OctCarb – A GNU *Octave* Script for the Analysis and Evaluation of Wide-Angle Scattering Data of Non-graphitic Carbons

Supporting information part A

Octave installation and examples

Authors

Oliver Osswald[a]* and Bernd M. Smarsly[a]

[a]Institute of Physical Chemistry, Justus-Liebig-University Giessen, Heinrich-Buff-Ring 17, Giessen, 35392, Germany

Correspondence email: oliver.osswald@phys.chemie.uni-giessen.de

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S1. Overview of all used parameters

In addition to 2.2, not the microstructure parameters itself, but parameters of distribution functions are refined (μ and β for interlayer parameters, v and α for intralayer parameters) from which the microstructure parameters are calculated (equations (7) – (14)). Therefore, a brief overview of all parameters used for the refinement and the received microstructure parameters as well as some other important units is given here.

For the number deviation of the average number of layers per stacks the following equations were found [1]:

$$n(N) = \frac{\beta^{\mu}}{\Gamma(\mu)} N^{\mu-1} \exp(-\beta N) \tag{7}$$

$$\langle N \rangle = \frac{\mu}{\beta} \tag{8}$$

$$L_c = \bar{a}_3 \frac{\mu+1}{\beta} \tag{9}$$

$$\kappa_c = \frac{1}{\mu} \tag{10}$$

Table S1 Overview of all used parameters for a refinement using *Octave*. The column “influence on scattering data” describes, if the parameter has a large (+), medium (o) or small (-) influence on the theoretical intensity.

Parameter	Parameter in *Octave*	Parameter used for refinement	Description	Influence on scattering data
q	q	Measured	Modules of the scattering vector (= $2 \cdot \pi \cdot s$)	
s	s	Measured	Modules of the scattering vector (= $2/\lambda \cdot \sin(\theta)$)	
λ	wavelength	Given constant	Wavelength of used radiation	
θ	theta	Measured	Scattering angle, half „Bragg-angle“	
$\overline{a_3}$	a3	$a3$	Average layer distance	+
$a_{3\,\mathrm{min}}$	a3min	$= \overline{a_3} - \mathrm{d}a_3$	Minimal layer distance	+ (only for visible (004) reflection, else -)
σ_3	sig3	σ_3	Disorder of the stacks (standard deviation of a_3)	+
L_a	La	$= (v + 1)/\alpha$	Average graphene layer size	+ (calculated parameter)
l_m	lm	$= v/\alpha$	Average chord length	+ (calculated parameter)
κ_a	kapa	$= 1/v$	Polydispersity of chord length	Depends a lot on the experiment, see 2.3
l_cc	lcc	$l$cc	Average C-C bond length	+
σ_1	sig1	σ_1	Disorder of the layers (i.e. stress and strain)	+
ε_1	eps1	-	Disorder of graphene layers due to local strains	-- (not implemented)
κ_r	kapr	$= 3\pi 2\ (1/v + 1)/32 - 1$	Polydispersity of the radius of the graphene layers	Currently not used

N	N	$= (\mu+1)/\beta$	Average number of graphene layers per stack	+ (calculated parameter)
L_c	Lc	$= \overline{a}_3 \cdot (\mu+1)/\beta = L_c \cdot \overline{a}_3$	Average stack height	+ (calculated parameter)
κ_c	kapc	$= 1/\mu$	Polydispersity of stack height	o (calculated parameter, but generally high error)
ε_3	eps3	$= a_3/a_{3\ \mathrm{min}}$	Disorder of stacks due to local strains	o (calculated parameter, but generally high error)
q	q	q	Preferred orientation	+
c_H	cH	c_H (constant)	Concentration of unorganized hydrogen	-
c_N	cN	c_N (constant)	Concentration of unorganized nitrogen	o (depends on value)
c_O	cO	c_O (constant)	Concentration of unorganized oxygen	o (depends on value)
c_S	cS	c_S (constant)	Concentration of unorganized sulfur	+
Δ_{an}	dan	Δ_{an}	Anisotropy of atomic form factor of carbon	-
k	k	k	Normalization constant for log10(k · Ie.u. + const1) + const2	+
$const_1$	const1	$const_1$	Constant shift for log10(k · Ie.u. + const1) + const2	+
$const_2$	const2	$const_2$	Non-constant (linear) shift for log10(k · Ie.u. + const1) + const2	+
g	g	g	Factor for exponential damping of the scattering intensity with Ie.u. = exp(g · s) · Ie.u.	+
Q	b	Q	Additional parameter for incoherent background (Q ≠ b, see 2)	-
ρ	density	ρ (constant)	Density of the sample	o

d	sampleThickness	d (constant)	Thickness of the sample	o
μ_{ab}	mue_ab	μ_{ab} (constant)	Absorption factor, calculated from density, sample thickness and wavelength	o
-	polarizedBeam	constant	Is the beam polarized?	+
Φ	polarizationDegree	Φ (constant)	Polarization direction of beam in °	+
r	par_r	r (constant)	Radius of the goniometer (in cm; fixed due to experiment)	o
δ	par_delta	δ (constant)	Divergence angle (in °; to choose by user)	o
l	par_l	l (constant)	Irradiated length (in cm; fixed during measurement)	o
R	R	R (constant)	Parameter for position correction according to D2thtx	-- (not available)
t	t	t (constant)	Parameter for position correction according to D2thtx	-- (not available)

Influence on scattering data is just the personal meaning of the author based on the experience of several refinements.

S2. Implementation and calculation time consumption of *v*

The implementation of v as described in 3.1 in the main article

$$P_\mathrm{L}(r) = \frac{1}{\Gamma(v+1)}[\Gamma(v+1, \alpha r) - \alpha r\Gamma(v, \alpha r)] \tag{11}$$

$$\langle l\rangle = \frac{v}{\alpha} \tag{12}$$

$$L_\mathrm{a} = \frac{\langle l^2\rangle}{\langle l\rangle} = \frac{v+1}{\alpha} \tag{13}$$

$$\kappa_\mathrm{a} = \frac{1}{v} \tag{14}$$

is used to calculate the profile shape of the intralayer reflections and therefore the intralayer parameters (e.g. the average layer extension L_a). However, v is not refined, but fixed to a constant depending on the maximum measured modules of the scattering vector s_max (see Table S2). Several calculations showed that this implementation is very useful to improve the speed of the refinement, since the influence of v is only very small on a single reflection. Also, the parameters α (for the calculation of the layer extension L_a) and σ_l (stress and strain of the layers ("disorder")) have a much higher influence on the profile shape. Hence, this usage of v does not degrade the physical assumptions and the resulting accuracy of the refinement. More precisely, it is not possible to determine ν and therefore the polydispersity of the graphene layers ($\kappa_\mathrm{a} = 1/v$) in an experimental way.

In the following, an overview of the recommended minimal value of v for the s-value is given. Table S2 is just a recommendation and for some samples or more special experiments, e.g. WANS with a low wavelength, it might be useful to use a larger value of v. In general, $v < 4$ is not recommended. Additionally, the calculation time using *Octave* is about 4 times faster than using the algorithm from Pfaff et al. [3] for $v = 4$.

Table S2 Recommended value of v for a maximum measured modules of the scattering vector s_{max}, regarding maximum polydispersity and the average calculation times using the algorithm from Pfaff et al. [3] and the improved used in the present study, respectively. In general, $v < 4$ is not recommended.

			Duration for a single calculation	
v	s_{max} / Å^{-1}	$\kappa_{\text{c, max}}$	Pfaff et. al (2018)	*Octave*
4	1.4	0.250	21.6 s	5.9 s
5	1.85	0.200	30.1 s	13 s
6	2.35	0.167	39.3 s	25.2 s
7	2.8	0.143	49.9 s	44.2 s
8	3.1	0.125	61.6 s	75.1 s
9	3.5	0.111	74.5 s	114.3 s
10	3.85	0.100	88.6 s	166.7 s

S3. Determination of the error bars

Generally, in non-linear fitting procedures the initial values of the fitting parameters may exhibit an undesired strong impact on the final fitting result. In order to evaluate this possible issue, two automatic refinements were performed for the four samples by means of *Octave*, using different starting parameter values. In the first one, the refined microstructure values taken from Pfaff et al. [3] were used as initial parameters (*Octave data set* Pfaff et al. see Figure 6 in the main article and Figure S1). The second one takes our default starting parameters as described in Table S6 (*Octave data set default values* , see Figure 6 in the main article and Figure S1). First tests using the already refined microstructure from Pfaff et al. [3] were done to validate, if the general refinement works in principle and leads to comparable results. The second test using the default starting parameters as described in Figure 6 in the main article and Figure S1 was performed to validate if the numerical refinement/minimalization of *Octave* works well and leads to the same (or similar) results. These default starting values are in principle mean-values of the aforementioned structural parameters of common NGCs.

More precisely, *Octave* uses the Levenberg-Marquardt non-linear fitting algorithm to perform the refinement. Hence, the influence of each (microstructure) parameter on each simulated fit value is calculated, so the refinement time depends on multiple effects: First, the size of the matrix as described before, second, the direct calculation time of a theoretical scattering curve and third, the number of scattering curves, which must be calculated in each refinement step (two for each refinement parameter - one with a greater and one with a lesser value). In the following steps, a so-called singular-value-decomposition (SVD) is performed, which leads to a covariance matrix of all refinement parameters. The square of the individual values of the diagonals of the matrix is then the standard deviation (σ deviation) of the corresponding refinement parameters. A deviation of 1σ means the probability that the "real" value of the parameter is in the range of ["calculated value – σ deviation"; "calculated value + σ deviation"] is ~ 68 %. In the same way, the so called $2\sigma/3\sigma$ deviations can be calculated: The probability of the value of a refinement parameter lying in the 2σ range (["calculated value – $2 \cdot \sigma$ deviation"; "calculated value + $2 \cdot \sigma$ deviation"]) is ~ 96.5 % and for the 3σ range the probability is ~ 99.7 %. In other words, the refined parameter is the mean value of a Gaussian distribution and σ its standard deviation.

This quantification of the refinement error is completely different than in prior studies [2–6], where the uncertainty of the individual microstructural parameters was determined by variating a single parameter until a significant visual change of the calculated scattering curve was observable. Applying this procedure, the typical error for each parameter was in the range of 10 – 15 %. By contrast, using *OctCarb* the error is systematically calculated within the fitting procedure, thus the resulting error is usually much smaller using *Octave*. However, the user has to decide whether to use the 1σ, 2σ or 3σ range as the refinement error, which refers to the importance of how exactly the relative parameter uncertainty should be known (2σ or 3σ).

We found that the uncertainties of the individual structural parameters defined in this way strongly depend on the number of data points in one scattering curve, which can now be nicely demonstrated by the samples used (Table S3): For example, the errors for the PIL WAXS data are small, because the data possess about 10-times more measurement points than the other samples. For the same reason, the calculated errors for AC Type H are quite high, because the WAXS data of this sample consist of only a low amount of measurement points (258 compared to 2392 of PIL) (for more information about the influence of the number of data points see S11).

