Instruction to PyXAS GUI

Contents

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Reference: J. Synchrotron Rad. 2020, 27, 567

Source code: https://github.com/gmysage/pyxas (installation instruction included)

Individual XANES image fitting

Lunch program: python pyxas_gui.py

Sample files (downloaded in github)

1. "NMC-LLZO.h5": 2D XANES image. This image stack derives from one slice of 3D xanes-tomography taken across the Ni absorption edge.
   The .h5 file has the following structure:
   - /img_xanes: flat-field normalized image stack
   - /X_eng: an energy list of energy associated with XANES image stack
2. ref_NiO.txt: reference spectrum of Ni^{2+} collected from NiO particles
3. ref_LiNiO2.txt: reference spectrum of Ni^{3+} collected from $LiNiO_2$ particles

Step-by-step of 2D XANES fitting

Data loading (Tab: Prep.) (see Fig. 1):

In the "Load image" section, select "hdf" as the file type.
In "Dataset for XANES", input "img_xanes", which is attribute name of 2D XANES image stack.
In "Dataset for Energy", input "X_eng", which is attribute name of XANES energy list.
Then click "Load image" button to load the example image file "NMC_LLZO.h5".
Note:

i. It supports image binning to reduce the image size, simply by clicking the "XANES Binning" button.
ii. Example of manual input of an energy list: self.xanes_eng = numpy.arange(8.2, 8.5, 0.001),

Image alignment (Tab: Prep.) (see Fig. 1):

1. Draw ROI: click "Draw ROI", and then use mouse to drag an area in the image display window to define the ROI. Multiple ROIs will be listed in the "ROI list", with names of "roi_0", "roi_1", etc.
2. To align image using ROI, in the "ROI index" field, input the index of ROI that will be used to align with, e.g., if using roi_0, input digit "0" in that field.
3. Select the reference image. Generally, the reference image should have good contrast. E.g., input "100" in the "Ref. Image" field, for which it will use the 101th image as reference. (Note: image index starts with 0, so "100" refers to the 101th image)
4. Choose the alignment algorithm, e.g., "StackReg"
5. Click the "Align Img (ROI)"

Note: most of the processed image data will be named as "Image updated" in the image display window.

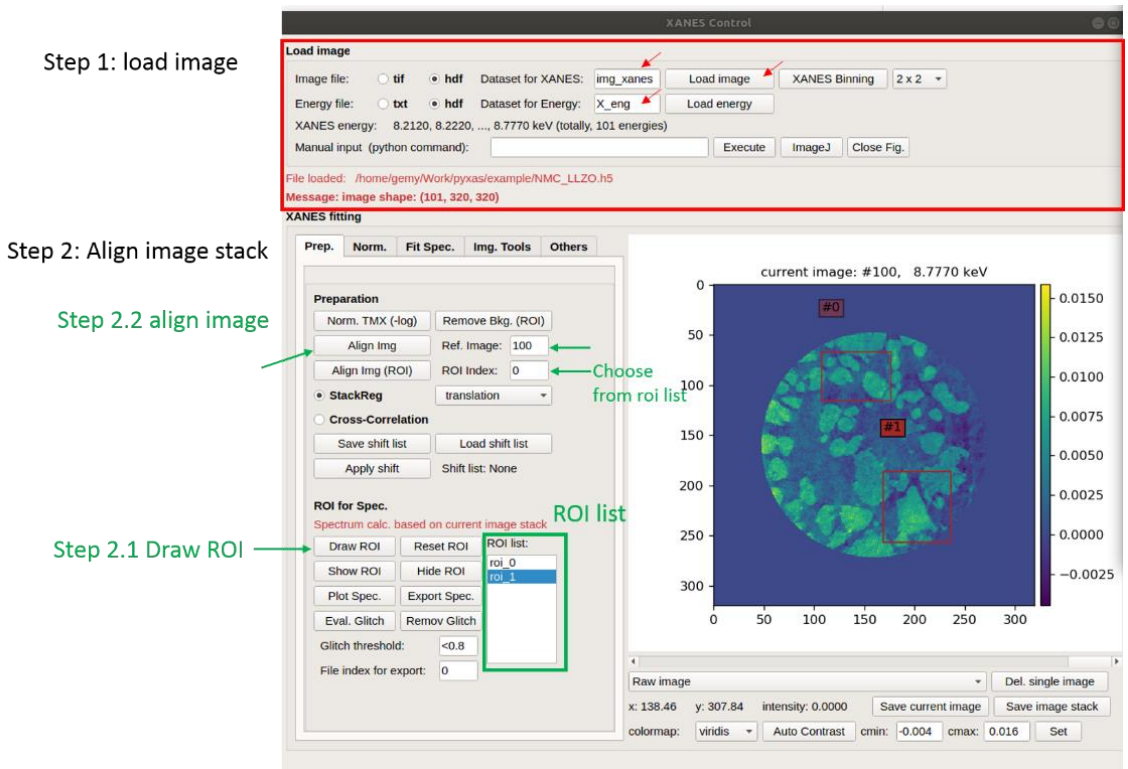


Fig. 1. Screen shot of "load image" and "align image stack"

Edge normalization (Tab: Prep. & Norm.) (Fig. 2, Fig. 3)

Special Note: if the XANES images are suffered from a lot of noises, e.g., pre-edge and post-edge is not smooth to fit with lines, you can skip the edge normalization, and directly to the next step to fit 2D XANES. There are parameters to fit the spectrum background.

