

# Example 13.7: Pyrite FeS<sub>2</sub>

Last update 29.6.2023

Data	Topic	Level
Electron diffraction	Complete procedure from data reduction to structure refinement using the dynamical approach	Intermediate

## Input data

### Data

Electron diffraction data were measured on a transmission electron microscope Tecnai 20 with a precession device DigiStar. Accelerating voltage 200 kV (wavelength = 0.0251 Å).

### Input files

directory “dp-100” with the set of experimental data (diffraction patterns)

pyrite.pts: input file for PETS2

pyrite\_reference.cif\_pets: this file is provided so that the structure analysis pat can be performed without the data processing part and to warrant reproducibility of the solution and kinematical refinement.

pyrite\_reference\_dyn.cif\_pets: result of the data processing for dynamical refinement. This file is provided so that the dynamical approach can be used without the processing part, and to warrant the reproducibility of the refinement.

Files pyrite.tut\* used to warrant a reproducible refinement in Jana2020.

## References

For further information about the data processing in PETS2 see:

- L. Palatinus et al. Specifics of the data processing of precession electron diffraction tomography data and their implementation in the program PETS2.0. Acta Cryst. B 75: 512-522 (2019).

-P. Brazda et al Accurate lattice parameters from 3D electron diffraction data. I. Optical distortions. IUCrJ 9: 735-755 (2022).

-PETS2 manual [pets.fzu.cz](https://pets.fzu.cz)

For further information about the dynamical refinement and associated parameters:

- L. Palatinus et al. Structure refinement using precession electron diffraction tomography and dynamical diffraction: theory and implementation. Acta Cryst. A71: 235–244 (2015).

- L. Palatinus et al. Structure refinement using precession electron diffraction tomography and dynamical diffraction: tests on experimental data. Acta Crystallogr B71: 740–751 (2015)

## Highlights

None

# Instructions

## PART 1 - Data treatment in PETS2

### 1. Prepare (check) input file for PETS2

#### Notes

*File pyrite.pts is the input for PETS2. The file was prepared by the software used for the data collection. However, you can easily prepare such input file for your own data.*

You can open the file pyrite.pts in any plain-text editor (Notepad, Wordpad, Notepad++, Vim,...) to view its contents.

#### Notes

*The beginning of the file gives you the experimental conditions as comments (#). These are irrelevant to PETS2. The important part starts here:*

```
lambda 0.0251
Aperpixel 0.002207
phi 1.01
omega 0.0
bin 2
reflectionsize 20
noiseparameters 3.5 38

imagelist
dp-100\001.tif -50.00 0.00
dp-100\002.tif -49.00 0.00
dp-100\003.tif -48.99 0.00
...
Endimagelist

reconstruction
hk0 1 0 0 0 1 0 0 0 0
hk1 1 0 0 0 1 0 0 0 1
...
hhl 1 0 0 0 1 0 0 0 0
h+1h 1 1 0 0 0 1 0 1 0 0
h+2h 1 1 0 0 0 1 0 2 0 0
endreconstruction
```

**lambda:** relativistic wavelength of the incident electrons in Angstroms (100 kV = 0.0370 Å, 120 kV = 0.0335 Å, 200 kV = 0.0251 Å, 300 kV = 0.0197 Å)

**Aperpixel:** Scale of the images given as the size of one pixel in reciprocal angstroms

**phi:** Precession angle in degrees used during the data collection. Can be zero, of course.

**omega:** orientation of the tilt axis of the sample holder with respect to the positive horizontal axis of the image. Must be estimated either by calibration, or by visual inspection of the series of images. Should be close to the correct value within +/-20 deg. Can be refined later in PETS2.

**bin:** Binning of the images prior to any treatment. Saves time, makes nicer pictures, and seems to make the whole analysis somewhat more robust. bin 2 is a good choice from our experience for most data sets. Important: All parameters in the input file that are given in pixels refer to the original, unbinned images. This concerns Aperpixel, center, reflectionsize, mask.

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**reflections***size*: Diameter of the spots on the images in pixels. Used for peak search and integration, should be large enough to encompass also the strong spots, but small enough to avoid overlaps of neighboring spots.

**noise***parameters*: two parameters for the determination of  $\sigma(I)$ . The first value is  $G\gamma$ , the second is  $\psi$ . The uncertainty on each pixel  $\gamma$  is then calculated as:

$$\sigma^2(p) = G\gamma p + \psi$$

For more information see Waterman, D. & Evans, G. (2010). *J. Appl. Cryst.* 43, 1356-1371.

**image***list* – **endimage***list*: Multiline keyword. Each line contains one image. The format is: name, alpha, beta. Name is the file name of the image (including possible relative or absolute path), alpha is the tilt angle, and beta is the possible second tilt value of the double tilt holder.

**Reconstruction** – **endreconstruction**: Multiline keyword to generate reciprocal space sections. Each line contains a specification of one layer in the reciprocal space that should be reconstructed by PETS2. The format is: filename, base vector 1, base vector 2, origin vector, where each vector is given by three reciprocal-space coordinates. The first two vectors define the orientation of the plane, and the third vector defines the origin of the plane. The standard set is  $hk0$ ,  $hk1$ ,  $hk2$ ,  $hk3$ ,  $h0l$ ,  $h1l$ ,  $h2l$ ,  $h3l$ ,  $0kl$ ,  $1kl$ ,  $2kl$ ,  $3kl$ . You can determine a specific set of sections later in the graphical interface or directly in the input file using the keywords “reconstruction” and “endreconstruction”.

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[Close pyrite.pts](#)

## 2. Run PETS2 and prepare files for indexation

Start PETS2 by double clicking the executable

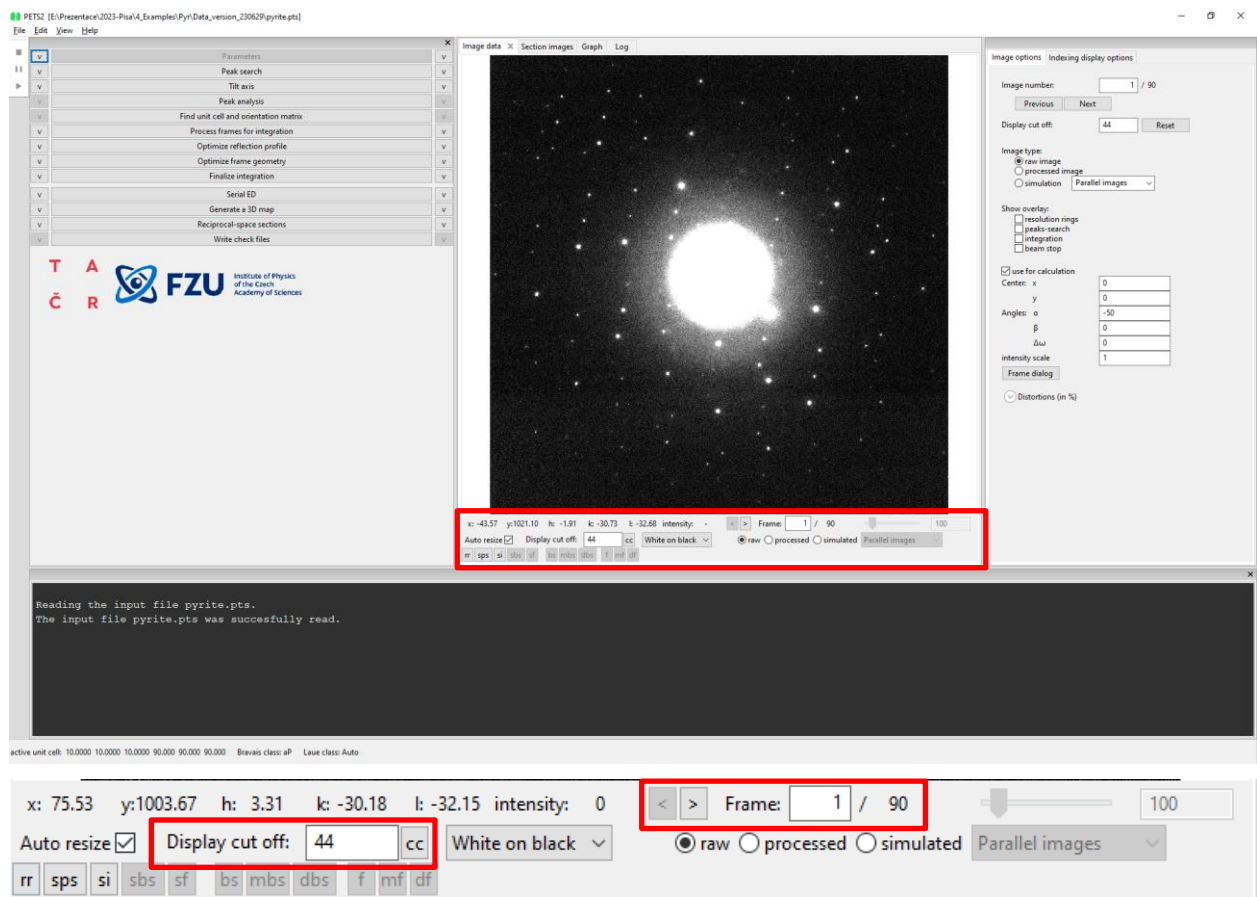
Go to File->Open in the upper menu

Open the file [pyrite.pts](#)

### Notes

In the Main panel (Image data tab) you can view individual diffraction frames of pyrite dataset stored in the directory “dp-100” using the arrows at the bottom of the panel. The contrast of the images can be adjusted by changing the value in the Cutoff-box (red on the picture). The parameters of the input files are visible in the console and in the menu of the tabs “Parameters” and “Peak search”. The arrows to the left of “Picture” (at the bottom controls of the Image data panel in blue) and the value next to it control the image that is shown. The radio buttons “raw” and “processed” show you the raw diffraction pattern as recorded and the processed frame (some background correction etc.), respectively.

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## Notes

Data reduction steps are run by clicking the respective action button. The action options can be modified in the panel which opens after clicking the small arrow next to the action button. The “Parameters” action button (first in the list of action buttons) is gray because there is no associated action. Note that you can adapt the size of the different window components (e.g. make the plot window more narrow or wider) and decouple the action bar or the console from the main window.

## 3. Setting parameters

Click on the arrows (v) next to “Parameters” and “Peak search”

to display the panels with parameters

Set the value max d\* for integration =  $2.2 \text{ \AA}^{-1}$  and max d\* for peak search =  $2.2 \text{ \AA}^{-1}$  (1.4 by default).

Set the value detector saturation limit to 13000 (linearity limit for the CCD camera).

Leave other settings unchanged.