S4. Results of the refined samples for the verification of the *OctCarb* (plots)

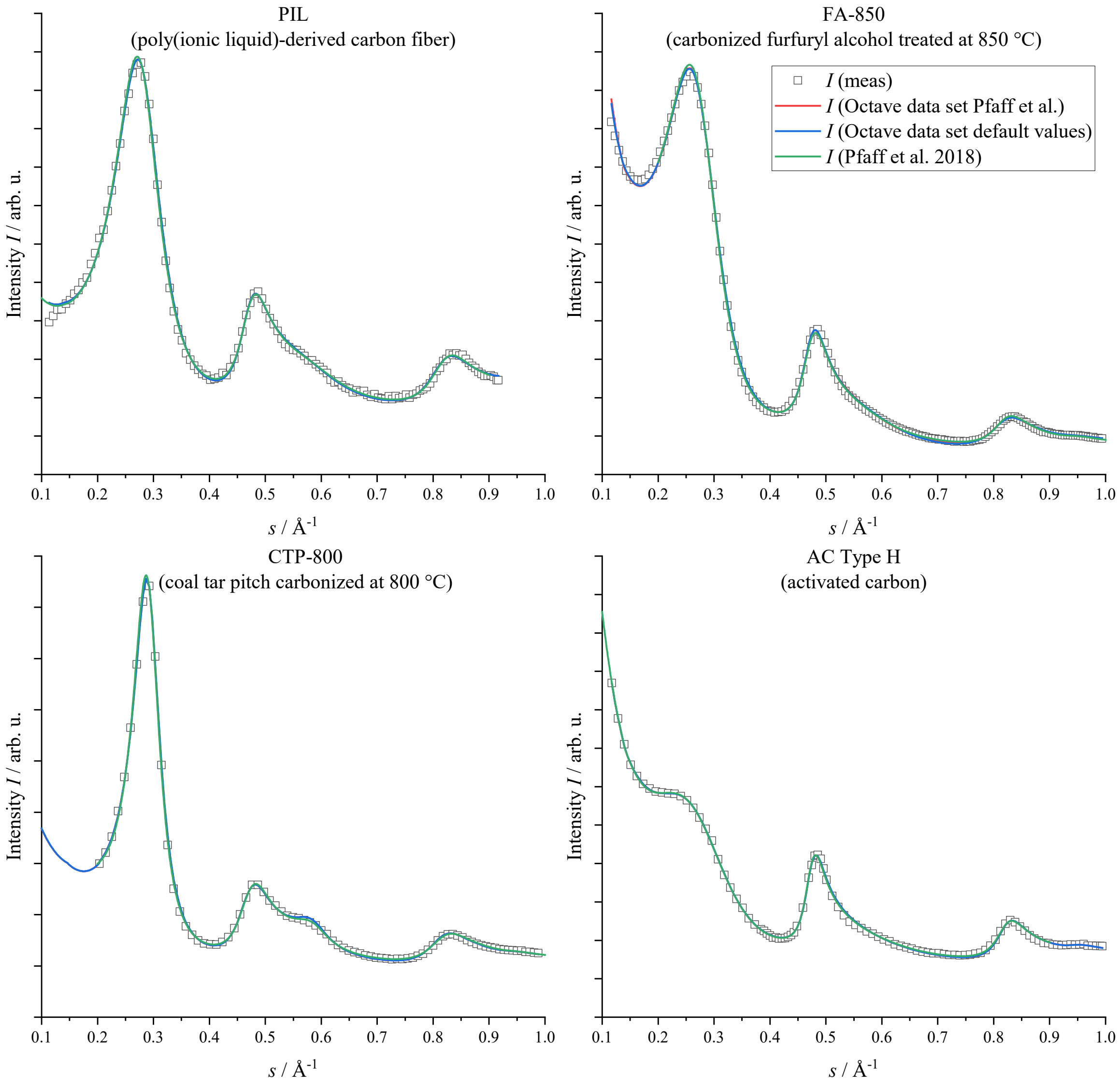

Figure S1 Validation of *CarbOct* using WAXS data of different well-evaluated samples (PIL = poly(ionic liquid)-derived carbon fiber, FA-850 = carbonized furfuryl alcohol treated at 850 °C, CTP-800 = coal tar pitch carbonized at 800 °C, AC Type H = an activated carbon). All three methods (automatic fits using *Octave* with the values of Pfaff et al. [3] as start values (red) and the recommended standard values as start values (blue) and the fit from Pfaff et al. [3]) lead to a similarly acceptable fitting and comparable structural parameters as reported in Pfaff et al. [3]. For improved visualization, for PIL only every 20th point and for all other samples only every 2nd point is shown. The evaluations of sample FA-850 by the different approaches are compared in further detail in Figure 6.

S5. Results of the refined samples for the verification *OctCarb* including the calculation times for the WAXS refinements

Table S3 Overview of the microstructure parameters of CTP-800 (carbonized coal tar pitch), AC Type H (activated carbon), FA-850 (carbonized furfuryl alcohol) and PIL (poly(ionic liquid) carbon fiber).

The parameters were rounded to the same number of digits as the values given in the original publications. *v* was fixed to 4, i.e. $\kappa_a = 1/v$ was fixed to 0.25. In Faber et al. [5] and Pfaff et al. [3], the errors of the other parameters were estimated by the influence of changing the parameter on the quality of the fit. For *Octave manual* and *Octave fitted* the error was calculated inside the fitting process. For this refinement, the error is a mathematical calculated error and more accurate than in the references.

Parameter	AC Type H [5]	AC Type H [3]	AC Type H (*Octave* manual)	AC Type H (*Octave* fitted)
L_a / Å	32 ± 4	31 ± 3.9	30 ± 7.3	30 ± 7.1
l_m / Å	26 ± 3.3	24 ± 3	24 ± 11.7	24 ± 11.4
σ_1	0.12 ± 0.014	0.12 ± 0.014 [1]	0.11 ± 0.006	0.11 ± 0.006
κ_a / Å	0.25	0.25	0.25	0.25
N_m	1.8 ± 0.23	1.8 ± 0.23	1.7 ± 0.37	2 ± 0.4
N	2.7 ± 0.27	2 ± 0.25	2 ± 0.2	2 ± 0.2
L_c / Å	7 ± 0.9	7 ± 0.9	7 ± 0.6	7 ± 0.6
κ_c / Å	0.15 ± 0.02	0.12 ± 0.02	0.15 ± 0.02	0.15 ± 0.02
$\bar{a}_3$ / Å	3.6 ± 0.054	3.56 ± 0.053	3.51 ± 0.057	3.52 ± 0.056
σ_3 / Å	0.57 ± 0.057	0.5 ± 0.05	0.45 ± 0.058	0.46 ± 0.058

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[1] Not published but known due to personal contact to the author.

Parameter	CTP-800 [5]	CTP-800 [3]	CTP-800 (*Octave* manual)	CTP-800 (*Octave* fitted)
L_a / Å	19 ± 2.4	19 ± 2.4	18 ± 1.3	18 ± 1.3
l_m / Å	16 ± 2	15 ± 1.9	15 ± 2.1	15 ± 2.1
σ_1	0.14 ± 0.016	0.13 ± 0.016 [2]	0.12 ± 0.02	0.12 ± 0.02
κ_a / Å	0.25	0.25	0.25	0.25
N_m	2 ± 0.25	2.2 ± 0.28	1.5 ± 0.59	1 ± 0.6
N	3.9 ± 0.49	3.8 ± 0.48	3.4 ± 0.39	3.4 ± 0.39
L_c / Å	13 ± 1.6	13 ± 1.6	12 ± 0.8	12 ± 0.8
κ_c / Å	0.96 ± 0.12	0.76 ± 0.1	1.27 ± 0.38	1.27 ± 0.38
$\overline{a_3}$ / Å	3.44 ± 0.052	3.44 ± 0.052	3.43 ± 0.006	3.43 ± 0.006
σ_3 / Å	0.26 ± 0.026	0.23 ± 0.023	0.2 ± 0.025	0.2 ± 0.025

Parameter	FA-850 [5]	FA-850 [3]	FA-850 (*Octave* manual)	FA-850 (*Octave* fitted)
L_a / Å	25 ± 3.1	23 ± 2.9	28 ± 1.6	28 ± 1.7
l_m / Å	20 ± 2.5	19 ± 2.4	23 ± 2.5	23 ± 2.8
σ_1	0.13 ± 0.016	0.13 ± 0.016 3	0.15 ± 0.012	0.16 ± 0.013
κ_a / Å	0.25	0.25	0.25	0.25
N_m	1.5 ± 0.19	1.6 ± 0.2	1.6 ± 0.28	2 ± 0.7
N	2.3 ± 0.29	2.3 ± 0.29	2.3 ± 0.17	2.8 ± 0.4
L_c / Å	8 ± 1	8 ± 1	8 ± 0.5	10 ± 1.3
κ_c / Å	0.51 ± 0.06	0.42 ± 0.05	0.44 ± 0.05	0.44 ± 0.1
$\overline{a_3}$ / Å	3.6 ± 0.054	3.61 ± 0.054	3.6 ± 0.025	3.74 ± 0.13
σ_3 / Å	0.43 ± 0.043	0.44 ± 0.044	0.44 ± 0.052	0.69 ± 0.217

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[2] Not published but known due to personal contact to the author.

[3] Not published but known due to personal contact to the author.

Parameter	PIL [7]	PIL [3]	PIL (*Octave* manual)	PIL (*Octave* fitted)
L_a / Å	20 ± 2.5	20 ± 2.5	22 ± 0.5	22 ± 0.5
l_m / Å	16 ± 1.9	16 ± 2	18 ± 0.8	18 ± 0.9
σ_1	0.25	0.13 ± 0.016 [4]	0.15 ± 0.005	0.14 ± 0.005
κ_a / Å	1.7 ± 0.21	0.25	0.25	0.25
N_m	3 ± 0.36	1.7 ± 0.21	1.5 ± 0.14	1 ± 0.1
N	11 ± 1.4	3 ± 0.38	2.6 ± 0.09	2.6 ± 0.09
L_c / Å	11 ± 1.4	11 ± 1.4	9 ± 0.3	9 ± 0.2
κ_c / Å	0.77 ± 0.1	0.77 ± 0.1	0.79 ± 0.05	0.78 ± 0.05
$\bar{a}_3$ / Å	3.63 ± 0.054	3.64 ± 0.055	3.57 ± 0.008	3.56 ± 0.007
σ_3 / Å	0.54 ± 0.054	0.56 ± 0.056	0.47 ± 0.015	0.46 ± 0.014

Table S4 Calculation times for the different samples and a different amount of measurement points.

HPC means high-performance computing cluster (JustHPC at the Justus Liebig University Giessen [8] using CentOS 7, typical node parameters are 2 x 12 sockets x cores/socket, 192 GB memory, 54.4 GFlops/Core and Intel Xeon Skylake 6126 or 6226 (1.7 Ghz or 1.9 Ghz) processors), *common PC* is a Windows computer using an Intel® Core™ i5-8400 CPU (4x 2.8 GHz) and 16 GB RAM *Raspberry Pi* is a Raspberry PI 4B Rev. 1.1 using an ARM v7 processor (4x 1.5 GHz) and 4 GB RAM running at Raspbian 11 (bullseye).. The main influence on the calculation time has the amount of measurement points and not the processor, due to the missing availability of parallel computing. Of course, a high-performance computing cluster is faster, if one want to refine multiple samples at once, because on these systems a high number of different programs can run simultaneously. All values are just examples and might vary due to different background processes on the different systems. The calculation time is given in minutes.

Sample	Measurement points	Calc. time HPC	Calc. time common PC	Calc. time Raspberry Pi
AC Type H	258	05:14	03:42	11:49
CTP-800	163	01:50	02:13	04:08
FA-850	300	04:36	07:56	05:13
PIL	2392	22:17	15:55	50:54

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[4] Not published but known due to personal contact to the author.

Table S5 Overview of the microstructure parameters of H-2100/H-2800 (phenol formaldehyde resorcinol) and LSPP-1200 (low softening point pitch) measured by wide-angle neutron scattering (WANS).

The parameters were rounded to the same number of digits as the values given in the original publications. v was fixed to 4, i.e. $\kappa_a = 1/v$ was fixed to 0.25. The error for each parameter for the references and the manual fitting is about 10-15 %.

Parameter	H-2100 2	H-2100 *Octave* automatic	H-2800 2	H-2800 *Octave* automatic
L_a / Å	40	34 ± 0.3	51	51 ± 1.2
l_m / Å	32	27 ± 0.5	43	41 ± 1.9
l_{cc} / Å	1.413	1.418 ± 0.0002	1.419	1.421 ± 0.0002
σ_1	0.067	0.056 ± 0.0025	0.047	0.052 ± 0.0024
L_c / Å	14	11 ± 0.4	23	19 ± 1
N_m / Å	3	2 ± 0.3	5	3 ± 0.7
N	4	3 ± 0.2	7	6 ± 0.4
κ_c / Å	0.37	0.43 ± 0.029	0.49	0.79 ± 0.032
$\overline{a_3}$ / Å	3.56	3.49 ± 0.005	3.48	3.46 ± 0.003
$a_{3,\,\mathrm{min}}$ / Å	3.16	2.49 ± 0.076	3.3	2.87 ± 0.037
σ_3 / Å	0.44	0.34 ± 0.009	0.25	0.2 ± 0.008
η	0.86	0.89 ± 0.003	0.93	0.96 ± 0.002

Parameter	LSPP-1200 2	LSPP-1200 *Octave* automatic	LSPP-1200 *Octave* manual
L_a / Å	27	23 ± 0.1	23
l_m / Å	22	18 ± 0.2	18
l_{cc} / Å	1.418	1.42 ± 0.0003	1.420
σ_1	0.062	0.066 ± 0.0037	0.066
L_c / Å	17	31 ± 3.3	15
N_m / Å	3	0 ± 0.7	3
N	5	9 ± 1.2	4
κ_c / Å	0.63	100 ± 559.241	0.67
$\overline{a_3}$ / Å	3.48	3.51 ± 0.006	3.51
$a_{3,\,\mathrm{min}}$ / Å	3.00	2.51 ± 0.1	3.21
σ_3 / Å	0.30	0.45 ± 0.009	0.33
η	1.00	1 ± 0.002	1

S6. Tests for fitting the (004)-region of the LSPP-1200 WANS-data

In section 4.2.3 of the main article, the problem of fitting the (004)-region of the WANS data of the sample LSPP-1200 was discussed. The main problem is the poor fitting of the (004)-region using the automatic fit from *OctCarb*. Hence, an automatic refinement using the whole data range must be adjusted manually afterwards (Figure 7 of the main article). To get deeper insights and a more detailed understanding of this issue, some other refinements were performed using only a smaller range of the measured data (Figure S2 - Figure S4).

