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

Supporting information part B

Correction/fine treatment of WAXS/WANS data
Mathematical background

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Supporting information part B – Correction/fine treatment of WAXS/WANS data and mathematical background

S14. Incoherent scattering and correction terms for WAXS

I_{incoh} is modified by different specific correlation factors for primary/secondary monochromatizing, e.g. Q_{abs} (specific absorption of Compton scattering), Q (secondary-monochromator) and the Breit-Dirac *recoil* factor. However, equation (15) is only valid for X-ray radiation and not for neutrons:

$$I_{incoh, c} = I_{com, c} \cdot recoil \cdot Q_{abs} \cdot Q \tag{15}$$

The influence of the different correction terms for incoherent x-ray scattering of NGCs is described in Figure S16. Further details about the incoherent scattering can be found in following parts. For WANS, in principle a similar equation like eq. (3) of the main article holds true, but it is not meaningful to calculate the inelastic scattering in an analytical way due to possible nuclear absorption resonance and possible similar values of the energy exchange and the incident energy. In essence, the possibly huge non-linear background of hydrogen, even present for small hydrogen contents of NGC, cannot be evaluated analytically. Hence, we treat WANS data in a different way, namely by fitting the incoherent background either by a polynomial or a Pseudo-Voigt function, depending on the amount of hydrogen in the sample as described in the works of Placzek [1] and Fischer et al. [2] as shown in S19. Thus, the pristine WANS data are corrected by subtraction of $I_{incoh,c}$ from the pristine WANS data prior to further evaluation, i.e. $I_{incoh, c}$ is not evaluated simultaneously together with the structural parameters, as performed for WAXS analysis.

Besides this description of the scattering of carbon, the scattering of foreign atoms (currently only H, N, O, S) can be considered if present in the sample. Since these foreign atoms cause an incoherent background, the following equations (16) and (17) are only valid for WAXS and not for WANS data. For the foreign atoms, the scattering intensity is given by (X = H, N, O, S)

$$I_{e.u., X} = I_{coh, X} + I_{incoh, X} = f^2{}_X + I_{com, X} \cdot recoil \cdot Q_{abs} \tag{16}$$

In this case, the intensity $I_{e.u., x}$ for every element is weighted by its concentration (c_H, c_O, c_N, c_S and $c_C = 1 - c_H - c_O - c_N - c_S$). Therefore, the overall resulting scattering of all atoms $I_{e.u.,}$ is calculated by

$$I_{e.u.} = c_C \cdot I_{e.u., c} + c_H \cdot I_{e.u., H} + c_N \cdot I_{e.u., N} + c_O \cdot I_{e.u., O} + c_S \cdot I_{e.u., S} \tag{17}$$

A detailed description about the calculation of the scattering intensity of the foreign atoms as well as the calculation of the atomic form factors including fits is given in S17.

S14.1. Compton scattering

The theoretical data for the atomic form factor of carbon is given in Milberg and Brailsford [3] for $s \leq$ 1.4 Å^{-1} (which is also used in Pfaff et al. [4]). A more detailed description is in Keating and Vineyard

[5]. For $s > 1.4$ Å^{-1} the data from Prince et al. [6] is used. Using a spline interpolation (5 parts, 3rd order polynomial), the intensity of Compton scattering of carbon is given by

$$I_{com,c}(s)$$
$$= \begin{cases} -33.525 \cdot s^3 + 26.296 \cdot s^2 + 2.160 \cdot s - 0.025, 0 < s \le 0.4 \\ 11.179 \cdot (s-0.4)^3 - 13.935 \cdot (s-0.4)^2 + 7.104 \cdot (s-0.4) - 2.901, 0.4 < s \le 0.8 \\ 0.222 \cdot (s-0.4)^3 - 0.520 \cdot (s-0.4)^2 + 1.322 \cdot (s-0.4) + 4.228, 0.8 < s \le 1.2 \\ 0.013 \cdot (s-0.4)^3 - 0.254 \cdot (s-0.4)^2 + 1.013 \cdot (s-0.4) + 4.688, 1.2 < s \le 1.8 \\ 0.023 \cdot (s-0.4)^3 - 0.230 \cdot (s-0.4)^2 + 0.722 \cdot (s-0.4) + 5.207, 1.8 < s \le 4 \end{cases} \quad (18)$$

S14.2. Breit-Dirac correction

The correction factor for the recoil effect as used in Ruland [7] and described in Pfaff et al. [4] is given by using the Breit-Dirac equation intensity of Compton scattering of carbon is given by

$$recoil = \left(\frac{1}{1 + 0.0485262 \cdot \lambda_A \cdot \frac{1}{4} \cdot s^2} \right)^3 \quad (19)$$

which can be reached using James [8] (20) and the Bragg's law (21) and the physical constants

$$B = 1 + \frac{2 \cdot h \cdot \lambda}{m \cdot c} \cdot \left(\frac{\sin(\theta)}{\lambda_m} \right)^2 \quad (20)$$

$$\lambda_A = 2 \cdot d \cdot \sin(\theta) \quad (21)$$

λ_m = Wavelength in m

λ_A = Wavelength in Å

B = Total scattered radiation ($recoil = B^3$)

h = Planck constant ($6.62607015 \cdot 10^{-37}$ J s) [1]

c = Speed of Light ($299792458 \cdot 10^{-37}$ m/s) [2]

m = Mass of electron ($9.1093837015(28) \cdot 10^{-31}$ kg) [3]

d = Lattice distance in Å ($d = 1/s$)

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[1] https://physics.nist.gov/cgi-bin/cuu/Value?me
[2] https://physics.nist.gov/cgi-bin/cuu/Value?c
[3] https://physics.nist.gov/cgi-bin/cuu/Value?h

S14.3. Absorption correction

The absorption correction of the incoherent scattering of the sample is given in Ruland [7] as

$$Q_{abs} = 1 + \frac{3 \cdot h}{m \cdot c \cdot \lambda_m} \cdot (\sin(\theta))^2 \tag{22}$$

and can be transformed into [4]

$$Q_{abs} = \frac{1}{1 + 0.0485262 \cdot \lambda_A \cdot \frac{3}{8} \cdot s^2} \tag{23}$$

S14.3.1. Secondary monochromator correction

While the recoil and absorption correction must be considered for each measurement, a secondary monochromator and the corresponding correction is optional. It is given by Ruland [9] as

$$Q = \frac{1}{\left(1 + \frac{\Delta\lambda}{b}\right) \cdot \left(1 + \frac{\pi^2}{(\Delta\lambda + b)^2} \Delta\lambda_C^2\right)} \tag{24}$$

(b = pass-band). For carbon $\Delta\lambda$ and $\Delta\lambda_C$ can be calculated using the following expressions

$$\Delta\lambda = \frac{\lambda_A^2}{c} \cdot s \cdot \Delta q(s) \tag{25}$$

$$\Delta q(s) = \Delta q_{max} \frac{s^2}{a^2 + s^2} \tag{26}$$

For graphite: $\Delta q_{max} = 3.05$, a $= 0.53$

$$\Delta\lambda_C = \frac{2 \cdot h}{m \cdot c} \cdot (\sin(\theta))^2 - D \cdot \lambda_A^2 \tag{27}$$