Otherwise, go through the following steps to perform edge-normalization:

1. Take -log of image if necessary. For the current example, we don't need to take negative natural log. For regular TXM XANES image, negative natural log is required, by clicking "Norm. TXM (-log)" button.
2. Check energy range of pre-edge and post-edge **(Fig. 2)**
   Select the one of the ROI listed in the "ROI list", click "Plot Spec.". For this example (see ROI spectrum below), pre-edge can be defined with energies below 8.33 keV, and post-edge can be defined for all energies above 8.4 keV.

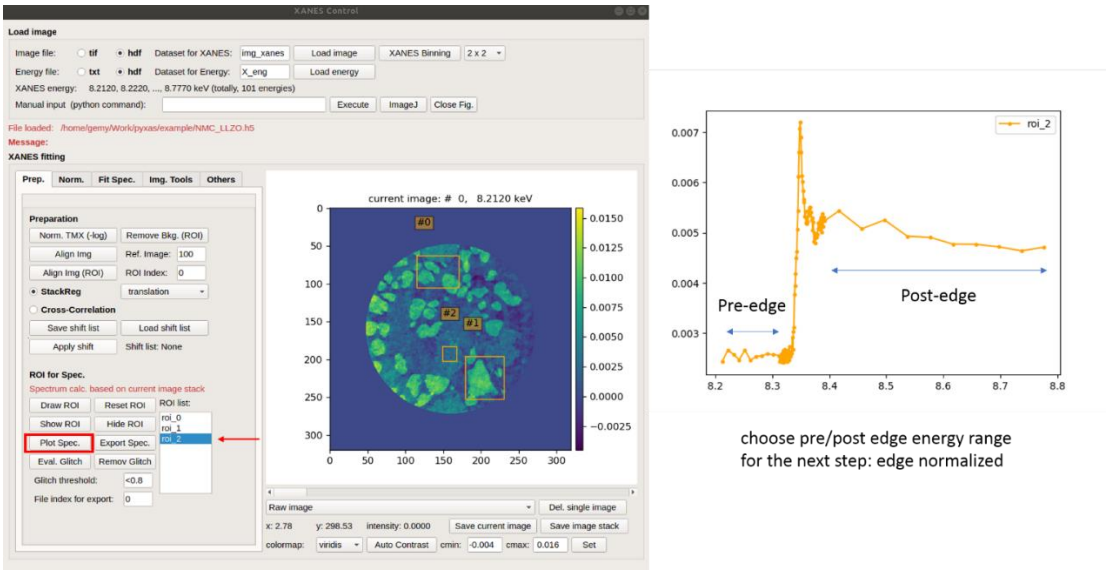


Fig. 2. Draw ROI and plot the spectrum, determine the energy range for pre/post absorption edges.

3. Under the "**Norm**." tab, input the pre and post-edge energy range in the relevant fields: "Pre-edge start/end" and "post-edge start/end" **(Fig. 3)**

4. To check how does the edge normalization work, click "Norm Spec (ROI)". It will plot the raw spectrum and edge-normalized spectrum averaged from the ROI region as selected.
   Note: if this is material that will be used as a reference material, click the "Save Spec" button to save the normalized spectrum for use as the reference spectrum required for the subsequent XANES fitting. (Fig. 3)

5. Once we are satisfied with the defined energy range of pre/post edge, we can perform the edge normalization for the whole image, simply by clicking "Norm Image" **(Fig. 3)**
   Note: we provide two methods for edge normalization.
   i. If image is noisy, use method 1. It will calculate slope of the absorption curve of pre/post range by averaging all the pixel values from areas that contains materials and keep the slope unchanged and then fit the offset of the absorption curve for individual pixel.
   ii. If image has good signals, use method 2. It will calculate the linear slope and offset of absorption curve individually for each pixel.