First, only the refinement range was reduced, and the resulting scattering curve was extrapolated over the entire range (red). Second, the normalization parameters were fixed, and the other microstructure parameter were refined and the result extrapolated (blue). In the next step, only the interlayer parameters with (green) and without the normalization parameters (purple) were refined and the result extrapolated. Since at higher values of s, only intralayer reflections are visible, a smaller range is sufficient to determine the interlayer structure parameters. Data at higher s-values do not lead to more accurate results regarding the interlayer structure parameters.

Three main conclusions can be drawn from these tests:

1. Looking at the refinements including the normalization parameters, it becomes evident that the normalization parameters were calculated wrong using only the smaller data range. On the other hand, the scattering data in this area including the (004)-region can be refined well. Overall, this indicates that the incoherent background of the scattering data was not subtracted properly.
2. Even if the measured data are hardly influenced by noise and the fitted curve lies almost perfectly on the data, the intralayer parameters cannot be determined exactly. For the higher order reflections ($s > 1.2$ Å^{-1}), slight differences are visible between the “without normalization” and “only interlayer” fits. This means, that *Octave* tries to use the overlapping intralayer reflections as small background correction to refine all data as best as possible. This results in slightly calculated intralayer reflections and therefore other intralayer parameters.
3. Another explanation for 2 can be found by the influence of the layer disorder on the scattering data: In general, a stronger layer disorder causes broader reflections, with higher order reflections becoming broader [1]. So the used range for the refinement might be too small to determine this disorder correctly, since the influence is too small on these lower ordered reflections. On the other hand, with higher values of s, no more interlayer reflections are visible, so an extension of the measurement range would not solve this problem.

However, overall the main problem for the automatic refinement regarding the (004)-reflection is its shape: It is too much a shoulder rather than a clearly visible reflection and therefore, it is too “insignificant” for the *Octave* fitting routine compared to the whole scattering curve, especially to the neighboring valleys/feet which makes the manual adjustment necessary.

As another test, in the ranges of $0.33\ \text{Å}^{-1} < s < 0.44\ \text{Å}^{-1}$ and $0.63\ \text{Å}^{-1} < s < 0.77\ \text{Å}^{-1}$ not all measured points were refined. Instead, some points were left out during the refinement and extrapolated afterwards (Figure S5 - Figure S7). This test confirms that for *Octave* the feet/valleys besides the (004) and (11) reflection are more relevant in the fitting algorithm as the (004) reflection itself. Figure S7 shows, that only every 30th point (in the s-ranges 0.33-0.44 & 0.63-0.77 Å^{-1}) should be considered in this case, while more points would still influence the results. Nevertheless, the results from the automatic refinement are different compared to the manual refinement, especially the stacks are now significant higher (L_c). At first glance, this sounds like a significant intrinsic flaw between these two methods. However, this difference can be explained on closer inspection of the individual parameters: With the manual fit, both the difference between a_3 and $a_{3\,\text{min}}$ and the absolute value for σ_3 are smaller. This higher order results in sharper interlayer reflections in WAXS/WANS data. To compensate for this effect, the crystallite size, i.e. the stack height, has to be smaller, which in turn leads to broader reflections. Overall, these effects balance each other out, so that both fits and both results are plausible.

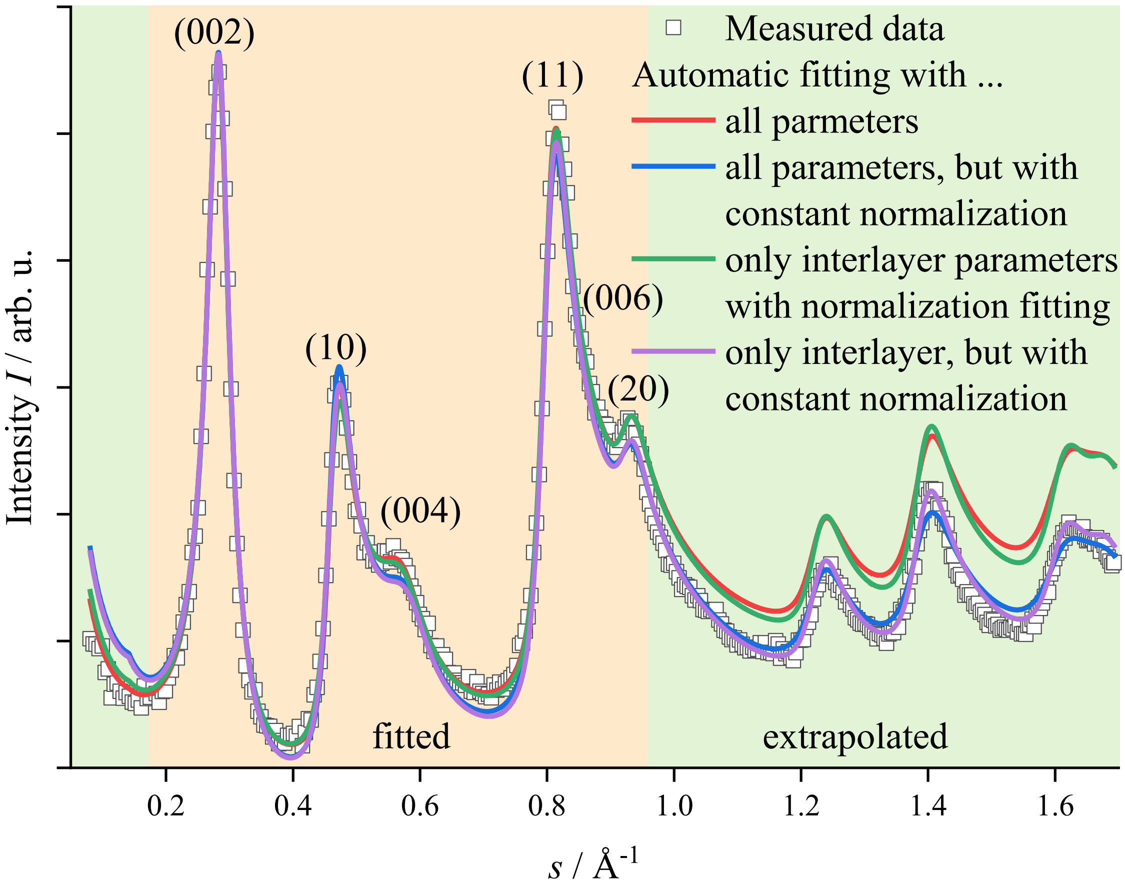

Figure S2 Tests for the automatic *Octave* refinement with special attention to the (004)-region of WANS data from LSPP-1200 (low softening-point pitch). For all tests, only the orange area was fitted, and the green areas extrapolated. Tests were performed with all parameters (red), the interlayer and intralayer parameters without the normalization (blue), the interlayer parameters with the normalization (green) and only with the interlayer parameters (purple). Only every 5th data point is shown.

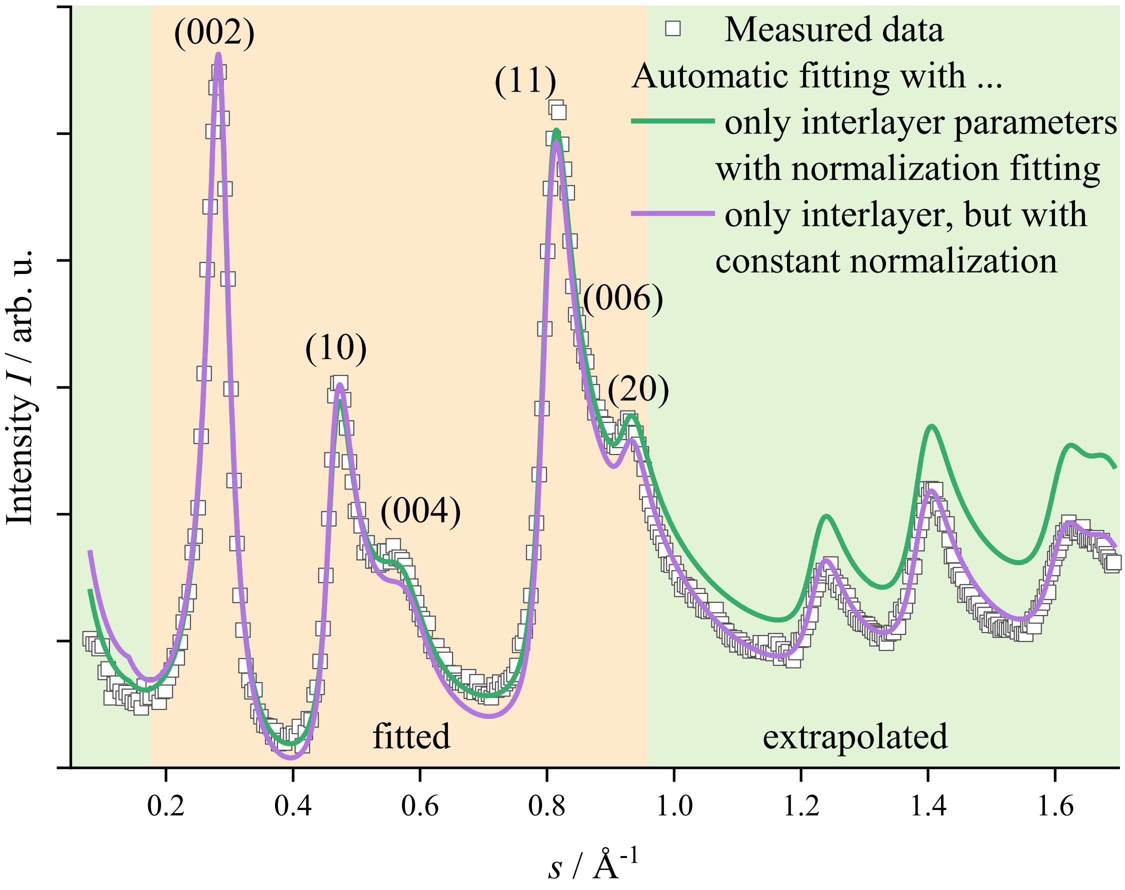

Figure S3 Tests for the automatic *Octave* refinement with special attention to the (004)-region of WANS data from LSPP-1200 (low softening-point pitch). For all tests, only the orange area was fitted, and the green parts extrapolated. Tests were performed with all parameters (red), the interlayer and intralayer parameters without the normalization (blue), the interlayer parameters with the normalization (green) and only with the interlayer parameters (purple).

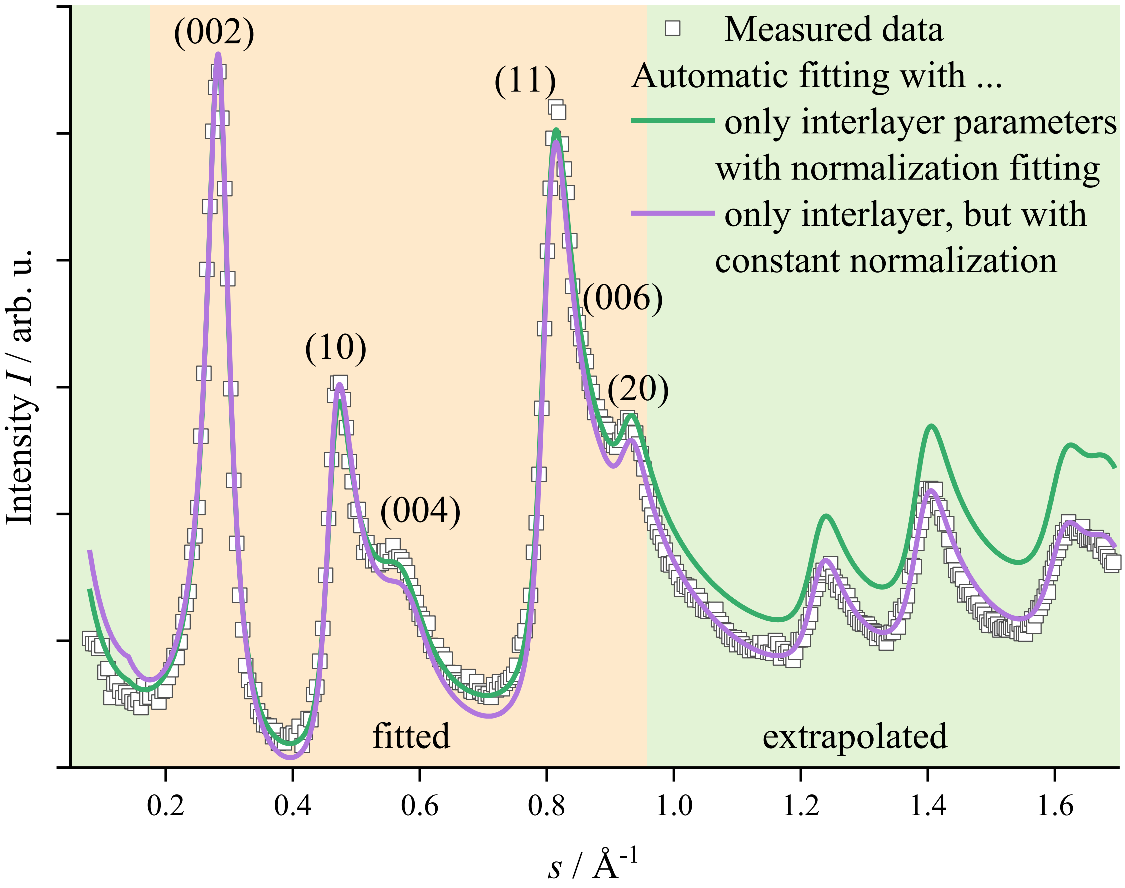

Figure S4 Tests for the automatic *Octave* refinement with special attention to the (004)-region of WANS data from LSPP-1200 (low softening-point pitch). For all tests, only the orange area was fitted, and the green parts extrapolated. Tests were performed with all parameters (red), the interlayer and intralayer parameters without the normalization (blue), the interlayer parameters with the normalization (green) and only with the interlayer parameters (purple).

