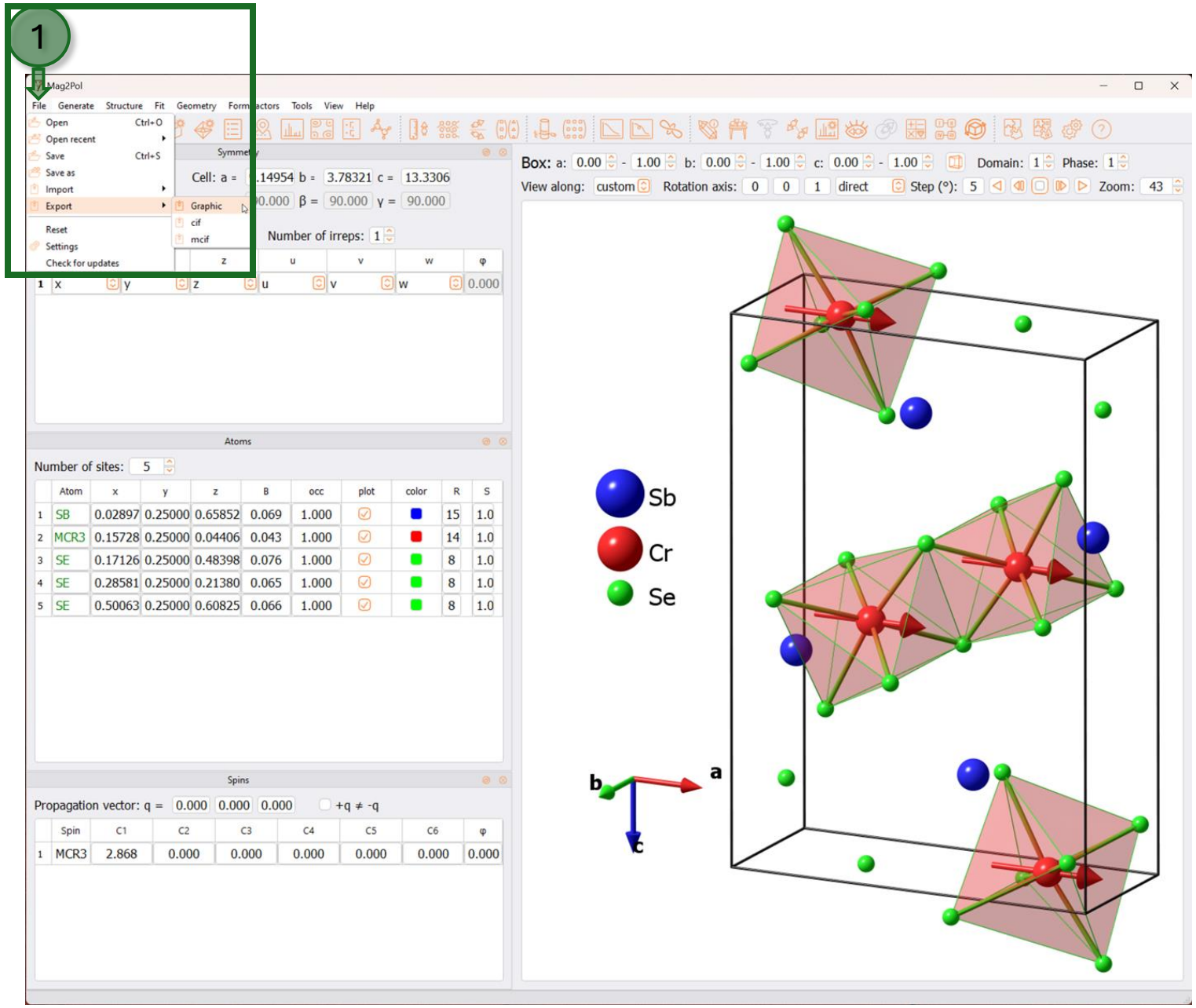
Step 7e: Export refinement results as latex tables



A variety of fit results can be copied directly in Latex format by following the menu tree:

