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Calibration of NRSF2 Instrument at HFIR

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1. INTRODUCTION

The Neutron Residual Stress Mapping Facility (NRSF2) at HB-2B is a new generation-diffraction instrument, adding many new Second Generation features, such as larger beam tube, large sample XYZ goniometer, and KAPPA orienter for a broad range of materials behavior studies. One key feature is the NRSF2 monochromator, which is a double focusing, double crystal monochromator system consisting of two sets of stacked Si crystal wafers [1] One set of wafers has Si [400] plane normal to the surface while the other set of wafers has the Si[500] normal to the surface.

The monochromator crystal diffracts at a fixed diffraction angle of 88° selecting a neutron wavelength determined by the monochromator d_{hkl}-spacing. This "Missouri" monochromator system has two independent monochromators, which enable diffraction from the following set of six diffraction planes: Si(511), Si(422), Si(331)AF (Anti-Fankuchen geometry), Si(400), Si(311), and Si(220). These diffraction planes can provide 6 different neutron wavelengths: approximately 1.45, 1.54, 1.73, 1.89 Å, 2.27, and 2.66 also incorporate seven position sensitive detectors located in a detector shield box.

To use this advanced instrument for scientific and engineering measurements, careful calibration needs to be performed to accurately calibrate the seven position sensitive detectors,neutron wavelength, and $2\theta_0$. Just as in the X-ray diffraction technique, neutron diffraction directly measures the diffraction angle (2θ) or diffraction peak position, then based on Bragg's law and a strain free lattice spacing, the strain can be calculated. Therefore anything that can affect the diffracting angle measurement can influence the accuracy of the strain measurements. The sources of difficulties in achieving acurate neutron diffraction peak positions can be classified into three categories:

(1) Instrument: These difficulties come from alignment of the monochromator, alignment of the incident and detector slits, leveling of the sample table, $2\theta_0$ offset, and response of the position sensitive detector.

(2) Counting statistics: If the peak profile count is too low, then the peak position derived from fitting a profile and background will have larger error. Therefore, adequate counting statistics and well-defined peaks are always good for precise peak position determination.

(3) Sample: Large grain size materials make it difficult to get enough diffracting grains, contributing to the different profile. With a low number he peak becomes "spot" and results in inaccuracy in peak position. Texture in the sample can change the effective elastic constants and also affect the peak intensity. Phase and composition inhomogeneity can make it difficult to determine an an accurate stress-free d_0 for strain calculation. A partially buried gauge volume due to proximity to the sample surface or buried interface can also shift the peak position.[ ]

The calibration method presented in this report will address the first two catergories of difficulties listed above. The FWHM can be minimized for each sample d-spacing by adjusting the horizontal bending of the monochromator crystal. For the monochromator, the optimum FWHM lies between 70 and 110 degree. This range is selected in order to maintain an approximately equiaxed gauge volume and avoid significant increases in peak breadth for the detectors above and below the horizontal plane. To adequately calibrate the position sensitive detectors, $2\theta_0$, and wavelength, a set of high purity reference powders were selected. Since the selected reference powders have define grain size is, the measurement errors from sample grain size and texture can be excluded, although there may still be micro-strain in the powders, which can broaden the reference peak.

In this report, the calibration procedure for the NRSF2 instrument will be presented and calibration results for five monochromator settings from HFIR cycle 403 will be presented. The monochromator

settings calibrated include Si(331)AF (Anti-Fankuchen geometry), Si(220), Si(511), Si(422), Si(400), and Si(311) The report presents calibration results for the single PSD that is in the horizontal plane defined by the center of the monochromator, sample, and PSD. Calibration for the out of plane detectors will require additional corrections related to the out of plane angle and finite height of each PSD detector

2. EXPERIMENTAL MATERIALS AND PROCEDURES

2.1 MATERIALS

Six reference powders were selected for the calibration of neutron wavelength and instrument angle offset, $2\theta_0$ The selected reference powders all have strong neutron diffraction lines and narrow FWHMS between 70 and 110 degrees 2θ for various wavelengths The purity and size distribution of the reference material powders are shown in Table 1.