Parameters	
geometry	precession
reflection diameter (binned, px)	10
calibration constant (rec. Å/pixel, binned)	0.004414
max d* for integration (rec. Å)	2.2
max d* for peak search (rec. Å)	2.2
min d* (rec. Å)	0.05
detector noise parameters: $G\gamma$	3.5
$\psi$	38
detector saturation limit	13000
beam stop	<input checked="" type="radio"/> No <input type="radio"/> Yes
	<input type="button" value="Browse"/>
bad pixels	<input type="button" value="Browse"/> <input type="button" value="Remove bad pixels list"/>

Peak search	
<input checked="" type="checkbox"/> Always accept center	
Center determination:	<input type="radio"/> use direct beam <input checked="" type="radio"/> use Friedel pairs <input type="radio"/> use saved centers
<input checked="" type="checkbox"/> Automatic initial direct beam coordinates	
x coordinate (pixels)	512
y coordinate (pixels)	512
I/sigma	10

#### 4. Peak search

##### Notes

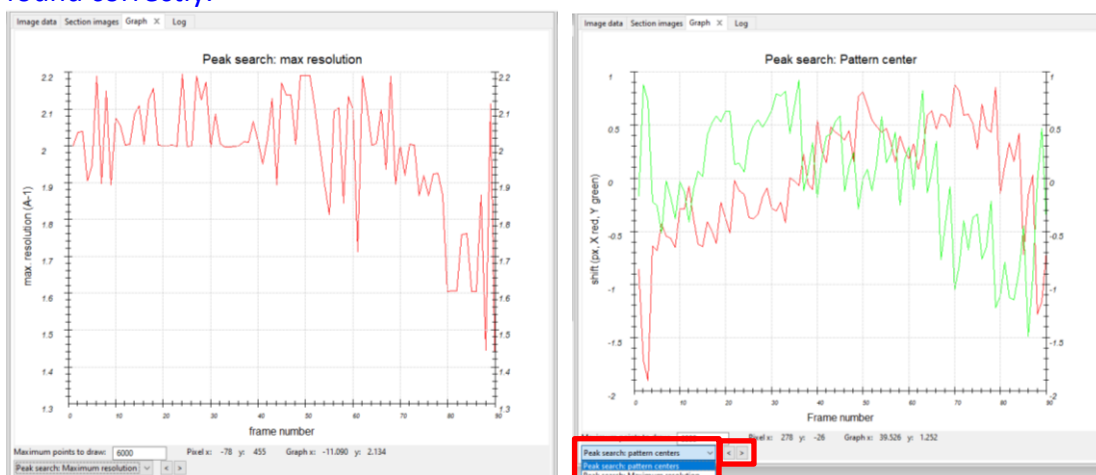
In Peak search parameters leave the default setting – check Always accept center, use Friedel pairs, Automatic initial direct beam coordinates checked, I/sigma 10.

Click the action button “Peak search” to start the procedure. The progress of the peak search can be followed in the Image data panel.

##### Notes

In the Main panel>Image data tab, diffraction images are sequentially displayed with circles at points found by PETS2 as significant peaks (above I/sigma specified in the Peak search parameters). Crosses appear in the if the peak's Friedel pair was detected

Click on Graph tab in Main panel. Inspect the “Peak search: Maximum resolution”. All frames have sufficient resolution. Then click on the drop-down menu to choose “Peak search: pattern centers” or use the arrows to the right to browse through the graphs. All the centers were found correctly.



## Notes

*In the console, for each frame the estimated position of the primary beam, the number of Friedel pairs and the number of significant peaks is given. The process ends with the following message in the console:*

```
Minimum, average and maximum position of the primary beam: 513.28 515.18
516.06 | 489.75 491.24 492.17
Finished reading 4766 peaks from the file pyrite_petsdata/pyrite.rpl.
```

## 5. Tilt axis

### Notes

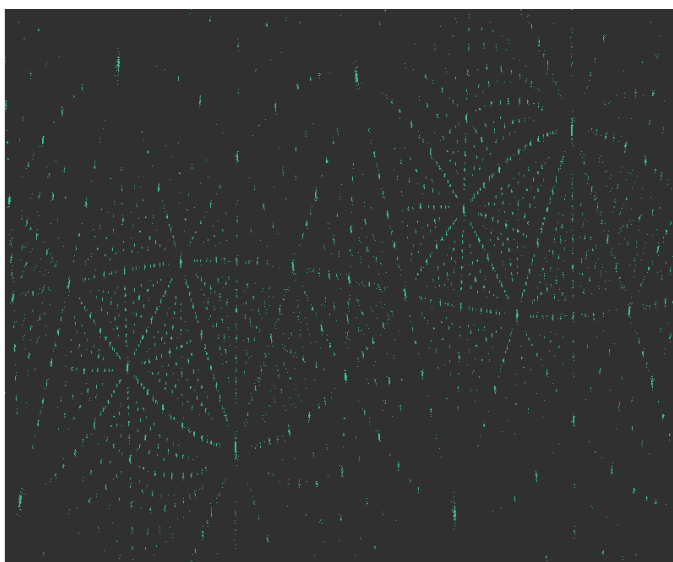
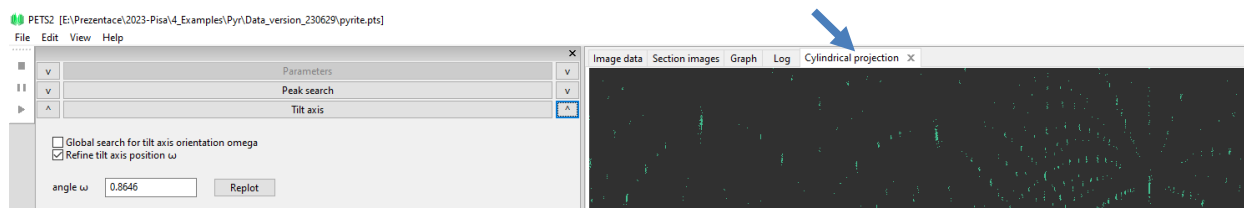
*The next step after peak hunting is the refinement of the angle between the horizontal axis (x axis of the diffraction image) and the projection of the rotation axis. This angle depends on the experimental setup and needs to be refined for each data set.*

[Click the action button “Tilt axis” to start the procedure.](#)

[Switch to the tab “Cylindric projection” in the main panel to see the progress of the refinement](#)

### Notes

*In the main panel “Cylindric projection”, a cylindrical projection of the difference space of the extracted peak positions is displayed. A correct azimuthal angle refinement results in the image containing sharp peaks aligned on sinusoidal curves. This step provides a first estimation of data quality. At the end of the refinement the omega angle is 0.8646 degree for a score of 6.48873.*

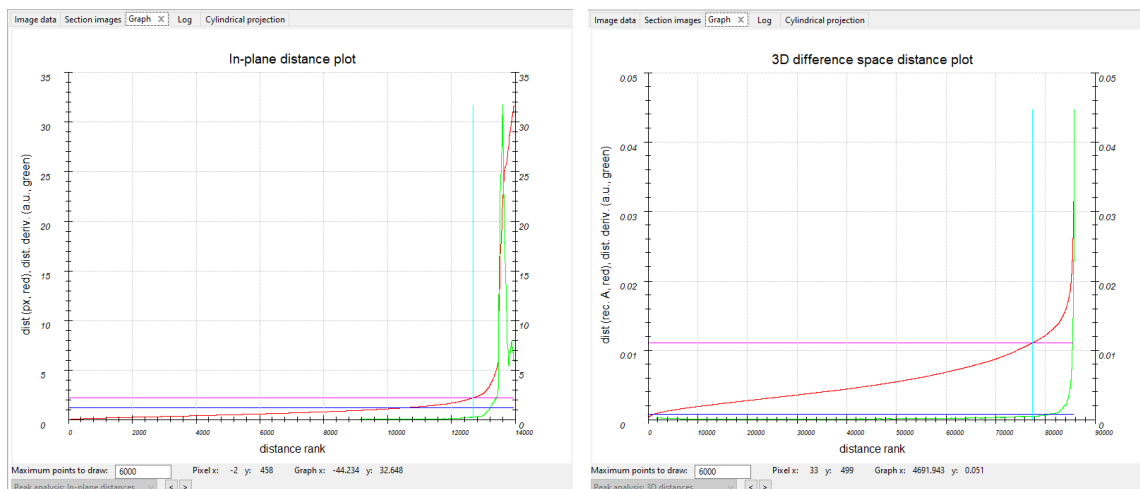


## 6. Peak analysis

[Click the action button “Peak analysis”.](#)

### Notes

*PETS2 starts the analysis of the peak positions obtained in the peak hunting procedure. In the first step, distance distribution between peaks in the image plane is analyzed. A sorted plot of inter-peak distances (red curve) is displayed in the Graph tab with its derivative (green):*



## Notes

*For good quality data set, the red curve contains distinct steps, and, consequently, the green curve has distinct peaks. The steps are at the highest distance for pyrite because the lattice parameters are very small.*

[Click “Peak analysis \(continue\)”](#)

## Notes

*In the next step, an auto convolution of the diffraction pattern (difference space) is analyzed, and the groups of peaks in the auto convolution (clusters) are replaced by the cluster centers. Again, a distance distribution is displayed (right screenshot), and clear-cut steps on the curve indicate a well-defined lattice.*

[Click “Peak analysis \(continue\)”](#)

## Notes

Now the first step is finished and PETS2 prepared the files needed for unit cell indexing. At the end of this step, you should get the following message in the console:

```
Threshold for considering two peaks the same: 0.0111 reciprocal angstroms.
14750 peaks merged into 3160 clusters, 1824 with more than one peak.
Peak analysis successfully finished.
```

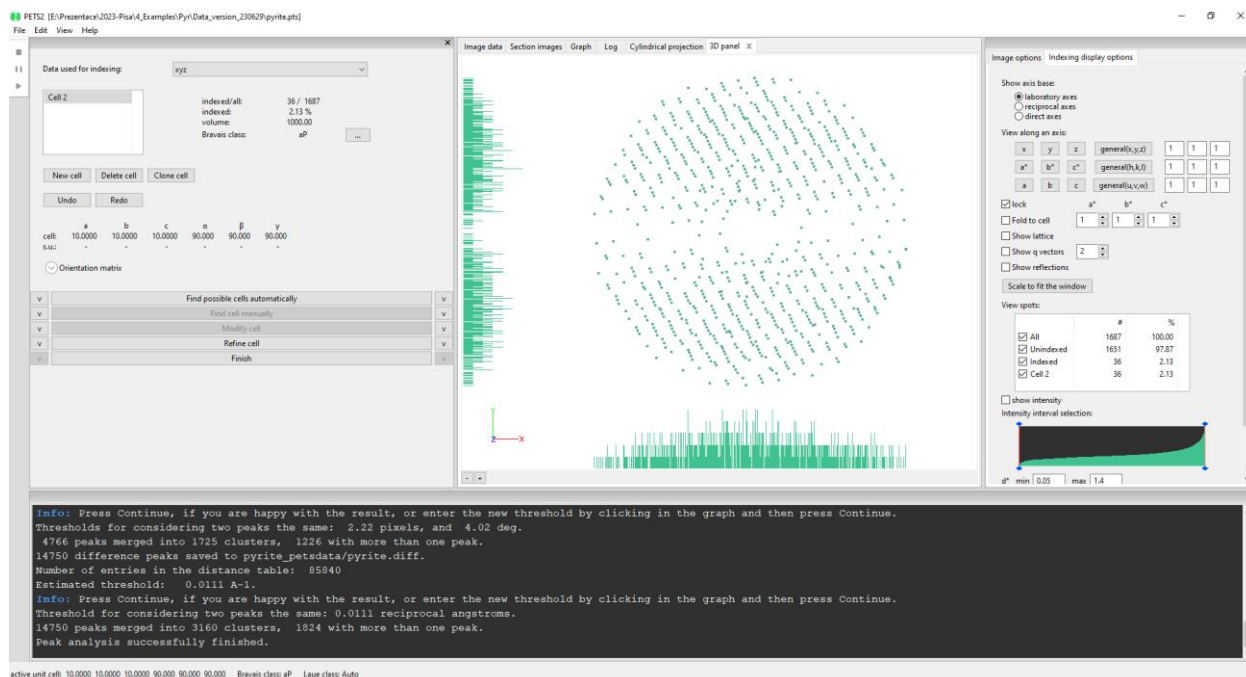
## 7. Find unit cell and orientation matrix

[Click “Find unit cell and orientation matrix”](#)

## Notes

New set of buttons with parameters opens as well as a new tab “3D panel” that displays the data used for indexing. The data can be enlarged by scrolling the wheel of your mouse and rotated by holding the left mouse button.

[In the drop-down list “Data used for indexing” select xyz \(should be set by default\).](#)



Open the menu below “Find possible cells automatically” by clicking on the arrow next to the action button.

## Notes

You can see the parameters used in the procedure as well as the possible unit cells found later by PETS2.

Check that the selected option is “from difference space” and keep the other parameters default.