Tools→LaTeX export

Step 7f: Export the Graphic and structure models



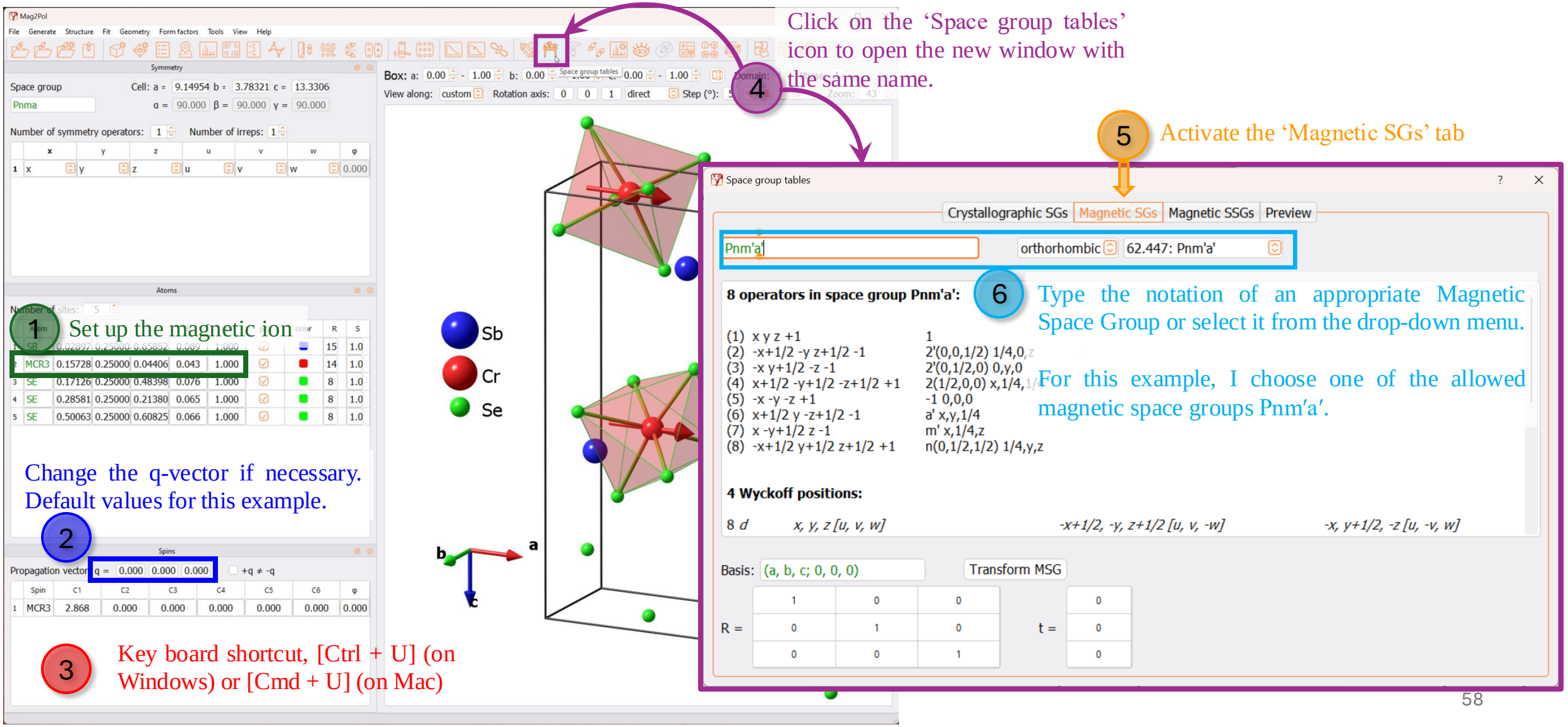
The structure model can be saved as *.png (Graphic) or *.cif or *.mcif, by following the menu tree:

File→Export

Refining magnetic structure using Shubnikov groups (Magnetic space groups) instead of Irreducible representations.