Table 1. Material powders used for instrument calibration

	Ni	Fe	Mo	Ge	CaF_2
Purity	99.9%	99.9%	99.9%	99.99%	99.7%
Size (μm)	<44	<10	<53	<44	<5

2.2 LATTICE PARAMETER MEASUREMENT OF THE CALIBRATION POWDERS

Each calibration powder was mixed with a small amount of SRM Si powder (640c) from NIST as an internal standard. The powder mixture was smeared on a zero background Si single crystal sample holder. A Scintag PAD-V X-ray diffractometer with a Cu K target x-ray tube$_\alpha$ radiation was used for the determination of the lattice parameter of each instrument calibration powder. The diffraction pattern was collected from 20 to 130 degrees 2θ with a step size of 0 01 degree and a dwell time of 6 seconds. The temperature of measurement was at 25 °C +/- 1 °C. The diffraction patterns were analyzed and lattice parameters were refined using the GSAS program [2] and keeping SRM Si powder's lattice parameter [3] as a constant. These are performed for 3 each of the seven position sensitive detectors (PSD):

2.3 NRSF2 INSTRUMENT CALIBRATION PROCEDURE

There are four steps in calibration: intensity response normalization; the change in 2θ per change in PSD channel number; the out of plane (OoP) angle for each detector relative to the horizontal detector, and finally the $2\theta_0$ and neutron wavelength calibration

2.3.1 Intensity response normalization

One ORDELA PSD with 512 channels and about 5 degrees 2 theta coverage was used for neutron diffraction test. The first single PSD test to calibrate the PSD intensity responses of all channels. For this test, a rectangular block of polyethylene material with mostly incoherent neutron scattering characteristics was used to scatter the neutron beam with a homogeneous neutron flux onto the PSDs. The PSD intensity response of all channels were measured. This is called the flood test, and the results will be used to correct the neutron diffraction intensity values.

2.3.2 Change in 2θ vs. change in PSD channel number

The second PSD calibration movement is performed to establish the channel number to 2θ angle conversion. First a detector step scan was done. Reference diffraction peak, such as Fe (211) is selected and the detector is moved so that the peak appears first at one edge of the PSD. The detector is then shifted peak was recorded repeatedly until the peak was at the other edge of the PSD output. The detector was moved about 3 degrees with a step size of 0 2 degree. The diffraction profiles were fitted to determine the channel number of the peak center for each setting of detector 2-theta. From this step scan, the Bragg angle change per PSD channel is obtained.

2.3.3. $2\theta_0$ and neutron wavelength calibration

The third calibration step is to determine the detector angular offset, $2\theta_0$, and the wavelength of the neutron beam at each monochromator setting. Then Bragg angle offset value $2\theta_0$ and neutron beam wavelength from a certain monochromator setting were calibrated from selected reference materials diffraction lines.

Each calibration material was put into a quartz tube (4mm I.D., 6mm O.D.) with one end open. Then a slip-fit quartz rod was inserted into the tube to compress the powder to a height of about 25mm. All five quartz tubes with calibration powders were vertically mounted on an Al block sample holder with 25mm distance between each sample. The calibration powder samples were mounted on the XY table that can move in both X (400mm) and Y (200mm) directions, and rotate omega angle in both clockwise and counterclockwise directions.

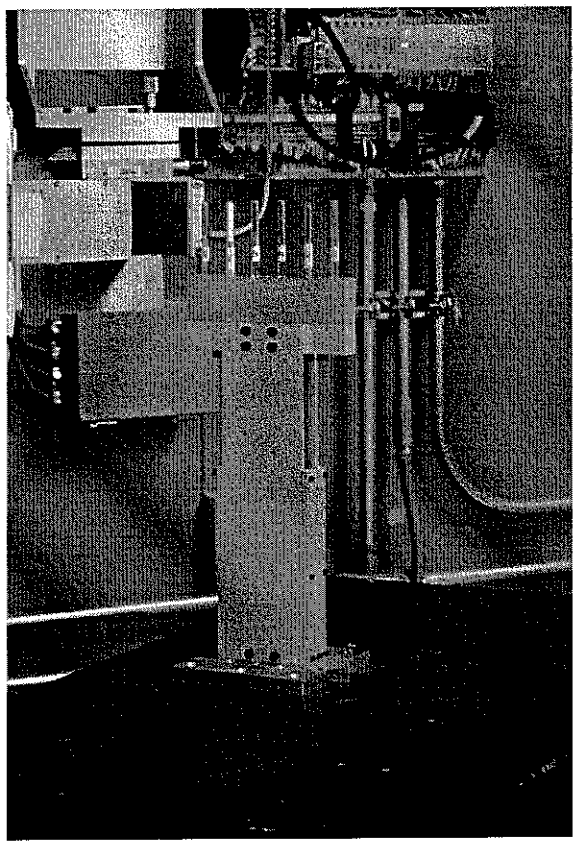

Figure 1. Reference calibration material powder samples were mounted on X-Y table.

