Supporting information for article:

Texture measurements on quartz single crystals to validate coordinate systems for neutron time-of-flight texture analysis

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S1. Detailed procedure for refinement of Quartz single crystal data.

These instructions expect some basic familiarity with the MAUD software, e.g. knowledge of the powder data analysis procedure described by Wenk, Lutterotti, and Vogel, Powder Diffraction **25** (2010) 283-296.

The data files and HIPPO parameter files used in this study are provide on the Berkely website: https://seismo.berkeley.edu/~wenk/TexturePage/MAUD.htm, click on the link next to "Download HIPPO-Coordinates:" for the file "HIPPO2023.zip". Unzip the files into a folder for this project.

1) In MAUD 2.9992 (some of the features described here are only available in this or later versions), go to File - New - HIPPO Wizard
2) Enter a sample name (e.g. "qtz sxtal"), load calibration file (hippo_sc_151003_45panels.prm)
3) Choose and load data files (select 30221.gda, hit shift, select 30223.gda highlighting all three files, then click Open)
4) For 30222 enter Omega(set) 67.5, For 30223 enter Omega(set) 90. Then "Next"
5) Choose d-range as 0.7-3.0 and disable bank 39 by making sure the checkbox next to "Bank 39 enabled" is not checked. Data for this sample was collected within a couple of minutes, therefore data above 3A in d-spacing is insufficient to be used. Make sure the box "treat rotations as separate instruments" is not checked, then click "Finish." After a few seconds, MAUD should show the raw data.
6) To compare the relative intensities between detector panels of a detector bank, click on Graphics – Plot Options, and make sure the checkboxes for "Calibration correction" is unchecked and the intensity scale mode is "sqrt". Click OK when done.
7) In the 2D Plot window of the raw data, inspect the data for each detector bank. Detector panels with substantial differences in observed intensity need to be excluded. These appear as dark stripes (much less intensity) with no peaks or orange/red stripes (much more intensity). For this dataset, exclude the data from panel 9 in the Bank90 detector bank: Highlight the Bank90 dataset, click on the eyeball icon or hit Ctrl-E. Click on the "Datafiles" tab, press the Ctrl key and mark all three occurrences of bank 9, then uncheck the "Enabled" checkbox in the top right corner of the dialog box. Click "OK" and now in the 2D display, the darker stripes without peaks originating from the panel #9 should be gone. Do the same for the data from panel 39 and 41. You may also check that eight background parameters are generated from step 1 in the "Background function" tab of each dataset.
8) In the background section, select "Chebyshev polynomials for background". Choose 3 general background parameters (shared by all histograms in a dataset or bank) and 0 individual parameters (>0 would enable additional histogram specific background parameters). Check that the number of individual background parameters is zero by highlighting an individual .gda file in the "Datafiles" tab and select "Additional par". There should be no parameters listed in the "background pol." tab.
9) In MAUD main window select the "Phases" tab and load the quartz phase file (barrel with left arrow). MAUD comes with a collection of crystal structures stored locally under Documents/maud/examples/structures.mdb. Pick the "Quartz, Si O2, trigonal" entry and click "Choose". Edit the phase after adding it using the eyeball icon or Ctrl-e. Under the "Microstructure" tab select "Options" next to Size-Strain model. Set "crystallite size" to 500Å and the "microstrain" to 0.001. Compared to the default values, these parameters make the peaks slightly broader to accommodate the broader observed peak shape from the single crystal.
10) Calculate the patterns using the calculator icon or hit Ctrl-m. In the "Plot" window, showing the integrated measured and calculated intensities for each dataset, it is obvious that the calculated intensity is far too high.

11) To compare the calculated intensities with the observed intensities in the next step, click on Graphics – Plot Options, and make sure the checkboxes for "Calibration correction" is checked and the intensity scale mode is "sqrt". Click OK when done.

12) Select each individual dataset and click the eyeball (edit), on the "General" tab in the Instrument panel click "Edit" and change the Incident intensity to 0.0001. Press OK on "LANSCE Hippo spectrometer". Switch to the "Background function" tab and change the value of the first background parameter "_riet_par_background_pol0" to 0.003. Check the box "Chebyshev polynomial" to switch the background from a polynomial to a Chebyshev polynomial similar to the GSAS background function #1. Adjusting the incident intensity and the background to reasonable and non-zero values is numerically more stable and increases the chances of convergence of the Rietveld fit. Click OK. Ensure this is changed for all Banks (144, 90,120, and 60). Calculate the pattern again with the calculator icon or Ctrl-m. Check for all datasets that the calculated intensities are within an order of magnitude of the observed intensities.