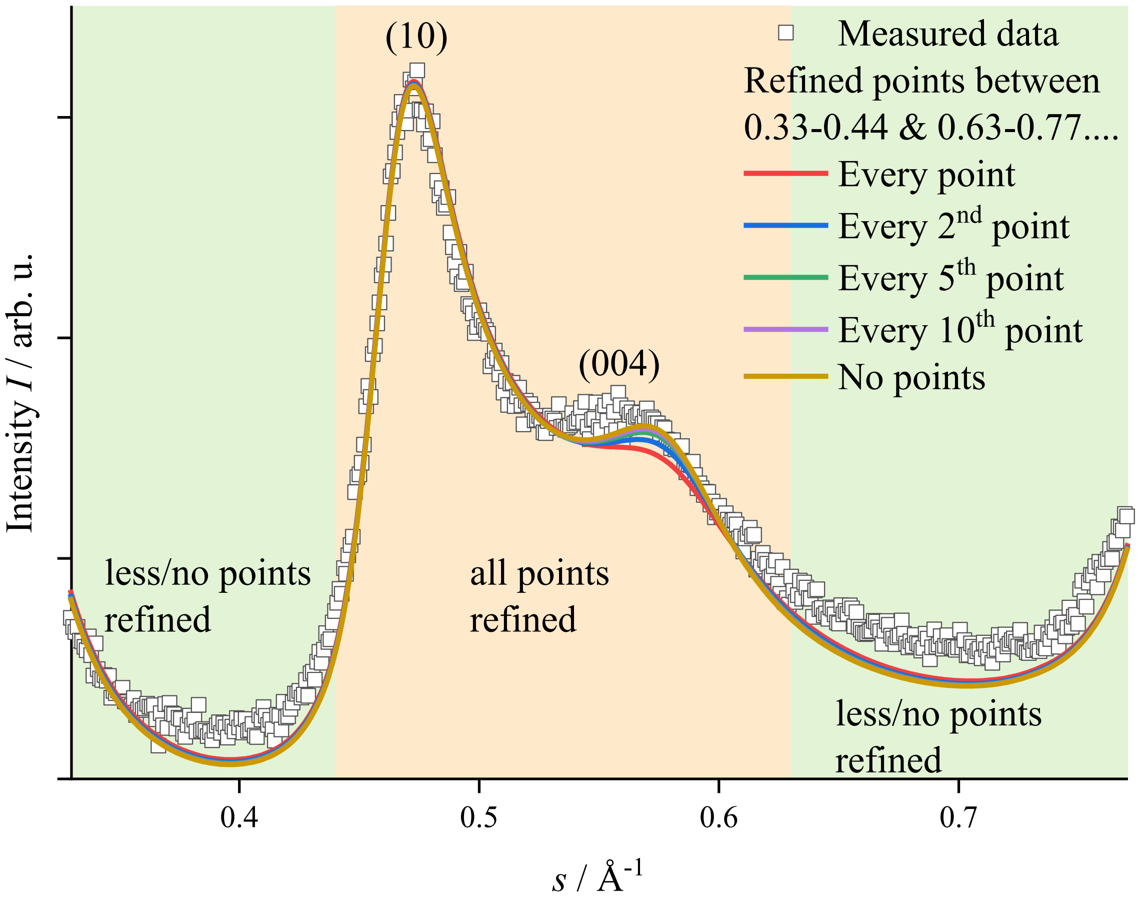

Figure S5 Tests for the automatic *Octave* refinement with special attention to the (004)-region of WANS data from LSPP-1200 (low softening-point pitch). For all tests, only the orange area was fitted, and the green parts extrapolated. In order to determine the influence of the feet/valleys on the (004)-reflection, first all (red) and then only every 2nd (blue), every 5th (green), every 10th (purple) and at last no point (brown) were used for the refinement. For more details, take a look at Figure S6. It becomes clearly, that the high number of data points in the feet/valleys influence the refinement. Compared to the (004)-reflection, these green areas are in mathematical terms more important. Less refined points in the green areas lead to a higher accuracy in the (004)-reflection.

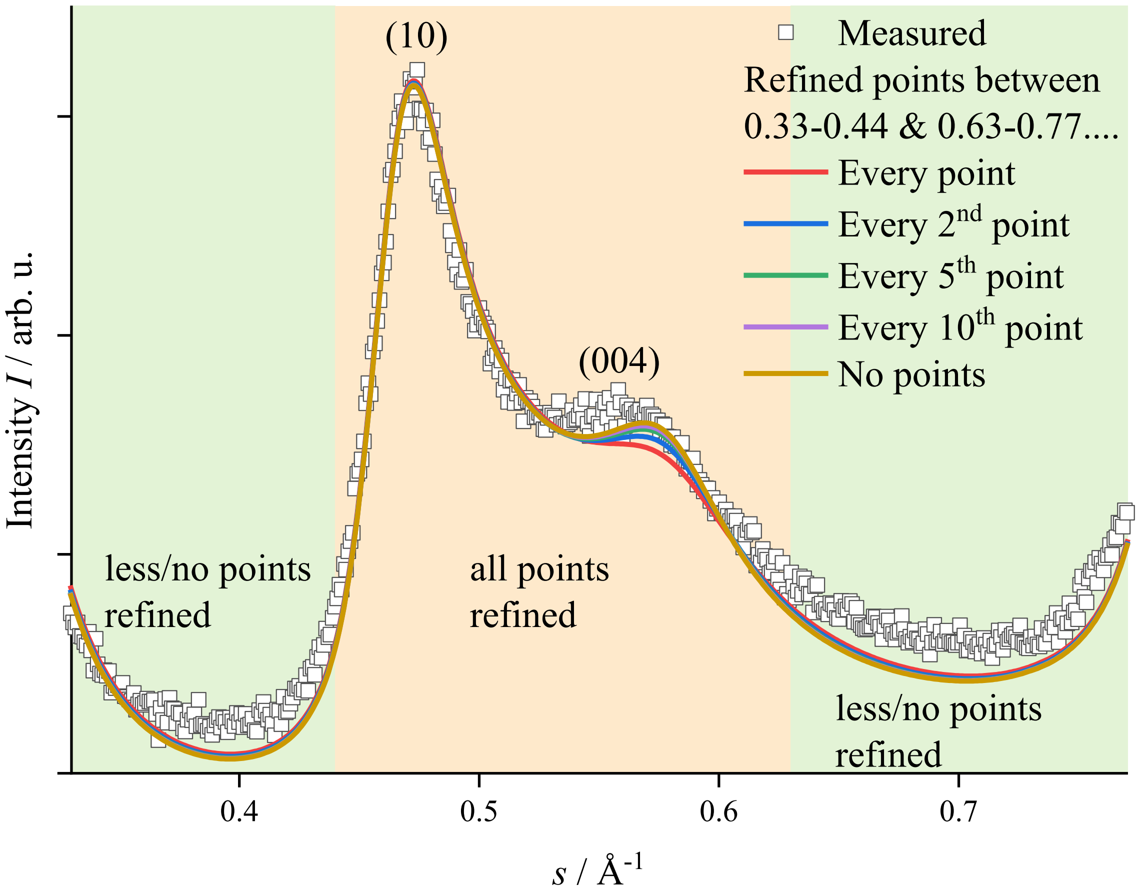

Figure S6 Tests for the automatic *Octave* refinement with special attention to the (004)-region of WANS data from LSPP-1200 (low softening-point pitch). For all tests, in the orange area all points and in the green area less points were fitted. Hence, in the ranges of 0.33 Å^{-1} < s < 0.44 Å^{-1} and 0.63 Å^{-1} < s < 0.77 Å^{-1}, not all points were used. In order to determine the influence of the feet/valleys on the (004)-reflection, first all (red) and then only every 2nd (blue), every 5th (green), every 10th (purple) and at last no point (brown) were used for the refinement. It becomes clearly, that the high number of data points in the feet/valleys influence the refinement. Compared to the (004)-reflection, these green areas are in mathematical terms more important. Less refined points in the green areas lead to a higher accuracy in the (004)-reflection.

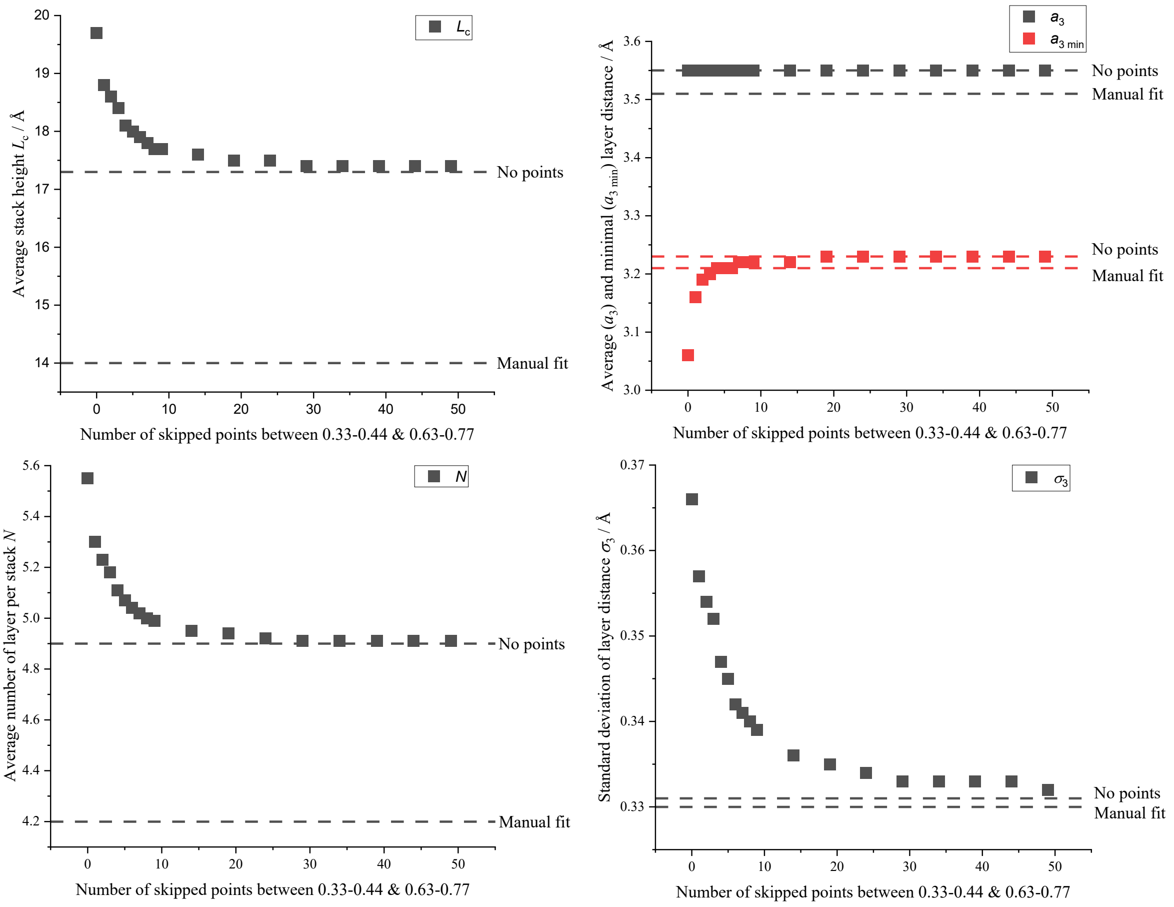

Figure S7 Tests for the automatic *Octave* refinement with special attention to the (004)-region of WANS data from LSPP-1200 (low softening-point pitch). For all tests, only the green range in Figure S5 was fitted. Hence, in the range of 0.33 Å^{-1} < s < 0.44 Å^{-1} and 0.63 Å^{-1} < s < 0.77 Å^{-1}, not all points were used. In order to determine the influence of the feet/valleys on the (004)-reflection, some points were skipped. Evidently, a high number of data points in the feet/valleys influence the refinement. Compared to the (004)-reflection, these regions of the scattering curve (green domains in the figures above) are in mathematical terms more important. Less refined points in the green areas lead to a higher accuracy in the (004)-reflection and therefore, the interlayer microstructure parameters are different.

