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V6

Neutron Texture Analysis with HIPPO at Your Fingertips

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One of the advantages of the multidetector neutron time-of-flight (TOF) diffractometer, HIPPO, at the Los Alamos Neutron Science Center (LANSCE), is the capability to measure preferred orientation of bulk materials. A routine method for measurements, both at ambient conditions as well as high temperature, has been established [1,2]. But only recently has the complex data analysis been streamlined to make it straightforward for a non-initiated user. Here we describe the Rietveld texture analysis of HIPPO data with materials analysis using diffraction (MAUD) as a step-by-step procedure, and illustrate it with a metamorphic quartz rock.

Introduction

The orientation of crystals in an aggregate has a profound influence on physical properties, most importantly, anisotropy. Texture has long been studied in metals as well as rocks (see [3] for details and applications). Various techniques are used to determine texture, most commonly x-ray, electron, and neutron diffraction. Neutron diffraction has unique advantages because of high penetration. The information derived from the diffraction data is averaged over a large volume (typically in the order of a cubic centimeter) and special sample environments are possible. TOF neutron diffraction is increasingly used to record textures of bulk materials, providing the additional advantage of determining textures of low symmetry phases and composites with complex diffraction patterns; TOF diffraction does not rely on single well-resolved peaks as x-ray pole figure goniometers do. Diffraction is most efficient with multidetector spectrometers such as HIPPO, the General Purpose Powder Diffractometer (GPPD) (at Argonne National Laboratory's Intense Pulsed Neutron Source (IPNS)), or SKAT (at the Joint Institute for Nuclear Research (JINR), Russia) where each detector records a spectrum of crystals with lattice planes in reflection orientation [4].

To advance from diffraction spectra to quantitative orientation distributions (OD) is not trivial. The Rietveld method, as implemented in the General Structure Analysis System (GSAS) [5] and MAUD [6], has proved to be a very powerful tool. Note that here we are not addressing neutron texture analysis or important applications of HIPPO data in general, but focus specifically on the Rietveld method with MAUD, as applied to HIPPO data, providing an efficient hands-on procedure. MAUD is written in Java and works with a variety of operating systems (Windows, MAC OS X, Linux, and Unix). For our demonstration we use Windows XP. We apply MAUD to a sample of quartz rock measured in the HIPPO automatic sample changer. The example with data and instrument parameter file can be downloaded from the MAUD web site, along with the latest version of the program.

Getting started

When measuring textures, it is of critical importance to know the orientation of the sample relative to the diffraction instrument, and thus the spectra. Samples need to be oriented and then mounted; for example, with an arrow pointing to a notch in the sample holder, in a systematic manner (Figure 1). This yields pole figures relative to the sample coordinate system; in this example, the cylinder axis and the arrow. Samples mounted on the sample holder's metal rod hang into the sample chamber. For texture measurements with HIPPO, typically 720 ^3He -detector-tubes, distributed over 30 detector panels, and arranged on 3 banks (150° , 90° , $40^\circ 2\theta$) are used. The user receives spectra where counts have been added for each panel and are stored in GSAS format (.gda files) (using the routine HDFBinWin at LANSCE). In addition, the instrument scientist provides an instrument parameter file (.prm) obtained by measuring a standard, containing information about the geometry and calibration of the diffraction instrument. With 30 detectors, HIPPO has a large pole-figure coverage (Figure 2a). Coverage can be improved by rotating the sample about a single axis into 4 different positions (Figure 2b). Typical rotations in the sample changer or furnace are $\omega = 0^\circ$, 45° , 67.5° , and 90° . Note that for the sample changer, rotation is counter clockwise, resulting in positive angles for MAUD, while for the furnace rotation is clockwise, resulting in negative angles.