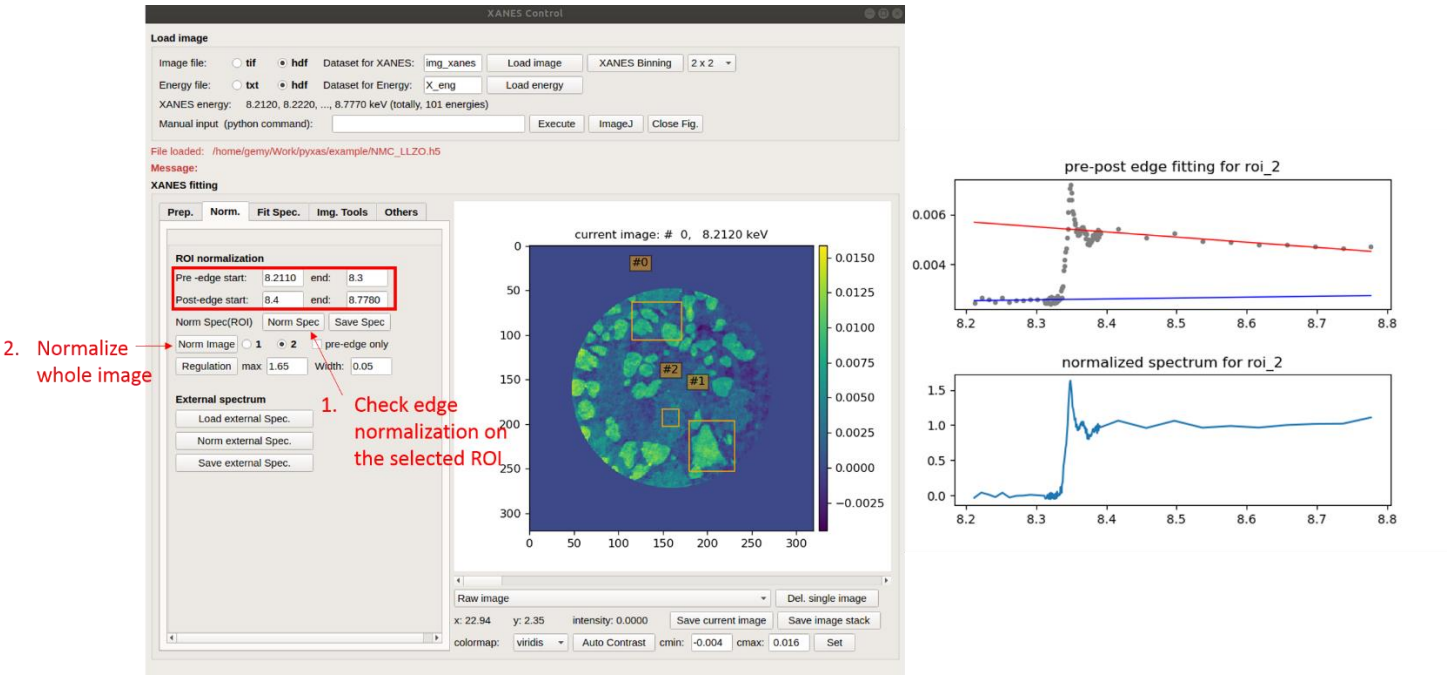


Fig. 3. Edge normalization on selected ROI (e.g., ROI2) and the whole image. Left: screen shot of package interface. Right: edge-normalization for ROI2

2D XANES fitting (Tab: Fit Spec.) (Fig. 4)

1. Load reference spectra: click "Load Ref." to load "ref_NiO.txt" and "ref_LiNiO2.txt"
   We can assign labels for these two ref. spectra. E.g, input "Ni2, Ni3" in the "Elem." filed.

2. Click "Fit 2D"
   Note 1: The fitting will use the image data which is currently displayed in the image display window. Make sure we selected the proper dataset. Mostly, we should use "Image updated".
   Note 2: "Baisc" uses least-square fitting. "NN-ADMM" performs non-negative iteration method, which is slower than "Basic" method.
   "Bkg. polynomial": choose the polynomial orders to fit the spectrum background. Suggest checking "1" and "0".
   "Maximum iteration" and "updating rate" apply to "NN-ADMM" method. Usually, we can leave to default values.
   After fitting, additional image datasets will be added in the image display window: "**XANES Fit thickness**", "**XANES Fit (ratio, summed to 1)**", "**XANES Fit (Elem, concentration)**" and "**XANES Fit error**".
   Note: XANES Fit (Elem, concentration) = XANES Fit (ratio, summed to 1) x XANES Fit thickness.

3. Fitting evaluation
   We can view the "XANES Fit error" to check the fitting quality.
   We can also check the fitting results from specific ROI region. E.g., if we would like to check ROI 0, put "0" in the "ROI #" field. Then click "Plot ROI fit". It will plot both the experimental data and fitted curve. A good match indicates a good fitting.