S7. Download and usage of *iObs*

The currently used C++ code for calculating *iObs* including some auxiliary files for compiling an *.oct file for *Octave* is available in the GitHub. [5]

S7.1. Compilation of an *.oct file for using *iObs* with *Octave*

An alternative way to use the calculations code in C++ is to compile it in an *.oct file, which can be used from *Octave*. A video describing the next step is available in the GitHub [6] and at YouTube [7] [8].The basic idea is, that *Octave* uses some code, which is precompiled for different operating systems. This code can be used as an additional library for C++, so the algorithm of calculating I_{obs} can be converted in an *.oct file on the currently used operating system. This *.oct file in turn can be opened and used by *Octave* to calculate I_{obs} inside *Octave*. This method allows to calculate I_{obs} and therefore to refine measured WAXS/WANS data without the directly usage of any C++ code. Hence, the command *mkoctfile* implemented in *Octave* is used to perform this compilation. In principle, beside the pure calculation file, an additional file performing the in- and output operations, which is basically a "connector" between *Octave* and C++ code, is needed (*iObs.cpp*).

Since the compilation is unique for each operating system, it is highly recommended to compile the file on every different operating system. While the compilation is different for each operating system, only a brief overview of the command and its usage can be given. A more detailed instruction how to build an *.oct file explaining these steps as well as some example files are available in the GitHub. [9]

For Microsoft Windows, two commands must be executed, whereas the paths must be adjusted, obviously:

```
cd C:\Octave\Octave-5.2.0\mingw64\bin
```

```
C:\Octave\Octave-5.2.0\mingw64\bin\mkoctfile  -LC:\Octave\Octave-5.2.0\mingw64\lib\Octave\5.2.0
-IC:\Octave\Octave-5.2.0\mingw64\include\Octave-5.2.0\Octave ‘C:\iObsOct\iObs.cpp’
```

For MacOS and Linux, only one command must be executed:

```
MacOS: mkoctfile -I/usr/local/bin/Octave ~//iObsOct/iObs.cpp
```

```
Linux: mkoctfile -I/usr/include/Octave-5.2.0/octave ~/iObsOct/iObs.cpp
```

The exact path for the include files can be found out using the following command:

```
which Octave
```

---

[5] https://github.com/osswaldo/NGCs/blob/master/Octave/oct-files/Self-compilation/iObsOct/calculations.cpp

[6] https://github.com/osswaldo/NGCs/tree/master/Instruction%20Videos

[7] English: https://www.youtube.com/playlist?list=PLTlnYDX5g1FylWfH8cSM_ZQUQpn3dSp6M

[8] German: https://www.youtube.com/playlist?list=PLTlnYDX5g1FwaOQkLXTxrpvHW9CRUwBrk

[9] https://github.com/osswaldo/NGCs/tree/master/Octave/oct-files

S8. Installation and updates

A video describing the next step is available in the GitHub [10] and at YouTube [11, 12].

S8.1. Installation under *Microsoft Windows*

For Microsoft Windows, an executable installation package can be downloaded directly from the developer website [13]. Afterwards the *optim* package including all needed dependencies can be installed using “pkg install -forge optim”.

S8.2. Installation under *MacOS*

For MacOS, no official-maintained installer or bundle is available, but, however, there are some *.app builds available from the developer [14], for newer versions, an installation guide can be found in the developer instructions. [15]

S8.3. Installation under *GNU/Linux*, *BSD* and other systems

For all other operating systems like *Linux*, *BSD* and distribution independent approaches like *Docker* it is impossible to give a brief installation guide due to the high number of different systems. In general, all guides can be found in the developer instructions. [16]

S8.4. Installation of *optim* for non-*Windows* builds

For some operating systems/installations (if *pkg install -forge optim* fails) the *optim* package and its dependencies must be installed manually using console commands. First the optim package and its dependencies (*structs*, *statistics* and *io*) must be downloaded from *sourceforge* [17]. Second, open *Octave* and navigate to the download directory, e.g. “cd C:\Users\<*Username*>\Downloads“. In the last step, install optim and its dependencies (order as followed):

pkg install io-<*version*>.tar.gz

pkg install statistics-<*version*>.tar.gz

pkg install struct-<*version*>.tar.gz

pkg install optim-<*version*>.tar.gz

---

[10] https://github.com/osswaldo/NGCs/tree/master/Instruction%20Videos
[11] English: https://www.youtube.com/playlist?list=PLTlnYDX5g1FylWfH8cSM_ZQUQpn3dSp6M
[12] German: https://www.youtube.com/playlist?list=PLTlnYDX5g1FwaOQkLXTxrpvHW9CRUwBrk
[13] https://www.gnu.org/software/octave/download
[14] https://octave-app.org/
[15] https://wiki.octave.org/Octave_for_MacOS
[16] https://wiki.octave.org/Category:Installation and https://wiki.octave.org/Octave_for_GNU/Linux
[17] https://octave.sourceforge.io/packages.php

S8.5. Links and updates

Due to the number of different programs, extensions and scripts that are used and the resulting update frequency that can be expected from them, it does not make sense to describe the update process in this static work. Current links, installation files can be found in the file “Useful links.txt” and scripts can be found in the GitHub. [18]

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[18] https://github.com/osswaldo/NGCs

S9. Usage of *Octave*

After loading the refinement script (*OctCarb*), only a few adjustments are necessary to perform the first fit. Stepwise, consecutive files for the next steps below can be found in the other SI files. A video explaining these steps is available in the in the GitHub [19] and at YouTube [20, 21].

0. Download and install *Octave*, *iObs* and the *CarbOct* (see above).
1. First, some initial parameters like the sample name, data path, radiation type, wavelength etc. must be adjusted (Figure S8). Additionally, a meaningful for the parameter *nSkip* should be set. *nSkip = 5* means, that only every 5th point will be calculated to make the calculations much faster, less accurate at the same time. The value should be higher the more measurement points are available. Also, a constant v must be chosen. For a common XRD measurement using Cu-K_α radiation in a range of $10\,° < 2\theta < 140\,°$, a value of $v = 4$ is sufficient for most of the samples (see Table S2).
2. As the first real refinement step, the normalization parameters (k, $const_1$, $const_2$) and the concentrations of foreign atoms (c_H, c_N, c_O, c_S) should be refined. While the last ones are known exactly form elemental analysis, the normalization parameters must be refined by hand. For the automatic adjustment, it can be done only roughly, but for the manual adjustment, it should be done more exactly. The normalization constants should be adjusted at every of the following steps.

Manual refinement:

3a. Generally, the manual refinement is divided into 3 parts: refinement of the interlayer structure, refinement of the intralayer structure and a final step with all parameters including the normalization. For the interlayer parameters, first μ or β can be left constant and only one of it should be variated. Also, $a_{3\ \mathrm{min}}$ should not be lower than 3 Å, otherwise it will dominate the whole curve too much, because the set value is no longer physically meaningful. σ_3 can be used to refine the broadness of the (002) and (004) reflections and has a high influence on the damping of the (004) reflection. If the (002) reflection is refined approximately, both μ and β should be further refined. It should be noted that the stack height $L_c \sim (\mu+1)/\beta$ should no longer vary too much. In this step, η (homogeneity of the stacks) can be refined by taking the left side of the (002) reflection and lower values of the modules of the scattering vector/scattering angle into account. q (preferred orientation) should be left constant in this step, because without a refinement of the intralayer structure, it is not possible to refine this parameter meaningfully.

---

[19] https://github.com/osswaldo/NGCs/tree/master/Instruction%20Videos
[20] English: https://www.youtube.com/playlist?list=PLTlnYDX5g1FylWfH8cSM_ZQUQpn3dSp6M
[21] German: https://www.youtube.com/playlist?list=PLTlnYDX5g1FwaOQkLXTxrpvHW9CRUwBrk

Figure 5 in the main article shows an exemplary WAXS pattern and a manually adjusted result that contains the priority of the various regions.

4a. For the most samples, this step is easier to perform than the refinement of the interlayer parameters due to the lower number of parameters. Since v is chosen as a constant, only α, σ_1 and l_{cc} must be refined. α influences both, the height and the broadness of the (10) and the (11) reflection, σ_1 influences more the (11) reflection. Using l_{cc}, the exactly position of both reflections can be refined. For some samples, the preferred orientation q must be refined at this step.

5a. As the last step for the manual refinement, all microstructural parameters including the normalization should be adjusted. The exact procedure depends strongly on the sample and can therefore not be described in general here. In general, the accuracy should focus on the valley between the (002) and the (10) reflection rather than on the values that lie left to the (002) reflection. For the (10) and (11) reflections it is important to assure that the ratio of the size and shape in relation to the background is correct. The (004) reflection needs to be adjusted precisely, even if it sometimes just becomes like a shoulder. The accuracy of the refinement of this reflection significantly influences the accuracy of the resulting disorder parameters of the stack structure (see Figure 5 in the main article).

Automatic refinement:

3b. For the basic automatic refinement, the default values from Table S6 can be used. These values are in principle mean values from the common range of these values. Of course, if other suitable start values are known, they should be used (i.e., for measurements from a temperature series). To start the automatic refinement, the switch *plotOnly* must be set to *false*. For some samples or measurements, the upper and lower bonds for the preferred orientation (q) must be adjusted to a range from 0 to 1 (ub3 & lb3), otherwise, a preferred orientation will not be refined.

4b. As a final refinement step, the parameter *nSkip* should set to 1. Now, every measured point will be refined, and the fitting result might improve. As start values, the values from the 3rd step (3a or 3b) should be used. In addition, the calculated error for the microstructure parameters will become a little bit lower.

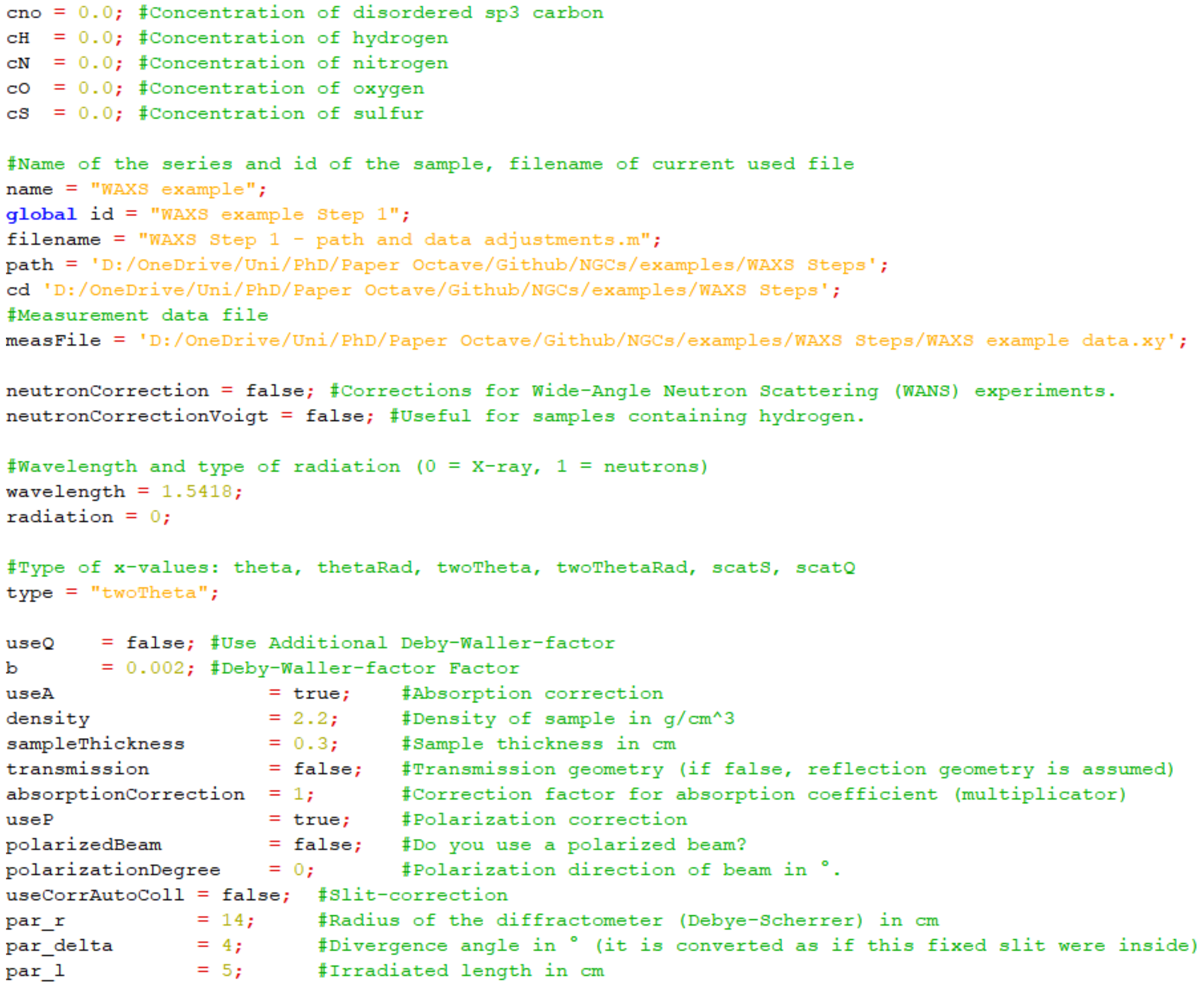
5b. As a last step, an additional manual adjustment can be performed. This step is often necessary for more disordered samples, where the (004) reflection is damped and broad.