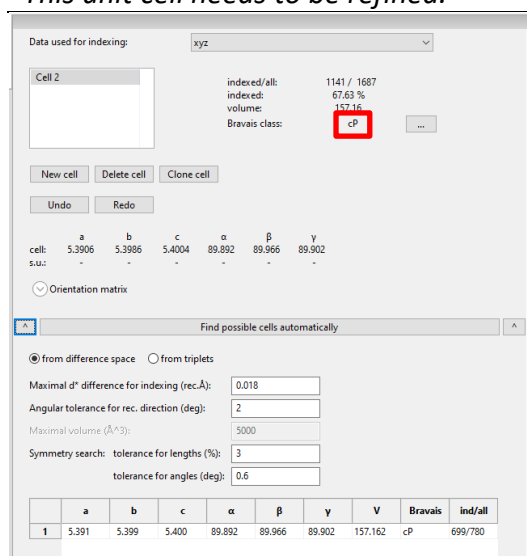
Run “Find possible cells automatically”

## Notes

Only one unit cell is found (see below) and the initial unit cell 10 10 10 90 90 90 is replaced by the new one  $a = 5.3906 \text{ \AA}$ ,  $b = 5.3986 \text{ \AA}$ ,  $c = 5.4004 \text{ \AA}$ ,  $\alpha = 89.892^\circ$ ,  $\beta = 89.966^\circ$ ,  $\gamma = 89.902^\circ$ .

PETS2 already suggests a Bravais class cubic primitive (cP) for the unit cell.

This unit cell needs to be refined.



Click on the arrow next to “Find possible cells automatically” to close the opts.

Click on the arrow next to “Refine cell” to access the options.



Leave all settings default and run the procedure by clicking on the “Refine cell” button.

#### Notes

Standard deviations will appear below the lattice parameters. The deviations from cubic cell are larger than three sigma. In the next step we will use calibrated distortion parameters to remove the optical distortions from the diffraction data.

Select the option “refine cell and distortions”,

In symmetry, keep “triclinic”

Select only “refine cell” and make sure that “refine distortions” is not activated now.

Click on the arrow next to “Distortions (in %)” to access the options.

Type in the values of the distortion coefficients in the boxes as indicated in the screenshot below.

Click “Refine Cell”.

Refine cell

☐ refine UB + cell  
☐ refine cell from d  
☒ refine cell and distortions

Maximal d\* difference for indexing[Inv(A)]: 0.018

☒ symmetry triclinic  
☐ fixed cell parameters  
10 10 10 90 90 90  
☐ fixed up to a scale factor

☒ refine cell ☐ refine distortions

☒ Distortions (in %)

☒ in % ☐ in pixels

Standard mode Expert mode

magnification: ☐ 0 Rerefine tilt axis  
barrel-pincushion: ☒ 0.1  
spiral: ☒ 0.33  
elliptical amplitude: ☒ 0.11 phase: ☒ 58  
radial SgPara: ☐ -0.07

#### Notes

The refined unit cell parameter is  $a = 5.4077(6) \text{ \AA}$ ,  $b = 5.4048(5) \text{ \AA}$ ,  $c = 5.4060(3) \text{ \AA}$ ,  $\alpha = 89.999(5)^\circ$ ,  $\beta = 89.973(10)^\circ$  and  $\gamma = 90.006(11)^\circ$ . Those parameters are just within three sigma compatible with a cubic cell.

	a	b	c	$\alpha$	$\beta$	$\gamma$
cell:	5.4077	5.4048	5.4060	89.999	89.973	90.006
s.u.:	0.0006	0.0002	0.0004	0.005	0.010	0.008

#### Notes

Because the data contained spiral distortion it is recommended to rerefine the tilt axis, because this distortion influences its position.

Click the button “Rerefine tilt axis”.

#### Notes

Optimized omega is now about  $0.15^\circ$  smaller. Finally, refine the cell again to fit the lattice into newly reconstructed reciprocal space (omega is influencing the reciprocal space reconstruction).

Click the button “Refine cell”.

#### Notes

The lattice parameters changed only marginally, but the orientation matrix changed.

Leave the indexing panel by clicking “Finish”

#### Notes

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The file `pyrite.cellist` is now created in folder `pyrite_petsdata`. This file contains the orientation matrix and the unit cell parameters.

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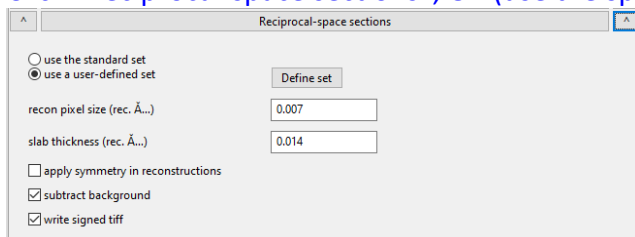
## 8. Create reciprocal-space sections

### Notes

PETS2 scans all images and reconstructs sections through reciprocal space at planes  $hk0$ ,  $hk1$ ,  $hk2$ ,  $h0l$ ,  $h1l$ ,  $h2l$ ,  $0kl$ ,  $1kl$ ,  $2kl$  (default set). These sections are used as an aid to determine the space group. Sections are saved in the directory `sections` as TIF-files. For pyrite, the lattice parameters are very close to be cubic and the complete analysis of the symmetry needs to include as well the sections of the type  $hhl$  ( $hhl$ ,  $(h+1)hl$  and  $(h+2)hl$ ). They were added to the standard set of layers directly in the input file to save some time but it is also possible to add them from the graphical interface.

In the parameters of the Reciprocal-space sections, you can change the size of pixels in the reconstruction images and the thickness of the slab that is projected into the section in  $\text{\AA}^{-1}$ . Too small numbers may result in “holey” reconstructions. We will proceed with the default values.

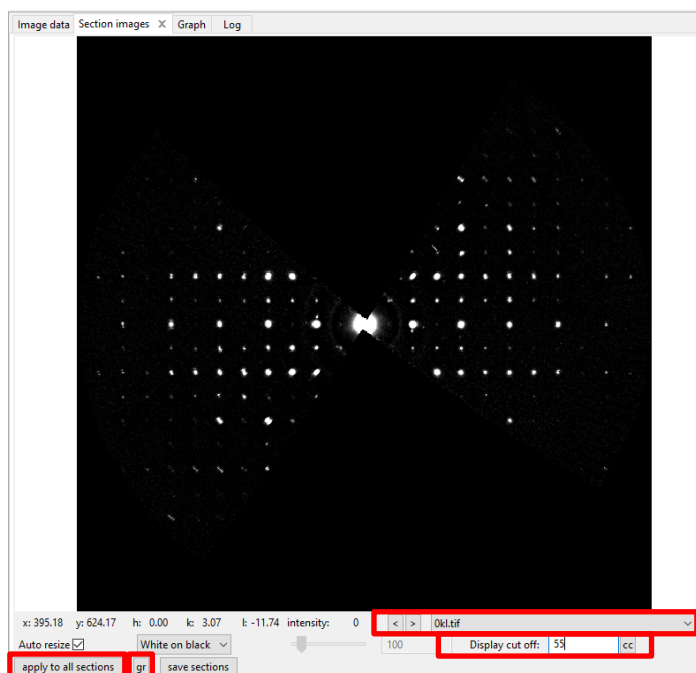
Click “Reciprocal-space sections”, OK (use the specific set)

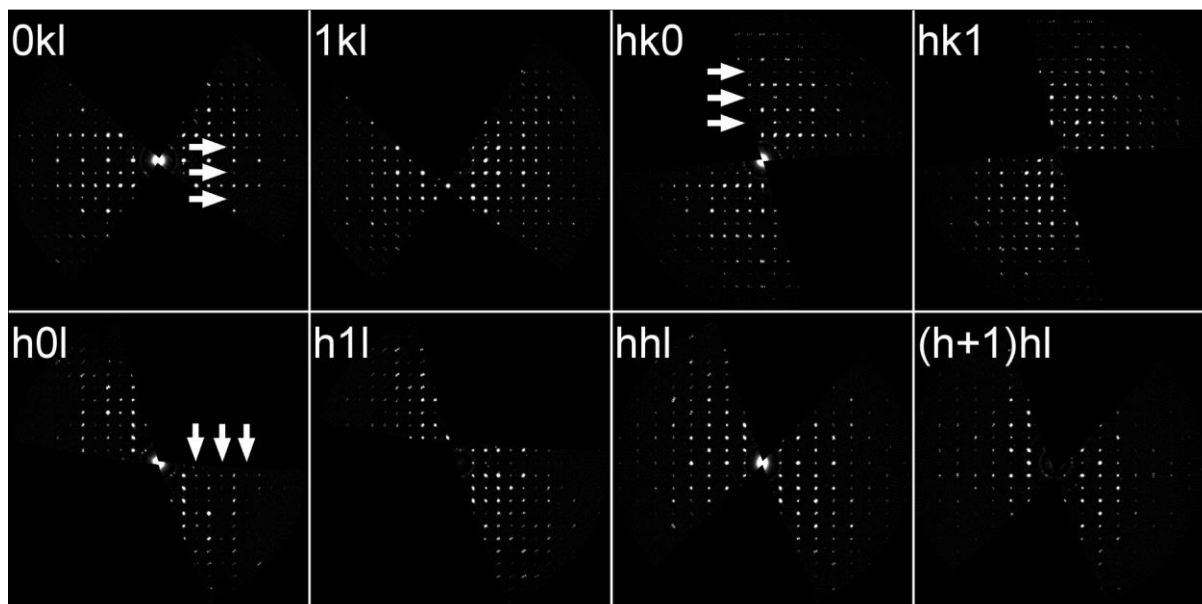


After the procedure is completed, go the Section images tab in Main panel, set “Display cut off” to 55 and click on “apply to all sections” button.

### Notes

You can visualize the sections in the tab “Section images” of the main panel and choose the section as shown on the picture below. Browse between the sections using arrows or drop-down menu. You can also click on the “gr” button, which will visualize the crystal lattice.





### Notes

From electron diffraction data, it is often difficult to select the correct space group. In particular, the presence or absence of glide planes and screw axes is hard to assess, knowing that kinematically absent reflections may have significant intensities due to dynamical diffraction effects. This example is a perfect illustration of this problem.

On the sections  $0kl$ ,  $h0l$  and  $hk0$ , you will see that the rows  $l = 2n+1$ ,  $k = 2n+1$  and  $h = 2n+1$ , respectively, present much lower intensities than the even rows (marked by white arrows). In the higher layers ( $1kl$ ,  $h1l$  and  $hk1$ ) these differences are no more visible. To detect the extinct reflections despite the dynamical effects, we compare the intensities in the layers 0 and 1. Further extinction condition is visible on the row  $00l$ :  $l = 2n$ . No further conditions are visible on  $hhl$  and  $(h+1)hl$ . If we assume that the weak reflections of the 0 layers are only due to dynamical effects, we can consider the conditions:  $l = 2n$  on  $0kl$ ,  $k = 2n$  on  $h0l$ ,  $h = 2n$  on  $hk0$  characteristic of glide planes  $c$ ,  $b$ ,  $a$  perpendicular to axes  $a$ ,  $b$  and  $c$ , respectively. The possible space groups are  $Pb3$  or  $Pb-3$ . If the space group is not obvious at this stage, it will be confirmed later during the structure solution steps.

