— Supplementary Information —

Discrimination of aluminum from silicon by electron crystallography with the JUNGFRAU detector

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1 Observed and calculated data for albite and zeolite A

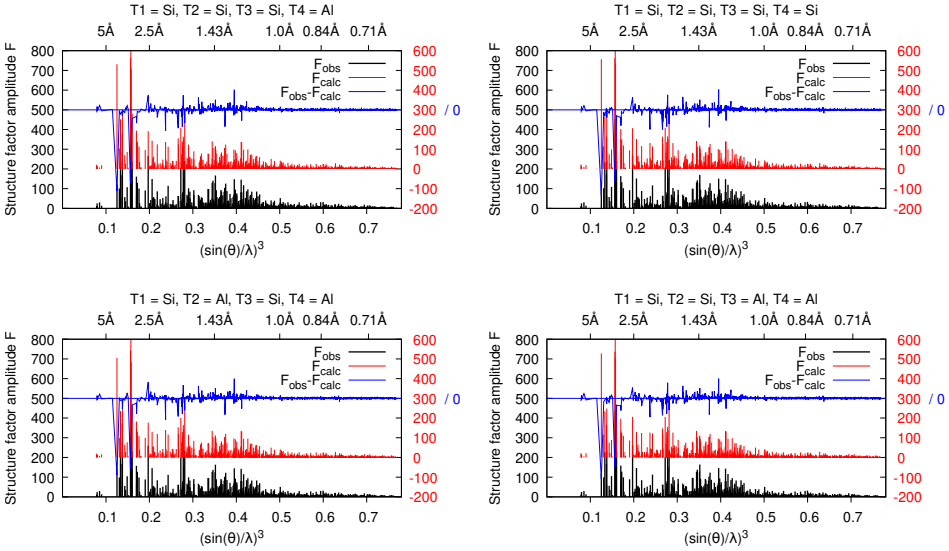

Figure S1: Data for albite structure depending on the T1-T4 compositions. Comparison between observed and calculated data. First graph top left represents the expected composition T1=Si, T2=Si, T3=Si, T4=Al.

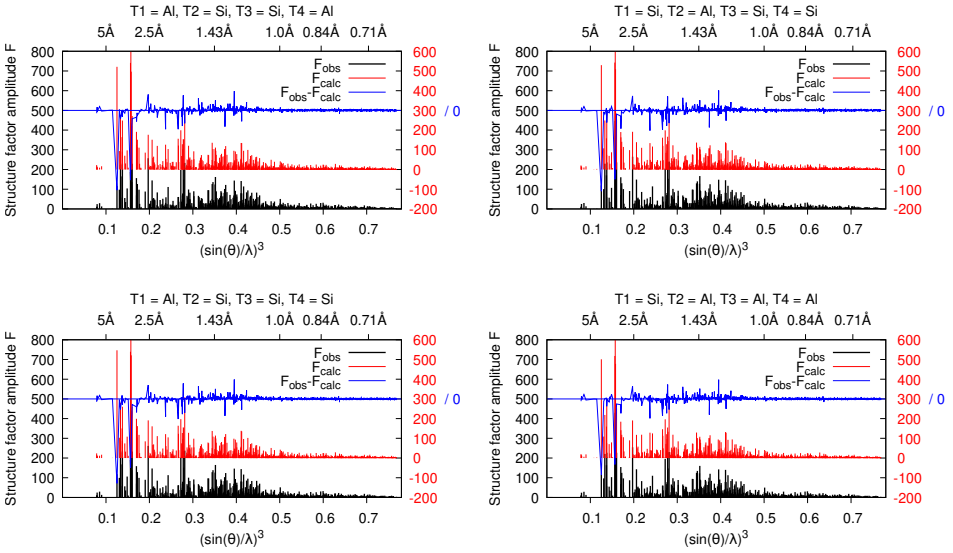

Figure S2: Cont'd: Data for albite structure depending on the T1-T4 compositions. Comparison between observed and calculated data.