The location of the sample to be measured must be moved to the rotation center of the Ω gonometer where neutron beam gauge volume is defined by incident and detector slits. By using two telescopes that were aligned to view the center of rotation, the X and Y coordinate values of each powder sample when it was at the center of rotation was determined The estimated error of positioning was less than 0.1mm. The rotation center of the gonometer was aligned with the center of incident neutron beam when instrument was installed. The incident and diffracted beam slits are aligned using a 1mm diameter Ni wire mounted on the XY table. By determine the location of maximum diffracted intensity for settings Ω and Ω +180, the incident slit position which places the beam over the center of rotation is determined.

By selecting a particular monochromator setting, the wavelength of the neutron beam was calibrated. The Si monochromator settings, such as the ^{m}X, ^{m}Y positions relative to the neutron beam guide, tilt of the Si crystal plane, and most importantly the horizontal bending curvature of Si wafers, were also optimized to achieve the highest diffraction neutron intensity, For calibration, incident slits of 2mm wide and 20mm high were used . A diffracted beam slit width of 2mm wide and no height limit slit used Both slits were offset approximately 25mm from the center of rotation . Since the calibration powder sample tube I.D. is 4mm, the width neutron gauge volume should be totally located inside the calibration powder sample when it is at the rotation center.

3. RESULTS AND DISCUSSION

3.1 LATTICE PARAMETER MEASUREMENT OF REFERENCE MATERIAL POWDERS The x-ray diffraction patterns of each reference powder mixed with SRM640c Si powder were analyzed using GSAS. Refined lattice parameters and the estimated standard deviation (e.s.d) The crystal structure of the materials, corresponding Powder Diffracton File (PDF) reference with lattice parameter values are also shown in the table. After GSAS refinement, the relative errors of all five lattice parameter values of reference material powders are within 10ppm

Table 2. Lattice parameters of reference material powders refinement results

Powder	Space Group	Refined by GSAS with Si 640c standard a=5.431195 Å λ=1.5405929 ($K\alpha_1$), 1.544427 ($K\alpha_2$) Lattice parameter ((n) .sd (n)(Å)	Literature values (Å) / (PDF file number)
Ni	Fm-3m (225)	3.523421 (0.000026)	3.5238 (#04-0850)
Mo	Im-3m (229)	3.147664 (0.000007)	3.1472 (#42-1120)
Fe	Im-3m (229)	2.866355 (0.000029)	2.8664 (#06-0696)
Ge	Fd-3m (227)	5.657554 (0.000017)	5.6576 (#04-0545)
CaF_2	Fm-3m (225)	5.464222 (0.000036)	5.46378 (#77-2093)

3.2 REFERENCE DIFFRACTION PEAKS FOR MONOCHROMATOR SETTINGS

The reference diffraction lines and their neutron count time used in HFIR cycle 403 are shown in Table 3. To assure that the peak position errors due to counting statistics are less than 0.003°, the count times were select3ed so that each reference lines"' peak intensity had about 1000 counts above background. The resulting total neutron count time for calibration ranges from 35 to 85 minutes. A Labview software program, NRSF2-View [5] was used for all neutron diffraction data collection, peak and background fitting, $2\theta_0$ and neutron wavelengths calculation

Table 3. Reference diffraction lines selected for calibration and their neutron count time used during calibration run at HFIR cycle403

Si (511)		Si(400)		Si(311)	
Reference lines	Count time (minute)	Reference lines	Count time (minute)	Reference lines	Count time (minute)
Ni(222)	10	Fe(200)	3	Ni(200)	5
Ge(422)	10	Ge(400)	20	CaF2(220)	10
Ge(511)	15	Mo(211)	2.5	Fe(200)	10
Ge(440)	20	Fe(211)	1	Mo(200)	20
Ge(531)	15	Ge(422)	10	Ge(400)	30
Ge(620)	15				

Si (331)AF		Si(220)	
Reference lines	Count time (minute)	Reference lines	Count time (minute)
Ni(311)	5	CaF2(220)	10
CaF2(422)	15	Mo(110_	5
Fe(200)	5	Ge(220)	12
Ge(400)	20	Ge(311)	20
Ge(331)	12		
Ge(422)	12		

3.3 FLOOD SEVEN DETECTORS TO DETERMINE THE CHANNEL INTENSITY RESPONSE

Figure 2 shows the result of 40 minute flood test used to calibrate the PSD intensity response. Generally the intensity response from whole PSD is quite flat, although there is noise non-linearity in the center channels of the PSD.