## 9. Integrate intensities

### (a) Process frames for integration

#### Notes

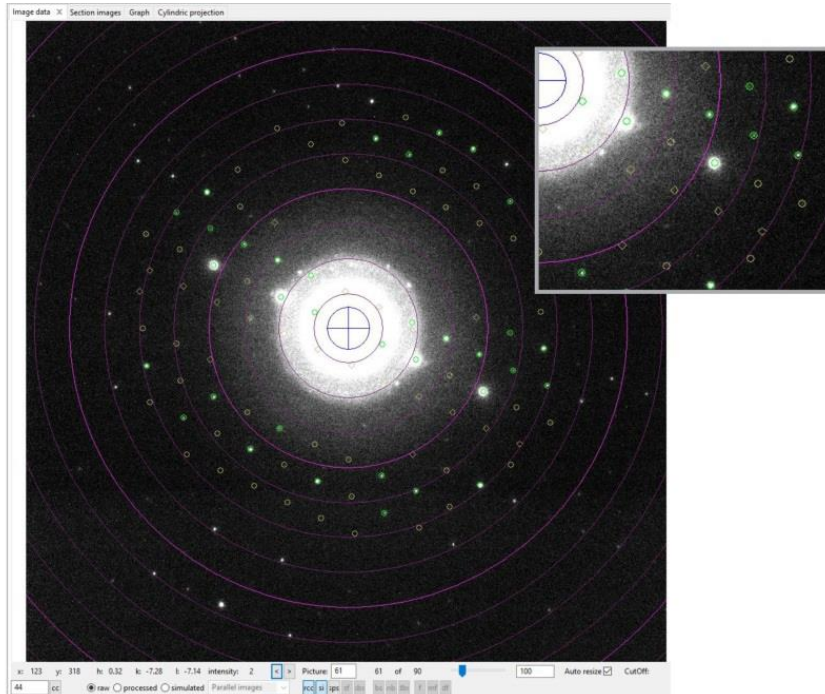
The intensity determination method is set to "sum counts", which is simple but fast. The box "shift integration box maximum intensity" is selected, which allows PETS to slightly shift the integration mask to better cover the diffraction and its background.

Leave all the settings default (see image below).

Start the procedure by clicking the action button "Process frames for integration".

## Notes

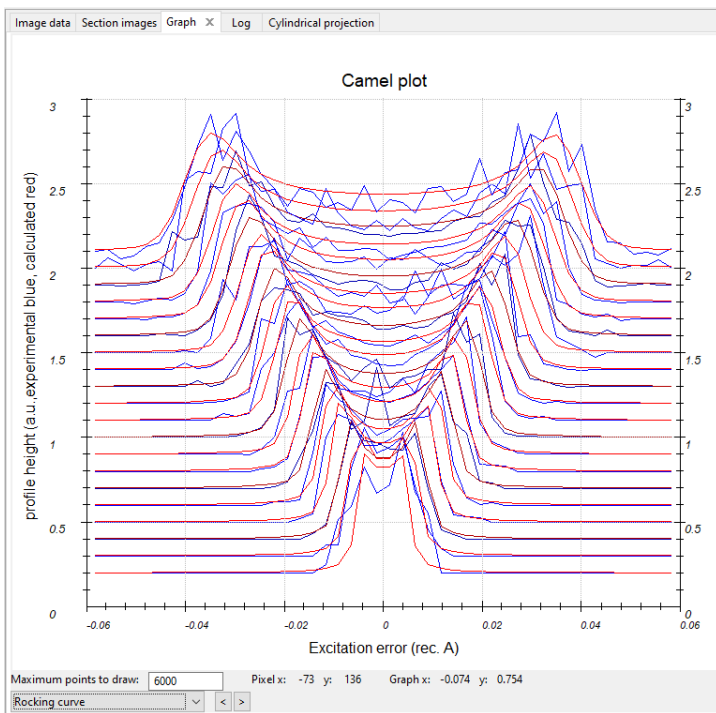
In the Image data tab, diffraction frames with circles drawn at positions predicted from the orientation matrix are displayed. A green circle means that a peak with  $I > 3\sigma(I)$  was found. A brown diamond means that a reflection is expected on upcoming frame(s) or was present on previous frame(s). To assess the resolution of your data, you can also display the resolution rings by selecting the option "rr" in the Image data panel (see below).



to display the resolution rings to display integration boxes

## Notes

At the end of the integration process, in the Graph tab, the camel plots (rocking curves profiles) are displayed (data - blue, fit - red). On the horizontal axis of the plot there is the excitation error of reflections. Each curve shows an average rocking curve of reflections in one resolution shell, starting from  $0.2-0.3 \text{ \AA}^{-1}$  in steps of  $0.1 \text{ \AA}^{-1}$ . Notice the splitting of the humps with increasing diffraction angle due to the precession. By changing the parameters, you can observe their effect on the curve. The reflections considered for the visualization of the camel plots are affected by the  $I/\sigma(I)$  threshold.



### Notes

*The Camel plot is not too bad but also far from optimal. The red curve represents the fit to the experimental (blue) rocking curve using RC width and mosaicity default setting. The fit can be improved by a refinement of these parameters.*

Click on the arrow next to “Optimize reflection profile” to open the options.

Leave the default settings.

Click on the “Optimize reflection profile” to run the refinement.

### Notes

*The values changed only marginally.*

## (b) Finalize integration

Open the parameters for “Finalize integration”.

Check that both “kinematical” and “dynamical” integration are checked.

Leave the radio button selection on “fit profile” next to “intensity estimation”.

### Notes

*For average to good data the “fit profile” option gives better results. “integrate profile” is more robust technique and works well for poorer data.*

Finalize integration

☐ skip saturated reflections

☒ kinematical

intensity estimation:

☒ fit profile  
☐ integrate profile  
☐ maximum intensity  
☐ Bragg position

integration parameters:

1 min. number of reflection measurements  
1 reflection width multiplier

Outlier detection limits:

1000 for profile fitting  
1.5 for symmetry averaging

☒ frame scaling

Laue class for scaling: m-3

Interframe correlation range: 3  
Interframe correlation weight: 1

☒ refine error model

s\_fac: 1  
s\_b: 0  
s\_add: 0

☒ dynamical

Change the “Auto” Laue class scaling to m-3, which we determined from the reciprocal space sections.

Run the procedure by clicking the action button.

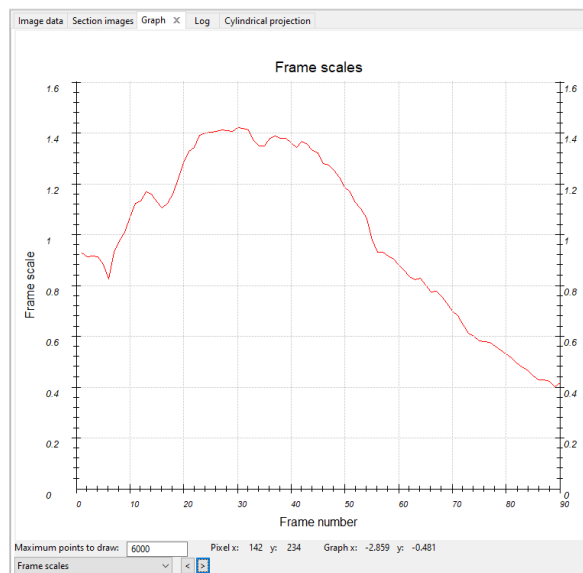
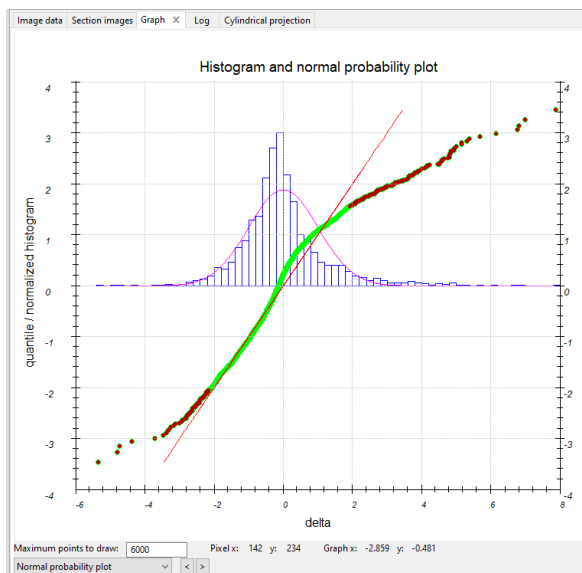
## Notes

*PETS2 will integrate the data, scale the frames using the Laue class symmetry, perform error model refinement (estimation of error modifiers to account for multiple scattering) and reject the reflections, which were identified to be too far from the group of symmetrically equivalent reflections (red dots in the “Normal probability plot”). 287 out of 3608 reflections (8%) were identified as outliers and were rejected. The number of rejected reflections depend on the level of the dynamical effects in the data. In this data, there are three near zone-axes regions (about frames 17, 34, and 56), where are the dynamical effects stronger. The material is relatively dense, which also increase the level of these effects.*

Inspect the “Normal probability plot” graph.

Choose the “Frame scales” graph.

Both graphs are acceptable.



Look at the console output.

```

Integrated intensities rescaled by a factor      100.00
Error model refinement...
Refined parameters of the error model: s_fac = 5.3163, s_b = 0.0000, s_add = 0.0175.
Info: 287 out of 3608 reflections identified as outliers at the rejection limit of 1.50 and rejected.

Intensity statistics:
Observed/Total number of integrated reflections: 859/ 3321
cwl Laue class Rint(obs) Rint(all) Rmeas(obs) Rmeas(all) Nobs/Nall redundancy
* -1 13.03 15.51 17.58 21.17 603/ 1754 1.893
* 2/m 15.80 18.25 19.70 23.03 496/ 1351 2.458
* 112/m 15.87 18.92 19.57 23.41 489/ 1238 2.683
* 2/m11 14.03 16.59 18.21 21.78 546/ 1539 2.158
* mmm 17.29 20.28 20.17 23.76 384/ 900 3.690
* 4/m 25.00 27.74 28.85 32.02 384/ 848 3.916
* 4/mmm 31.94 34.00 34.97 37.19 275/ 499 6.655
* -3 27.77 28.59 33.54 35.03 528/ 1428 2.326
* -31m 40.70 41.57 46.34 47.54 390/ 853 3.893
* -3m1 37.18 38.69 41.90 43.85 368/ 858 3.871
* 6/m 36.27 37.29 41.05 42.40 387/ 855 3.884
* 6/mmm 43.84 44.89 47.66 48.83 271/ 505 6.576
* m-3 22.10 24.64 23.30 26.00 185/ 333 9.973
* m-3m 37.38 38.51 38.75 39.92 144/ 207 16.043
Info: Estimated Laue class: m-3

Statistics for Laue class m-3
d*-range d-range Nobs Nall Nthr compl. rdnd. I/s Rint(obs) Rint(all)
0.00-1.02 Inf-0.98 36 39 46 0.85 7.85 6.87 15.15 15.15
1.02-1.28 0.98-0.78 23 27 33 0.82 10.11 4.46 34.59 35.39
1.28-1.47 0.78-0.68 26 34 37 0.92 9.71 3.54 20.90 21.80
1.47-1.62 0.68-0.62 24 34 36 0.94 9.53 2.59 23.75 25.49
1.62-1.74 0.62-0.57 15 30 31 0.97 10.03 1.82 31.56 34.59
1.74-1.85 0.57-0.54 15 34 36 0.94 10.47 1.63 28.21 33.86
1.85-1.95 0.54-0.51 17 35 37 0.95 10.89 1.41 36.93 40.72
1.95-2.04 0.51-0.49 10 30 30 1.00 10.87 1.05 27.52 37.32
2.04-2.12 0.49-0.47 9 38 38 1.00 10.24 0.81 28.98 42.35
2.12-2.20 0.47-0.46 10 32 32 1.00 10.47 0.91 29.28 39.16
-----
0.00-2.20 Inf-0.46 185 333 356 0.94 9.97 2.94 22.10 24.64

*** Report from dynamical integration ***

```

## Notes

*In our case, we used Laue class m-3, which resulted in Rint(obs) of 22.10, which is consistent with the space group derived earlier from the reciprocal-space sections (Pb-3).*

## (c) Orientation optimization

### Notes

*At this stage we can try to further improve the integration by optimizing the frame orientation and the distortions. Ideally, some correction of the distortions (zero order) should be done before the frame orientation optimization. For this example, calibrated distortions values are already considered. We will proceed directly to the frame orientation optimization.*

In “Optimize frame geometry” select “integrated intensity”.

Select smoothing of correction angles to “polynomial” and type 3 in the “Order” box.

Leave all other settings default (see picture).