13) Select "Parameter list" from the "Analysis" drop down menu or hit Ctrl-l. First select "Fix all parameters" and then "Free backgrounds."

14) In the "Sample tab" select the sample ("Sample x" or whatever was typed in the HIPPO wizard in step 3) and edit it by selecting the eyeball. Select the "Sample position" tab. Next to "Precession error", use the drop down menu to select "Hippo precession". Click the "Options" button next to it and input "4" for "Precession radius" and 110 for "Precession starting angle." These values are estimated from the precession of the crystal. In general, starting at 0 is numerically worse than starting at a non-zero value. Right click both fields and set these parameters to refine.

15) In the "Phases" tab edit the quartz phase and set both "a" and "c" lattice parameters to refinable by right clicking in the fields with the lattice parameter values. In the "Advanced models" tab of the phase menu set "Texture:" to "arbitrary tex". Click OK.

16) Click Analysis, Options, Algorithm Options and set the number of iterations to 6.

17) Before starting the refinement, save the analysis.

18) Start the refinement by selecting the hammer icon or hit Ctrl-r. You should see a reasonable agreement between data and model once the fit has ended.

19) In the "Phases" tab, select and edit the quartz phase. Under "Advanced Models" change the "Texture" model to "E-WIMV." Select "Options" next to it and set "ODF resolution in degrees" to 3.

20) In the parameter list (Ctrl-l), click on "Refine TOF SF" to refine the time-of-flight histogram scale factors.

21) Refine again for 6 iterations. Check the "Wgt'd ssq" and "Wss" values by scrolling up in the output window (below Datasets/Phases/Sample window). If the values are changing by more than 1%, the fit is not converged, and one should refine another 6 iterations.

22) Refining the diffractometer constants, essentially refining the sample position, using the "Refine all DIFC/shifts" button in the parameter list could be done, but does not reduce the residuals significantly since the HIPPO sample precession model captures the majority of the detector panel specific shifts with much less parameters.

23) In "Graphic" on main page you can now with "Texture plot" plot pole figures, e.g. the 200, 003, and 202 pole figures.

24) In "E-WIMV" Options you can export ODF in MTEX format or BEARTEX format for further processing.

25) To inspect the ODF, e.g. to identify the Euler angles of the main texture components, click Graphics – Texture Plot or hit Ctrl-t. In the "ODF Map Plot" section, chose for alpha/gamma the range 0 to 120 degrees in 5 degrees increment and for the beta the range 0 to 90 with a 5 degree increments. Click "Plot ODF" with "2D" selected next to the button. In each panel, the alpha angle in on the horizontal axis and the gamma angle on the vertical axis. For each panel, the bottom left corner is (0,0) and the range for each axis is 0 to 120 degrees as selected

before plotting. The panels are the beta angle sections in 5 degree steps as selected starting from zero in the lower left corner and the moving left to right and up. Ignore the alpha and gamma degrees axis which are a artifact of the library used to create this plot which does not support sub-figures. The main intensity should be in the last beta panel, therefore at beta=90. The main component is therefore approximately alpha=115 degrees, beta=90 degrees, gamma=0 (or 120) degrees. A second component occurs at alpha=115 degrees, beta=90 degrees, gamma=60 degrees. This second component is only 60 degrees off from the first one and therefore could correspond to a Dauphine twin. A similar analysis could be conducted in MTEX by exporting the ODF (in BEARTEX format) and using the MTEX function identifying the main texture components.

26) To model the ODF as a standard function, which is much more appropriate for a single crystal, change the texture model for the quartz phase to "Standard functions" in the Advanced models tab and click the "Options" button. In the Spherical Component subpanel, click "add term". Change the name to "main" and enter intensity=1, alpha=115, beta=90, gamma=0, FWHM=3 and leave Gaussian at 0.5 (making the split between Gaussian and Lorentzian component 50:50). Right click on all fields and set to refine except for the intensity field. Add a second component called "twin" but do not change any values. Click OK to close the Standard Functions panel and the phase panel.

