Correlating Atom Probe Crystallography Measurements with Transmission Kikuchi Diffraction data (Supplementary information)

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# Mass spectrums and ranging.

The following mass spectrums and corresponding ranging were those used for the datasets presented in the main text.



**Figure 1** Mass spectrum and ranging used for Al-0.5Ag (wt.%) sample with Si impurities (Run id: R18\_55344). Data was collected at 40 K using voltage pulsing (20% pulse fraction, 2000 Hz, 0.5% evaporation rate).

/Users/admin/Documents/projects/2016/Tom/mass spectrums/Mo/Mo mass spec - Version 04 - Xaxis line to front and units updated.pdf

**Figure 2** Mass spectrum and ranging used for technically pure Mo sample (run id: R21\_07344). A specimen temperature of 60 K, target evaporation rate of 1% and laser pulsing using a green laser (λ = 532 nm) with 0.6 nJ laser energy at 250 kHz pulse frequency were used during experimental acquisition.

Worked example for calculating disorientation between two grains in an atom probe reconstruction:

The following calculations show how the minimum misorientation, or disorientation, was calculated for the nanocrystalline Al-0.5Ag dataset collected on the straight flight path instrument. The calculation was done based on the information shown in Figure 1 of the manuscript. That is, for a ~ 1 million atom slice of the reconstruction that has been crystallographically calibrated using the methods described by (Gault, et al., 2011; Gault, et al., 2009). It is highly recommend that a spreadsheet or computer script is used to determine the minimum misorientation given the length of the calculation and repeatability of the problem.

From the plane orientation extraction (POE) maps in Figure 1, the normal (pixel with highest intensity) to two sets of planes in each grain can be observed in polar angles.

|  |  |  |
| --- | --- | --- |
| Crystallographic direction (Cc frame) | Observed normal direction (Cs frame) | |
|  | theta () ° | phi ( ° |
| Grain 1 (g1) | | |
| 111 | 16.6 | 6.2 |
| 113 | 10.6 | -20.5 |
| ^ | -15.0 | -5.4 |
| Grain 2 (g2) | | |
| 111 | -25.8 | 10.5 |
| 113 | -12.5 | -19 |
| \* | 57.8 | 50.4 |

^experimentally determined but not shown in manuscript

\*calculated normal vector to the other 2 using the cross product. 202 in grain

From these polar angles the directions can be calculated using the following equations

|  |  |  |
| --- | --- | --- |
|  |  | 1 |

|  |  |  |  |
| --- | --- | --- | --- |
| Crystallographic direction (Cc frame) | Observed normal direction (Cs frame) | | |
|  | u | v | w |
| Grain 1 (g1) | | | |
| 111 | -0.286 | 0.103 | 0.953 |
| 113 | 0.259 | -0.091 | 0.962 |
|  | -0.184 | -0.344 | 0.921 |
| Grain 2 (g2) | | | |
| 111 | 0.435 | 0.164 | 0.885 |
| 113 | 0.216 | -0.318 | 0.923 |
| \* | -0.846 | 0.411 | 0.340 |

\*calculated normal vector to the other 2 using the cross product

The orientation matrix of each grain is defined using the following equation:

|  |  |  |
| --- | --- | --- |
|  |  | 2 |

Where and are the crystal frame and specimen frame respectively and is the orientation matrix. can be thought of as the rotation required to get the directions in aligned with . However, since there are slight errors in the measured orientation of directions in the frame, due to inaccuracies in the tomographic reconstruction, Cs must be calibrated first. To do this, a new set of directions were fitted to those measured within the reconstruction using two restraints:

1. The angles between the fitted directions are equal to the theoretical angles between those crystallographic directions.
2. The residual sum of squares (RSS) of the angles ( between the measured and fitted directions in the frame was minimised.

|  |  |  |
| --- | --- | --- |
|  |  | 3 |

Angle between crystallographic directions (grain 1)

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| V1 | V2 | angle ° (theory) | angle ° (measured) | diff ° |
| 111 | 113 | 29.5 | 26.6 | 2.9 |
|  | 202 | 35.3 | 33.6 | 1.7 |
| 113 | 2 | 31.9 | 29.7 | 1.8 |

RSS (initial)= 2.92 + 1.72 + 1.82 = 14.5

Angle between crystallographic directions (grain 2)

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| V1 | V2 | angle ° (theory) | angle ° (measured) | diff ° |
| 111 | 113 | 29.5 | 30.8 | 1.3 |
|  |  | 90 | 90 | 0 |
|  | 113 | 90 | 90 | 0 |

\*since -220 was calculated using the cross product the angle with the other directions exactly matches the theoretical

RSS (initial)= 1.32 = 1.7

A solver function (such as those found on Microsoft Excel® or MATLAB®) to find a solution with the two defined restraints above.