In this case, c is the velocity of light given in astronomical unit ($c = 137$) and $D = 1.5 \cdot 10^{-3}$ for carbon. Since this equation is only valid for high Compton energies, it must be corrected for low energies, especially for copper radiation, where the $\Delta\lambda_c$ would be negative for $\theta < 23\,°$. It can be assumed, that D correlates with $\Delta q(s)$ for low energies. In this way, an additional term like equation (26) can be used to consider this effect. Finally, the correction term for the secondary monochromator is [4]:

$$Q = \cfrac{1}{\left(1 + \cfrac{\cfrac{\lambda_A^2 \cdot s}{137} \cdot \left(\cfrac{3.05 \cdot s^2}{0.53^2 + s^2}\right)}{b}\right) \cdot \left(1 + \cfrac{\pi^2 \cdot \left(\cfrac{0.0485262}{4} \cdot s^2 \cdot \lambda_A^2 - \cfrac{1.5 \cdot 10^{-3} \cdot \lambda^2 \cdot s^4}{(0.53^2 + s^2)^2}\right)^2}{\left(\cfrac{\lambda_A^2 \cdot s}{137} \cdot \left(\cfrac{3.05 \cdot s^2}{0.53^2 + s^2}\right) + b\right)^2}\right)} \tag{28}$$
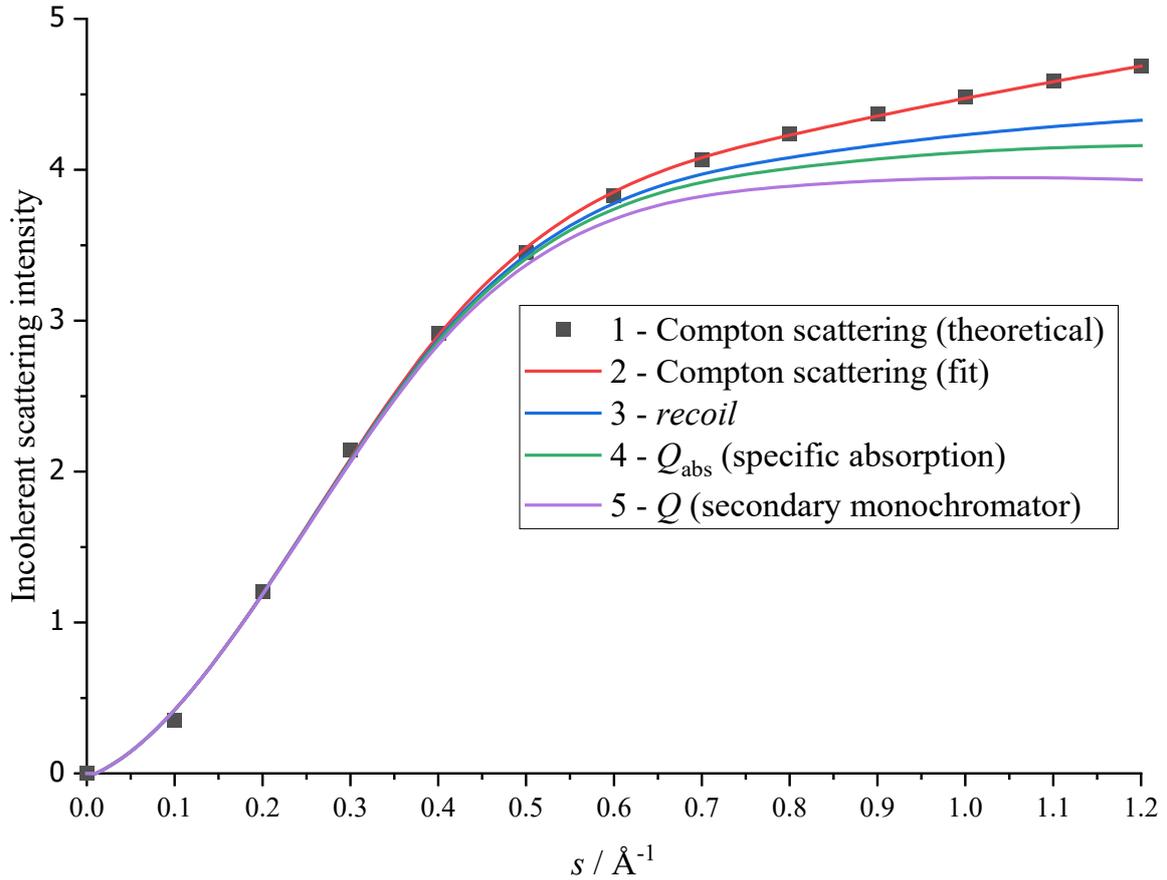

Figure S17 Theoretical (1) intensity and spline interpolation (2) of the Compton scattering of carbon. This scattering must be corrected with the Breit-Dirac recoil factor (*recoil*) and the specific absorption (Q_{abs}). In addition, the absorption of a possible secondary monochromator can also be considered (Q).

S15. Implementation of ν

The last difference in the code used in this study compared to Pfaff et al. [4] is the implementation of ν, which together with α serves for modelling the intralayer scattering I_{intra}. Since this is a very special feature, it is not handled in the main text, but in the SI in part … ν is used to calculate a polydispersity parameter for the chord-length distribution of the graphene layers (κ_a).

In Ruland and Smarsly's model [10], the correlation function $P_L(r)$ of the intralayer scattering is calculated as a gamma function Γ with the parameters ν and α:

$$P_L(r) = \frac{1}{\Gamma(\nu+1)} [\Gamma(\nu+1, \alpha r) - \alpha r \Gamma(\nu, \alpha r)] \tag{29}$$

$\Gamma(x)$ is the complete, and $\Gamma(a, x)$ the incomplete gamma function. ν and α are related to the layer size (L_a) and its polydispersity (κ_a) as follows:

$$L_a = (\nu + 1)/\alpha \tag{30}$$

$$\kappa_a = 1/\nu \tag{31}$$

Thus, both parameters ν and α are needed to calculate the resulting intralayer parameters by this approach. Equation (32) is used to calculate the shape profile of the intralayer reflections (J_{hk}):

$$J_{hk}(s) = \frac{1}{s} \text{Im} \left[\int_0^\infty P_L(r) J_0(2\pi r s_{hk}) \exp(2\pi i r s) \, dr \right] \tag{32}$$

It should be noted that equation (32) does NOT include the disorder of the layers. s_{hk} is the position of the hk reflection, which should be calculated and J_0 the Bessel function of the first kind of order zero. We found that a numerical calculation of J_{hk} by eq. (32) takes inacceptable time. Since equation (32) cannot be solved analytically for real values of ν, but for whole numbers, ν is set to a fixed whole number for one particular fit in our approach.

Since ν generally has also only a very small influence on the resulting WAXS pattern for the same layer sizes (Figure S17), it is not meaningful to refine this parameter. Note that we had already proposed in a previous study (Faber et al. [11]) that the value of κ_a (ν) cannot be reliably determined. We found the average layer extension (L_a) is predominantly determined by α, and the disorder of the layers (σ_1) has a much higher influence on the profile shape than ν. Hence, setting ν *constant* does not compromise the accuracy of the refinement and the physical assumptions in the model at the same time.