27) Open the parameter list and navigate to the quartz phase objects. In the Standard Functions object, find the "twin" object. Change the Status of the twin object by clicking on the "*****" in the Status column and select "Equal To". In the subsequent dialog, select the "main" component from the tree by first navigating to the quartz phase object again. Click "Set bound" and then
"Close". When you now close and open the twin tree, the values for the twin parameters should be identical to the ones of the main object. Modify the status of the "Intensity" parameter of the twin object from "Equal To" to "Refine". This allows to refine the weight fraction of the twin component. Since the twin component has a much smaller volume fraction than the main component, set the value for the intensity of the twin component to 0.02. Click on the "Equal To" status of the gamma angle of the twin and click on "Equal To" in the list that appears. In the tree that opens, the value of the twin is already bound to the value of the main component. However, it is set to be identical. It needs to be 60 degrees off, therefore, select 1.0 as multiplier and 60 as offset. Click "Set Bound" and "Close". If you open and close the twin tree or scroll around, the gamma value for the twin should be updated to 60 degrees.

28) Close the parameter list and calculate the patterns. The agreement of the calculated patterns should be very close to the experimental ones. Refine for 6 iterations.

29) The agreement should be good, but not perfect. The differences between calculated and experimental data now require further investigation e.g. whether calculated diffraction intensity is missing in the experimental data because the direction of diffracted neutrons points between [3]He detector tubes. Most discrepancies are additional calculated intensities that are missing in the experimental data. Furthermore, MAUD assumes detectors as points rather than the 2D objects they are on the HIPPO instrument. The parameter in the model capturing the spatial extent of the detector panels is the FWHM in the standard functions. This parameter has to match all different geometries of the HIPPO detector panels and therefore some observed intensity might be missed. Such effects can explain why experimental intensity is missing in the calculated data.

Table S1 Convention used by MAUD for definition of crystal lattice parameters with respect to cartesian coordinate system.
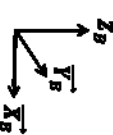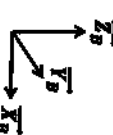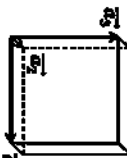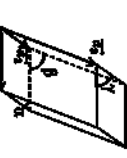
CS	basic vectors lengths $|a_1|\,|a_2|\,|a_3|$	angles $\alpha_{23}\,\alpha_{13}\,\alpha_{12}$	Starting set of K_B	
cubic	a a a	$\frac{\pi}{2}\ \frac{\pi}{2}\ \frac{\pi}{2}$	$e_x \parallel a_1$, $e_y \parallel a_2$, $e_z \parallel a_3$	
tetragonal	a a c ($a \neq c$)	$\frac{\pi}{2}\ \frac{\pi}{2}\ \frac{\pi}{2}$	$e_x \parallel a_1$, $e_y \parallel a_2$, $e_z \parallel a_3$	
orthorhombic	a b c ($a < b < c$)	$\frac{\pi}{2}\ \frac{\pi}{2}\ \frac{\pi}{2}$	$e_x \parallel a_1$, $e_y \parallel a_2$, $e_z \parallel a_3$	
monoclinic	a b c ($a < b$)	$\frac{\pi}{2}\ \frac{\pi}{2}\ \gamma\ (\gamma < \frac{\pi}{2})$	$e_x \parallel a_1$, $e_y = e_z \times e_x$, $e_z \parallel a_3$	

CS	basic vectors lengths $|a_1|\,|a_2|\,|a_3|$	angles $\alpha_{23}\,\alpha_{13}\,\alpha_{12}$	Starting set of K_B	
triclinic	a b c ($a < b < c$)	$\alpha\ \beta\ \gamma$ $(\alpha,\beta,\gamma < \frac{\pi}{2})$	$e_x = e_y \times e_z$, $e_y \parallel a_3 \times a_1$, $e_z \parallel a_3$	
hexagonal	a a c	$\frac{\pi}{2}\ \frac{\pi}{2}\ \frac{2\pi}{3}$	$e_x \parallel a_1$, $e_y \parallel a_3 \times e_x$, $e_z \parallel a_3$	
trigonal	a a a	$\alpha\ \alpha\ \alpha$ $(\alpha < \frac{2\pi}{3})$	$e_x \parallel a_1' = a_1 - a_2$, $e_y \parallel e_z \times e_x$, $e_z \parallel a_3' = a_1 + a_2 + a_3$ trig: a_1' hexa: a_1'	

Table S2   Parameters added to Maud to properly represent HIPPO data in the MAUD coordinate system.

Parameter	Value
PlotPF.NWSE	N
Testing.HippoInvertDetzmInImport	true
Testing.HippoShiftDetzmInImport	180
Testing.invertEta	false
Testing.useNewRotationMatrices	true