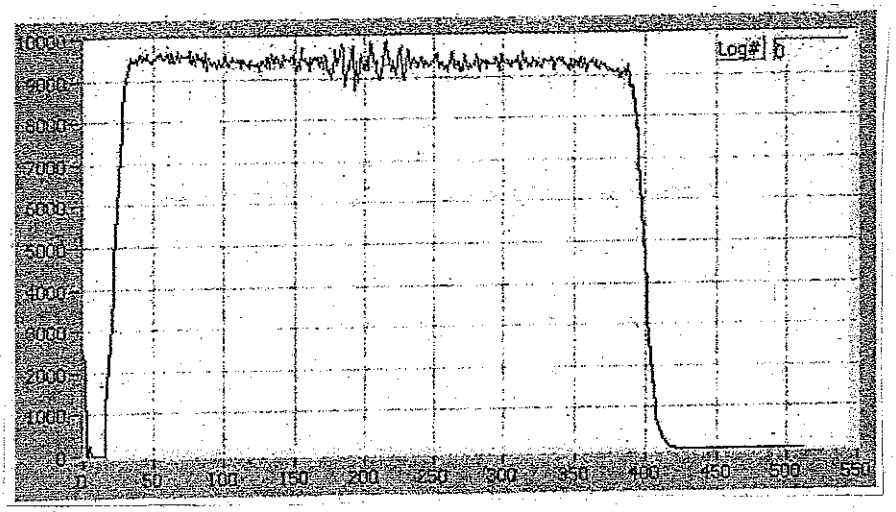

Figure 2. Flood test on PSD intensity response with40- minute count time. The x-axis is channel number and the y-axis is neutron counts

The intensity for a measured diffraction peak is corrected for counting non-linearity,

$$I_{corr}(i) = \frac{I_{obs}(1)}{I_{flood}(I)}$$

Where I is the channel number

3.4 STEP SCAN TO DETERMINE 2θ ANGULAR CHANGE PER PSD CHANNEL

Figure 3 shows the step scan and the linear fit results for the relationship between the angular channel and channel for a single PSD detector. The linearity of the step scan test is very good, and from the slope of the linear fit, we can see 2θ changes 0.01777 degrees per channel.

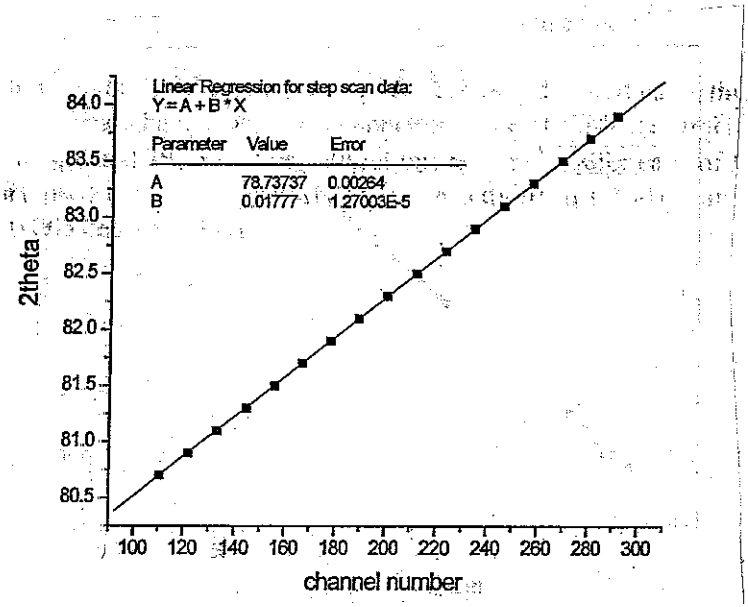

Figure 3. Step scan to determine 2θ angular change per PSD channel.