4. The fitting results can be saved in a .h5 file by clicking "Save 2D Fit".

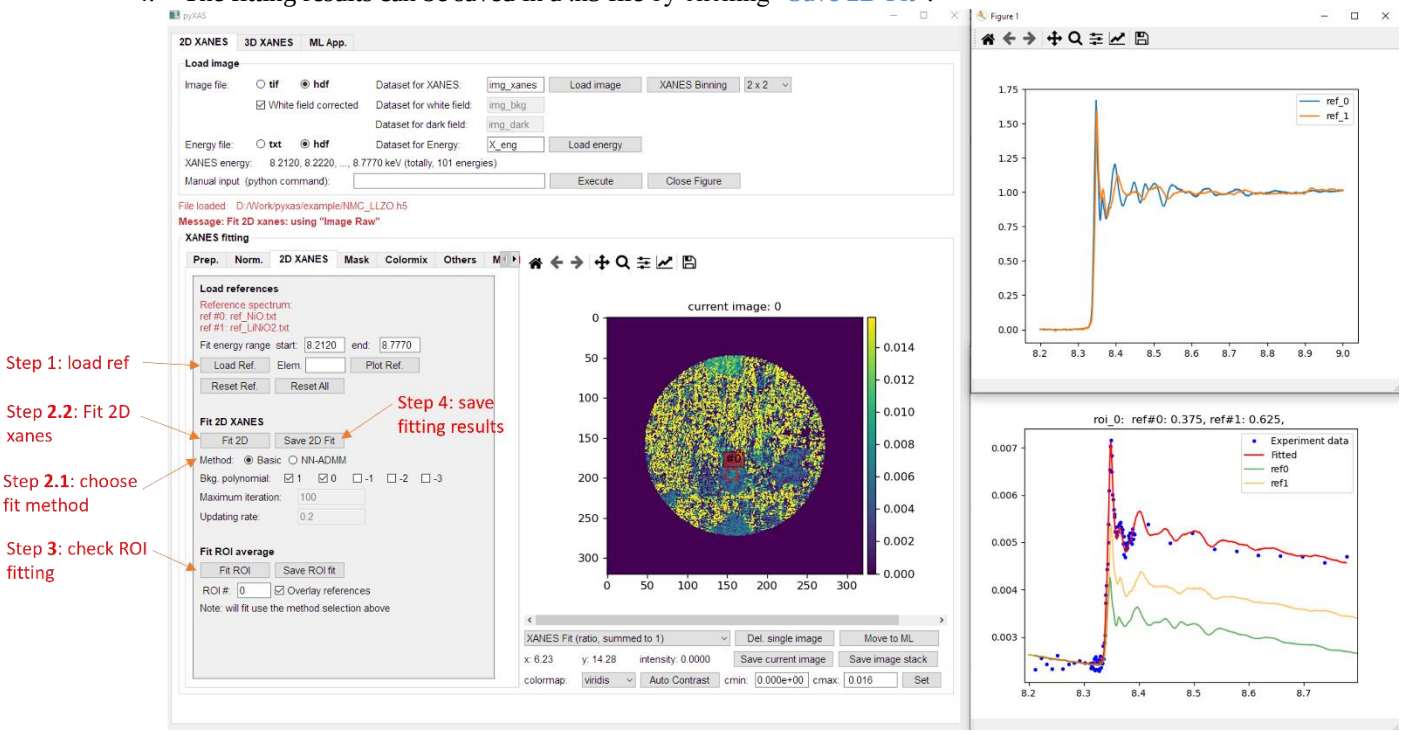


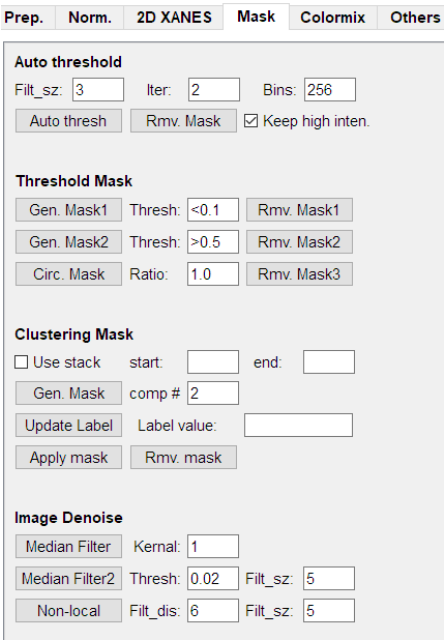
Fig. 4. XANES fitting

Segmentation/Mask (Tab: Img. Tools)

1. **Auto threshold:** using otsu method to generate a threshold mask
2. **Threshold mask**:

   E.g., generate mask from "XANES Fit error": display the "XANES Fit error" and select a threshold value. E.g., we would like to remove regions with fitting error larger than 0.3. Then we can write ">0.3" in the field of "Gen. Mask1(2) Thresh:", then click "Gen. Mask1(2)" button.

   Note: the mask will be applied to all the images in the package. To remove the mask, simply by clicking "Rmv, Mask1(2)"



3. **Clustering mask**:
   i. Choose the "Raw image" in the image display window.
   ii. Under "Clustering Mask", check the checkbox "Use stack". If we would like to use part of image stack, e.g., from image #20 to #40, put "20" in "start:" and "40" in "end:".
   iii. Put the desired number of components, e.g., "4" in the field of "comp #"
   iv. Click "Gen. Mask"
   v. A new image dataset "Smart Mask" will be added to the image display window for visualization.
   vi. For the 4 masks generated (because we set comp# to be 4), if we would like to apply the second mask to the image, in the image display window, navigate the slider-bar to the second mask, then click "Apply mask". It will propagate the mask to all images. We can remove it by click "Rmv. mask".

Colormix (Fig. 5)

After XANES fitting, we can use colormix to generate a colored image to represent the distribution of element with different oxidation states, e.g., Ni^{2+} and Ni^{3+}.

i. In the droplist in image display window, select the fitted results, e.g., "**XANES Fit (Elem, concentration)**" or "**XANES Fit (ratio, summed to 1)**"

ii. click "Color Mix". The colored image will be added to image display window. We can save the color image by clicking "Save current image" to save it.

iii. We can also perform batch colormix by loading the tiff files in a folder. Colors will be scaled by assigning CMin and CMax, corresponding to the minimal value you want the image displaying black, and the maximum value you want the image displaying white.