The parameter is now rather used as empirical limit for the number of (hk) reflections to be calculated and based on eq. (32) indirectly specify an upper limit for the polydispersity:

$$k \leq h \leq v \text{ and } h^2 + h \cdot k + k^2 < (v+1)^2 \tag{33}$$

$$\kappa_a \leq 1/v \tag{34}$$

This implementation and the relationships (33) and (34) have no direct physical basis, rather it is a practical implementation for a better software-handling based on empirical knowledge. Yet, this procedure is also justified with respect to the NGC structure, as a high amount of observable (hk) reflections means a small layer disorder (κ_a) and therefore a high value of v. More details about the implementation and calculation times for different values of v can be found in the SI file under S2.
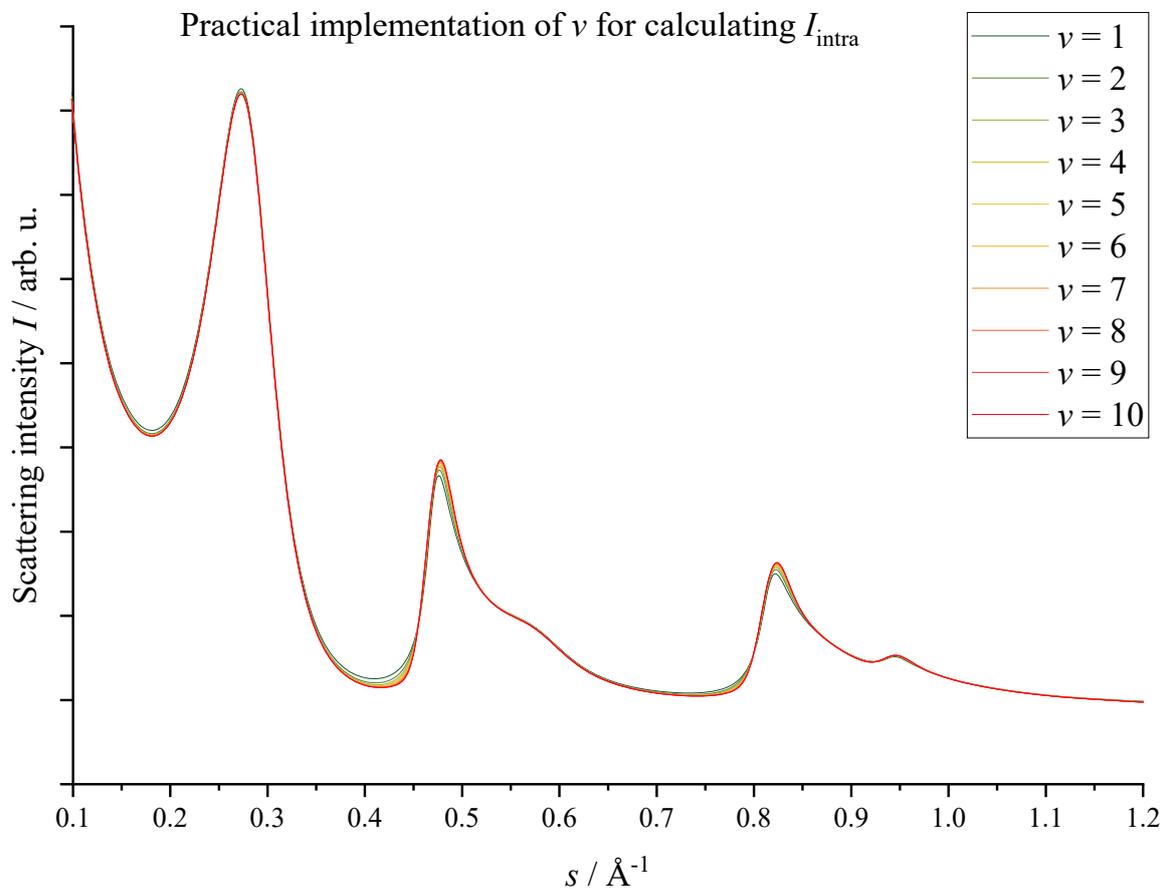
Figure S18 Influence of v on a typical WAXS scattering pattern. v is one of the two parameters used for defined a chord-length distribution of the graphene layers, according to [10]. Since this parameter influences only the intralayer scattering, the (002) and (004) reflections are not affected. The calculations are done for a constant $L_a = 30$ Å. Hence, it is seen that v only minor affects the shape of "classic" scattering patterns of NGCs. It is thus sufficient to use a fixed value of v in the fitting routine.

S16. General intensity correction terms

S16.1. General remarks

The following correction terms are dependent on the sample and the measurement geometry. There is no generally applicable guideline which corrections must be used and which not. All correction terms except the experimental broadening widening are implemented in *OctCarb* and can be optionally used. Overall, the correction for the fixed irradiated length (if used), the polarization and the Polarization correction must be used for every measurement, but note that these corrections might already be performed by the measurement software of the device. *gFact* is a general scaling correction term and is not directly related to a physical phenomenon, so it should be used in the refinement only, if refining the structural parameters does not result in a reasonable fitting. The absorption correction can be used, but in many of the cases studied, the correction is comparably small in its effect on the refinement. Only for very thin samples in reflection geometry using X-ray radiation, e.g. for thin film measurement, the absorption correction has a significant to the measured intensity.

S16.2. Experimental device broadening

For the instrumental broadening, no generally applicable procedure can be given. The best way is to measure the instrumental broadening using a reference sample and then to correct the measured data before refining it. However, there are some approaches for such data correction, which are explained in the SI file of Pfaff et al. [4]. However, the influence of the device broadening is only very small for materials with broad reflections, i.e. for quite disordered carbons (see Figure S18). The black dots are a typical (002) reflection profile, which can be roughly fitted through a Gaussian profile (red, standard deviation is 0.01 Å^{-1}). For a XRD instrument with a medium resolution of ca. 100 nm (domain size), which means a standard deviation of 0.0007413 Å^{-1} for an integral width of 0.001 Å^{-1}, the resulting reflection broadening is shown in the blue line. The resulting corrected gauss profile of the (002) reflection is included as green line. The difference is evidently small and therefore, the device broadening can be ignored for most of the cases.
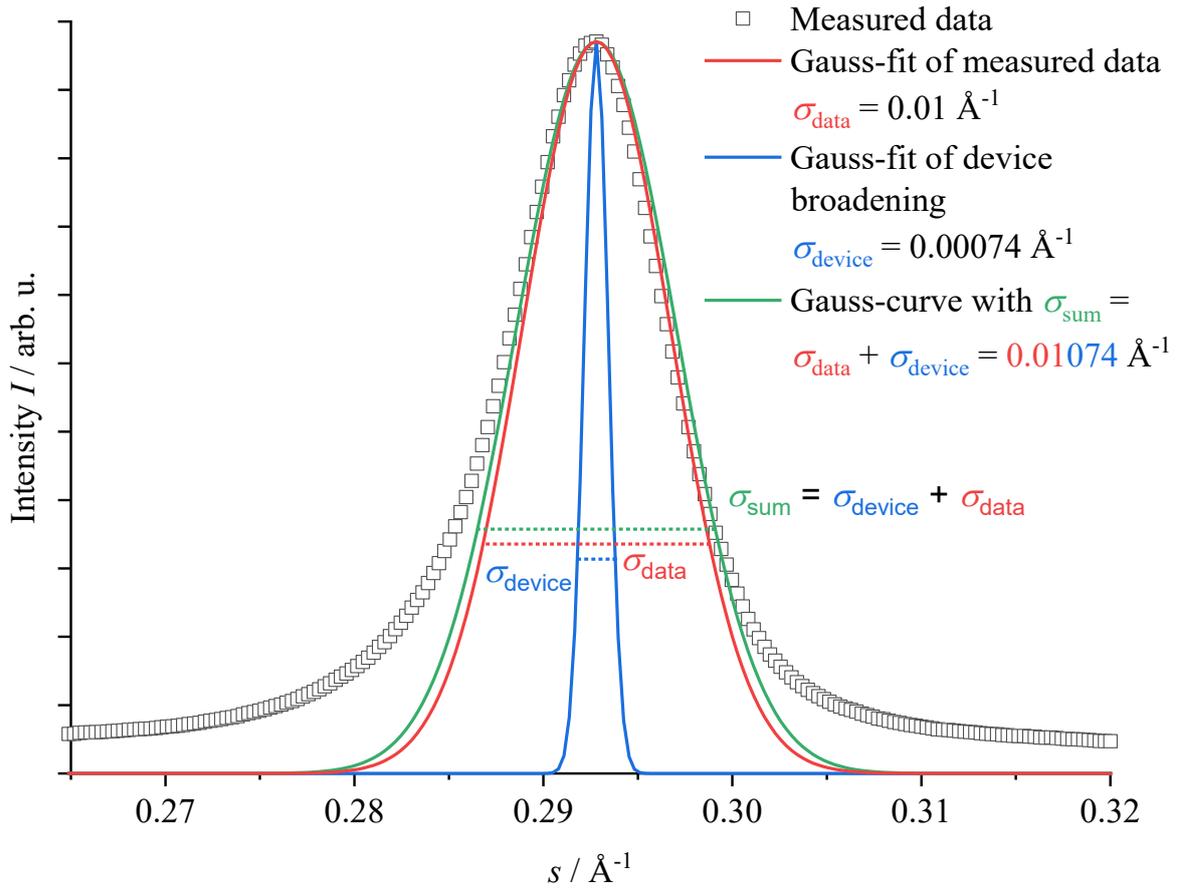
Figure S19 Overview of the influence of the device broadening effect. The (002) reflection is so wide that the device broadening does not have to be considered.