Entering data

1. Download MAUD from the web and install it on your computer (<http://www.ing.unitn.it/~maud/>). You also need a recent version of Java (<http://java.sun.com/javase/downloads/index.jsp>; JVM 1.5 or higher preferred). Java is not installed by default in the Windows operating system. Also, download the demo data set into your computer. Then start MAUD. The menu option "File → New → Hippo wizard" opens a window (Figure 3) that allows you to conveniently import the experimental data and prepare it for the automatic analysis. In the Hippo wizard window:
2. Input a title.
3. Select a parameter file (for this demonstration we use hippo_sc_050819_30panel.prm) by clicking on the "Browse" button next to the "Choose calibration file" field and then select data files (14445.gda to 14448.gda).
4. Assign each data file a rotation angle (in the case of the test sample 0° for 14445.gda, 45° for 14446.gda, 67.5° for 14447.gda and 90° for 14448.gda). The bulk rotation, "Omega offset," (-61.7°) accounts for the HIPPO sample changer oddity that the sample holder notch is displaced from the incident beam direction. Applying this rotation realigns the sample, with the arrow pointing at the incident beam in the reconstructed pole figures. Click on each file again to verify that the angles were assigned correctly.
5. Click "Next" and choose the d-ranges to be considered for each detector bank. This range depends on the number of peaks in your patterns. For instance, if the maximum reflection d-spacing is at 2.3 \AA , choose a maximum d-spacing of 2.5 \AA since a lot of data represents only background. For this example, we use 1 to 2.75 \AA . Data at lower d-spacings do not improve the analysis (in this case) because of the large number of overlaps. Restricting the range also speeds up the

- computation. Generally, you consider each bank and each rotation as a different instrument and refine instrumental parameters (namely the scaling factors and conversion factors for TOF to d-spacing), accordingly. Hence, leave the corresponding option in the dialog checked.
6. “Finish” the wizard. In MAUD you may now inspect your data using the “Plot 2D” panel and by clicking on each “Dataset.” Next click on the “Phases” panel and load phases from a data base ( structures.mdb or CIF compliant files containing crystal structures), or entering your own structure information (space group, lattice parameters, and atomic coordinates). For this example, use “Quartz” from the MAUD data base (Figure 4).
 7. Edit () each phase and in “Advanced models” choose E-WIMV as a texture method. Under the texture options select an ODF resolution of 10° or 15°, which approximates the angular resolution of HIPPO [7].
 8. If you need the sample rotation axis in the center of the pole figure, edit the “Sample” and for “Sample position” choose $\chi = -90^\circ$. This orients the system so that the rotation axis points to the bottom and the incident neutron beam enters from the south of the pole figure. In the final pole figure (Figure 7) you are looking down on the sample as in Figure 1. In the sample-editing window you can enter the volume fractions for polyphase materials in the “Layers” dialog.
 9. Data are displayed on the MAUD main window either as average profile (Plot (Figure 5) or as a color stack of all spectra for a given “Instrument” (Plot 2D) (Figures 4,6). In the Plot 2D panel you can check all “Datasets” by selecting them one at a time in the list.
 10. Note the bottom (first) spectrum of the 90° bank has a very low intensity (Figure 4) and you need to remove it from the analysis. Edit “Datasets,” go to “Datafiles,” and “disable” the first spectrum on the list (#9).
 11. At this point you should save your analysis in your data directory (“File,” “Save analysis as”) as a .par file. Now you are ready to start the refinement.

Rietveld refinement

For simple materials you can use an automated procedure. Under “Analysis” select “Wizard”. In the appearing window choose “Texture analysis” in the right panel and press the button “Go!” You may watch the progress of the refinement in the graphics window (Figure 4). This will refine the most important parameters in four cycles. Select no more than five iterations (this can be adjusted as the refinement proceeds). To guarantee convergence you may, at the end, perform 3–5 additional iterations (selecting from the menu “Analysis”, “Refinement”). The texture wizard works well for single phase materials and clearly defined compounds. For complex materials you need to proceed in individual steps, using the menu at the bottom of “Analysis”/“Parameter list”. Fix all parameters first, then free backgrounds and scale parameters and perform a refinement. Next refine “spectra monitors” and “TOF bank SF” and finally “texture”, depending on how the procedure converges. Below we only consider the “Texture wizard”.