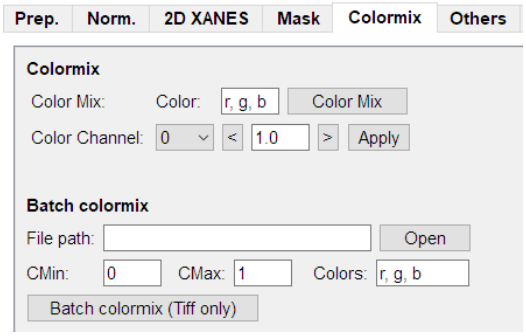


Fig. 5

Fit XANES peak position (Tab: Others) (Fig. 6)

In case we don't have reference spectrum at hand, it is convenient to look at the peak position of XANES spectrum as a first approximation to the variation of chemical state (oxidation state).

1. In the droplist in image display window, select "Image update"
2. Under section of "Other", input the estimated peak position range. See Fig. 6 left.
3. Here, it uses spline curve to fit individual spectrum. It is recommended to test the fitting parameter "smooth" and "order" by fitting ROI spectrum first.
    a. Input the ROI # (assume you have created ROI already, check Fig. 1)
    b. Click "Fit ROI", it will generate a figure showing the fitting results and indicate the peak position. (Fig. 6 right)

    Note: if the "Image update" has very low intensity, e.g., pixel value around 0.001, it is recommended to scale the image intensity by e.g, x1000.
4. To fit the whole image, click "Fit image"
5. Fitted image will be added to the droplist in the image display window
6. (optional) it can scale the fitted image to (0, 1) by assigning the min/max value of peak position.

Note: it also supports to fit external spectrum, by "loading curve" and then "Fit curve" using the parameters entered above.

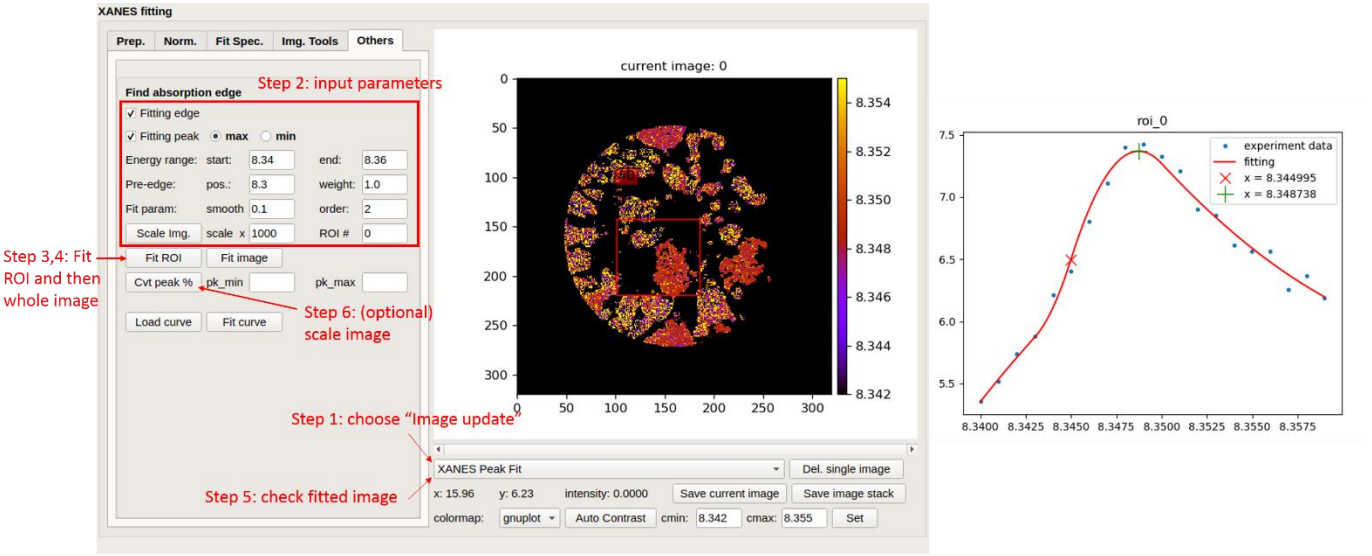


Fig. 6. Fitting peak position of XANES spectrum

3D XANES analysis

Basic steps to analyze 3D XANES:

1. Reconstruct 3D tomography at each energy
2. Align the 3D object
3. Extract the XANES spectrum at each slice of aligned tomography
4. Batch fitting the XANES

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1. Reconstruct 3D tomography at all energies

Refer to the section of "Tomography reconstruction"

2. Align the 3D object

This is the most important and tricky part. In most cases, we may need process the data including:

e.g., crop the 3D image to retain the region of interest (isolate the particle)

e.g., denoise the 3D image or remove the outlier if necessary

In the following, we provide some basic function to do 3D alignment, assuming: only translation shift (no rotation) is involved for 3D object across all energy measurements.