3.5 NEUTRON WAVELENGTH AND $2\theta_0$, CALIBRATION RESULTS

Calibrated neutron wavelength and $2\theta_0$ for five monochromator settings are shown in Table 4. All calibrated neutron wavelengths are close to the nominal wavelength values of each Si monochromator reflection. The mean square error (MSE) of the neutron wavelength and $2\theta_0$ calibration fitting are all below 1.3×10^{7}, and the root mean square (RMS) of the difference between the observed and calculated 2-theta value is below 0.003 degrees, which means the uncertainty in the peak position determination is less than +/-0.003 degree 2θ.

For the Si(511) wafer side, the monochromator settings Si(511), Si(400), and Si(311) show a consistent difference in the calibrated $2\theta_0$ if about 0 23 degree. However, on the Si (220) wafer side, the monochromator settings, Si(331)AF and Si(220) show a large $2\theta_0$ difference . One possible source of this $2\theta_0$ difference is a neutron flux spatial distribution. By changing the monochromator's position (MX, MY) and orientation (MROT), it is possible to shift all the $2\theta_0$ values close to zero. Future studies are planned to explore the interplay of MX and MY with λ nd $2\theta_0$ as well as the optimization of MBend for each powder's 2θ.

Table 4. Calibration results for five monochromator settings

	$2\theta_0$	Wavelength (Å)	MSE	RMS	MBend	MX	MY
Si(511)	0.211196	1.4450074	8.647933E-8	0.002146	80	10	11 20
Si(400)	0.269667	1.882517	6.861375E-8	0.001717	110	12.64	7.40
Si(311)	0.211210	2.272084	2.401707E-8	0.001690	80	8.40	12.10
Si(331)AF	0.038219	1.730152	2.544385E-8	0.001363	120	12.40	4.00
Si(220)	0.501460	2.659219	1.294359E-7	0.002936	90	12.00	10.60

Mbend, MX, MY: Values of the monochromator horizontal bending level and translation settings used.

4. Summary

The horizontal PSD detector for NRSF2 was calibrated using three steps: flood, step scan to determine the $2\theta_0$ angular change per PSD channel, and use of calibrated reference powders to deterine the neutron wavelength and $2\theta_0$. The five calibrations reported, for each of five wavelengths, showed that the mean square error of the calibrations were better than 1.3×10^{7}, and the $2\theta_0$ root mean square error is less than 0.003 degrees 2theta.

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APPENDIX A

X-RAY POWDER DIFFRACTON CALIBRATON OF THE LATTICE PARAMETERS FOR THE FIVE REFERENCE POWDERS USING GSAS

APPENDIX A. X-RAY POWDER DIFFRACTION CALIBRATION OF THE LATTICE PARAMETERS FOR THE FIVE REFERENCE POWDERS USING GSAS

Table A-1. Fe powder GSAS refinement output

Reduced CHI^2	GU	GV	GW	GP
2.027	2.0	-2.0	50.33	0.4
LX	**LY**	**S/L**	**H/L**	**shft**
11.6	0	0.0026	0.0066	-11.5

Table A-2. Ni powder GSAS refinement output

Reduced CHI^2	GU	GV	GW	GP
2.429	2.0	-2.5	39.47	0.4
LX	**LY**	**S/L**	**H/L**	**shft**
5.733	9.813	0.0026	0.0066	-20.55

Table A-3. Mo powder GSAS refinement output

Reduced CHI^2	GU	GV	GW	GP
5.602	2.0	-2.5	6.315	0
LX	**LY**	**S/L**	**H/L**	**shft**
3.398	0.7766	0.0026	0.0066	3.617

Table A-4. Ge powder GSAS refinement output

Reduced CHI^2	GU	GV	GW	GP
4.82	2.0	-2.5	23.17	0
LX	**LY**	**S/L**	**H/L**	**shft**
3.213	0.2606	0.0026	0.0066	-14.04

Table A-5. CaF_2 powder GSAS refinement output

Reduced CHI^2	GU	GV	GW	GP
3.5	2.0	-2.5	25.36	0
LX	**LY**	**S/L**	**H/L**	**shft**
3.064	3.569	0.0026	0.0066	-4.04

APPENDIX B

NEUTRON WAVE LENGTH AND 2θ ANGLE CALIBRATION FROM REFERENCE DIFFRACTION LINES FOR EACH MONOCHROMATOR SETTING

APPENDIX B. NEUTRON WAVE LENGTH AND 2θ CALIBRATION FROM SELECTED REFERENCE DIFFRACTION LINES FOR EACH MONOCHROMATOR SETTING