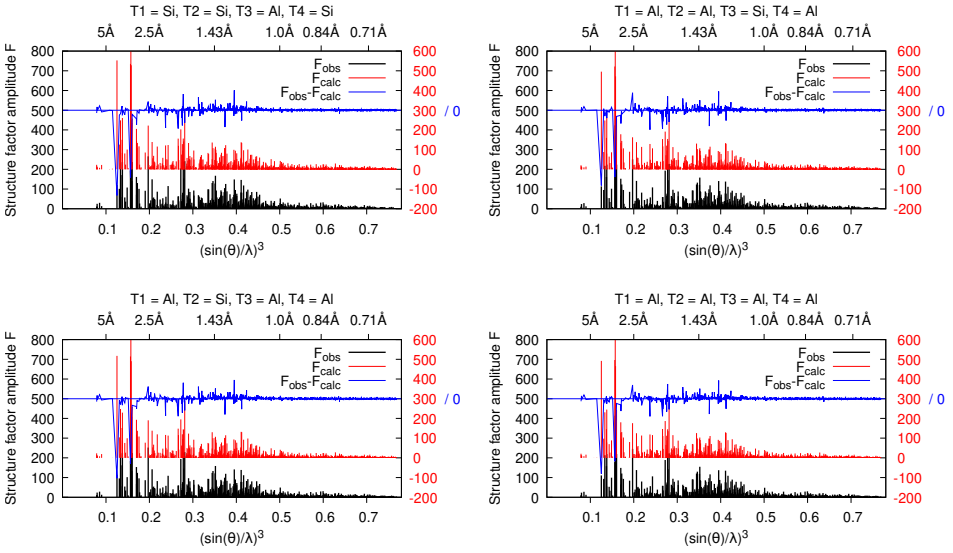

Figure S3: Cont'd: Data for albite structure depending on the T1-T4 compositions. Comparison between observed and calculated data.

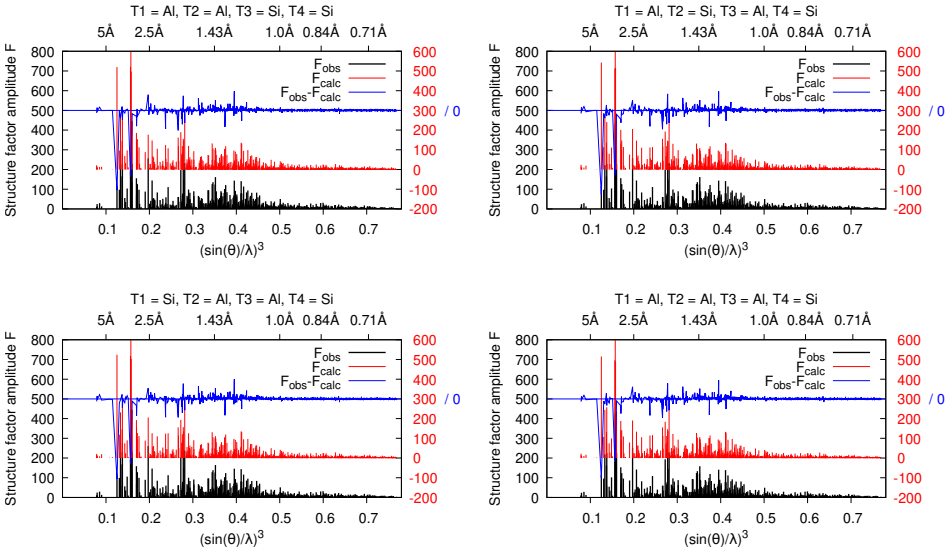

Figure S4: Cont'd: Data for albite structure depending on the T1-T4 compositions. Comparison between observed and calculated data.