Grain 1

|  |  |  |
| --- | --- | --- |
| V1 | V2 | degrees |
| 111 (original) | 111 (new) | 1.5 |
| 113(original) | 113 (new) | 1.6 |
| 202 (original) | 202 (new) | 0.9 |

RSS(final) = 1.52 + 1.62 + 0.92 = 5.4

Grain 2

|  |  |  |
| --- | --- | --- |
| V1 | V2 | degrees |
| 111 (original) | 111 (new) | 0.64 |
| 113(original) | 113 (new) | 0.64 |
| -220 (original) | -220 (new) | 0 |

RSS(final) = 0.642 + 0.642 + 0.002 = 1.3

|  |  |  |  |
| --- | --- | --- | --- |
| Crystallographic direction (Cc frame) | Calibrated normal directions using solver (Cs frame) | | |
|  | x | y | z |
| Grain 1 (g1) | | | |
| 111 | -0.294 | 0.127 | 0.947 |
| 113 | 0.273 | -0.086 | 0.958 |
|  | -0.187 | -0.369 | 0.910 |
| Grain 2 (g2) | | | |
| 111 | 0.435 | 0.164 | 0.885 |
| 113 | 0.216 | -0.318 | 0.923 |
| \* | -0.846 | 0.411 | 0.340 |

Grain 1:

|  |  |  |
| --- | --- | --- |
|  |  |  |

|  |  |  |
| --- | --- | --- |
|  |  |  |

Grain 2:

|  |  |  |
| --- | --- | --- |
|  |  |  |

|  |  |  |
| --- | --- | --- |
|  |  |  |

The misorientation, i.e. the transformation necessary to rotate from one crystal orientation to the next, can then be calculated as:

|  |  |  |
| --- | --- | --- |
|  |  | 4 |

To determine the minimum misorientation, or disorientation, between the grains, all orientation variants of one of the grains must be considered. These variants can be found in texture and crystallographic texts such as (Randle & Engler, 2000). For a cubic system, 24 orientation variants exist.

|  |  |  |
| --- | --- | --- |
|  |  | 5 |

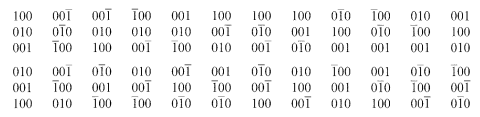


Figure 3. The 24 orientation variants Ti for a cubic crystal. Taken from (Randle & Engler, 2000)

The minimum misorientation angle, i.e., the grain boundary angle, can be calculated from the following equation:

|  |  |  |
| --- | --- | --- |
|  |  | 6 |

The misorientation axis is the real eigenvector of M12, a 3x1 column vector , corresponding to the real eigenvalue :

|  |  |  |
| --- | --- | --- |
|  |  | 7 |

It turns out that for this particular case, orientation variant i = 4 gives the minimum misorientation angle.

Using equation 7

=

Therefore the misorientation axis (in the crystal frame) is:

x = [-0.168, 0.054, 0.984]

Or an equivalent crystallographic direction to match up with the outputted value from TKD:

x = [-0.168, 0.984, 0.054]

Gault, B., Loi, S.T., Araullo-Peters, V.J., Stephenson, L.T., Moody, M.P., Shrestha, S.L., Marceau, R.K.W., Yao, L., Cairney, J.M. & Ringer, S.P. (2011). Dynamic reconstruction for atom probe tomography. Ultramicroscopy **111**(11), 1619-1624.

Gault, B., Moody, M.P., de Geuser, F., Tsafnat, G., La Fontaine, A., Stephenson, L.T., Haley, D. & Ringer, S.P. (2009). Advances in the calibration of atom probe tomographic reconstruction. Journal of Applied Physics **105**(3), 034913.

Randle, V. & Engler, O. (2000). *Introduction to texture analysis : macrotexture, microtexture and orientation mapping*. Amsterdam, The Netherlands: Gordon & Breach.