2.1 Load reconstructed 3D tomography (Fig. 7)

- specify the prefix of all files. For example, if all the reconstruction files are named as "recon_xxx", then write "recon" in the text editor of "file_prefix"
- Click the button "Load FXI tomo(s)". If load successfully, the loaded files will be displayed in the box of "Loaded files"

2.2 Align the 3D tomo files (Fig. 7)

- First, specify the reference 3D object to be aligned. Put the index of that file to the edit box "Ref. File". For example, "Ref. File" = 0 means it will use the 3D reconstruction from first file as the reference, and all other 3D reconstructions will be aligned with it. "Ref. File" = -1 means it will use the last one as reference.
- Decide whether it needs bin the 3D image. Write the binning in the edit box "binning"
- Decide whether it needs to apply a circle mask
- Select the aligning method. "Method" = 1 is align using mass center of object. "Method" = 2 is using 3d cross-correlation
- Click "Align tomo files" to start aligning. The progress will be displayed in the terminal.

Aligned file will be saved in the same directory of the raw data with file name as "ali_recon_xxx.h5"

3. Extract the XANES spectrum at each slice of aligned tomography

- First, load the aligned 3D tomography files by clicking the button "Load aligned tomo".
- Specify the range of slices that need to be assembled into xanes image, in the edit box "slices:". Leave it as "[]" if want to assemble all slices
- Specify the "Ref. File" and "circle mask ratio" (similar to the previous section 2.2)
- Click "Assemble XANES" to start. Progress will be displayed in the terminal. Results will be saved in a sub-directory named as "xanes_assemble"

4. Batch fitting the XANES

Open the tab "Fitting". See following section of "Batch fitting" for detailed instruction.