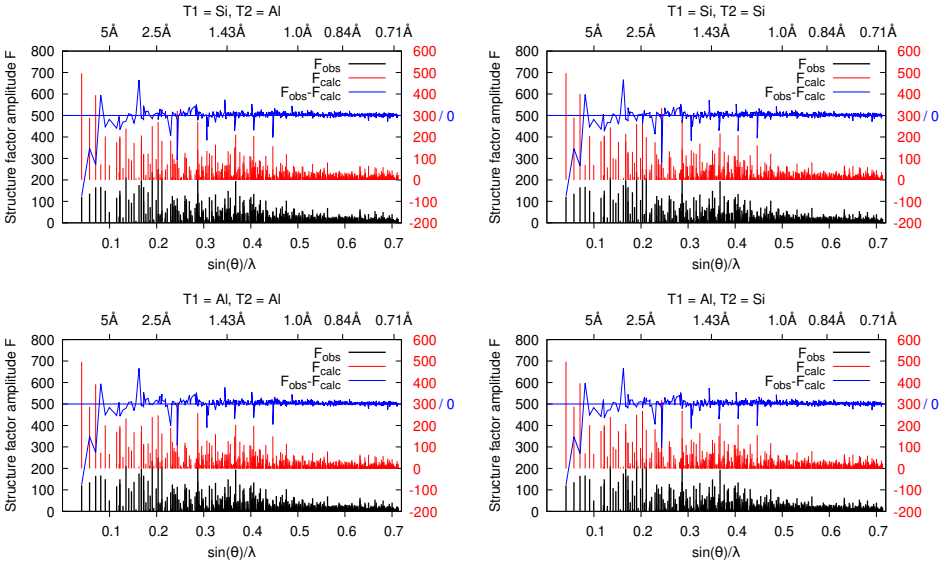

Figure S5: Data for Zeolite A structure depending on the T1/T2 composition. Comparison between observed and calculated data.

2 T-O Bond lengths

The following tables shows T-O bond lengths for the albite model and one selected zeolite A model. For T-O bond statistics for the remaining zeolite A models, we refer to the CIF files published together with this work.

Table S1: T-O bond lengths for the refined structure of albite. Oxygen atoms marked '*' refer to symmetry equivalent positions with respect to the CIF file.

T-site	Oxygen	length [Å] (std. unc.)
Si1	O1	1.616(10)
	O2	1.592(10)
	O3	1.595(14)
	O4*	1.627(15)
Si2	O3	1.617(10)
	O4	1.597(13)
	O5	1.590(13)
	O6	1.583(12)
Si3	O1*	1.639(12)
	O6	1.621(11)
	O7	1.595(15)
	O8	1.580(12)
Al4	O8	1.710(9)
	O2*	1.727(12)
	O7*	1.732(14)
	O5*	1.740(15)

Table S2: T-O bond lengths for the refined structure of zeolite A, crystal 3. Oxygen atoms marked '*' refer to symmetry equivalent positions with respect to the CIF file.

T-site	Oxygen	length [Å] (std. unc.)
Si1	O1	1.623(19)
	O2	1.607(18)
	O3*	1.604(13)
	O3	1.604(13)
Al2	O1	1.699(19)
	O2*	1.747(19)
	O3*	1.670(13)
	O3*	1.670(13)

3 Data set statistics

Table S3: Information for data sets from 9 crystals merged for albite. All datasets scaled to dataset "x014-D190" (bold). Crystals are grouped. 'D500' refers to the low resolution datasets (not measured for each crystal), 'D190' refers to high resolution data set. Number of reflections in dataset as "# refl.". K and B: Datasets were scaled with $K\exp(B*(\sin\theta/\lambda)^2)$ to put intensities on common scale. ISa: asymptotic $I/\sigma(I)$ [1]. Individual data sets were not scaled because of the low symmetry spacegroup, i.e. ISa0 = 50 [2].

Dataset	# refl.	K	B	ISa [1]	Δ
x000-D190	1163	0.284	-0.243	3.73	275.3
x001-D500	150	1.495	-0.149	2.59	772.6
x002-D190	2164	1.034	-0.110	6.66	270.4
x003-D500	129	0.687	-0.447	6.10	770.1
x006-D190	2104	0.333	-0.002	2.16	269.4
x008-D190	1690	1.942	-0.039	5.02	268.9
x009-D500	134	2.150	-0.384	6.50	754.1
x010-D190	1060	1.363	-0.104	7.68	269.9
x011-D500	75	1.636	-0.972	2.14	746.6
x014-D190	2119	1.000	0.000	5.14	268.6
x015-D500	131	0.997	0.336	4.67	742.7
x016-D190	483	0.697	0.181	3.58	272.0
x018-D190	468	1.015	0.144	3.43	264.0
x019-D190	268	0.876	0.108	2.68	264.2
x021-D190	2074	0.646	-0.057	3.46	266.6
x036-D190	1090	3.473	-0.083	5.34	270.9

Table S4: Data statistics for albite after merging of data sets from 9 crystals. The unit cell dimensions are calculated as weighted mean from 9 high resolution data sets with 6909 reflections in total. Numbers in brackets for unit cell parameters are standard uncertainties. Numbers in brackets for I/σ_I, $CC_{1/2}$, and completeness refer to the low resolution shell 6.36 Å– 2.86 Å.