S16.3. AutoColl

The correction term *AutoColl* can convert scattering data from Bragg-Brentano geometry at the scattering angle θ using a fixed irradiated length to a fixed slit geometry with the divergence angle δ. The goniometer radius r and the irradiated length l must have the same dimension [4].

$$AutoColl = \frac{r \cdot \sin(\theta) \cdot \sin^2(\delta)}{l \cdot \left(\sin^2(\theta) - \sin^2\left(\frac{\delta}{2}\right) \right)} \tag{35}$$

S16.4. gFact

gFact can be used to perform an exponential damping or increasing of the overall scattering curve due to scattering effects which we have not considered so far, e.g. for a possible influence of a small-angle X-ray/neutron (SAXS/WANS) intensity contribution at low s values choosing a meaningful parameter g [4]. Since it does not have any physical meaning, the factor should only be used for special reasons and not by default.

$$gFact = \exp(g \cdot s) \tag{36}$$

S16.5. Lorentz-factor

The Lorentz factor as described in [12,13] is considered in the model of Ruland and Smarsly [10] and therefore, there is no need to consider a further Lorentz factor as an additional correction term.

S16.6. Polarization

In general, the polarization correction can only be used for X-rays, but not for neutron radiation due to the different orientated electron orbitals. For a polarized radiation source with the polarization direction Φ, the following equation can be used [14]:

$$P(\theta, \Phi) = 1 - \sin^2(2\theta) \cos^2(\Phi) \tag{37}$$

For a non-polarized radiation, the polarization factor can be calculated using the average of Φ from 0 to 2π [4,14]:

$$P(\theta) = \frac{1}{2\pi} \int_0^{2\pi} P(\theta, \Phi)\, \mathrm{d}\Phi = \frac{1}{2}(1 + \cos^2(2\theta)) \tag{38}$$

S16.7. Absorption

For the absorption correction, both, the measurement geometry as well as the type of radiation (X-ray or neutrons) must be considered. The most common cases for wide-angle powder scattering are the reflection geometry for X-rays and transmission geometry for neutrons and therefore, this will be discussed here. The discussion and formulars are based on scripts from W. Ruland (1995) and the work of Sivia [14].

S16.7.1. Absorption coefficients

For X-ray radiation, μ_{ab} is the mass attenuation coefficient, which can be fitted using the data from [15]:

$$\mu_{ab} = \lambda^{-3.089} \cdot 10^{1.081} \cdot \rho \tag{39}$$

ρ is the density of the sample in g/cm^3. For neutron radiation, the absorption coefficient is given by

$$\mu_{ab} = n \cdot \sigma_{abs} \tag{40}$$

n is the number of atoms per unit cell:

$$n = \rho \cdot N_A/m = \rho \cdot 6.02214076 \cdot 10^{23}/12.011 \text{ cm}^3/\text{g} \tag{41}$$

σ_{abs} can be calculated using the wave vector ω (in Å^{-1}) and the incoherent neutron cross section σ_{inc}:

$$\sigma_{abs} = 4\pi/\omega \cdot \sigma_{inc} \tag{42}$$

$$\omega = 2\pi/\lambda \tag{43}$$

For carbon, $\sigma_{inc} = 0.001 \cdot 10^{-24}$ cm^2 [16]. Per definition, σ_{abs} is measured and tabulated for neutrons with a velocity of $v = 2200$ m/s, i.e. a wavelength of $\lambda = 1.8$ Å, but in most of the cases, σ_{abs} must be calculated for the used wavelength.

S16.7.2. Reflection geometry

For reflection geometry, i.e. Bragg-Brentano geometry, the case of an infinitely thick sample with a thickness smaller than the maximum depth of penetration of X-ray radiation in a carbon sample can be used. The covered track of the X-ray radiation in the sample with a thickness d in cm is not constant. Therefore, also the absorption correction is not constant:

$$I(\theta, \mu_{ab}, d) = I_0(\theta) \cdot \frac{1}{2 \cdot \mu_{ab}} \cdot \left(1 - \exp\left(\frac{-2 \cdot d \cdot \mu_{ab}}{\sin(\theta)}\right)\right) \tag{44}$$

S16.7.3. Transmission geometry

The average way of a neutron through a cylindric sample is not the diameter but smaller, so it must be calculated. For a beam bigger than the sample diameter, die average length is

$$l = \frac{\int_0^d \sqrt{d^2 - 4x^2}\ dx}{d} = \frac{\pi}{4}d \qquad (45)$$

where $\sqrt{d^2 - 4x^2}$ is the chord length of a circle with the diameter d with a distance x to the center. For a beam smaller than the diameter of the cylindric sample ($a < d$), the average length is

$$l = \frac{\int_0^{\frac{a}{2}} \sqrt{d^2 - 4x^2}\ dx}{\frac{a}{2}} = \frac{a\sqrt{-a^2 + d^2} + d^2 \mathrm{ArcTan}\left[\frac{a}{\sqrt{-a^2 + d^2}}\right]}{2a} \qquad (46)$$

In *Octave*, only the mainly used first case is considered, so if you want to use an endless beam width smaller than the diameter, you have to calculate the resulting irradiated length by your own using

$$d = \frac{4}{\pi}l \qquad (47)$$

The resulting absorption can be calculated by

$$I(\theta,\ \mu_{ab},\ l) = I_0(\theta) \cdot \frac{l}{\cos(\theta)} \cdot \exp\left(\frac{-l \cdot \mu_{ab}}{\cos(\theta)}\right) \qquad (48)$$

S17. Atomic form factors

In general, the atomic form factor can be calculated solving the Schrödinger equation or relativistic wavefunction for heavier atoms than hydrogen. As this is procedure very complex and time-consuming to calculate, these values have already been calculated and tabulated [6]. In addition, the atomic form factors can be fitted using

$$f(s) = \sum_{i=1}^{4} a_i \cdot \exp\left(-b_i \cdot s^2\right) + c \tag{49}$$

s = modules of the scattering vector (in Å)