Optimize frame geometry

for simulation use:  
☐ uniform intensities  
☒ integrated intensity

☒ frame orientation angles  
☒ center of the diffraction patterns  
☐ RC width  
☒ apparent mosaicity  
☐ distortions  
☒ magnification  
☒ elliptical  
☐ parabolic  
☐ Replace global distortions

Reset to default

Smoothing of correction angles:  
☐ none  
☒ polynomial  
☐ moving average

Order: 3

### Notes

*The option “integrated intensity” can be used only if a kinematical integration has been performed before and gave acceptable results. This is our case now. (When the camel plot*



---

or the integration is not satisfying, use the option “uniform intensity” that does not require to finalize the integration).

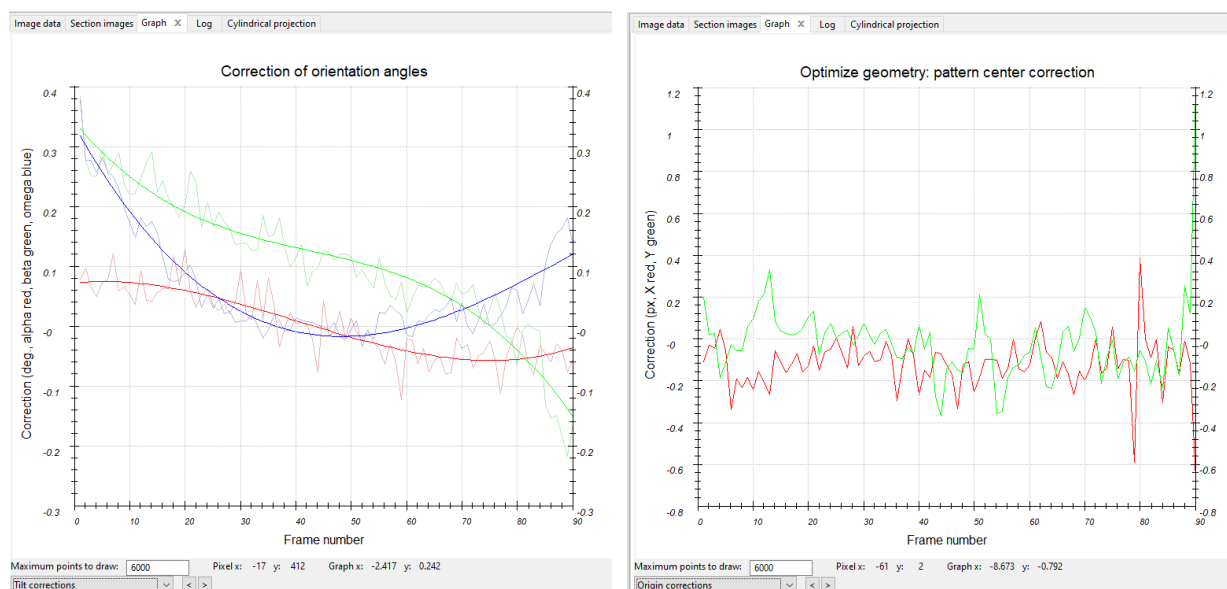
---

Press the action button “Optimize framer geometry” to proceed.

## Notes

The frame orientation refinement optimizes the rotation angles  $\alpha$ ,  $\beta$  and  $\omega$  together with the position of the diffraction pattern centers and the apparent mosaicity of each frame. The routine generates diffraction patterns based on the previous finalized integration. The least-squares minimization routine tries then to reduce the pixel-wise differences of the generated pattern to the background-subtracted experimental diffraction pattern. You can find more details about the method in Palatinus, L. et al. (2019). *Acta Cryst B* 75, 512-522.

The results are visible on the Graph tab selecting the panel “Tilt corrections” on a curve  $\Delta\alpha$ ,  $\Delta\beta$ ,  $\Delta\omega$  (pixel) as a function of the frame number. We can consider that the optimization is successful.



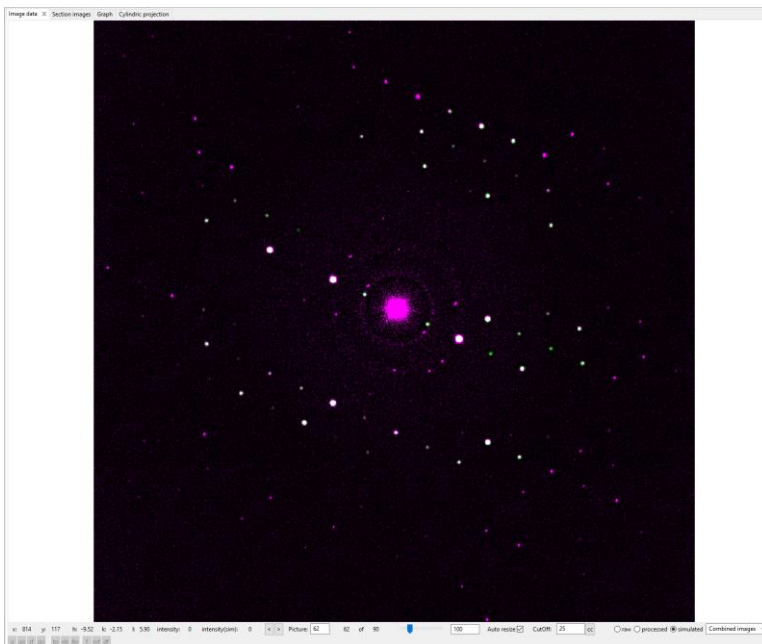
## Notes

In the Graph tab, the origin corrections are also presented on a curve as a function of the frame number. A last panel “Profile parameters” is available to visualize the mosaicity refinement as well as, if included, the RC width and the spot size.

Another way to visualize the results is to open the Image data tab and select simulated and combined images. This view shows a superposition of the calculated and experimental image. The green color corresponds to the calculated intensities in excess of the observed ones, the pink color is the observed intensity in excess of calculated, and the white parts correspond to the good overlap of both.

---





## Notes

*Because the frame orientations have changed, we need to redo some of the data processing steps, namely “Process frames for integration” and refinement of the rocking curve profile.*

Choose fit “profile” option in “Process frames for integration”.

## Notes

*Fit profile uses the average profile of the reflections, which is created during peak search. The average profile is then fitted into each reflection and scaled. The extracted intensity is usually better than that from the “sum counts” procedure.*

Click the action button “Process frames for integration”.

Switch to the tab Graph to view the new camel plot.

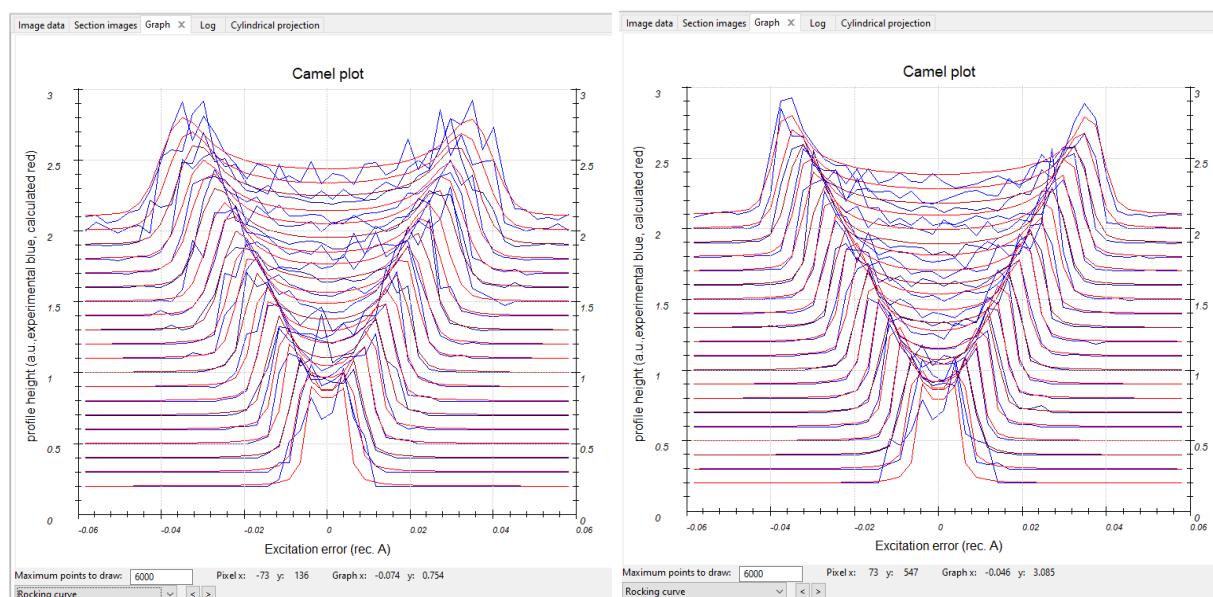
## (d) Optimize reflection profile

Go to the options of “Optimize reflection profile”.

Start the fitting procedure by clicking on the action button.

## Notes

*The optimized values for the camel plot are now RC width =  $0.00095 \text{ \AA}^{-1}$  and apparent mosaicity =  $0.0566 \text{ deg}$ . The camel plot is visibly improved (image below, right) compared to the plot before frame orientation (image below, left). It is sharper, less noisy and can be described with smaller RC width and mosaicity parameters.*



In the Finalize integration options, check that fit profile is still selected as well as the frame scaling for the Laue class m-3.

Run Finalize integration.

## Notes

*Rint(obs)* for Laue class m-3 decreased to 20.27% and the number of rejected reflections decreased to 7%.

```
Integrated intensities rescaled by a factor      100.00
Error model refinement...
Refined parameters of the error model: s_fac = 4.5213, s_b = 0.0000, s_add = 0.0214.
Info: 262 out of 3596 reflections identified as outliers at the rejection limit of 1.50 and rejected.

Intensity statistics:
Observed/Total number of integrated reflections: 1004/ 3334
cwl Laue class Rint(obs)  Rint(all)  Rmeas(obs)  Rmeas(all)  Nobs/Nall  redundancy
* -1          11.27      12.78      14.99      17.23      698/ 1757  1.898
* 2/m         15.18      16.50      18.39      20.31      549/ 1337  2.494
* 112/m       13.53      15.61      16.31      18.94      553/ 1236  2.697
* 2/m11       12.19      13.64      15.55      17.63      626/ 1538  2.168
* mmm        15.56      17.41      17.79      20.05      418/  887  3.759
* 4/m        23.85      25.50      27.06      29.01      428/  840  3.969
* 4/mmm      30.96      31.86      33.60      34.60      305/  489  6.818
* -3         26.65      26.95      32.02      32.73      600/ 1441  2.314
* -31m       41.36      41.48      46.90      47.20      450/  865  3.854
* -3m1       38.66      38.88      43.36      43.80      423/  867  3.845
* 6/m        35.44      35.76      39.86      40.38      431/  864  3.859
* 6/mmm      45.59      45.67      49.32      49.45      315/  510  6.537
* m-3        20.27      21.51      21.31      22.63      222/  335  9.952
* m-3m       33.74      34.10      34.87      35.25      165/  205 16.263
Info: Estimated Laue class: m-3

Statistics for Laue class m-3
d*-range  d-range  Nobs  Nall  Nthr  compl.  rdnd.  I/s  Rint(obs)  Rint(all)
0.00-1.02  Inf-0.98   37   40   46   0.87   7.95   7.33   16.76   16.76
1.02-1.28  0.98-0.78  28   29   33   0.88   9.69   4.98   26.58   26.57
1.28-1.47  0.78-0.68  28   33   37   0.89  10.27   4.28   19.41   19.78
1.47-1.62  0.68-0.62  28   35   36   0.97   9.20   3.12   17.67   18.39
1.62-1.74  0.62-0.57  20   29   31   0.94  10.21   2.11   25.80   27.26
1.74-1.85  0.57-0.54  23   34   36   0.94  10.44   2.12   24.25   26.68
1.85-1.95  0.54-0.51  20   36   37   0.97  10.67   1.83   24.01   27.03
1.95-2.04  0.51-0.49  12   30   30   1.00  10.77   1.42   21.66   29.15
2.04-2.12  0.49-0.47  14   38   38   1.00  10.24   1.06   31.61   38.79
2.12-2.20  0.47-0.46  12   31   32   0.97  10.55   1.18   27.02   33.64
-----
0.00-2.20  Inf.-0.46  222  335  356   0.94   9.95   3.67   20.27   21.51

*** Report from dynamical integration ***
```

Close PETS

Answer “Yes” to the question about saving the project

## Notes

The current state of the project is saved in the file pyrite.pts2. Opening this file with PETS next time allows you to continue from the current state of processing. If you open

---

*pyrite.pts, you must start from scratch again, because pyrite.pts is not overwritten by PETS and still stores the initial settings. In this case the calibrated distortion parameters are very close to the parameters that would be obtained by refinement. In this introductory example, the distortion parameters are not refined, because it does not bring any visible improvement to the result, and it makes the data reduction longer and more complicated. All the refined parameters and graphs are saved also in the Log file, which are accessible after the job is again opened or you can access them in pyrite\_petsdata\log folder.*

---

## PART 2 - Structure solution and kinematical refinement in Jana2020

### 1. Create new structure

#### Notes

**Important!** The data-processing procedure is almost never perfectly reproducible. Small differences in the indexing and cell refinement procedure may result in small differences of integrated intensities. If you want to be sure that you can reproduce the following part of the tutorial, do not use the file *pyrite.cif\_pets* that you have just created, but use the file *pyrite\_reference.cif\_pets* provided with the example files. Using your own *cif\_pets* file is also possible, but your results may slightly differ from the results described in this tutorial. Make also sure that the *pyrite.tut\** files are present in the folder with the Jana2020 files, so that the tutorial mode can be used.