Spacegroup	$P1$
Unit cell parameters	
a	7.127(59)
b	7.380(53)
c	7.641(41)
α	115.17 (11)
β	107.20(28)
γ	100.60(19)
Resol. $[d_{\max} - d_{\min}]$	6.36 - 0.64
$\langle I/\sigma(I)\rangle$	3.3 (9.5)
$CC_{1/2}$ [3]	97.7 (97.6)
Completeness	96.0 (93.5)

Table S5: Data statistics for zeolite A, crystal x1 measured at room temperature. The first part of the high resolution run moved out of beam and was continued as part 2. The individual data sets were processed in Laue group $m\bar{3}m$ for stability reasons during refinement. Δ: refined detector distance; # refl: number of reflections to determine unit cell parameters. Numbers in brackets for I/σ_I, $CC_{1/2}$, and completeness refer to the low resolution shells 8.7 Å–2.19 Å, 8.6 Å–2.19 Å, 12.2 Å–4.54 Å, and 12.2 Å–2.12 Å respectively. Data were cut where $CC_{1/2}$ [3] drops below 70 %.

$\Delta[mm]$	307.5	294.4	830.6	n/a
Spacegroup	$Fm\bar{3}c$	$Fm\bar{3}c$	$Fm\bar{3}c$	$Fm\bar{3}c$
Unit cell parameters				
a	24.47(32)	24.44(28)	24.47(42)	24.458(14)
b	24.47(32)	24.44(28)	24.47(42)	24.458(14)
c	24.47(32)	24.44(28)	24.47(42)	24.458(14)
α	90	90	90	90
β	90	90	90	90
γ	90	90	90	90
# refl.	2671	1360	868	n/a
Resol. $[d_{\max}$ - $d_{\min}]$	8.7 - 0.75	8.6 - 0.75	12.2 - 4.54	12.2 - 0.75
$\langle I/\sigma(I)\rangle$	4.7 (8.3)	4.4 (8.3)	8.5 (9.9)	5.7 (14.9)
$CC_{1/2}$ [3]	98.8 (98.4)	98.9 (98.1)	99.6 (99.9)	99.2 (99.4)
Completeness	100.0 (96.6)	94.1 (87.9)	93.0 (66.7)	99.6 (97.8)

Table S6: Data statistics for zeolite A, crystal x2 measured at room temperature. Δ: refined detector distance; # refl: number of reflections to determine unit cell parameters. Numbers in brackets for unit cell parameters are standard uncertainties. Numbers in brackets for I/σ_I, $CC_{1/2}$, and completeness refer to the low resolution shells 7.38 Å–4.04 Å, 14.13. Data were cut where $CC_{1/2}$ [3] drops below 70 %.

$\Delta[mm]$	300.8	822.4	n/a
Spacegroup	$P1$	$P1$	$Fm\bar{3}c$
Unit cell parameters			
a	17.26(12)	17.335(85)	24.4559(3)
b	17.248(96)	17.310(79)	24.4559(3)
c	17.21(12)	17.282(84)	24.4559(3)
α	60.28(14)	60.252(46)	90
β	60.008(63)	60.006(53)	90
γ	59.969(46)	59.924(41)	90
# refl.	10167	976	n/a
Resol. $[d_{\max} - d_{\min}]$	7.38 - 0.70	14.13 - 1.62	12.23 - 0.70
$\langle I/\sigma(I)\rangle$	2.3 (4.1) 5.9 (7.0)	5.2 (11.0)	
$CC_{1/2}$ [3]	97.3 (97.8)	99.2 (98.9)	98.9 (99.0)
Completeness	39.7 (38.0)	33.7 (34.1)	99.2 (100.0)

Table S7: Data statistics for zeolite A, crystal x3 at cryo-temperatures ($-189^{\circ}C$). The log-file for the oscillation width of the low resolution pass got corrupted so that this data set was not processed. Δ: refined detector distance; # refl: number of reflections to determine unit cell parameters. Numbers in brackets for I/σ_I, $CC_{1/2}$, and completeness refer to the low resolution shell 8.6 Å–2.38 Å. Data were cut where $CC_{1/2}$ [3] drops below 70 %.