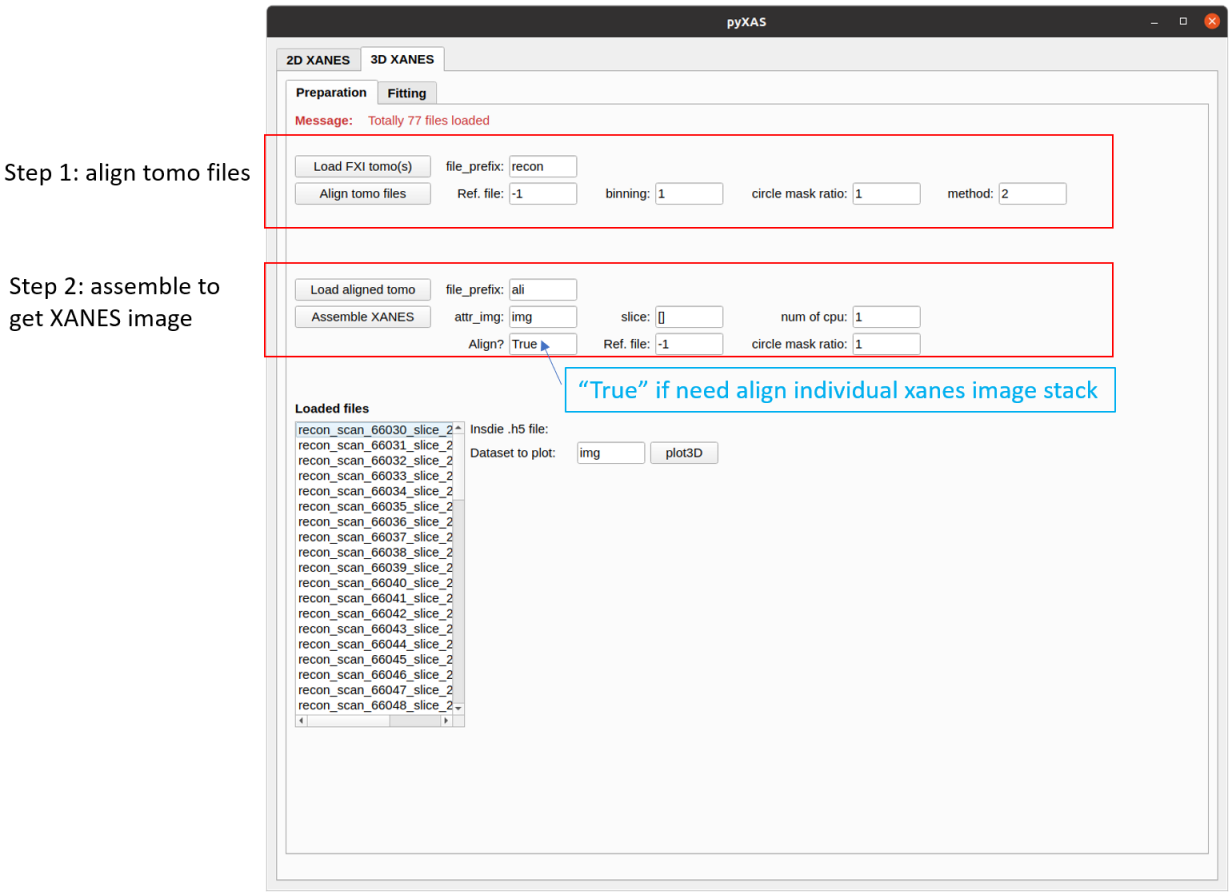
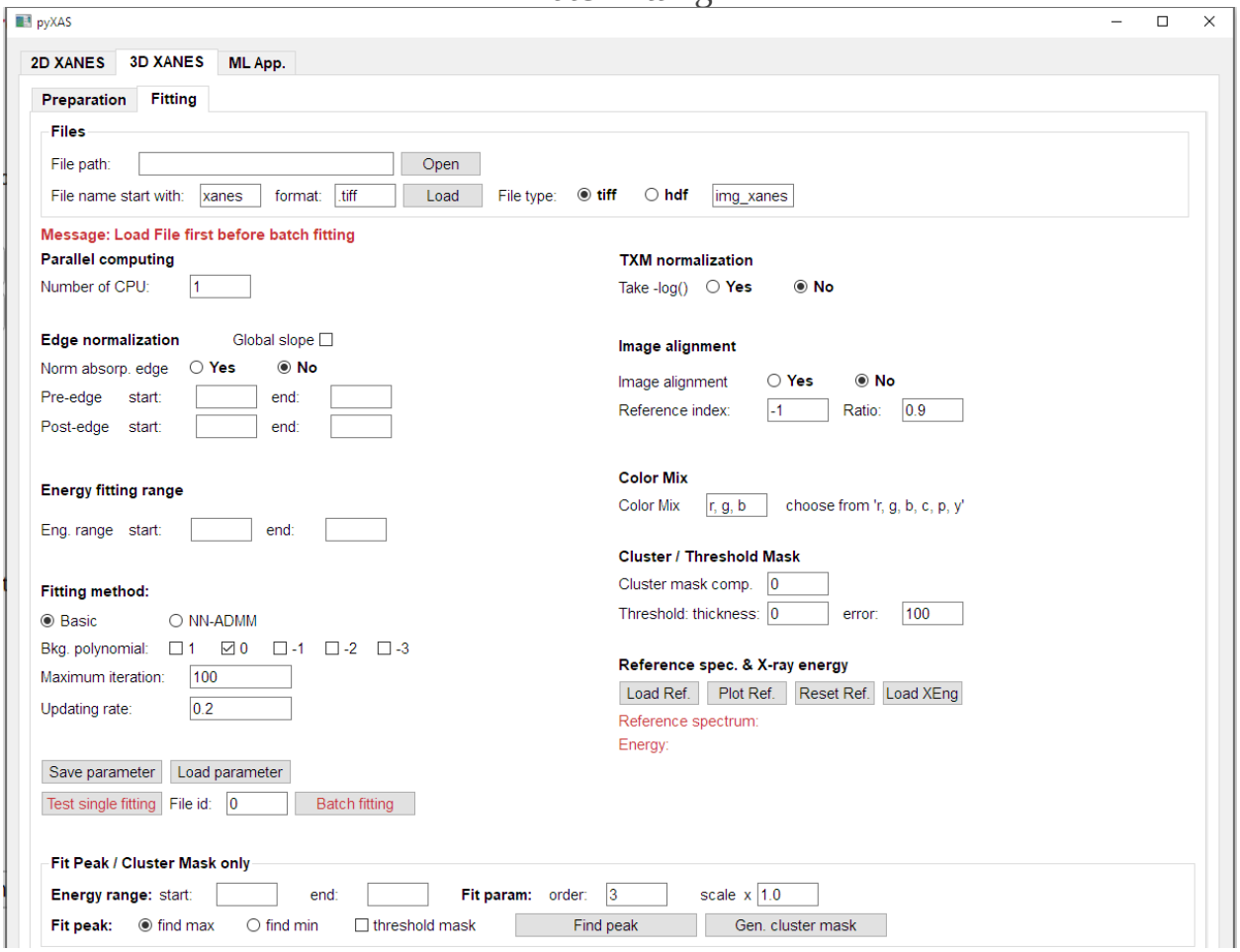


Fig. 7 Tomo alignment and xanse assembling

Batch fitting



Data preparation

Image file:

All 2D xanes image stacks (e.g., xanes1.tiff, xanes2.tiff, … xanes_1001.tiff) are saved in one folder.

Energy list:

Energy list save in a txt file. Assume all xanes images are collected using same x-ray energy list

Reference spectrum:

2-column data (energy vs. intensity) saved in txt files, e.g., Ref_1.txt, Ref_2.txt.

Step-by-step

Load files

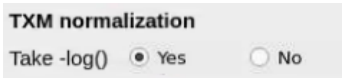
1.1. Click “Open” to direct into the file directory, select one of the xanes image stack (e.g., xanes_1.tiff), then click OK.