#### Start Jana2020

In the Main menu bar, use “Structure → New” and open new structure “pyrite” in the directory of this example.

### 2. Import Wizard

[On the screen: Specify type of the file to be imported]

Select “Single crystal: known diffractometer formats”; NEXT

[On the screen: Data reduction file from]

Select “Pets electron diffractometer”

For “Input file”, browse the file *pyrite\_reference.cif\_pets* (or your file)

Make sure the option “make the refinement file for dynamical refinement” is unselected

This file produced by PETS2 contains all important information, including the list of intensities.

NEXT

[On the screen: Complete/correct experimental parameters]

Replace the temperature by 100, Leave all the other settings unchanged; NEXT

Leave all settings unchanged; NEXT

The program reads 3334 reflections from the file

OK

For absorption correction select “None or done before importing” (default); NEXT

FINISH; OK to accept the data set

#### Notes

You just read in the cell parameters and the intensity list. If you do not have the data in the *cif\_pets* format, you can read them in from a general *hkl* file using the option “reflection file corrected for LP and absorption” at the beginning of the import wizard. You then have to input the radiation type, wavelength and cell parameters by hand.

---

### 3. Symmetry Wizard

NEXT to close the information window and start the symmetry wizard.

#### Notes

Symmetry Wizard can be started separately by expanding “Reflection file” in the Command tree and selecting “Make space group test”. The default settings are prepared for x-ray diffraction. For electron diffraction we have to increase the tolerances. Change the tolerances according to this screenshot:

The screenshot shows the Symmetry Wizard dialog box. It has two radio buttons at the top: "Start with data collection cell" (selected) and "Start with actual cell". Below them is a checkbox "Search for higher symmetrical supercell (recommended)" which is checked. There are input fields for cell parameters: 5.407, 5.405, 5.406, 90.00, 89.98, 90.00. Under "Tolerances for crystal system recognition:", there are three input fields: "Maximal deviation for cell lengths in Ang:" (0.02), "Maximal deviation for cell angles in degs:" (0.2), and "Maximal deviation for modulation vector:" (0.01). Under "Tolerances for space group recognition:", there are two input fields: "Maximal ave(1/sig(I)) for systematic extinctions induced by cell centering:" (5) and "Maximal ave(1/sig(I)) for systematic extinctions induced by other symmetry:" (25). At the bottom are "Back", "Next", and "Cancel" buttons.

NEXT; OK

[On the screen: Select Laue point group]

The screenshot shows the "Select Laue point group" dialog box. It contains a table with the following columns: PointGroup, Axes, Rint(obs/all), #averaged(obs/all), and Redundancy. The table lists various point groups and their corresponding statistics. The "m-3" point group is highlighted in blue. Below the table, there is a text label "Averages made from 1004/3334 reflections", a "Details" button, and a checkbox "Introduce t<sub>g</sub>in law for subgroups". At the bottom are "Back", "Next", and "Cancel" buttons.

PointGroup	Axes	Rint(obs/all)	#averaged(obs/all)	Redundancy
2/m	(-b+c,b+c,-a)	21.98/23.39	567/1205	2.767
2/m	(-b-c,b-c,a)	10.12/11.95	698/1691	1.972
2/m	(a-c,-a-c,b)	18.77/20.24	619/1372	2.43
2/m	(a-b,-a-b,-c)	21.17/22.96	532/1154	2.889
2/m	(-a-b,a-b,c)	20.21/22.16	606/1297	2.571
mmm	(c,a,b)	11.19/13.39	446/950	3.509
mmm	(a+c,a-c,b)	24.02/25.35	453/866	3.85
mmm	(-b-c,b-c,-a)	22.47/24.00	512/999	3.237
mmm	(-a-b,-a-b,-c)	25.57/27.12	436/775	4.302
4/m	(a,b,c)	20.11/22.06	456/897	3.717
4/m	(a,b,c)	20.88/22.40	448/916	3.64
4/m	(a,b,c)	23.07/24.80	480/930	3.585
4/mmm	(a,b,c)	27.15/28.30	330/531	6.279
4/mmm	(c,a,b)	26.50/27.50	337/559	5.964
4/mmm	(-c,-b,-a)	25.09/26.40	342/560	5.954
-3	(-b+c,a+b,-a+b+c)	9.94/12.20	513/1135	2.937
-3	(-b-c,-a+b,-a+b-c)	10.75/12.96	494/1088	3.064
-3	(a-c,b-c,-a-b-c)	10.23/12.74	534/1173	2.842
-3	(a-c,-a-b,-a+b-c)	11.05/13.31	415/914	3.648
-3m	(-b+c,a+b,-a+b+c)	25.50/26.84	388/649	5.137
-3m	(-b-c,-a+b,-a+b-c)	26.75/27.95	381/662	5.036
-3m	(-a+c,b-c,-a-b-c)	26.29/27.86	407/673	4.954
-3m	(a-c,-a-b,-a+b-c)	26.15/27.20	320/536	6.22
m-3	(a,b,c)	14.14/15.70	234/356	9.365
m-3m	(a,b,c)	29.44/29.91	179/222	15.018

Select Cubic m-3 with no axis transformation; NEXT

#### Notes

The internal R-values would be rather high for x-rays, but are acceptable for electron diffraction data.

[On the screen: Select cell centering]

**Select cell centering**

Centering	#obs/#all	ave(I/sig(I))	
<input checked="" type="radio"/> P	0/0	0.000/0.000	<a href="#">Details</a>
<input type="radio"/> A	n.a.	n.a.	<a href="#">Details</a>
<input type="radio"/> B	n.a.	n.a.	<a href="#">Details</a>
<input type="radio"/> C	n.a.	n.a.	<a href="#">Details</a>
<input type="radio"/> I	479/1701	5.613/2.399	<a href="#">Details</a>
<input type="radio"/> R-obverse	n.a.	n.a.	<a href="#">Details</a>
<input type="radio"/> R-reverse	n.a.	n.a.	<a href="#">Details</a>
<input type="radio"/> F	491/2469	5.086/1.897	<a href="#">Details</a>
<input type="radio"/> X (1/2,0,0), ...	-----/-----	-----/-----	<a href="#">Details</a>
<input type="radio"/> X (1/3,0,0), ...	-----/-----	-----/-----	<a href="#">Details</a>

[Show/modify X centering](#)

Warning: The cell centering need not be one you expect from collection as the program first transform the cell to the reduced form.  
Moreover after your selection the program makes another transformation whenever the centring is not the standard one.

[Back](#) [Next](#) [Cancel](#)

## Notes

The statistics clearly excludes a centered cell. The choice of a primitive cell is also in agreement with our visual inspection of the reciprocal space sections.

Select primitive unit cell; NEXT

## Notes

[On the screen: Select space group]

The window shows possible space groups.

The list of the strongest reflections contradicting the selected space group can be displayed by "Details" button.

**Select space group**

Characteristics for systematically absent reflections

Space group	#obs/#all	ave(I/sig(I))	Figure of merit
Pb-3	37/285	3.989/1.520	0.03451
P213	0/12	0.309	0.07936
Pa-3	78/292	5.736/2.318	0.20733
Pn-3	77/283	5.689/2.347	0.21384
Pm-3	0/0	-----	1.00000
P23	0/0	-----	1.00000

[Details](#)

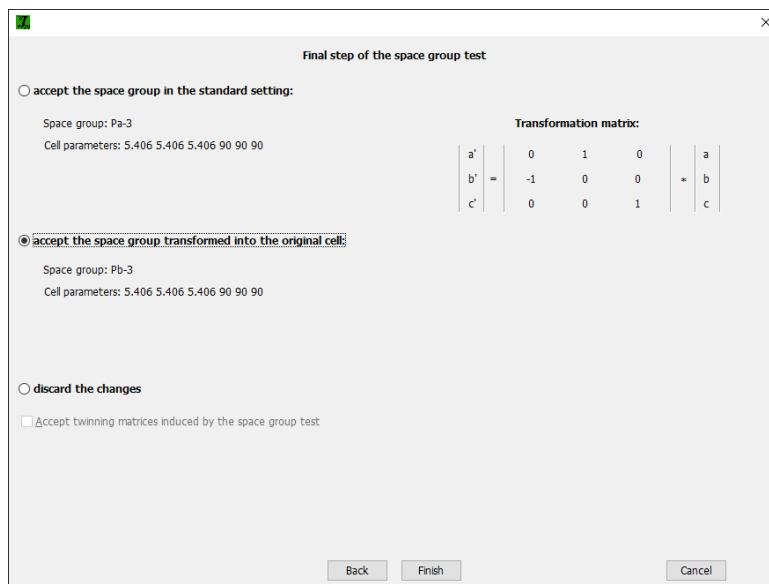
[Back](#) [Next](#) [Cancel](#)

## Notes

In Section 7 of the data reduction (Create reciprocal-space sections), we saw that the space group determination was difficult. Despite some violations, JANA2020 offers the expected space groups Pb-3.

Note that a wrong choice of the space group is not detrimental for the structure solution, because we will use the program Superflip for structure solution. This program makes additional check for the most probable space group.