Table S6 Overview about the default starting values for the refinement with *Octave*.

These values are in principle mean values of the common range of the fitting values and can be regarded as recommendations. Of course, every additional information like results from an elemental analysis should be used to choose as best start values as possible. The *microstructural parameters* are partly calculated from the *parameter for Octave*.

Parameter for *Octave*	Value
μ	4
β	0.5
$\overline{a}_3$ / Å	3.5
$\mathrm{d}a_3$ / Å	0.4
σ_3 / Å	0.25
u_3	0
η	1
ν	4
α	0.2
l_{cc} / Å	1.412
σ_1	0.1
q	0
Δ_{an}	0
k	500
$const_1$	0
$const_2$	0

Microstructural parameter	Value
L_a / Å (= $(\nu + 1)/\alpha$)	25
l_m / Å (= ν/α)	20
κ_a / Å (= $1/\nu$)	0.25
l_cc / Å	1.412
σ_1	0.1
L_c / Å (= $(\mu + 1)/\beta \cdot \overline{a}_3 = N \cdot \overline{a}_3$)	35
κ_c / Å (= $1/\beta$)	0.25
$\overline{a}_3$ / Å	3.5
$a_{3\,\mathrm{min}}$ / Å	3.1
σ_3 / Å	0.25
N (= $(\mu + 1)/\beta = N$)	10
u_3	0
η	1
q	0
Δ_an	0



```
cno = 0.0; #Concentration of disordered sp3 carbon
cH  = 0.0; #Concentration of hydrogen
cN  = 0.0; #Concentration of nitrogen
cO  = 0.0; #Concentration of oxygen
cS  = 0.0; #Concentration of sulfur

#Name of the series and id of the sample, filename of current used file
name = "WAXS example";
global id = "WAXS example Step 1";
filename = "WAXS Step 1 - path and data adjustments.m";
path = 'D:/OneDrive/Uni/PhD/Paper Octave/Github/NGCs/examples/WAXS Steps';
cd 'D:/OneDrive/Uni/PhD/Paper Octave/Github/NGCs/examples/WAXS Steps';
#Measurement data file
measFile = 'D:/OneDrive/Uni/PhD/Paper Octave/Github/NGCs/examples/WAXS Steps/WAXS example data.xy';

neutronCorrection = false; #Corrections for Wide-Angle Neutron Scattering (WANS) experiments.
neutronCorrectionVoigt = false; #Useful for samples containing hydrogen.

#Wavelength and type of radiation (0 = X-ray, 1 = neutrons)
wavelength = 1.5418;
radiation = 0;

#Type of x-values: theta, thetaRad, twoTheta, twoThetaRad, scatS, scatQ
type = "twoTheta";

useQ    = false; #Use Additional Deby-Waller-factor
b       = 0.002; #Deby-Waller-factor Factor
useA                  = true;     #Absorption correction
density               = 2.2;      #Density of sample in g/cm^3
sampleThickness       = 0.3;      #Sample thickness in cm
transmission          = false;    #Transmission geometry (if false, reflection geometry is assumed)
absorptionCorrection  = 1;        #Correction factor for absorption coefficient (multiplicator)
useP                  = true;     #Polarization correction
polarizedBeam         = false;    #Do you use a polarized beam?
polarizationDegree    = 0;        #Polarization direction of beam in °.
useCorrAutoColl = false;  #Slit-correction
par_r           = 14;     #Radius of the diffractometer (Debye-Scherrer) in cm
par_delta       = 4;      #Divergence angle in ° (it is converted as if this fixed slit were inside)
par_l           = 5;      #Irradiated length in cm
```

Figure S8 Overview about the basic instrumental parameters, which should be checked and changed for every different measurement.

S10. Example refinement

All files described in this section are also available under in the GitHub[22] and in the file "Example refinement.zip".

S10.1. Step 0 – initial situation

First, you have to download and install *Octave* and an *iObs* file as described below. Depending on your system, you might have to compile the *iObs* file by your own. For this case, the Numerical Recipes for C must be downloaded[23]. Generally, the lines 89 – 134 should be checked for each measurement or measurement group. In these lines, some parameters like the polarization or absorption as well as mathematical fit parameters like the function tolerance, upper and lower bounds and the weight can be tweaked. The initial (downloaded) refinement script (WAXS Fit-Routine-IUCr.m) can be found in the "Excerpt from Github.zip" file. In general, all other lines from 136 onwards should not be changed. The final modified script is also shown in S10.11.

S10.2. Step 1 – path and data adjustments

As first steps, v (line 14) must be tweaked. In general, for a common XRD using Copper-radiation (1.54 Å), $v = 4$ is sufficient. If you use another wavelength, you can look at Table S2 to get a suitable value for v. Additionally, the lines 41 – 87 must be checked. "name" means the name of your sample series or the current date or whatever you want to choose. The resulting refinements will be saved in this directory. The "global id" is a unique name for your current sample or step. You can also use a consecutive number to not override old refinements. "filename" must be the name of the currently used refinement scripts and can be found on top of the editor. "<path_to_filename>" must be replaced by the path, where the "filename" AND the *iObs.oct* file is placed. The *iObs.oct* file must be in the same directory as the refinement script. The complete path of your measurement file (x, y without headers) is stored in the variable "measFile". If you use neutron scattering or another wavelength, you must check the lines 54, 57, 60 and 61, otherwise you can skip these lines. If you want to plot only the coherent or incoherent scattering, you can change the values in the lines 64 and 65. Line 74 ("type") is important if you are not using 2 θ in ° as x-values. The parameters "nStart", "nEnd", "nSkip" and "nUp" can be used to skip some points at the beginning or the end or to plot only every nth point ("nSkip"). To prevent negative values of the intensity, you can use "nUp" to add a constant background to move the WAXS/WANS pattern up.

Regarding the measurement geometry, there are some correction terms, you can use (absorption, polarization and variable slit, lines 121 – 134). These parameters are set to Bragg-Brentano geometry using a unpolarized incidence beam and absorption correction for a 3 mm graphite sample. You should

---

[22] https://github.com/osswaldo/NGCs/tree/master/Example%20refinement/WAXS%20Steps

[23] http://numerical.recipes/com/storefront.html

check the parameters “density” and “sampleThickness” in lines 122 and 123 and change it to the values of your samples. It is not necessary to know the exact values, but you should put in the correct tendency.

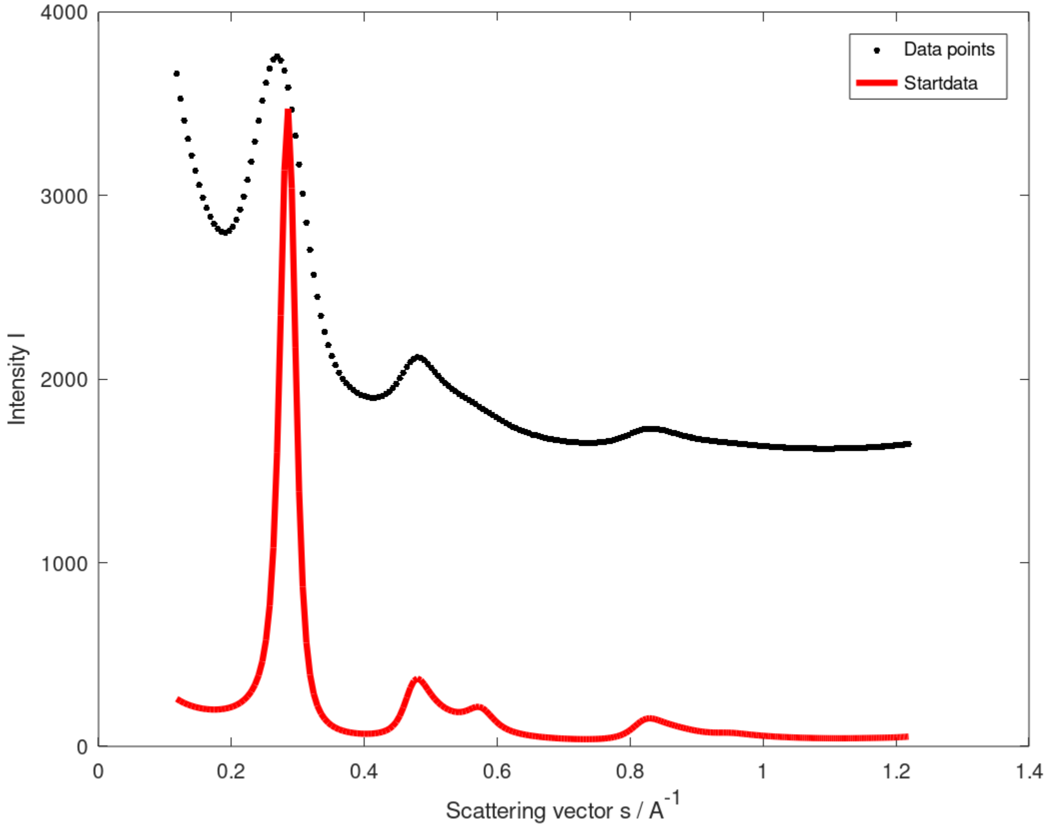


Figure S9 Step 1 - path and data adjustments without any refinements of the normalization or the microstructure.

S10.3. Step 2 – manual background

Second, the background must be corrected in order to start to refine the microstructure. “k” is a normalization constant depending on the amount of the sample, intensity of the radiation etc. and “const1” is used a constant offset of the scattering intensity caused by several effects like cosmic background radiation, incoherent scattering by your sample holder and other general measurement effects. The concentrations of foreign atoms (in atomic %) of hydrogen, nitrogen, oxygen and sulfur known from an elemental analysis can be considered in the lines 29 – 32 (parameters “cH”, “cN”, “cO” and “cS”).

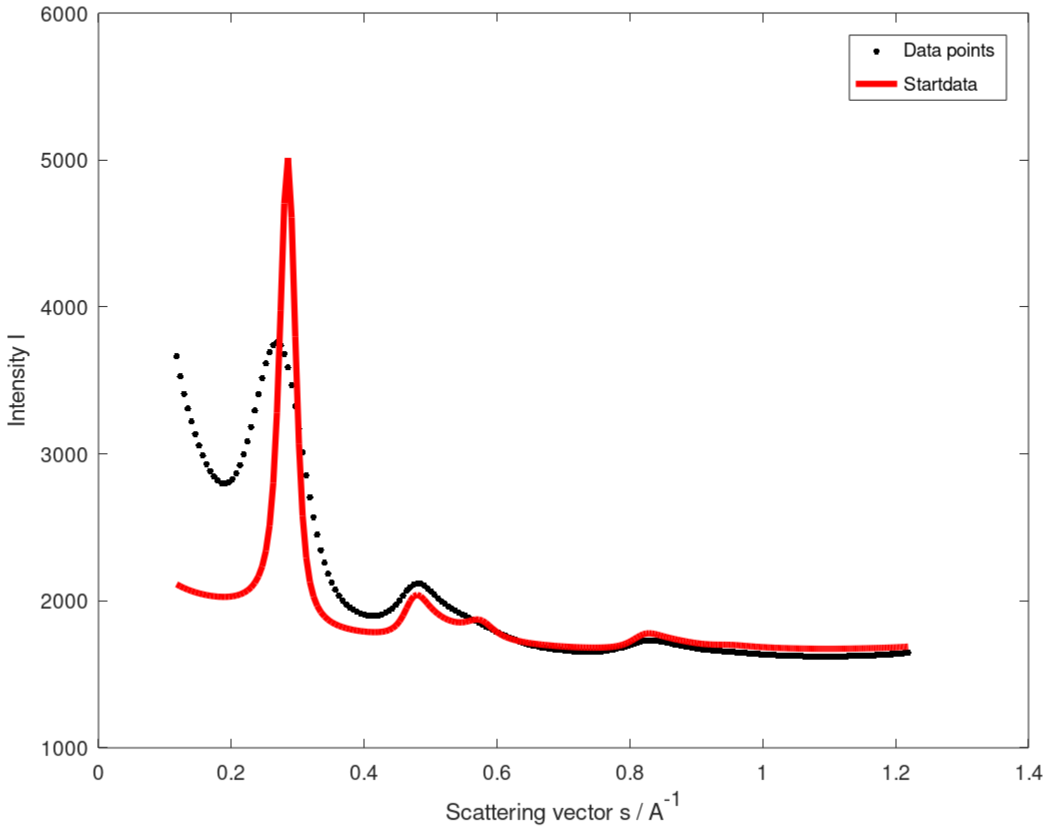


Figure S10 Step 2 – manual background. Only “k” and “const1” are refined.