Since this equation diverges to c instead to 0, Fox et al. [17] proofed the following formular as more accurate for higher values of s, which correspondences to relativistic Hartree-Fock wavefunctions:

$$f(s) = \exp\left(\sum_{i=0}^{3} a_i \cdot \left(\frac{s}{2}\right)^i\right) \tag{50}$$

S17.1. Atomic form factor of carbon

Regarding equation (49) and (50) and Prince et al. [6], the atomic form factor of carbon is given by

$$f_{c,perp}(s) = \begin{cases} 2.31 \cdot \exp(-20.84339 \cdot s^2) + 1.02 \cdot \exp(-10.2075 \cdot s^2) \\ +1.5886 \cdot \exp(0.5687 \cdot s^2) + 0.865 \cdot \exp(-51.6512 \cdot s^2) + 0.2156, s < 3.74 \\ \exp\left(1.7056 - 1.5676 \cdot \dfrac{s}{2} + \dfrac{1.1893}{10}\left(\dfrac{s}{2}\right)^2 - \dfrac{0.42715}{100}\left(\dfrac{s}{2}\right)^3\right), \text{else} \end{cases} \tag{51}$$

This equation is a little bit different to Pfaff et al. [4] because of the introduction of the 4th exponential term and the correction for high s-values, which make the fit a little bit more accurate

S17.1.1. Anisotropy

For the anisotropy of the atomic form factor, i.e. the anisotropy in the interlayer scattering caused of the parallel stacking of the graphene layers, the parameter Δ_{an} is used. While the parameter itself is physically meaningful, the implementation is done only in an empirical mathematical way. Therefore and because of the generally small influence, it is recommended to fix this parameter to 0. However, the resulting atomic form factor for the parallel graphene sheets can be calculated through:

$$f_{c,para}(s) = \begin{cases} 2.31 \cdot \exp(-(20.84339 + \Delta_{an}) \cdot s^2) + 1.02 \cdot \exp(-10.2075 \cdot s^2) \\ +1.5886 \cdot \exp(0.5687 \cdot s^2) + 0.865 \cdot \exp(-51.6512 \cdot s^2) + 0.2156, s < 3.74 \\ \exp\left(1.7056 - 1.5676 \cdot \dfrac{s}{2} + \dfrac{1.1893}{10}\left(\dfrac{s}{2}\right)^2 - \dfrac{0.42715}{100}\left(\dfrac{s}{2}\right)^3\right), else \end{cases} \quad (52)$$
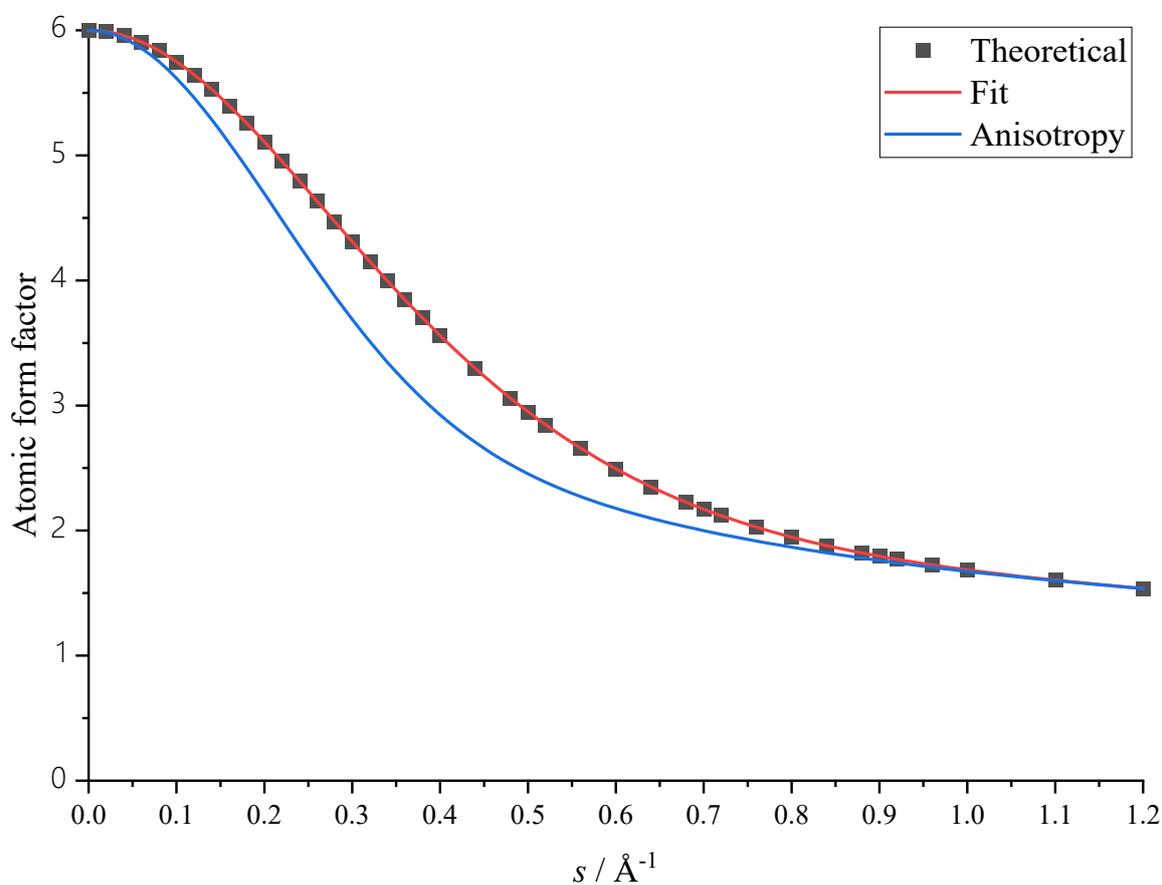

Figure S20 Theoretical and fitted atomic form factor and the influence of the anisotropy ($\Delta_{an} = 25$).

S17.2. Atomic form factor of foreign atoms (H, N, O, S)

The atomic form factor of hydrogen, nitrogen, oxygen and sulfur can be calculated like the atomic form factor of carbon. All data is taken from Prince et al. [6].

S17.2.1. Hydrogen

$$f_{c,\text{perp}}(s)$$
$$= \begin{cases} 0.493002 \cdot \exp(-10.5109 \cdot s^2) + 0.322912 \cdot \exp(-26.1257 \cdot s^2) \\ +0.140191 \cdot \exp(3.14236 \cdot s^2) + 0.04081 \cdot \exp(-57.7997 \cdot s^2) + 0.003038, s < 3.81 \\ 0, \text{else} \end{cases} \tag{53}$$

S17.2.2. Nitrogen

$$f_{c,\text{perp}}(s)$$
$$= \begin{cases} 12.2126 \cdot \exp(-0.0057 \cdot s^2) + 3.1322 \cdot \exp(-9.8933 \cdot s^2) \\ +2.0125 \cdot \exp(28.9975 \cdot s^2) + 1.1663 \cdot \exp(-0.5826 \cdot s^2) - 11.529, s < 3.88 \\ \exp\left(1.5494 - 1.2019 \cdot \dfrac{s}{2} + \dfrac{0.51064}{10}\left(\dfrac{s}{2}\right)^2 + \dfrac{0.2472}{100}\left(\dfrac{s}{2}\right)^3\right), \text{else} \end{cases} \tag{54}$$