Select Pb-3; NEXT



Choose “accept the space group transformed into the original cell”.  
[FINISH](#)

#### Notes

*Symmetry is saved in file pyrite.m50.*

*The data files are produced in the Pb-3 settings, so we have the non-standard setting.*

*In general, if you realize during the analysis that the unit cell found by PETS2 leads to a non-standard setting, you can go back to the indexing, change the setting, and redo all the other steps to integrate the data. It is planned that it would be possible in future to transform the structure to the standard setting later in Jana2020. However, now due to certain technicalities, this transformation would make it impossible to proceed from kinematical refinement to the dynamical refinement. It is therefore better to use the cell setting as set in PETS2.*

## 4. Creating refinement reflection file

#### Notes

*In this step, the program creates file pyrite.m90 containing the data set merged by symmetry and with discarded forbidden reflections. This file will be used for refinement.*

[Proceed with the wizard, leaving all settings default. NEXT](#)

#### Notes

*The Import statistics-obs/all gives 1004/3334 reflections read from input file, 973/3049 reflections written to output file and 31/285 reflections rejected as symmetrically extinct.*

[OK,](#)

[OK again; NEXT](#)

*The Rint(obs/all) = 13.83%/15.21% for 210/298 independent reflections.*

[OK; FINISH](#)

## 5. Structure Solution Wizard

[OK to close the information window and start the solution wizard](#)

#### Notes

*[On the screen: window of Structure solution]*

*Structure solution wizard can be executed separately from the Command tree. Jana2020 does not contain solution procedures, it calls external programs. By default, Jana uses*

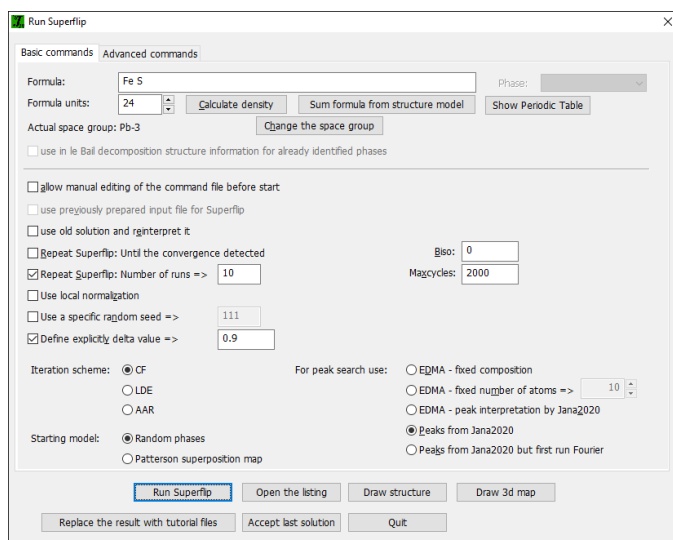
*Superflip (using charge flipping as the solution method). Superflip is distributed with Jana2020.*

In “Formula” textbox type the list of elements: Fe S

### Notes

*Superflip does not need information about the chemical composition but it will be required for the peak assignment based on the electrostatic potential map calculated by Superflip. If Jana2020 is used for peak assignment, a list of expected chemical elements is sufficient. If you know the expected formula, you may enter it and use EDMA for peak search (at the bottom of the window). If the formula is correct, EDMA often gives cleaner and more complete interpretations of the solution.*

Leave other settings default; “Run Superflip”.



### Notes

*Window of Superflip appears with iteration running. After ten runs, Superflip stops and Jana2020 shows a table with the basic information, especially the determined space group symmetry:*

INFORMATION	
R-value after noise suppression: 22.401	
HM symbol derived by Superflip: Pb-3	
Fit of user-defined symmetry elements	
Symbol	R-value
b	0.52
a	4.59
c	0.46
3	1.74
Formula: Fe0.167 S0.333	
OK	

OK to close the table.

### Notes

**Important!** *If you want to ensure perfect reproducibility of your refinement with this tutorial, you need to replace the result of your Superflip run with the reference run. Superflip starts from random starting points and, therefore, each solution may be slightly different. The differences are small, but may lead to imperfect reproducibility of the numbers. Therefore, it is advised that you replace your Superflip results with the reference stored in the pyrite.tut\* files. You can do this by clicking the appropriate button:*

Click the button “Replace the result with tutorial files”

### Notes

*A new information table appears.*

*The listing of Superflip contains much more information about the iteration and you can view it by pressing the button “Open the listing”.*



---

The space-group selection  $Pa-3$  was confirmed. In cases of doubts or difficult problems, it is worth checking the statistics, especially the symmetry analysis, for each of the ten runs, and seeking for the most probable one. We can close the log file. Note that it is saved as *pyrite.sflog*, so you can also open it with a text editor.

---

Click "Draw 3D map"

#### Notes

VESTA opens with a 3d electrostatic potential shown as isosurfaces, and an atomic model overlaid (in the default settings the shown isosurface level corresponds to the  $2\sigma$  level based on the statistics of the 3D electrostatic potential map). Rotate the view to inspect the structure (Fe - brown, S - yellow). You may switch to polyhedral model (in the left panel). The model looks very good and should contain only 2 atomic sites, 1 Fe and 1 S. The correct model present  $\text{FeS}_6$  octahedra and  $\text{S}_2$  pairs.

---

Close VESTA (do not save the file).

Click "Draw structure".

Now the structure is displayed using the plotting program of Jana2020 called JanaDraw  
In the lower toolbar, press build molecule.

Close Vesta.

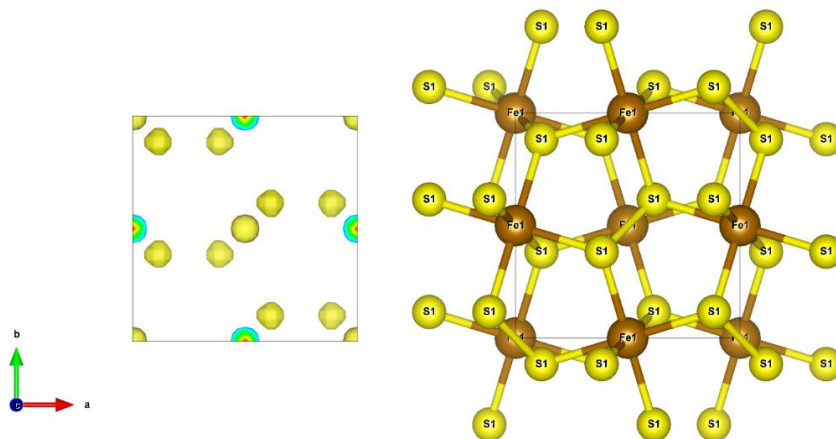
Press "Accept last solution".

#### Notes

The program creates file *pyrite.m40* with atoms of the structure model

To access the list of atoms with their positions and atomic displacement parameters, you can open the *pyrite.m40* using "Edit file" menu in Command tree, or using "Edit structure parameters → Edit atomic parameters", or using the quick button "Edit atoms".

---



## 6. Kinematical refinement

[On the screen: basic window of Jana]

Right-click the quick button "Run Refinement".

[On the screen: refinement options]

Change the number of cycles to 100 the damping factor at 0.1;

Uncheck "Disable atoms having too large isotropic ADP parameter".

Uncheck "Refinements on  $F(\text{obs})^2$ ".

OK.



Choose “Yes+start”

## Notes

*Refinement converges with R values of  $R(\text{obs}) = 21.76\%$ ,  $wR(\text{all}) = 23.49\%$  for 210/298 observed over all reflections and 4 refined parameters. At this moment you may check the refinement listing (Command tree  $\rightarrow$  Refinement  $\rightarrow$  View refinement listing”) with a lot of information about the refinement procedure and the results. The ADPs of both Fe and S are slightly negative, which is due to the dynamical effects.*

RFactors overview

R factors : [298=210+88/4], Damping factor: 0.1000  
GOF(obs)= 5.24 GOF(all)= 4.56  
R(obs)= 21.76 wR(obs)= 22.70 R(all)= 25.10 wR(all)= 23.49  
Last wR(all): 23.49 23.49 23.49 23.49 23.49 23.49 23.49 23.49  
Maximum change/s.u. : -0.0469 for Uiso[Fe1]

Question

Serious warnings in the listing!  
Regular end of REFIN program

Open the listing?

Yes No

It remains 3 sec - press this button to continue

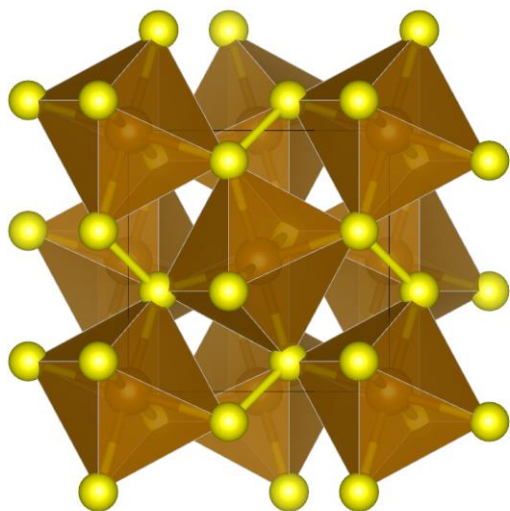
## 7. The resulting structure

Change focus to JanaDraw

Press “Build molecule” on the lower toolbar to see part of the resulting structure

In Command tree, expand “Draw structure” and double click “User defined” to start “Vesta”

Here is a polyhedron view by VESTA:



## 8. Difference Fourier map

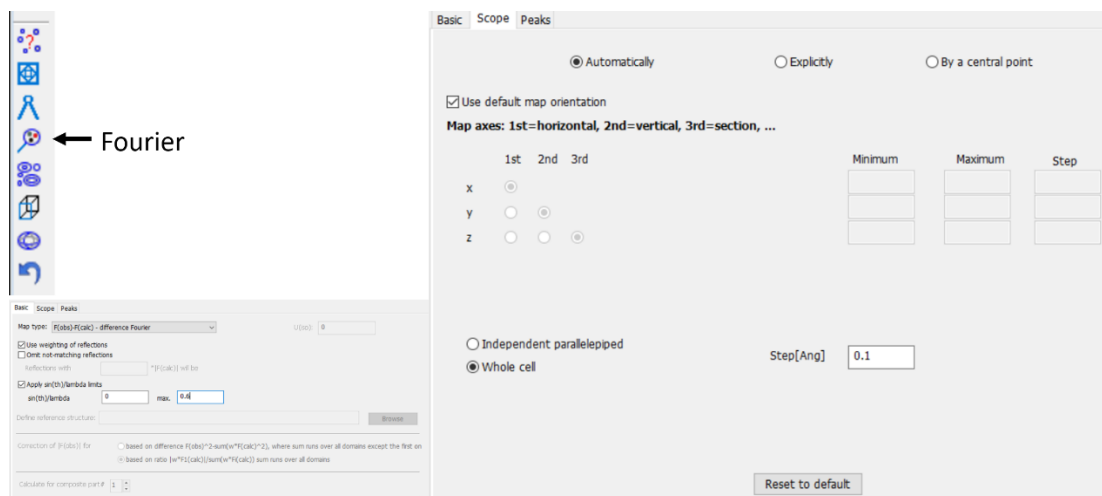
### Notes

Difference Fourier map is used to check if there is any missing electrostatic potential that has not been described by the structure model yet. This is not the case here, the structure is very simple, but in general you have to check it.

[On the screen: basic window of Jana]

Right-click the quick button “Run Fourier synthesis”

Select the following parameters to calculate the Fourier map.



OK; “Yes+start”

Choose “No” to avoid the procedure for including new atoms.

Choose “No” to avoid opening the listing.

In Command Tree, double-click “Run Contour”

In Command Tree of Contour, expand “Use old Fourier map” and start “Draw maps as calculated”

[On the screen: Contour program]

In the right pane, use “Go to map” to display next/previous two-dimensional section

In bottom toolbar, press the button “Run Vesta”

### Notes

To adjust the display of the potential map, go to Properties in VESTA, tab Isosurfaces. Click on isosurface No 1 and change it to Positive and its level. To display the isosurface

---

*corresponding to  $3\sigma$ , multiply the value of the isosurface level by 1.5. No significant isosurface are visible that would indicate a missing atom in the structure.*

---

[Close Contour](#)

## PART 3 - Dynamical refinement in Jana2020

### 1. Preparation of reflection file for the dynamical refinement

#### Notes

*During the integration procedure in PETS2, an input file for dynamical refinement was created. The integration for the dynamical refinement considers each diffraction image separately. The result is a list with the hkl indices, the measured intensity with the respective  $\sigma(I)$  and the number of the frame in which the intensity is present. PETS2 creates the file pyrite\_dyn.cif\_pets with all this information, which will be used in Jana2020. Note that the same hkl reflection might be present in several frames, with different intensity due to the multiple scattering relative to different crystal orientation and thickness at this condition.*

In the main menu bar, use "Structure → Save as" and save a new structure pyrite\_dyn for the dynamical refinement

YES to Continue with the new structure.