S10.4. Step 3a – manual fitting – interlayer

To start the refinement of the microstructure, you should start with the interlayer scattering, but you can also start with the intralayer scattering (Step 4a). For the interlayer scattering, the parameters “mu”, “beta”, “a3”, “da3”, “sig3” and “eta” must be refined. In addition, “q” (preferred orientation), “k” and “const1” must be refined parallel (lines 7 – 22). The important and significant parts of the WAXS data are shown in Figure 5 in the main article. “mu” and “beta” are parameters to refine the stack height ($L_c = (\mu+1)/\beta \cdot \overline{a}_3$) and polydispersity ($\kappa_c = 1/\mu$), “a3” is the average layer distance ($\overline{a}_3$), “da3” is the difference between the average and the minimal layer distance ($a_{3\ \mathrm{min}} = \overline{a}_3 - \mathrm{d}a_3$), “sig3” the standard deviation of it (σ_3) and eta means the homogeneity of the stacks (η).

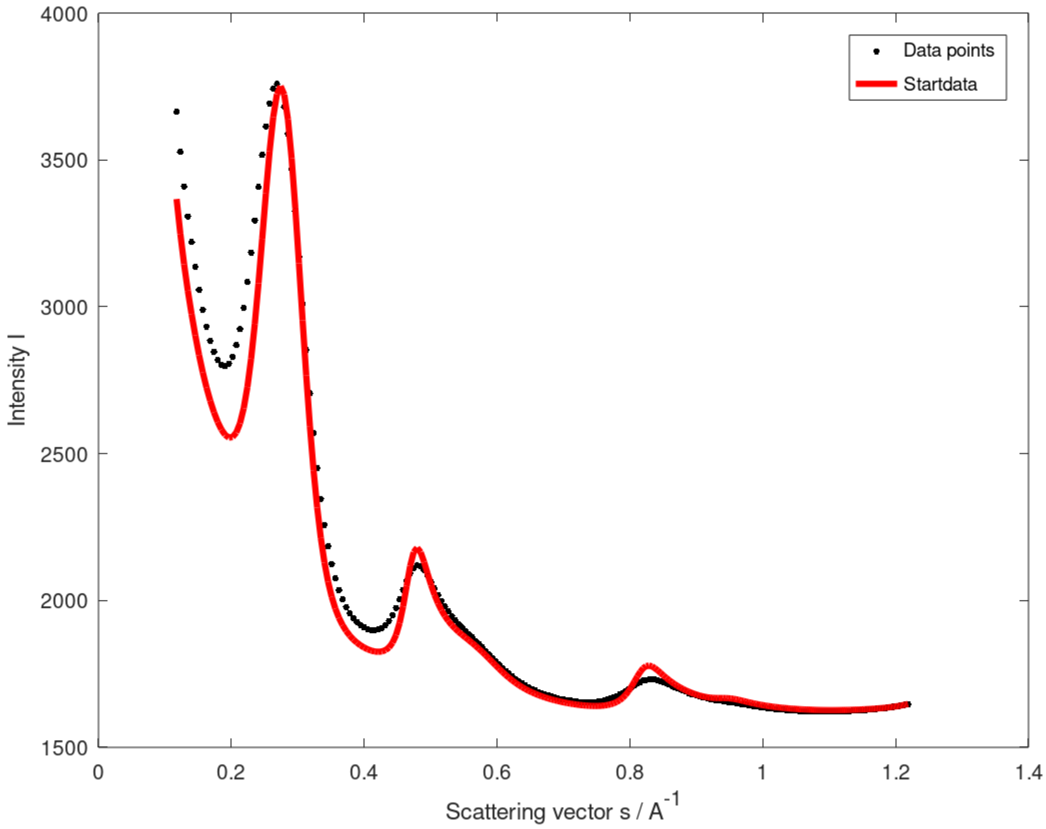


Figure S11 Step 3a – manual fitting – interlayer. The microstructure parameters for the interlayer scattering (“mu”, “beta”, “a3”, “da3”, “sig3” and “eta”) and “q” (preferred orientation) as well as “k” and “const1” were refined.

S10.5. Step 4a – manual fitting – intralayer

Beside the normalization, the parameters "alpha", "lcc" and "sig1" (lines 15 – 17) must be refined in this step. Analogous to the interlayer scattering, "alpha" is used to calculate the average layer extension ($L_a = (v+1)/\alpha$), l_{cc} is the average bond length and σ_1 the disorder of the layers (i.e. stress and strain).

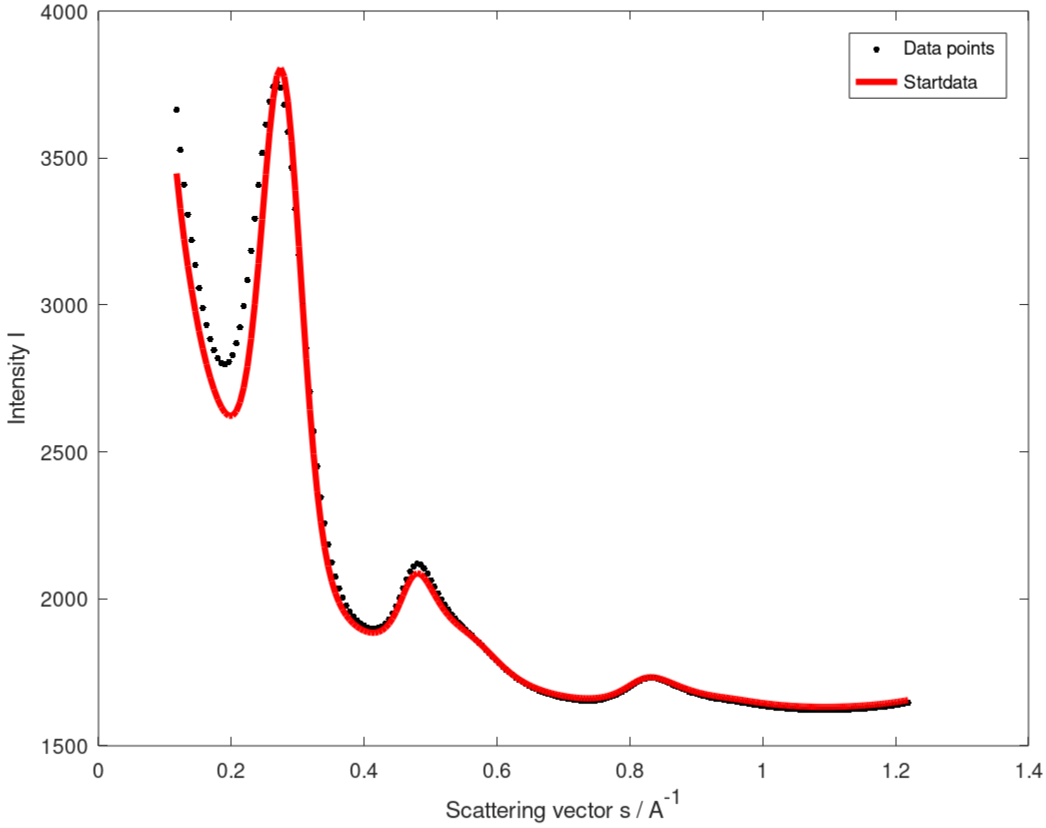


Figure S12 Step 4a – manual fitting – interlayer. The microstructure parameters for the intralayer scattering ("alpha", "lcc", "sig1") and "q" (preferred orientation) as well as "k" and "const1" were refined.

S10.6. Step 5a – manual fitting – all

For the final step of the manual fitting, you should set “nSkip” to 0 (line 84). Afterwards, try to refine all parameters as described above. You should start with the normalization constants, q and η. As an alternative, you can use these parameters as new start parameters for step 2. In principle, you should repeat the steps 2 – 5 multiple times to get a result, which is as good as possible.

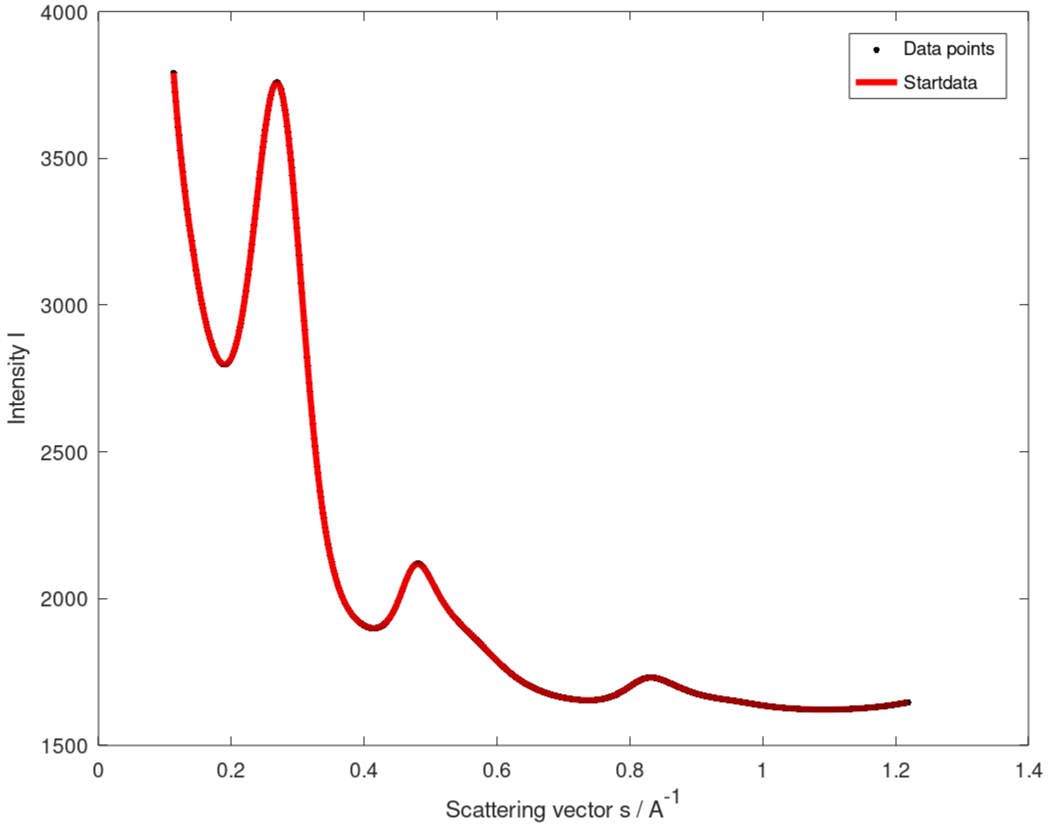


Figure S13 Step 5a - manual fitting – all. In this step, all microstructure and normalization parameters were refined at once.

S10.7. Step 3b – automatic fitting – initialization

To make things easier, you do not have to refine the scattering data manually. There is an automatically fitting routine, which can fit the scattering data and give out the microstructure data. To use this method, you need to roughly refine the background (step 2) set “plotOnly” to “false” (line 35). Basically, that is all what you have to do (of course, you have to press the “run” button on top of the *Octave* GUI). Beside this, you can set the lower and upper bonds for the different microstructure parameters (89 – 104). This might be useful, if you have a temperature series and some parameters have to increase or decrease, but in general, it is not necessary to use it.