Cycle 1: Refining the scale, background parameters and phase fractions for multiphase systems. The intensity calibration (12 parameters) are directly loaded

from the HIPPO calibration file and then kept constant. A single scale factor is refined for each detector. Backgrounds are expressed with a polynomial. Three coefficients are refined for each bank and in addition two for each panel. Backgrounds start to look reasonable and so does the overall intensity (Figure 5). Note that the high-angle 150° bank detectors (Figure 5a) have a much higher resolution than the 40° bank (Figure 5b), but lower counting statistics. Texture is not reflected in the recalculated spectra (top of Figure 6a). Also, the wavy line is not matched due to miss-positioning of the sample, which is particularly serious for the 40° bank.

Cycle 2: In addition to the previous parameters peak positions are adjusted in during this cycle by refining cell parameters and instrument errors based on observed d-spacings. The waviness is corrected (Figure 6b).

Cycle 3: The microstructural parameters (crystallite size and microstrains) are added to the refinement to adjust peak shapes. Only one overall temperature factor, B , is used for all atoms of a phase to avoid correlations and minimize possible errors. Other structural parameters such as atomic coordinates are not refined with the Texture Wizard. You may refine them separately at the end.

Cycle 4: The orientation distribution is refined. Intensities of individual peaks start to match (Figure 6c). Warning: The interactive Plot 2D display only shows the real calculated pattern at the end of each cycle. In between it may indicate a perfect match, but these are Le Bail factors, not true texture intensities.

The procedure with the Wizard does not automatically refine all possible parameters for all phases. Crystallite size, and microstrains are not refined for phases that are below a certain threshold, and that can be selected in "Analysis → Preferences." After this refinement several checks need to be performed: the overall quality of the least-squares refinement is expressed in R- or Rwp-factors, with 5–15% for a good convergence. These numbers are included in the file "Analysis → Results." There are separate R-factors for texture and overall refinement because the WIMV and EWIMV methods use their own algorithm to fit the experimental texture data extracted with the modified Le Bail procedure [8]. Compare observed (bottom) and calculated (top) intensities for all detectors on the multiplots (Plot 2D, Figure 6c). In the "Datasets" editing window under "Datafiles" select the spectra, one at time, and press the "View" button to get a closer look at observed and calculated intensity spectra. If the results appear reasonable, close the window and from the main menu select "Graphic" then "Texture Plot" to check the pole figures (Figure 7). In this window you can also confirm that you have the correct pole figure coverage (Figure 2).

The Wizard refinement is often sufficient to capture the main texture features. You may wish to continue to refine additional parameters from all the many possibilities offered by MAUD. They include lattice parameters for accessory phases, atomic coordinates, temperature factors, anisotropic grain size, and residual stresses. This can be done in "Analysis → Parameter List," fixing the ones you don't want to refine and refining those

you do by selecting them in the list and changing their status to “Refinable.” Or, you can edit the individual parameters in the individual edit windows (for phases, datasets, instruments, and sample), and selecting them to refine (by pressing the right mouse button when over any of the parameter text fields).