S17.2.3. Oxygen

$$f_{c,\text{perp}}(s) = \begin{cases} 3.0485 \cdot \exp(-13.2771 \cdot s^2) + 2.2868 \cdot \exp(-5.7011 \cdot s^2) \\ +1.5463 \cdot \exp(0.3239 \cdot s^2) + 0.867 \cdot \exp(-32.9089 \cdot s^2) + 0.2508, s < 3.79 \\ \exp\left(1.3053 - 0.83742 \cdot \dfrac{s}{2} + \dfrac{-0.16738}{10}\left(\dfrac{s}{2}\right)^2 + \dfrac{0.475}{100}\left(\dfrac{s}{2}\right)^3\right), \text{else} \end{cases} \tag{55}$$

S17.2.4. Sulfur

$$f_{c,\text{perp}}(s) = \begin{cases} 6.9053 \cdot \exp(-1.4679 \cdot s^2) + 5.2034 \cdot \exp(-22.2151 \cdot s^2) \\ +1.4379 \cdot \exp(0.2536 \cdot s^2) + 1.5863 \cdot \exp(-56.172 \cdot s^2) + 0.2156, s < 5.86 \\ \exp\left(1.104 - 0.40325 \cdot \dfrac{s}{2} + \dfrac{0.20094}{10}\left(\dfrac{s}{2}\right)^2 + \dfrac{-0.26058}{100}\left(\dfrac{s}{2}\right)^3\right), \text{else} \end{cases} \tag{56}$$
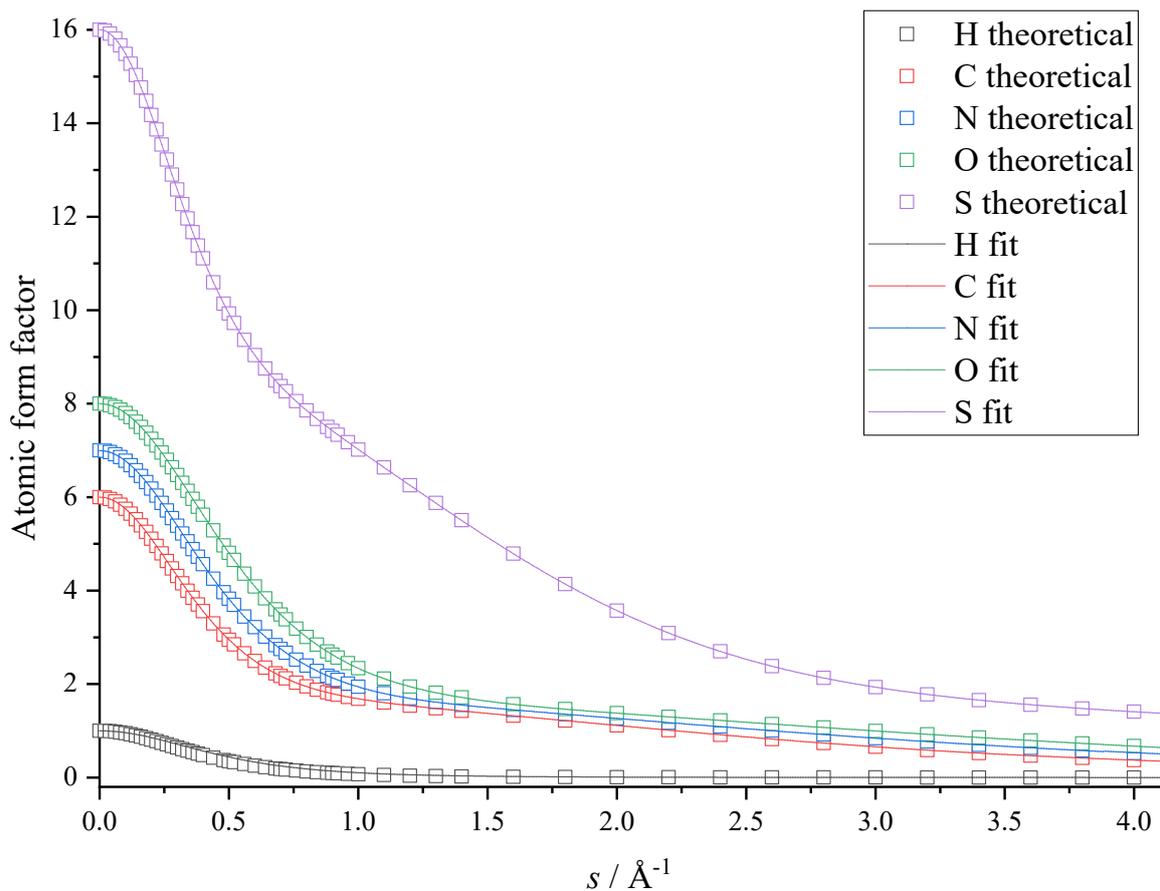
Figure S21 Theoretical and fitted atomic form factors of carbon and the most common (hydrogen, nitrogen, oxygen, sulfur).

S17.3. Atomic form factor for neutron scattering

For wide-angle neutron scattering (WANS), the atomic form factor is directly related to the coherent nuclear scattering length b_{coh} [14]:

$$f(\lambda, \theta) = - b_{coh} \tag{57}$$

For b_{coh}, the values are tabulated in Table S7 [18] and online available under [16]. The scattering length densities required for the purpose here are taken from the NIST center.

Table S7   Coherent nuclear scattering lengths for C, H, N, O, S. The values are mean values from the natural isotopes.

Atom	Coherent scattering length / fm
C	6.6460
H	-3.7390
N	9.36
O	5.803
S	2.847

S18. Incoherent scattering – theoretical and calculated data

Analogous to S14, the Compton scattering for carbon and the foreign atoms (hydrogen, nitrogen, oxygen, sulfur) can be fitted through spline interpolation. Since the commonly lowest used wavelength is $\lambda = 0.72$ Å (Mo K-α) and therefore, the maximum scattering vector is about $s = 2.75$ Å^{-1}, the tabulated theoretical values for the Compton scattering up to $s = 4$ Å^{-1} are more than sufficient [6] for atoms with only a few amount of electrons (C, H, N, O). For sulfur, the data from Cromer [19] up to $s = 16$ Å^{-1} were used to get more accurate data for possible synchrotron measurements.

S18.1. Carbon

$$I_{com,c}(s)$$
$$= \begin{cases} -33.525 \cdot s^3 + 26.296 \cdot s^2 + 2.160 \cdot s - 0.025, 0 < s \leq 0.4 \\ 11.179 \cdot (s - 0.4)^3 - 13.935 \cdot (s - 0.4)^2 + 7.104 \cdot (s - 0.4) - 2.901, 0.4 < s \leq 0.8 \\ 0.222 \cdot (s - 0.8)^3 - 0.520 \cdot (s - 0.8)^2 + 1.322 \cdot (s - 0.8) + 4.228, 0.8 < s \leq 1.2 \\ 0.013 \cdot (s - 1.2)^3 - 0.254 \cdot (s - 1.2)^2 + 1.013 \cdot (s - 1.2) + 4.688, 1.2 < s \leq 1.8 \\ 0.023 \cdot (s - 1.8)^3 - 0.230 \cdot (s - 1.8)^2 + 0.722 \cdot (s - 1.8) + 5.207, 1.8 < s \leq 5 \\ 6, 5 < s \end{cases} \quad (58)$$

S18.2. Hydrogen

$$I_{com,H}(s)$$
$$= \begin{cases} -8.009 \cdot s^3 + 5.645 \cdot s^2 + 0.925 \cdot s, 0 < s \leq 0.4 \\ 3.378 \cdot (s - 0.4)^3 - 3.966 \cdot (s - 0.4)^2 + 1.596 \cdot (s - 0.4) + 0.760, 0.4 < s \leq 0.8 \\ -0.202 \cdot (s - 0.8)^3 + 0.088 \cdot (s - 0.8)^2 + 0.045 \cdot (s - 0.8) + 0.980, 0.8 < s \leq 1.2 \\ 0.217 \cdot (s - 1.2)^3 - 0.155 \cdot (s - 1.2)^2 + 0.018 \cdot (s - 1.2) + 0.999, 1.2 < s \leq 1.8 \\ 1, 1.8 < s \end{cases} \quad (59)$$