Step 8a: Set up a Magnetic Space Group

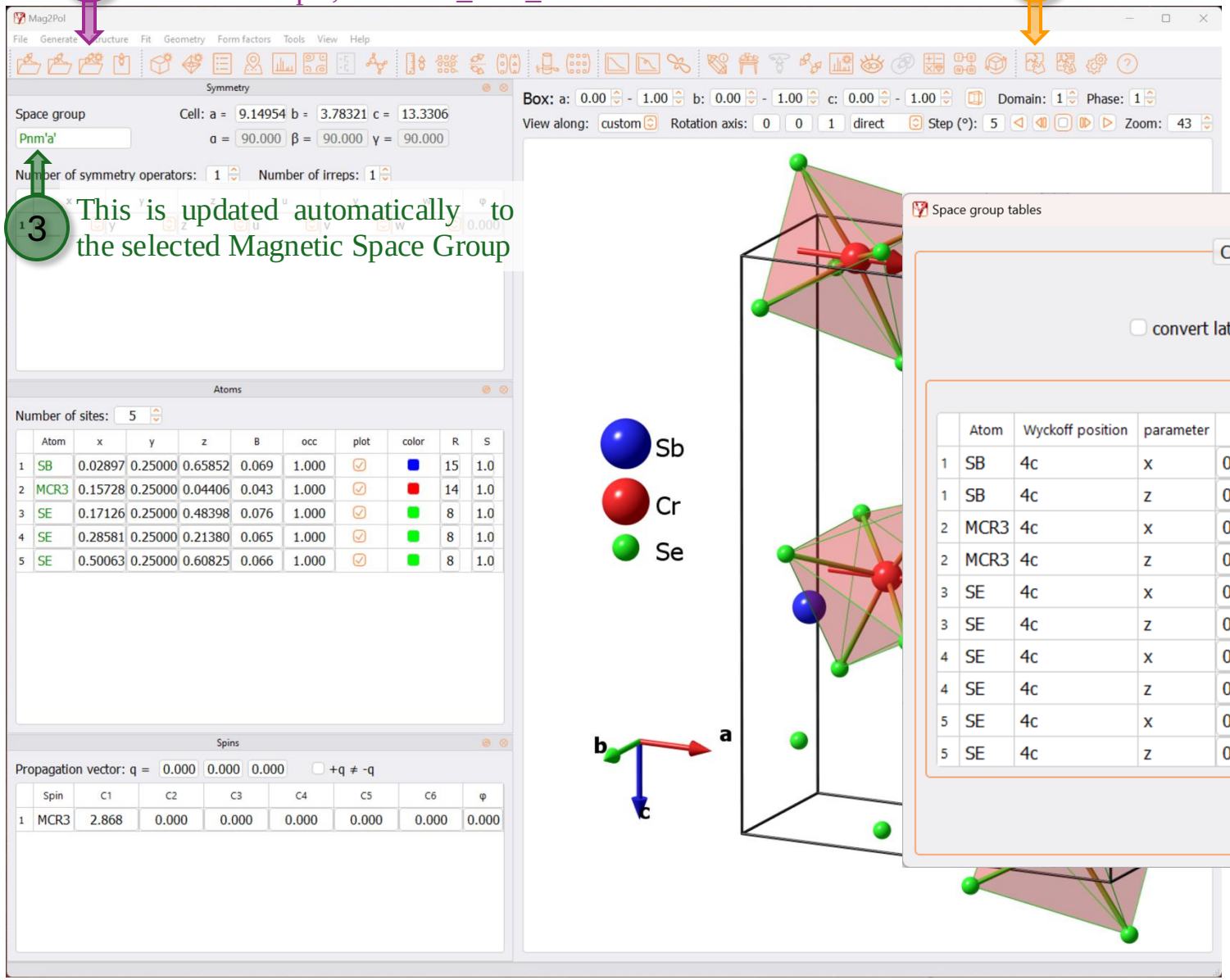
One way to start this procedure is to prepare a *.mcif file on the Bilbao crystallographic server, download it, and load that *.mcif directly in Mag2Pol. The alternate starting point for this procedure would be to use the final version of the Mag2Pol project with lattice-only contribution. For this example, we will start with the saved project with lattice-only contribution, CrSbSe3_10K_Lattice.xml