Exporting data

For further processing of the texture data the orientation distribution (OD) can be exported (in the EWIMV window) in Beartex format (.mod) [9,10]. Then, in Beartex the MAUD format is first converted to the standard binary format routine (CMAU). Then it is smoothed to alleviate the rigid cell structure by applying a 7.5° Gauss filter (SMOO). With this smoothed OD you can then calculate pole figures (PCAL) and plot them as shown in Figure 8a (PING). Also you can use this OD to calculate polycrystal physical properties (TENS). As a comparison, we analyzed the same quartzite sample with the scanning electron microscope [using the](#) electron back-scattered diffraction (EBSD) method. For EBSD, 5824 orientations were used, then smoothing with a 15° filter and rotating the EBSD-OD to get it into the same orientation as the neutron pole figures (Figure 8b). The two sets of pole figures are similar, including pole densities, but EBSD data that rely on much smaller numbers of grains show stochastic effects—and are statistically less relevant, underlining the main advantage of neutron texture analysis. An interesting feature of this quartz texture is that 101 poles align perpendicular to the schistosity (center of the pole figure). ~~This feature is caused by mechanical twinning~~[Mechanical twinning causes this feature](#) and the pattern can be used as a paleopiezometer to unravel the stress history. Also, in the resolution of positive (101) and negative rhombs (011), which are required for such analysis, neutron diffraction excels over EBSD. With EBSD some of the pseudohexagonal orientations that define the trigonal symmetry are misindexed.

Conclusions

Rietveld texture analysis of HIPPO-TOF neutron diffraction data with MAUD has been applied to several samples; metals [7] as well as rocks [11,12], including high-temperature conditions inside a furnace [13-15].

No doubt procedures will be further refined, and we hope that this brief introduction will make texture analysis more palatable for uninitiated HIPPO users. Remember the 4 steps: (1) load the HIPPO data with the HIPPO Wizard, (2) enter phase, and (3) refine with the Analysis Wizard (Texture), export the orientation distribution in EWIMV, and calculate pole figures in Beartex. A sample without complexities is finished in 1–2 hours.

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Figures:



Figure 1. Cylinder of rock, marked with an arrow to indicate orientation, mounted on a sample holder.

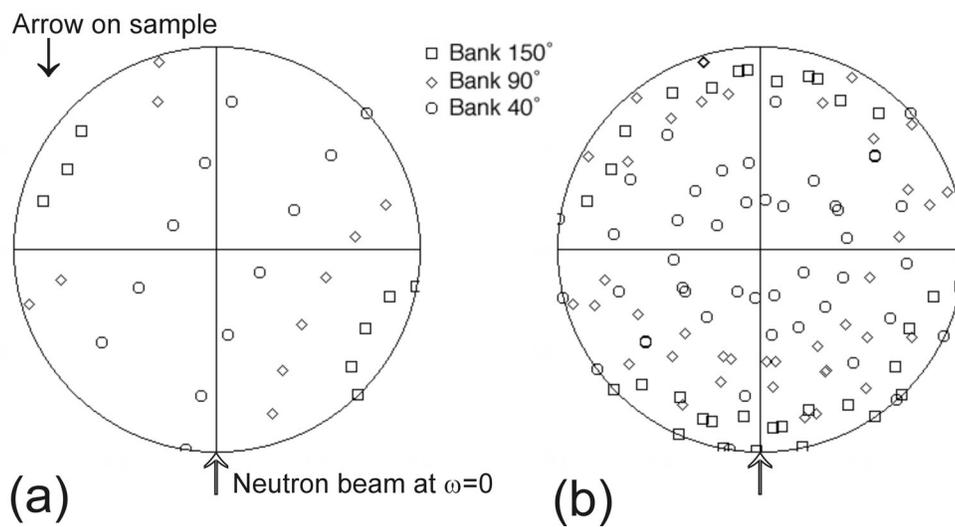


Figure 2. Pole figure coverage for (a) initial orientation ($\omega = -61.7$) and (b) for all 4 orientations to improve coverage. Equal area projection.