Table B-1. Calibration data for Si(511)

	Reference d-spacing	Measured $2\theta_{obs}$	ESD $2\theta_{obs}$	Corrected $2\theta_C$	Reference $2\theta_R$	$\Delta 2\theta$
Ge[422]	1.1548	77.9927	0.0029	77.7815	77.7828	-0.0013
Ge[511]	1.0888	83.7169	0.0027	83.5057	83.5035	0.0022
Ni[222]	1.0171	91.1475	0.0016	90.9363	90.9341	0.0022
Ge[440]	1.0001	93.1382	0.0025	92.9270	92.9318	-0.0098
Ge[531]	0.9563	98.8211	0.0018	98.5927	98.5927	-0.0048
Ge[620]	0.8945	108.5080	0.0020	108.2968	108.2994	-0.0025

$2\theta_{obs}$: Measured Bragg angle from reference diffraction lines,
$2\theta_C$: Corrected Bragg angle, and $2\theta_C = 2\theta_{obs} - 2\theta_0$,
$2\theta_R$: Bragg angle calculated from reference diffraction lines d-spacing,
$\Delta 2\theta$: Bragg angle difference between $2\theta_C$ and $2\theta_R$, and $\Delta 2\theta = 2\theta_C - 2\theta_R$,
ESD: Estimated standard deviation.

Table B-2. Calibration data for Si(400)

	Reference d-spacing	Measured $2\theta_{obs}$	ESD $2\theta_{obs}$	Corrected $2\theta_C$	Reference $2\theta_R$	$\Delta 2\theta$
Fe[200]	1.4332	82.3784	0.0016	82.1088	82.1054	0.0034
Ge[400]	1.4144	83.7052	0.0026	83.4355	83.4387	-0.0032
Mo[211]	1.2850	94.4602	0.0009	94.1905	94.1922	-0.0017
Fe[211]	1.1702	107.3675	0.0008	107.0978	107.0964	0.0014
Ge[422]	1.1548	109.4613	0.0018	109.1917	109.1912	0.0004

Table B-3. Calibration data for Si(311)

	Reference d-spacing	Measured $2\theta_{obs}$	ESD $2\theta_{obs}$	Corrected $2\theta_C$	Reference $2\theta_R$	$\Delta 2\theta$
CaF_2[220]	1.9319	72.2464	0.0018	72.0352	72.0366	-0.0014
Ni[200]	1.7617	80.5239	0.0020	80.3127	80.3097	0.0030
Mo[200]	1.5738	92.6252	0.0020	92.414	92.4143	-0.0003
Fe[200]	1.4332	105.0786	0.0014	104.8674	104.8698	-0.0024
Ge[400]	1.4144	107.0846	0.0024	106.8734	106.8730	0.0005

Table B-4. Calibration data for Si(331)

	Reference d-spacing	Measured $2\theta_{obs}$	ESD $2\theta_{obs}$	Corrected $2\theta_C$	Reference $2\theta_R$	$\Delta 2\theta$
Fe[200]	1.4332	74.2921	0.0013	74.2481	74.2502	-0.0021
Ge[400]	1.4144	75.4540	0.0017	75.4101	75.4076	0.0023
Ge[331]	1.2979	83.6368	0.0013	83.5928	83.5912	0.0005
Ge[422]	1.1548	97.0645	0.0015	97.0206	97.0186	-0.0009
CaF_2[422]	1.1154	101.7505	0.0013	101.7065	101.7049	-0.0020
Ni[311]	1.0624	109.0702	0.0017	109.0262	109.0189	0.0023

Table B-5. Calibration data for Si(220)

	Reference d-spacing	Measured $2\theta_{obs}$	ESD $2\theta_{obs}$	Corrected $2\theta_C$	Reference $2\theta_R$	$\Delta 2\theta$
Mo[110]	2.2257	73.8662	0.0015	73.3647	73.3663	-0.0016
Ge[220]	2.0002	83.8257	0.0018	83.3242	83.3245	-0.0003
CaF_2[220]	1.9319	87.4894	0.0015	86.9880	86.9818	0.0062
Ge[311]	1.7058	102.9192	0.0037	102.4177	102.4230	-0.0005

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