S18.3. Nitrogen

$$I_{com,N}(s)$$
$$= \begin{cases} -31.81 \cdot s^3 + 764.875 \cdot s^2 + 1.124 \cdot s, 0 < s \leq 0.4 \\ 5.846 \cdot (s - 0.4)^3 - 10.409 \cdot (s - 0.4)^2 + 8.066 \cdot (s - 0.4) + 2.856, 0.4 < s \leq 0.8 \\ 2.573 \cdot (s - 0.8)^3 - 3.393 \cdot (s - 0.8)^2 + 2.545 \cdot (s - 0.8) + 4.791, 0.8 < s \leq 1.2 \\ 0.055 \cdot (s - 1.2)^3 - 0.305 \cdot (s - 1.2)^2 + 1.066 \cdot (s - 1.2) + 5.431, 1.2 < s \leq 1.8 \\ 0.020 \cdot (s - 1.8)^3 - 0.206 \cdot (s - 1.8)^2 + 0.759 \cdot (s - 1.8) + 5.972, 1.8 < s \leq 5.5 \\ 7, 5.5 < s \end{cases} \quad (60)$$

S18.4. Oxygen

$I_{com,O}(s)$

$$= \begin{cases} -30.697 \cdot s^3 + 29.263 \cdot s^2 + 0.183 \cdot s - 0.002, 0 < s \le 0.4 \\ 2.011 \cdot (s - 0.4)^3 - 7.570 \cdot (s - 0.4)^2 + 8.860 \cdot (s - 0.4) + 2.789, 0.4 < s \le 0.8 \\ 3.591 \cdot (s - 0.8)^3 - 5.157 \cdot (s - 0.8)^2 + 3.769 \cdot (s - 0.8) + 5.251, 0.8 < s \le 1.2 \\ 0.379 \cdot (s - 1.2)^3 - 0.849 \cdot (s - 1.2)^2 + 1.367 \cdot (s - 1.2) + 6.163, 1.2 < s \le 1.8 \\ 0.013 \cdot (s - 1.8)^3 - 0.166 \cdot (s - 1.8)^2 + 0.759 \cdot (s - 1.8) + 6.960, 1.8 < s \le 6 \\ 8, 6 < s \end{cases} \quad (61)$$

S18.5. Sulfur

$I_{com,S}(s)$

$$= \begin{cases} -47.563 \cdot s^3 + 37.813 \cdot s^2 + 5.116 \cdot s - 0.058, 0 < s \le 0.4 \\ 16.192 \cdot (s - 0.4)^3 - 19.262 \cdot (s - 0.4)^2 + 12.536 \cdot (s - 0.4) + 4.963, 0.4 < s \le 0.8 \\ -1.502 \cdot (s - 0.8)^3 + 0.168 \cdot (s - 0.8)^2 + 4.898 \cdot (s - 0.8) + 7.989, 0.8 < s \le 1.2 \\ 0.355 \cdot (s - 1.2)^3 - 0.168 \cdot (s - 1.2)^2 + 4.311 \cdot (s - 1.2) + 9.853, 1.2 < s \le 1.8 \\ 0.143 \cdot (s - 1.8)^3 - 0.996 \cdot (s - 1.8)^2 + 2.733 \cdot (s - 1.8) + 11.928, 1.8 < s \le 4 \\ 0.00186 \cdot (s - 4)^3 - 0.04921 \cdot (s - 4)^2 + 43412 \cdot (s - 4) + 14.64861, 4 < s \le 16 \\ 16, 16 < s \end{cases} \quad (62)$$
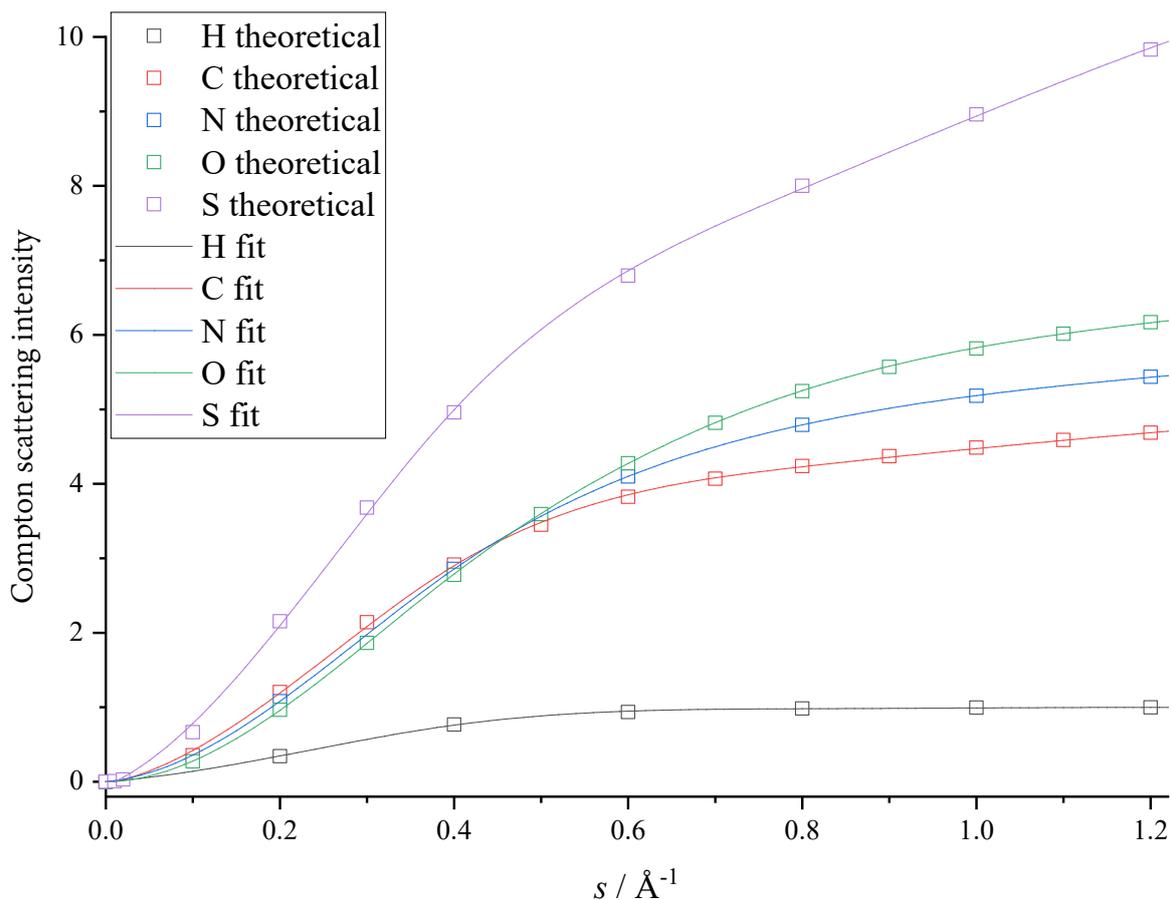
Figure S22 Theoretical and fitted Compton scattering intensity of carbon and the most common foreign atoms (hydrogen, nitrogen, oxygen, sulfur).