$\Delta[mm]$	272.9	
Spacegroup	$P1$	$Fm\bar{3}c$
Unit cell parameters		
a	17.358(90)	24.33(32)
b	17.15(12)	24.33(32)
c	17.19(11)	24.33(32)
α	60.01(16)	90
β	60.084(86)	90
γ	59.850(72)	90
# refl.	781	781
Resol. $[d_{\max} - d_{\min}]$	8.60 - 0.82	8.60 - 0.82
$\langle I/\sigma(I)\rangle$	3.3 (10.4)	6.1 (16.3)
$CC_{1/2}$ [3]	99.5 (99.6)	99.3 (99.2)
Completeness	20.7 (19.7)	94.2 (78.8)

Table S8: Data statistics for zeolite A, crystal x4 at cryo-temperatures ($-189^{\circ}C$). Δ: refined detector distance; # refl: number of reflections to determine unit cell parameters. Numbers in brackets for I/σ_I, $CC_{1/2}$, and completeness refer to the low resolution shells 14.22 Å–4.6 Å, 7.38 AA–1.98 Å, and 14.22 Å–2.46 Å respectively.

	low resolution pass	high resolution pass	merged
$\Delta[mm]$	759.6	279.1	n/a
Spacegroup	$P1$	$P1$	$Fm\bar{3}c$
Unit cell parameters with standard uncertainties			
a	17.418(86)	17.372(35)	24.6084(16)
b	17.401(76)	17.322(34)	24.6084(16)
c	17.371(78)	17.299(35)	24.6084(16)
α	59.996(47)	59.993(27)	90
β	59.932(24)	59.798(30)	90
γ	60.027(67)	60.078(62)	90
# refl.	491	6021	n/a
Resol. $[d_{\max} - d_{\min}]$	14.22 - 1.61	7.38 - 0.72	8.70 - 0.72
$\langle I/\sigma(I)\rangle$	7.3 (6.3)	3.0 (6.7)	5.1 (10.4)
$CC_{1/2}$ [3]	99.2 (99.9)	99.3 (99.4)	99.3 (99.3)
Completeness	23.0 (21.2)	26.9 (23.7)	98.4 (95.1)

Table S9: Data statistics for zeolite A, crystal x5 at cryo-temperatures ($-189^{\circ}C$). Δ: refined detector distance; # refl: number of reflections to determine unit cell parameters. Numbers in brackets for I/σ_I, $CC_{1/2}$, and completeness refer to the low resolution shells 14.25 Å–4.6 Å, 7.38 AA–2.03 Å, and 14.25 Å–2.43 Å respectively.

	low resolution pass	high resolution pass	merged
$\Delta[mm]$	765.5	285.0	n/a
Spacegroup	$P1$	$P1$	$Fm\bar{3}c$
Unit cell parameters			
a	17.389(97)	17.386(49)	24.5898(106)
b	17.427(93)	17.347(49)	24.5898(106)
c	17.305(99)	17.241(52)	24.5898(106)
α	60.179(68)	60.16(15)	90
β	60.156(98)	60.288(28)	90
γ	60.11(21)	60.005(37)	90
# refl.	759	6021	n/a
Resol. $[d_{\max} - d_{\min}]$	14.25 - 1.63	7.41 - 0.70	14.25 - 0.70
$\langle I/\sigma(I)\rangle$	9.8 (12.2)	7.8 (3.7)	6.8 (13.3)
$CC_{1/2}$ [3]	99.6 (99.8)	99.4 (99.6)	99.2 (98.7)
Completeness	29.6 (30.6)	33.3 (31.2)	99.6 (100.0)

References

[1] K. Diederichs. ‘Quantifying instrument errors in macromolecular X-ray data sets’. In: *Acta Crystallogr* D66 (2010), pp. 733–740.

[2] G. M. Assmann, M. Wang and K. Diederichs. ‘Making a difference in multi-data-set crystallography: simple and deterministic data-scaling/selection methods’. In: *Acta Crystallogr* D76 (2020), pp. 636–652.

[3] P. A. Karplus and K. Diederichs. ‘Linking Crystallographic Model and Data Quality’. In: *Science* 336 (2012), pp. 1030–1033.