Step 8b: Transfer the Magnetic Space Group for the refinement

4 Save the project with an appropriate name.
For this example, CrSbSe3_10K_Pnm'a'.xml

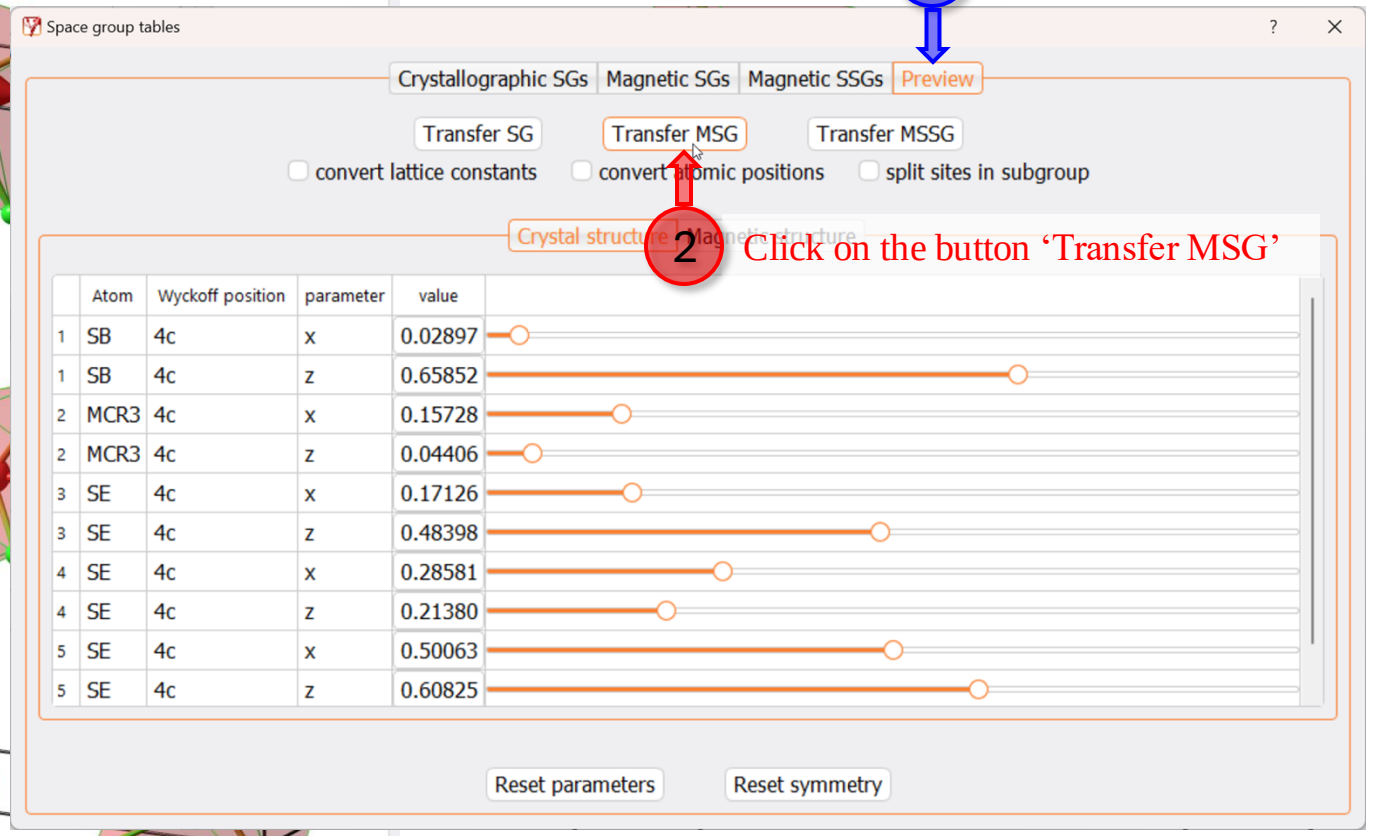
5 Click this icon to open the 'Fit' window.