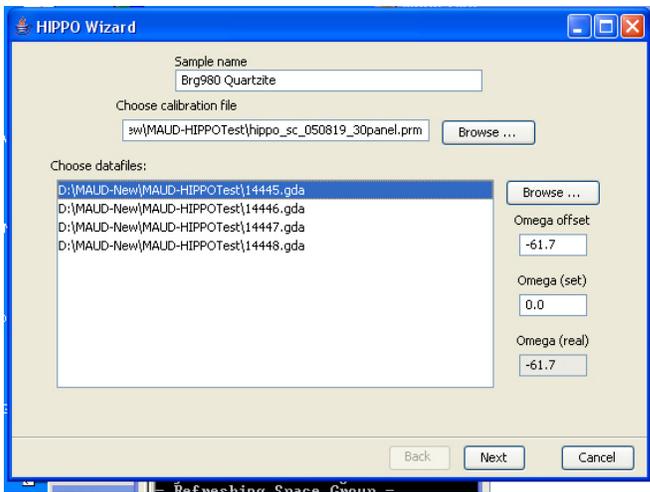


Figure 3. HIPPO Wizard to load instrument parameter file and data files.

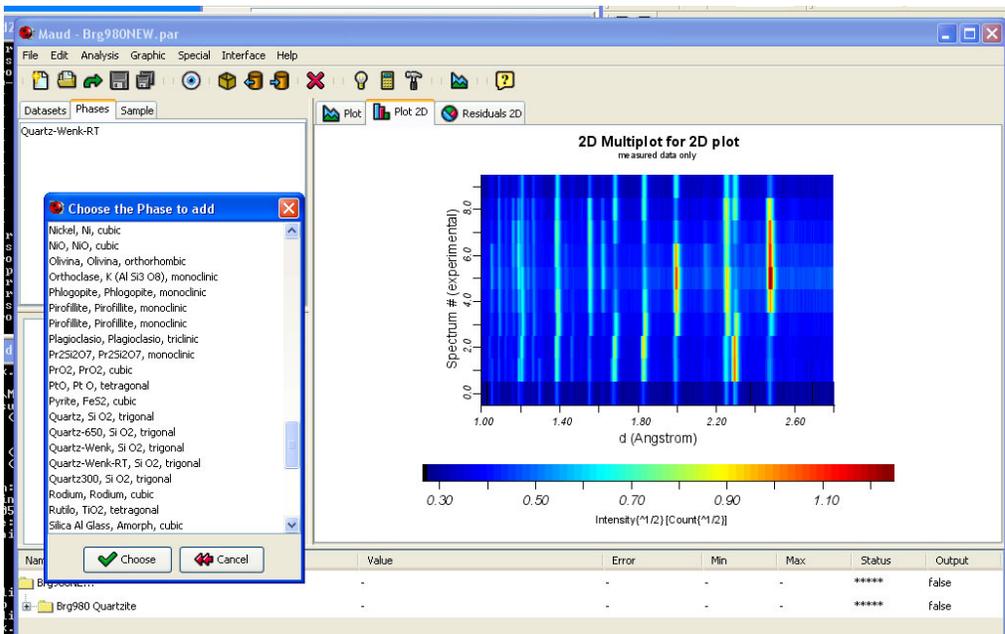


Figure 4. Main MAUD page with Plot2D display for 90° detectors: enter the datasets, disable faulty spectra such as the one at bottom (#9), then enter phases (the data base window is shown) and edit the sample, if needed.