#### Notes

*This step saves the structure you refined kinematically as a new job, which will be used for dynamical refinement.*

*Before starting, the Uiso should be changed to positive values to start from a reasonable structure.*

Click the quick button "Edit atoms"

Select both atoms: hold CTRL and click on the atoms

Press "Action" and select "Define/Edit atom parameters"

In the page "Edit", click the Uiso textbox

#### Notes

*Yellow colour indicates the textbox is locked as it corresponds to atoms with different values of Uiso*

Click "Unlock" button in the lower left corner of the form

Replace the value of Uiso with 0.005.

OK; OK; Yes to rewrite changes.

In Command Tree, expand „Reflection file“ and start „Import/modify reflection file“.

Click „Reimport“.

#### Notes

*Again, if you want to be sure that you can exactly reproduce the following part of the tutorial, use the file pyrite\_reference\_dyn.cif\_pets provided with the example files.*

Select the pyrite\_reference\_dyn.cif\_pets as the new reflection file

Select the option "make the refinement file for dynamical refinement"

NEXT,

Set the temperature in "Data collection details:" to 100 K

NEXT, NEXT.

Now, Jana reads 17785 reflections from all the frames.

OK, NEXT, FINISH, OK

[On the screen: Processing refinement reflection file ... ]

NEXT, OK, OK, FINISH

#### Notes

*Notice that 8539 reflections are observed out of 17785 reflections read. From these, 520 were rejected because they violate the systematic extinctions. You can find this information in the file \*.rre (Edit/View → Reflection report).*

## 2. Dynamical refinement parameters

In the command tree expand “Edit structure parameters” and double click “Edit parameters for electron diffraction”

[On the screen: Parameters for electron diffraction data]

Change the parameters according to the following image (g(max), RSg and Number of integration steps):

Orientation matrix: U11 -0.0706 U12 0.08881 U13 0.14613  
U21 -0.12561 U22 -0.1342 U23 0.02087  
U31 0.11592 U32 -0.0913 U33 0.11149

Go to reblock# 1

Maximal diffraction vector g(max): 1.7  
Maximal excitation error (Matrix): 0.01  
Maximal excitation error (Refine): 0.1  
RSg(max): 0.5  
DSg(min): 0  
Number of threads: 4  
Dyno commands:

Number of integration steps: 100  
Use dynamic approach: ☒  
Use twin version: ☐  
Apply correction for crystal tilt: ☒  
For Fourier rescale to Fcalc: ☐

Geometry: ☒ precession  
☐ rotation

Select zones for refinement Define zones of equal thicknesses

Run optimizations except of scale, optimize also: ☒ Thickness ☐ Orientation Show thickness plots

Zone#: 1 uvw: (1.000,0.255,0.323) R(all)= Select zones for editing

Precession angle 1.01 Alpha -49.927 Beta 0.33

EDScale 899.9999 EDThick 860 EDPhi 0 EDTheta 0

Esc OK

### Notes

You can set the number of threads to the number of cores on your processor to speed up the calculation.

## 3. Sample thickness

### Notes

The sample thickness is an important parameter of the dynamical refinement. The default value is 400 Å.

Enable “Except of scale, optimize also: Thickness”

Click “Run optimizations”

Orientation matrix: U11 -0.0706 U12 0.08881 U13 0.14613  
U21 -0.12561 U22 -0.1342 U23 0.02087  
U31 0.11592 U32 -0.0913 U33 0.11149

Go to reblock# 1

Maximal diffraction vector g(max): 1.7  
Maximal excitation error (Matrix): 0.01  
Maximal excitation error (Refine): 0.1  
RSg(max): 0.5  
DSg(min): 0  
Number of threads: 4  
Dyno commands:

Number of integration steps: 100  
Use dynamic approach: ☒  
Use twin version: ☐  
Apply correction for crystal tilt: ☒  
For Fourier rescale to Fcalc: ☐

Geometry: ☒ precession  
☐ rotation

Select zones for refinement Define zones of equal thicknesses

Run optimizations except of scale, optimize also: ☒ Thickness ☐ Orientation Show thickness plots

Zone#: 1 uvw: (1.000,0.255,0.323) R(all)= Select zones for editing

Precession angle 1.01 Alpha -49.927 Beta 0.33

EDScale 1000 EDThick 400 EDPhi 0 EDTheta 0

Esc OK

Edit electron diffraction parameters for reblock#1

Orientation matrix: U11	-0.0706	U12	0.08881	U13	0.14613
U21	-0.12561	U22	-0.1342	U23	0.02087
U31	0.11592	U32	-0.0913	U33	0.11149

Go to reblock# 1

Maximal diffraction vector  $g(\max)$ : 1.7  
 Maximal excitation error (Matrix): 0.01  
 Maximal excitation error (Refine): 0.1  
 $RSg(\max)$ : 0.5  
 $DSg(\min)$ : 0  
 Number of threads: 4  
 Dyno commands:

Number of integration steps: 100  
 Geometry: ☒ precession  
☐ rotation  
 Use dynamic approach: ☒  
 Use twin version: ☐  
 Apply correction for crystal tilt: ☒  
 For Fourier rescale to  $E_{\text{calc}}$ : ☐

Select zones for refinement Define zones of equal thicknesses

Run optimizations except of scale, optimize also: ☐ Thickness ☐ Orientation Show thickness plots

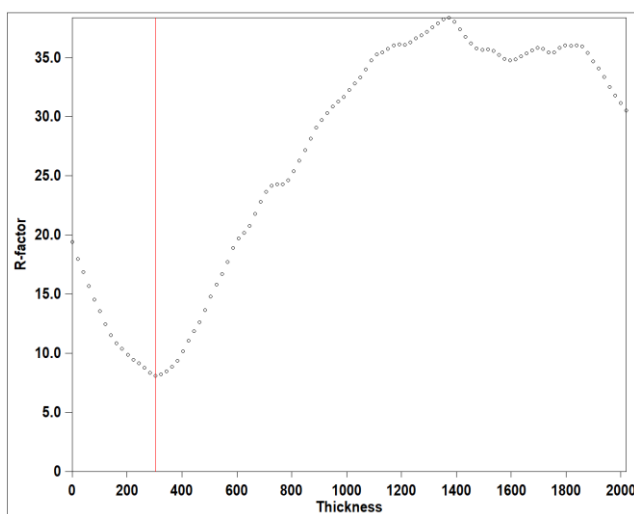
Zone#: 1 uvw: (1.000,0.255,0.323) R(all)= Select zones for editing

Precession angle 1.01 Alpha -49.927 Beta 0.33  
 EDScale 1000 EDThick 400 EDPhi 0 EDTheta 0

Esc OK

Dyngo calculates  $wR$ -factor for the thickness varying from 0 to 2000 Å.

“Show thickness plots”. Check the variation of the thickness of the crystal for the frames. Quit.



## Notes

Most frames have well defined  $wR$ -factor minima for one thickness. However, few frames either have poorly defined minimum or the minimum is at a very different thickness. You can check the  $wR(\text{all})$  going through the zones in the main window of “parameters for electron diffraction data” or while viewing the graphs. Looking at the typical position of the minimum across the frames you can see that 400 Å is usually a reasonable starting point for the refinement.

“Select zones for editing → Select all → OK”.

Click in the field EDThick, click the button Unlock and type 400 for “EDThick”.

“Select zones for editing → Refresh → OK”.

Uncheck “Except of scale, optimize also: Thickness”.

Click “Run optimizations” (neither the Thickness nor the Orientation check box should be checked!).

## Notes

This optimizes scale factors of individual frames with the new common thickness and sets a good starting point for the least-squares refinement.

Click OK and YES to rewrite M42 file and return to the main screen of Jana2020.

## 4. Initial refinement

Right-click on quick button “Refine”, set the number of cycles is set to 100 and the damping factor to 1.

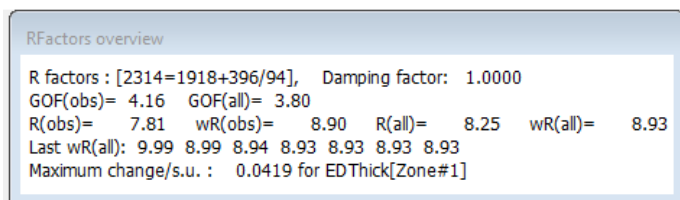
OK; “Yes+Start”.

### Notes

*The refinement converges after 8 cycles, each cycle takes a few seconds (depending on your computer).*

*You can see the details of the refinement in the file pyrite\_dyn.ref on the screen or in refinement listing (Command Tree → Refinement → View refinement listing). The refinement converges with  $R(\text{obs})/wR(\text{all}) = 7.81\%/8.93\%$ . It is important to check the ratio observed reflections to refined parameters, which is around 20 in the present case (1918/94). It is recommended to keep this ratio above 10. It can be increased by increasing  $RSg(\text{max})$  (selecting more reflections for the refinement).*

*Another important aspect to check is the number of observed reflections per zone (frame). Also, in the pyrite\_dyn.ref file (refinement listing), you can see “R-factors for individual zones”, where the list of zones with the respective R-factors and number of reflections is shown.*



RFactors overview

R factors : [2314=1918+396/94], Damping factor: 1.0000  
GOF(obs)= 4.16 GOF(all)= 3.80  
R(obs)= 7.81 wR(obs)= 8.90 R(all)= 8.25 wR(all)= 8.93  
Last wR(all): 9.99 8.99 8.94 8.93 8.93 8.93 8.93  
Maximum change/s.u. : 0.0419 for EDThick[Zone#1]

*Note that the displacement parameters of both Fe and S are now positive.*

## 5. Optimization of orientation

[On the screen: Main window of Jana2020].

Save a new structure “Structure→Save as→pyrite\_dyn\_orient”; OK; YES

In Command Tree, expand “Edit structure parameters” and start “Edit parameters for electron diffraction”

[On the screen: Parameters for electron diffraction data]

Enable “Except of scale, optimize also: Orientation”

Click “Run optimizations”

[On the screen: Intensity calculation for optimizing of orientation].

Using “Zone #” box, check the resulting EDTheta for the individual frames.

### Notes

*EDTheta is the amplitude of the correction of the frame orientation. It should be a small number, a few tenths of a degree at most, because the orientation was already corrected in PETS during the data reduction. A deviation of  $0.5^\circ$  is already high and signifies a failure of the optimization procedure for this individual frame. In such case, the frame should be removed from the refinement. In the current case no value of ED Theta exceeds  $0.5^\circ$  and most values are below  $0.2^\circ$ .*

OK; YES to save file M42

Start the refinement.

### Notes

The refinement converges after 3 cycles with  $R(\text{obs})/wR(\text{all}) = 7.04\%/7.85\%$  for 1905/2297 observed/all reflections and 94 refined parameters (3 structural parameters + crystal thickness + 90 scales).

The refined thickness is 458(2) Å.

Information from refinement:

RFactors overview

R factors : [2297=1905+392/94], Damping factor: 1.0000  
GOF(obs)= 3.65 GOF(all)= 3.35  
R(obs)= 7.04 wR(obs)= 7.79 R(all)= 7.61 wR(all)= 7.85  
Last wR(all): 7.91 7.85  
Maximum change/s.u. : 0.0387 for EDThick[Zone#1]

Check the final result, calculate the difference Fourier map, evaluate the structure.

The difference Fourier map is almost empty with very low residual potentials.

## 6. The resulting structure

### Notes

You can view the resulting structure in JanaDraw or by using your favorite program associated with Jana2020 - see "Draw structure" in the Command Tree.

The structure is very simple, no big change will be observed in the positional parameters with respect to the kinematical refinement. However, the dynamical refinement leads to much better figures of merit, positive Uiso values and a better accuracy of the parameters.

		Kinematical		dynamical	
	Wyck.	Uiso [Å <sup>2</sup> ]	x/a	Uiso [Å <sup>2</sup> ]	x/a
Fe1	4a	-0.0012(7)		0.0013(1)	
S1	8c	-0.0029(7)	0.1156(5)	0.0016(2)	0.1152(1)