1.2. Based on file type, choose “tiff” or “hdf” accordingly. If it is hdf file, write the attribute name pointing to the image dataset, e.g., “img_xanes”, to the textbox on right side of “hdf”

1.3. Click “Load”. All files loaded with success will be displayed on the “Message” line

TXM normalization

Depending on type of data to choose whether need to take -log(). E.g., for regular TXM xanes, we need to take -log(). For 3D XANES tomography (e.g., 3D reconstructed data taken at different x-ray energies) does **NOT** need to take -log().



Edge-normalization

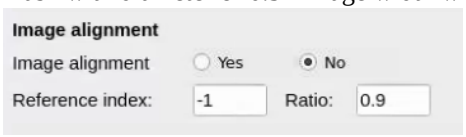
Choose "yes" if need to perform edge normalization. Input the energy range for pre-edge and post-edge. Unit is keV.

Note: If choose "Global slope", it is equivalent to use edge normalization method "1" in the previous related section in 2D xanes analysis using xanes_2D.py. (page 3)

Image alignment

Choose "Yes" if need to align the image stack. Input the which image will be used as reference. "-1" means the last image in each image stack will be used as reference, and all other images in the image stack will be aligned with respect to it.

You can also apply a circular mask to the images, using parameter "Ratio". Ratio=0.9 means a circular mask with diameter of 0.9x image width will be applied



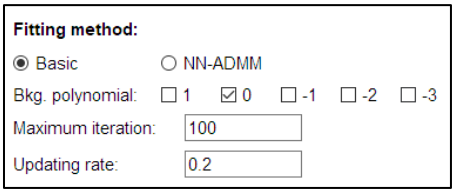
Fitting

Parameters used for fitting. Choose from "Basic" or "NN-ADMM" method.

Specifically, "**Baisc**" uses least-square fitting. "**NN-ADMM**" performs non-negative iteration method, which is slower than "**Basic**" method.

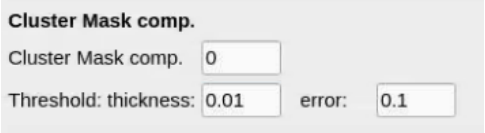
"**Bkg. polynomial**": choose the polynomial orders to fit the spectrum background. Suggest checking "1" and "0".

"**Maximum iteration**" and "**updating rate**" apply to "**NN ADMM**" method. Usually, we can leave to default values showing here.



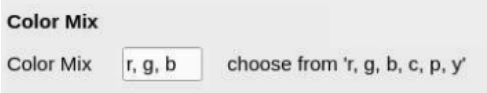
Mask

1. If "Cluster Mask comp." > 0 (e.g., 2), it will use clustering method to create mask to segment the image into (e.g., 2) masks. Check the paper (J. Synchrotron Rad. 2020, 27, 567) for details. The clustering segmentation will be applied based on the aligned images (if image alignment is chosen) otherwise on the very raw images loaded.
2. Threshold mask: mask be calculated by the threshold value on the "thickness image" and "fitting_error" image. See previous "Segmentation" section on page 4.



Color mix

Two types of colormix images will be generated based on “xanes_fit_ratio” and “xanes_fit_concentration”



Reference and energy list

Load the reference spectrum and energy list.

Note: all xanes files must have the same energy list during experiment.

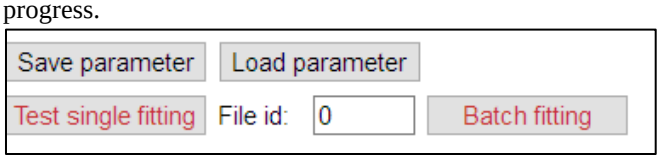
Save parameter and batch fitting

Suggest saving the parameter first, before click “Batch fitting”

We can also load previously saved parameter file.

“**Test single fitting**”: since we have loaded multiple 2D xanes files, we can fit the individual one to check the fitting quality, by assigning the “File id”. File-id=0 will fit the first loaded 2D xanes file.

“**Batch fitting**”: once we are satisfied with the parameters, we can do batch fitting. Check terminal for progress.



File saving

Finally, all fitting files are saved in the subfolder “Fitted”.

Note: keep eye on the output in the command terminal. If you want to terminate the program, using “ctrl + C”

Fit peak position only

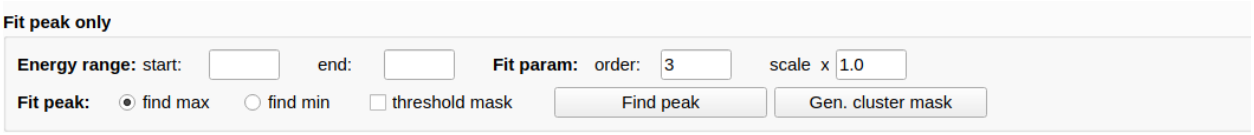
You can also choose to fit the peak position without using reference spectrum.

Currently, it use polynomial with specified order to fit the peak profile

Specify the energy range (start and end).

Specify the order of polynomial. Then click “Find peak”

As a separate function, you can create a mask using clustering method, simply by clicking “Gen. cluster mask”. Note that it will use the dataset within the specified energy range.


Fit peak only
Energy range: start:
end:
Fit param: order: 3
scale x 1.0
Fit peak: find max
find min
threshold mask
Find peak
Gen. cluster mask