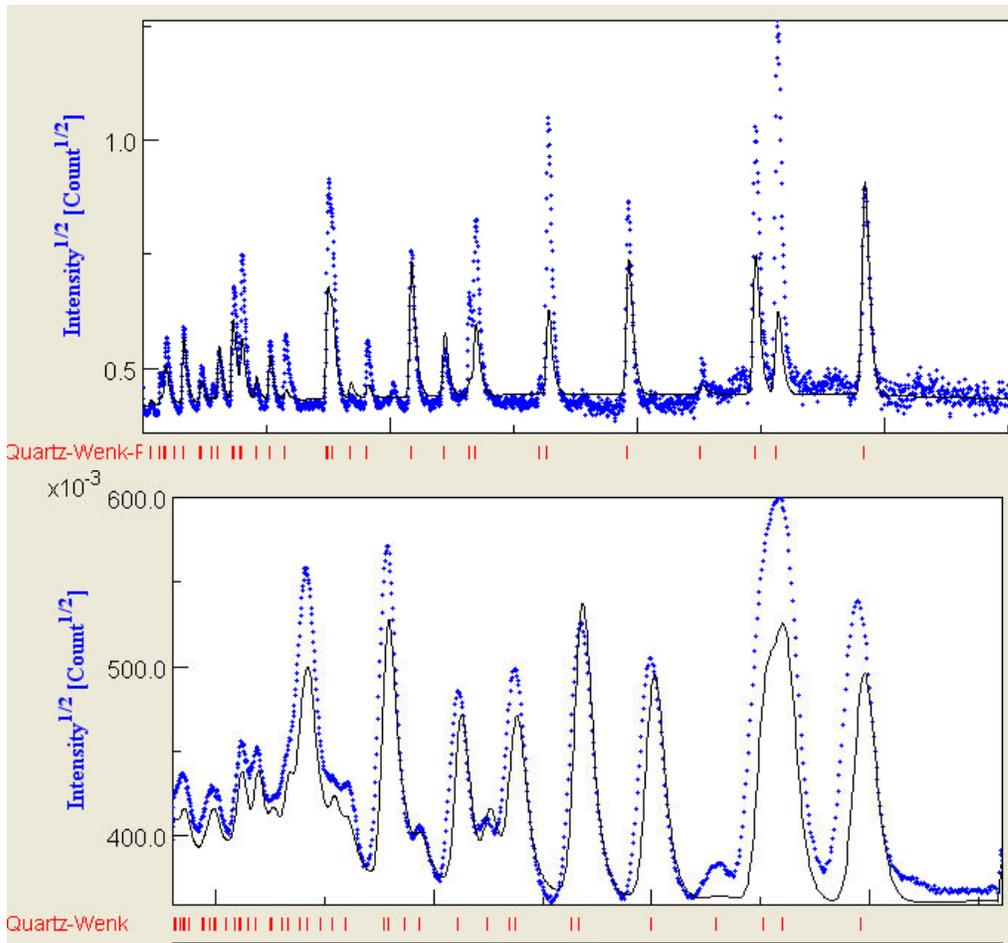
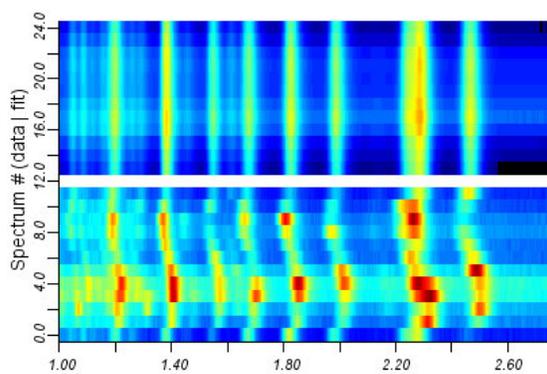


Figure 5. The fitting scale factors and backgrounds for a 150° (top) and 40° (bottom) bank average spectrum (Cycle 1). Note the much higher angular-resolution of the high-angle detector, but poor counting statistics.