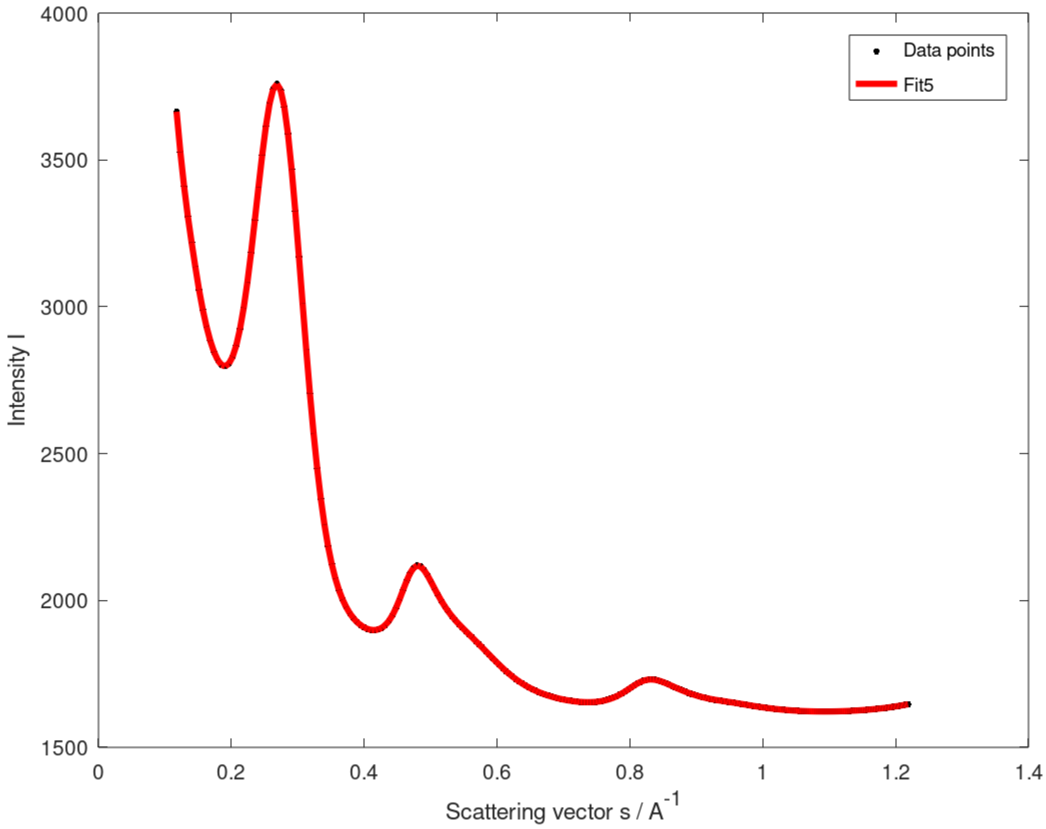


Figure S14 Step 3b – result of the automatic refinement. No further refinement must be done.

S10.8. Step 4b – automatic fitting - fine adjustment

If everything works well, you do not need this step (like in this example). For other samples or insufficient start values or a too bad refinement of the normalization in step 2, you have to make a manual refinement after the automatically fit. To do so, change “plotOnly” back to “true” and perform the steps 3a – 5a. As an alternative, use the resulting parameters as new start parameters and repeat step 3b.

S10.9. Common warnings/errors during the refinement and how to fix them

A list of common errors and their solutions can be found in the GitHub [24] and in the SI under Octave/README.pdf.

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[24] https://github.com/osswaldo/NGCs

S11. General influence of the amount/noise of data points and start parameters on the fitting routine

The influence of the number of data points and the noise must be considered in a differentiated manner. Evidently, data noise should be as low as possible, especially for the parts of low scattering intensities in a WAXS/WANS curve.

We find that the number of points and the spread of the measurement data influence the time required for the refinement (Table S4), which is notably also dependent on the computing hardware used. In general, *Octave* uses only one core for computing, especially on Microsoft Windows. For Linux distributions, parallel computing can be performed using *OpenBLAS*, it is thus sufficient to use a cheap Raspberry Pi running on Linux. For high-performance computing clusters (HPCs), the option to perform several refinements simultaneously is a main advantage, which can decrease the refinement time for one WAXS/WANS data set. For a single refinement, the HPC might not even be faster than a commonly used desktop computer, since *Octave* mainly performs computation only with one CPU core. HPCs are mainly built to perform multiple parallel computations, and they might have slower cores than other commercially available PCs. On the other hand, HPCs can run 50 or more *Octave* scripts (*OctCarb* instances) at once, so that the overall time requirement is lower for a high number of samples/data sets.

Another point is the available RAM, which is needed to save and perform the huge matrices calculations (covariance and correlation matrices). The N x N matrix dimension increases quadratically with the number of data points (N), thus HPCs are advantageous for data sets with a high number of measured points (10.000 and above). Another important point is the range of the measured data. For maximum values of the modules of the scattering vector $s_{\max} > 4$ Å^{-1}, a lot of (*hk*) reflections must be calculated and saved in between (in the RAM), here HPCs offer a clear benefit.

Last but not least, we found that the start parameters used as input for launching the refinement are not that relevant with regard to attaining a decent fitting, with the exception of the normalization parameters k, $const_1$ and $const_2$ (see eq. (6) from the main article). These scaling parameters need to be refined first, prior to the microstructure parameters, aiming at a reasonable match between the fit function and the WAXS/WANS curve with respect to the global scaling. For all parameters, the standard input parameters from Table S6 can be used for the automatically refinement. Using the automatic refinement, first a refinement of the normalization parameters is done, and afterwards the physical microstructure parameters will be refined. Hence, the interlayer and intralayer parameters were refined separately, so these steps need only a short computation time, due to the only small number of simultaneously refined parameters. Only the last step, in which all parameters are refined altogether, takes a long time, because the calculation time is roughly proportional to the square of the number of the number of parameters to be refined.

S12. *Octave* cannot always calculate parameter errors – what to do

The following issue is important for determining the error range of the refinement parameters: In some cases, *Octave* is not able to calculate the correlation and covariation matrices correctly, generating "NaN" as output values for the errors. However, this does not disturb the quality of the fitting, i.e. the resulting structural parameters themselves are still reasonable. If the errors are, however, relevant, in such cases there are basically 2 ways to work around the problem, described in the SI this section and Figure S15. First, the resulting parameters obtained from the fitting can be used as input parameters for another automatic refinement (variant 1 in Figure S15). This procedure might already allow *Octave* to calculate the matrices and therefore the errors correctly. If not, the errors from the intralayer- and interlayer-refinement (steps 1 and 2 of the refinement script) can be used as errors. Second, the "NaN" error often occurs for the normalization parameters k and $const_1$. Hence, the influence of the variation of these parameters on the resulting scattering curve is negligible (< 0.1 %). Second, another possibility to avoid the NaN problem and to obtain error bars is to add an additional refinement step after all other steps, in which all microstructure parameters except for the normalization parameters are refined and therefore the errors of the parameters are calculated (variant 2 in Figure S15). To do so, some other changes are necessary and already implemented in the file "WAXS Fit-Routine-IUCr with additional step all-without-normalization.m".

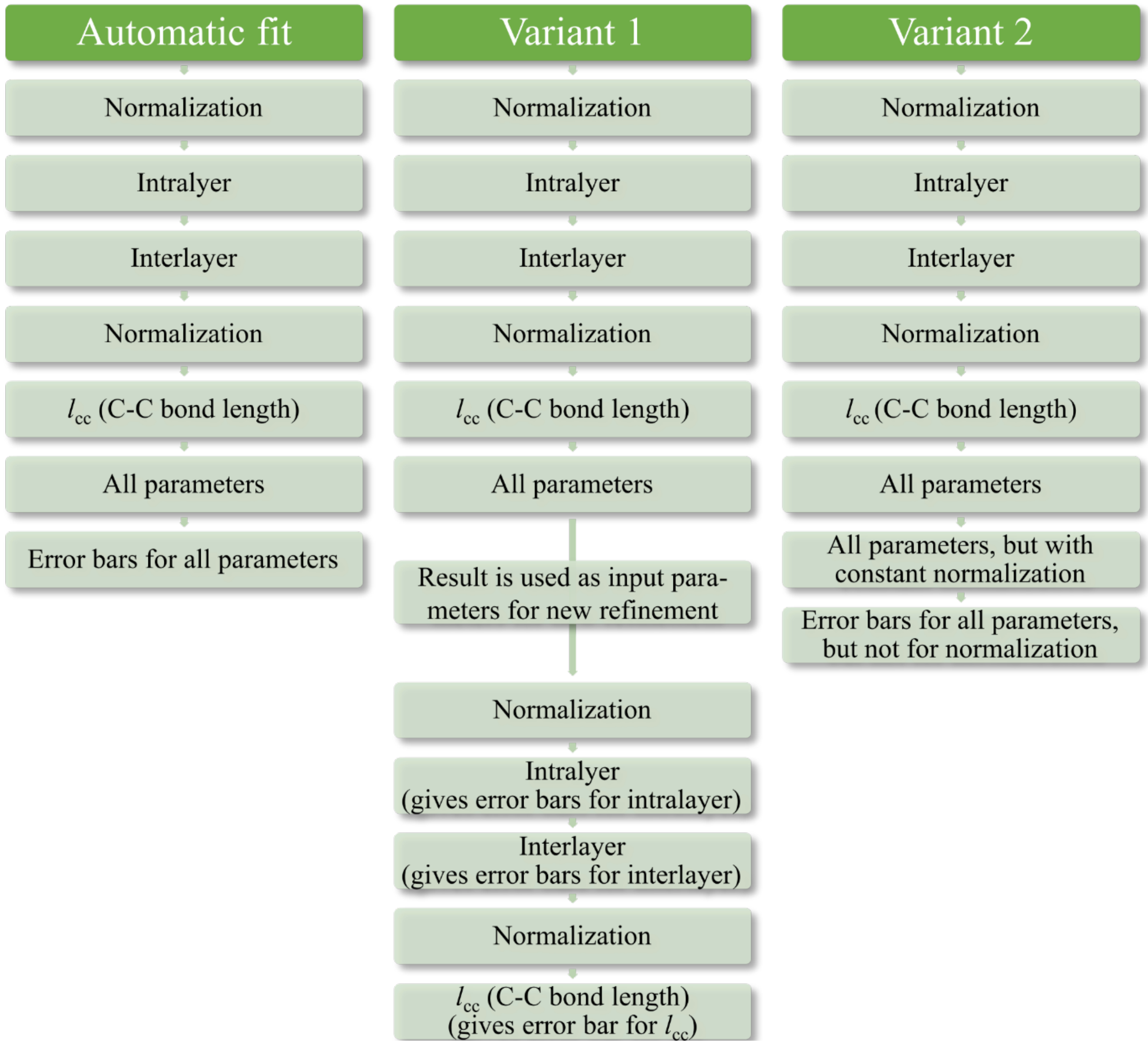
Automatic fit
Normalization
Intralayer
Interlayer
Normalization
l_{cc} (C-C bond length)
All parameters
Error bars for all parameters
Variant 1
Normalization
Intralayer
Interlayer
Normalization
l_{cc} (C-C bond length)
All parameters
Result is used as input parameters for new refinement
Normalization
Intralayer (gives error bars for intralayer)
Interlayer (gives error bars for interlayer)
Normalization
l_{cc} (C-C bond length) (gives error bar for l_{cc})
Variant 2
Normalization
Intralayer
Interlayer
Normalization
l_{cc} (C-C bond length)
All parameters
All parameters, but with constant normalization
Error bars for all parameters, but not for normalization

Figure S15 Schematic representation for calculating the error bars for the refined parameters. If the automatic fit cannot calculate the errors, there are basically 2 options for how to proceed: Variant 1 can be used with the existing script. The result of the fit is used there as input parameters for a new fit. In this new fit, the error from the individual steps for the intralayer and interlayer parameters can be determined individually. For mathematical reasons, the error here is generally higher than if all parameters were calculated at the same time. Variant 2 describes the procedure with a modified script. In this case, after the automatic fit, a fit of intra- and interlayer parameters is performed while the now known normalization parameters are kept constant. This allows the error bars to be calculated, but it is possible that the structure parameters will change very slightly as a result.

S13. Whole refinement script (*OctCarb*)

The final refinement scripts (*OctCarb*), which can used with *Octave* are in the SI in the directory "Example refinement". In principle, the file "WAXS Fit-Routine-IUCr.m" should be used to perform the whole refinement. In some cases, it might happen, that the errors of the refined parameters cannot be calculated (see S11). This is often caused by the refinement of the normalization parameters (which one could not be determined so far). In this case and if the error values of the refined parameters needed to be known (and only in this case), the file "WAXS Fit-Routine-IUCr with additional step all-without-normalization.m" should be used. Hence, an additional refinement step including the intralayer and interlayer but without the normalization parameters will be performed. This will reduce a bit the quality of the whole fit, but *Octave* will calculate the parameter errors correctly.

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