S18.6. Incoherent neutron scattering

For the incoherent, i.e. the inelastic scattering, of neutrons with the nucleus, is related to the spin of the nucleus [14]. For ^1H (i.e. a proton), the scattering lengths for the triplet and singlet states are $b^+ = 10.85$ fm and $b^- = -4.750$ fm, respectively. Therefore, a hydrogen nucleus has an average scattering length of

$$\langle b \rangle = \frac{3}{4}b^+ + \frac{1}{4}b^- = \text{-3.738 fm} \tag{63}$$

The standard, i.e. the root mean square, deviation Δb of it is the so-called incoherent scattering length:

$$b_{\text{inc}} = \Delta b = \sqrt{\langle b^2 \rangle - \langle b \rangle^2} = \text{25.266 fm} \tag{64}$$

using

$$\langle b^2 \rangle = \frac{3}{4}(b^+)^2 + \frac{1}{4}(b^-)^2 = 652.354 \text{ fm} \tag{65}$$

The value measured in [18] are close similar to the value calculated above and therefore, they can be used for the present calculations. The data in Table S9 is also available in [16].

Table S8 Incoherent nuclear scattering lengths for C, H, N, O, S. The values are mean values from the natural isotopes.

Atom	Incoherent scattering length / fm
C	-0.00572
H	25.271
N	1.99
O	0.000068
S	0.011

For the calculation of the incoherent scattering, a superposition of the coherent and incoherent scattering lengths is assumed:

$$I_{\text{incoh, x}} = c_X \cdot (b_{\text{coh}} + b_{\text{inc}})^2 \tag{66}$$

for X = C, H, N, O, S

However, since these values are constants, it is not necessary to use exact values of the concentrations or the scattering lengths. Furthermore, it is possible to set I_{incoh} to 0 since a constant offset is applied and refined using the normalization constants in equation (6). Nevertheless, this approach of a constant background is only applicable for an energy exchange, $\hbar\omega$, is NOT comparable to the incidence E_0, otherwise, the self-scattering will "fall off" with an increasing value of s and a non-constant correction is necessary. Additionally, neutron absorption resonances can lead to a non-constant background [2]. For these reasons, there are different models for taking these into account [1,20]. The finally used corrections are described in the following chapter.

S19. Background correction for wide-angle neutron scattering

As described in the chapter before, the case of constant inelastic scattering is only applicable, there as the $\hbar\omega$, is comparable to the incidence E_0. Especially for hydrogen (^{1}H, i.e. a proton), the a huge damping of the incoherent scattering is measured, whereas for ^{113}Cd there are strong neutron absorption resonance effects in the commonly used wavelength, range around 0.5 to 1 Å [2].

Overall, the measured data is "fitted" over the whole q range (NOT θ) using two different equations. The resulting data oscillates around 1 for high values of q, so in principle $S(q)$ data, which can also be used for a pair-distribution-function calculation, is generated (without a correct normalization) [21,22].

For samples containing significant amounts of hydrogen (^{1}H), either deuterium (^{2}H) must be used or a pseudo-Voigt function, i.e. a linear combination of a Gaussian and a Lorentzian function with equal full-with at half-maximum (FWHM) values, must be applied (equation (67)). For all other cases, a correction using the method of the Placzek falloff, i.e. a quadratic equation with only even powers is sufficient [1,2] (equation (68)).

$$S(q) = I_{obs}(q) - k \cdot (\eta \cdot L(q, \omega) + (1 - \eta) \cdot G(q, \omega)) + 1 \tag{67}$$

$$S(q) = I_{obs}(q) - (a \cdot q^2 + b) + 1 \tag{68}$$

$$L(q, \omega) = \frac{1}{1 + \left(\frac{q}{\omega}\right)^2} \tag{69}$$

$$G(q, \omega) = \exp^{-\ln(2) \cdot \left(\frac{q}{\omega}\right)^2} = 2^{-\left(\frac{q}{\omega}\right)^2} \tag{70}$$

I_{obs} = observed (measured) intensity
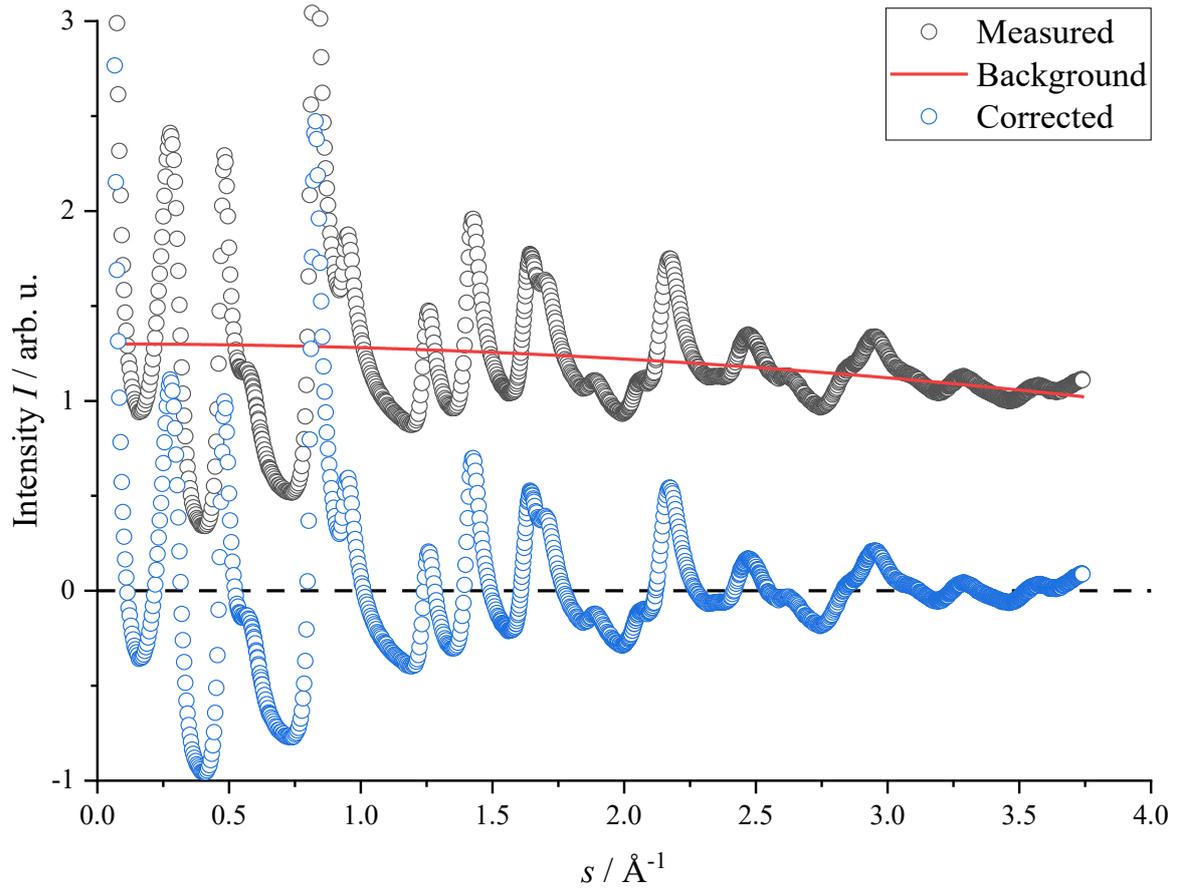
Figure S23 Example for a background correction of WANS data using the method from sufficient [1,2] (equation (68)). Black: measured data. Red: Assumed background. Blue: Corrected data.

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