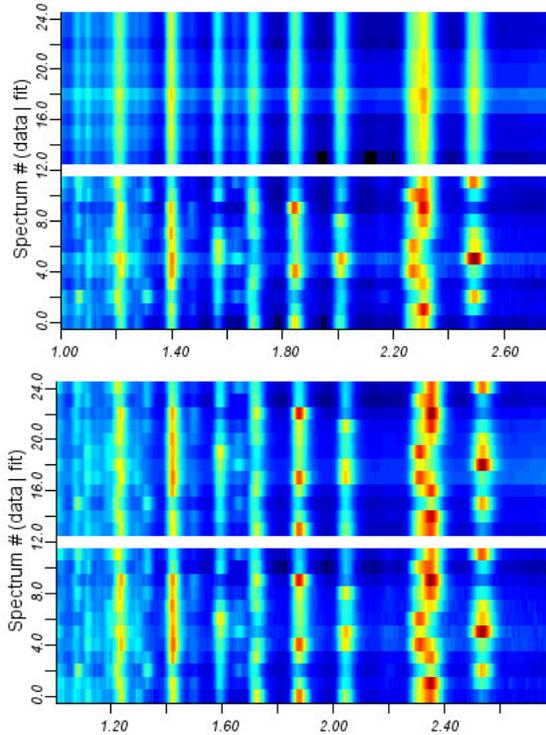


Figure 6. Texture Wizard with multiplots illustrating refinement stages in 3 of the cycles for 40° detectors. Top calculated, bottom observed.

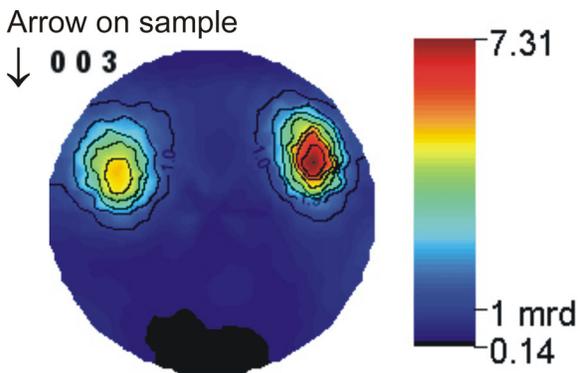
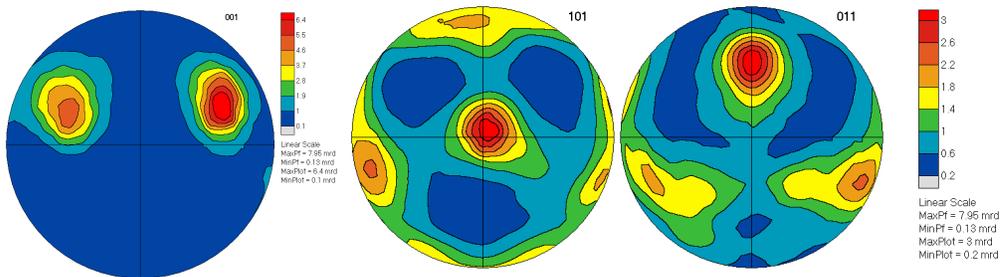


Figure 7. MAUD quartz 0001 (c-axis) pole figure, looking down on the sample as in Figure 1, with the arrow as indicated. Equal area projection, upper hemisphere.



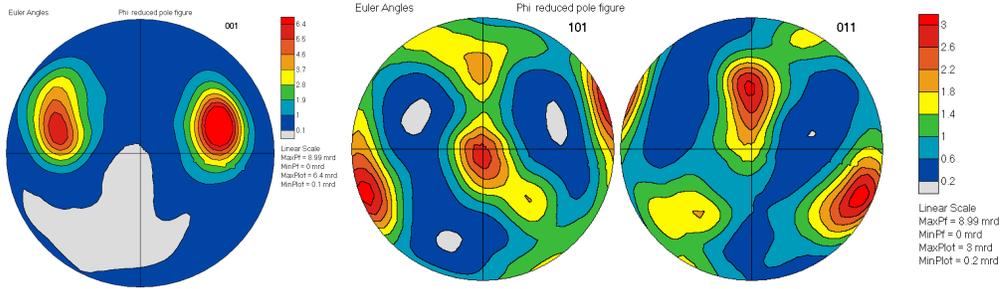


Figure 8. Pole figures of c-axes and positive and negative rhombs of trigonal quartz, recalculated from the OD and plotted with Beartex. HIPPO data (top) and EBSD data (bottom) for the same sample: equal area projection, upper hemisphere.