3 This is updated automatically to the selected Magnetic Space Group

Switch to the 'Preview' tab

1



2 Click on the button 'Transfer MSG'

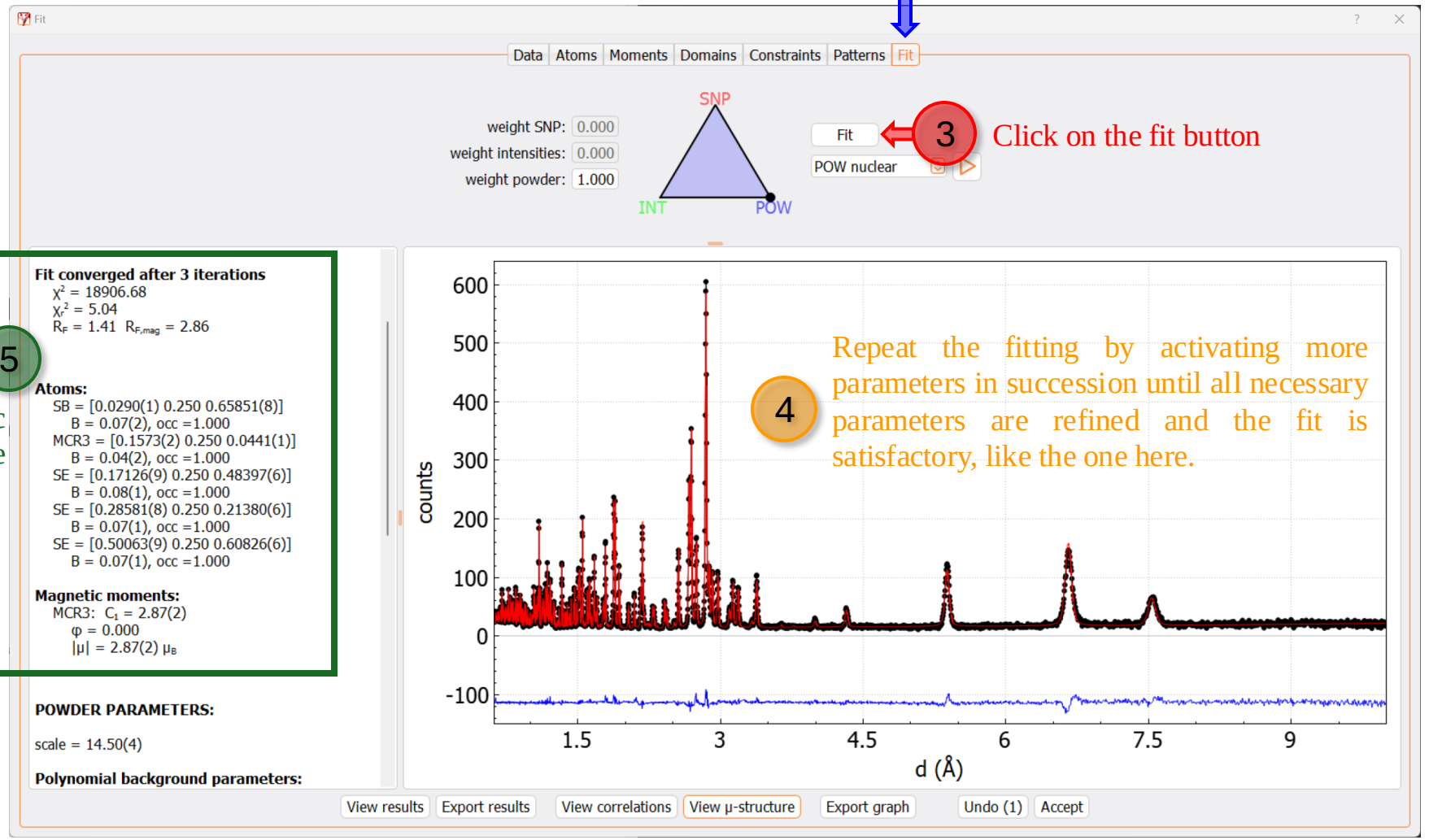
Step 8c: Perform the refinement using the chosen Magnetic Space Group

1

Fix all parameters except magnetic moments and scale parameter.

Also, make sure both 'nuclear' and 'magnetic' phases are active and have the same Peak profiles, '***PV * B2B exponential (Tof1)***'

2



5

The refinement results using the Magnetic Space Group Pnm′a′ are identical to those obtained using irrep Γ7.

If you have any questions:

Regarding the experiments and data collected at the instrument POWGEN, SNS

Contact:

Qiang Zhang
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Regarding Mag2Pol and need to report bugs.

Contact:

Navid Qureshi
Institute Laue Langevin,
Grenoble, France
Email: qureshi@ill.fr

Regarding this tutorial

Contact:

Naveen Kumar Chogondahalli Muniraju
Institute of Nuclear Physics, PAN
Krakow, Poland
Email: naveen.chogondahalli@ifj.edu.pl

Good luck with your next refinement project!